PRINCIPAL WORKING GROUP N° 3
ON PRIMARY CIRCUIT INTEGRITY

Specialist meeting on
IRRADIATION EMBRITTLEMENT AND
OPTIMISATION OF ANNEALING

Paris (France)
20-23 September 1993

Jointly organised by the
OECD Nuclear Energy Agency
and the
International Atomic Energy Agency

COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS
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PRINCIPAL WORKING GROUP N°3 ON PRIMARY CIRCUIT INTEGRITY

Specialist meeting on Irradiation Embrittlement and Optimisation of Annealing - Paris (France), 20-23 September 1993

ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

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The primary objective of NEA is to promote co-operation among the governments of its participating countries in furthering the development of nuclear power as a safe, environmentally acceptable and economic energy source.

This is achieved by:

- encouraging harmonization of national regulatory policies and practices, with particular reference to the safety of nuclear installations, protection of man against ionising radiation and preservation of the environment, radioactive waste management, and nuclear third party liability and insurance;
- assessing the contribution of nuclear power to the overall energy supply by keeping under review the technical and economic aspects of nuclear power growth and forecasting demand and supply for the different phases of the nuclear fuel cycle;
- developing exchanges of scientific and technical information particularly through participation in common services;
- setting up international research and development programmes and joint undertakings.

In these and related tasks, NEA works in close collaboration with the International Atomic Energy Agency in Vienna, with which it has concluded a Co-operation Agreement, as well as with other international organisations in the nuclear field.

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COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of scientists and engineers. It was set up in 1973 to develop and co-ordinate the activities of the Nuclear Energy Agency concerning the technical aspects of the design, construction and operation of nuclear installations insofar as they affect the safety of such installations. The Committee's purpose is to foster international co-operation in nuclear safety amongst the OECD Member countries.

CSNI constitutes a forum for the exchange of technical information and for collaboration between organisations which can contribute, from their respective backgrounds in research, development, engineering or regulation, to these activities and to the definition of its programme of work. It also reviews the state of knowledge on selected topics of nuclear safety technology and safety assessment, including operating experience. It initiates and conducts programmes identified by these reviews and assessments in order to overcome discrepancies, develop improvements and reach international consensus in different projects and International Standard Problems, and assists in the feedback of the results to participating organisations. Full use is also made of traditional methods of co-operation, such as information exchanges, establishment of working groups and organisation of conferences and specialist meeting.

The greater part of CSNI's current programme of work is concerned with safety technology of water reactors. The principal areas covered are operating experience and the human factor, reactor coolant system behaviour, various aspects of reactor component integrity, the phenomenology of radioactive releases in reactor accidents and their confinement, containment performance, risk assessment and severe accidents. The Committee also studies the safety of the fuel cycle, conducts periodic surveys of reactor safety research programmes and operates an international mechanism for exchanging reports on nuclear power plant incidents.

In implementing its programme, CSNI establishes co-operative mechanisms with NEA's Committee on Nuclear Regulatory Activities (CNRA), responsible for the activities of the Agency concerning the regulation, licensing and inspection of nuclear installations with regard to safety. It also co-operates with NEA's Committee on Radiation Protection and Public Health and NEA's Radioactive Waste Management Committee on matters of common interest.
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Foreword

This Specialists Meeting was jointly sponsored by the IAEA International Working Group on Life Management of Nuclear Power Plant and by the Principal Working Group 3 (PWG-3) of the NEA CSNI, and hosted by the CEA. PWG-3 deals with Reactor Component Integrity, and has a joint secretariat with the Commission of the European Communities. The activities of PWG-3 fall into three main areas: Non-Destructive Examination (NDE), fracture analysis and ageing materials degradation. In NDE, the main activity has been the Programme for the Inspection of Steel Components (PISC), jointly with the CEC. In fracture analysis, the activities are organised by the Fracture Analysis Group, and include the round robins on Fracture Analysis of Large Scale International Reference Experiments (FALSIRE). In the area of ageing/materials degradation, PWG-3 considers case studies of operational experience, and sponsors Specialists Meetings and Workshops.

The proceedings of the meeting were issued in February 1994 as a CSNI report. This was considered at a number of meetings of PWG-3 where further consideration was given to the Conclusions and Recommendations agreed at the Specialists Meeting. This consideration has led to a change of emphasis and these current PWG-3 revisions are reproduced here.

Note for the 1997 issue

In 1996 the mandate of PWG-3 was widened to consider the integrity of components and structures, with an emphasis on ageing. There are now three sub-groups, dealing with metal structures and components, ageing of concrete structures, and the seismic behaviour of structures.

This document is published under the responsibility of the Secretary-General of the OECD.
FOREWORD

This joint International Atomic Energy Agency (International Working Group on the Life Management of Nuclear Power Plants) and the OECD, Nuclear Energy Agency (Principal Working Group No. 3) Specialists Meeting on the Irradiation Embrittlement and Optimization of Annealing was held in Paris on the 20-23 September 1993.

The meeting was attend by 60 delegates from 16 countries (See Appendix 1) and 2 International Organizations.

The Agenda for the meeting is shown in Appendix 2.

The Organizing Committee for the meeting was composed of:

Mr. P. Petrequin (Committee Chairman)  
CEN-SACLAY

Acad. L.M. Davies  
Chairman, IAEA IWG-LMNPP

Pr. K. Törrönen  
Chairman, OECD/NEA PWG-3

Mr. L. Ianko  
IAEA

Dr. A. Miller  
OECD Nuclear Energy Agency

and the Chairman of the meeting was: Eur. Ing. Acad. L.M. Davies.

The Scientific Secretaries of the Meeting were Mr. L.Ianko (IAEA) and Mr. A. Miller (OECD).

The meeting was organized into 6 Sessions and the Session Chairmen were:

Session A. Mr. Petrequin (France), Mr. Törrönen (Finland)
Session B. Ms. Horowitz (France), Mr. Gillemot (Hungary)
Session C. Mr. de Keroules (France), Mr. Rosinsky (USA)
Session D. Mr. English (UK), Mr. Houssin (France)
Session E. Mr. Brumovsky (Czech Rep.), Mr. Miannay (France)
Session F. Mr. Soulat (France), Mr. Leitz (Germany)
OPENING CEREMONY
OPENING SPEECH

Dr. P. Petrequin, Chairman of the Organizing Committee

Ladies and Gentlemen,

It is very impressive to speak first, but the most important thing that I would like to tell you is that I am very glad to welcome you, here, in Paris for this Specialists Meeting on Irradiation Embrittlement and Optimization of Annealing, on behalf of the IAEA and the OECD.

At its last meeting, the International Working Group on Life Management of Nuclear Power Plants of the IAEA, suggested to have this meeting and the French Atomic Energy Community, including the Atomic Energy Commission (CEA), Electricité de France (EDF) and FRAMATOME proposed to organize it in France, in Paris.

As Chairman of the Organization C69, I have to apologize for the relatively late announcement of the Meeting, due to some administrative delays in France, out of my control, but, the efficiency of the Chairman of the Conference and of the working group, Academian Myrddin Davies, of our IAEA Scientific Secretary, Mr. Leonid Ianko and of some colleagues made that, after all, we are glad to welcome today, 61 participants, representing 17 different countries and 3 organizations for 28 papers.

This success is due too, to the very strong interest of the subject that we have deal with. The irradiation embrittlement of the pressure vessel materials of nuclear reactor is a very important topic which has to be taken into account in all the countries embarked in nuclear energy production and the international cooperation is a necessity to progress efficiently.

I remember that more than 20 years ago, the Agency started coordinating activities related to irradiation embrittlement by different International Working Groups promoting Coordinated Research Programmes, Conference and Specialists Meetings, but the subject is so tough that the International cooperation remains necessary to make progresses, and that is the reason why the Agency is still organizing Specialists Meetings and why we are together today for that.

During this time, the topics related to irradiation embrittlement evoked in the Specialists Meeting changed progressively.

In the seventies, as many reactors were under erection, the more significant subjects were related to the role of the chemical composition on irradiation embrittlement, the establishment of specification of pressure vessel materials, the comparison of the different national licensing rules, the preparation of surveillance programmes, and the development of testing techniques, in the field of fracture mechanics.

In the eighties, as the role of chemical compositions appeared to be under control, some surprising results pushed the laboratories to try to understand the irradiation hardening and embrittlement on a more physical basis, introducing sophisticated physical
examinations besides the mechanical testing of irradiated materials.

In the nineties, as many reactors have been in operation for long times, their life management becomes one of the more important features of our activities, and that is a major issue for our meeting.

During those 20 years, France was developing an important nuclear programme, and by the way, participated efficiently to all the different activities of the IAEA in the field but, surprisingly, France never had the opportunity to host such a Specialists Meeting.

That is the reason why, again, I am so glad to welcome you here in Paris. I hope that the practical organization of the meeting - I will tell you words about it at the end of this ceremony - will allow all of us to work efficiently and comfortably, in order to produce a fruitful contribution to the progress of the safety and economy of the nuclear energy.

Now, Mr. Leonid Ianko, Scientific Secretary and Academician Myrddin Davies, Chairman of the meeting, would like to say some words to you, and I leave the microphone to Mr. Ianko.

Thank you very much.
OPENING SPEECH

L. Ianko - Scientific Secretary

Specialists Meeting on Irradiation Embrittlement and Optimization of Annealing

Ladies and Gentlemen,

On behalf of the IAEA I would like to welcome all the participants of this joint IAEA, OECD/NEA Specialists Meeting.

Many technical, economic and safety aspects of NPPs ageing and life extension have been discussed at different IAEA meetings and have been included in the IAEA Programme of Activity. The main problems which have been discussed are: safety analysis methods for time-dependent phenomena, classification of components, methodology for effective management of the lives of critical components, establishment of data bank from information available in all countries. The activities planned by the IAEA International Working Group on Life Management of Nuclear Power Plants include information exchange, meetings of several kinds, development of publications and a data base. The purpose is to enable Member States with an interest in using nuclear power in the future to learn and gain a better understanding of the means available to protect and enhance the economic operating life and safety of their plants. Successful and safe performance of NPPs during their lifetime depends, to a large extent, on the reliability of the steel pressure vessels. The effect of the radiation on the pressure vessel steel is manifested by an increase in yield strength, hardening, a shift in brittle-ductile transition temperature and a decrease in ductility. Reactor pressure vessels have thick walls and are subject to stress gradients arising from design, operation, accidents, and so on.

The Specialists Meeting is organized by the IAEA International Working Group on Life Management of Nuclear Power Plants in cooperation with OECD Nuclear Energy Agency Committee for the Safety of Nuclear Installations.

The purpose of the Meeting is to provide an international forum for discussion on recent results in research and utility experience on:

- mechanism of radiation damage,
- effects of operating parameters (flux, temperature, time etc.),
- results from surveillance programmes and their analysis,
- results from research reactor investigations on irradiation effects,
- fracture mechanics testing and evaluation (mainly of small specimens),
- annealing and optimization of the process,
- re-embrittlement after annealing.

Our Meeting will be divided into 6 sessions and will be chaired by a chairman and co-chairman, as you can see in the working programme. It is expected that each session
will result in conclusions and recommendations which will be reviewed during the final session on Thursday.

To conclude, please allow me in the name of the participants and the IAEA to express high appreciation and thanks to Mr. Petrequin of CEN-SAACLAY, France, for his decision to convene our Meeting here, in Paris, and for the very good working conditions provided to us.

I believe that our Meeting will not only be interesting but also very useful and successful and that discussions and exchange of opinions on this particular topic will be very fruitful.

Thank you very much for your kind attention.
Opening Remarks at Irradiation Embrittlement and Annealing, Paris, France

Ladies and Gentlemen,

I would like to reiterate Leonid Ianko’s welcome to Paris, this time on behalf of the OECD Nuclear Energy Agency. This meeting on Irradiation Embrittlement and Annealing Optimization is co-sponsored by IAEA and NEA Principal Working Group-3 on Reactor Component Integrity. The chairman of this group is Professor K. Törrönen, who is co-chairman of the first session. The proceedings of the meeting will be presented as a state-of-the-art report to the NEA Committee on the Safety of Nuclear Installations. OECD headquarters is in Paris, but last summer NEA moved from Paris to Issy-les-Moulineaux, which is about three kilometers from here, should any of you come to visit us. I would like to thank Mr. Peterequin and the CEA for their hospitality and for making the local arrangements, and I wish you all a successful meeting.

Alex Miller
Opening Remarks by L.M. Davies, Chairman IWG-LMNPP

Ladies and Gentlemen. It is a pleasure to be here with you in Paris for this meeting on "Irradiation Embrittlement and Annealing". It is a pleasure to meet longstanding acquaintances and I look forward to working with you again.

I thank the IAEA for arranging this timely meeting and the French hosts, in particular Pierre Petrequin the Chairman of the Organization Committee, for organizing and arranging matters.

It is also a pleasure to note that this is a joint meeting with the Principal Working Group-3 of the Nuclear Energy Agency of the OECD. It is a pleasure to work with Kari Törrönen, the Chairman of PWG-3 and Alex Miller, its Secretary.

The problems associated with irradiation embrittlement continue as plants get older and plant life assurance becomes important and plant life extension also comes to the fore. Plant life management is also a subject of action for 'new' plants - which have benefitted from the use of improved steels. Revised surveillance schedules and the requirements for specimens for longer life plants also presents problems-albeit welcome ones!

The interest in annealing has made a resurgence - both by the annealing of vessels in Eastern and Central Europe and also by the development of the studies in the USA. Re-irradiation effects are also then the subject for current study.

There is a perceived international need for a more comprehensive database and it is interesting to note the developments in this area. Allied to database developments is the proliferation of "Guides" which are used to predict irradiation response of materials. They are utilised by various countries and organizations in various ways but they suffer the limitation that they are empirically based but they are easy to use.

I anticipate that the subject of this meeting will be getting a good "airing" in the next couple of days.

In keeping with the practice at these meetings 'masters' of papers have to be handed to Pierre Petrequin of Leonid Ianko during this meeting for publication. Session Chairmen have been identified and "volunteered" for the tasks of Chairing half day sessions and writing a brief (less than one page) report summarising their session and drawing any conclusions and recommendations. If any of you wish to make a recommendation, to be considered by the IAEA IWG-LMNPP, then please get them to Leonid Ianko before the end of play on Wednesday when we hope to have all the material for the discussion on Thursday morning.

I wish you all a successful meeting and a pleasant stay in Paris.
SESSION A

NEUTRON IRRADIATION EFFECTS
1.1 INTRODUCTION

Of those 496 reactors of 400,000MWe (net) capacity in operation and under construction at the end of 1992 [1] the commonest type is the Pressurised Water Reactor (PWR) (Slide 2). In this paper we will also be commenting upon the WWERs (Vodo-Vodianyi Energeticheskiy Reactor). WWERs are pressurised water reactors and are generally located in Central and Eastern Europe. Three WWER models have been developed [2]. The first, the 440/230 has a capacity of 440 MWe and was developed in the 1960s and produced exclusively in the then USSR. The second, the 440/213 has the same capacity but an improved design mainly with regard to safety equipment. The third type is the WWER 1000 with a capacity of 1000 MWe and developed in the 1980s.

This paper is about the effects of neutron irradiation on the steel and welds used for the pressure vessels (PV) which house the reactor cores in light water reactors. There have been earlier overviews, for example [3] which can be used to expand upon the historical basis of the topic. The Reactor Pressure Vessel (RPV) is a key component in most Nuclear Power plants and, as it is generally considered to be irreplaceable, its operating life can therefore determine the lifetime of the plant. Its integrity is paramount in avoiding the release of contamination in the event of an, albeit remote, accident. It is therefore necessary to understand those features which affect its mechanical properties and which are used to predict its integrity and lifetime. The lifetime of a nuclear power
plant is usually determined by those factors which determine an acceptable margin of safety and allow the production of electricity at an acceptable cost. The objective is to operate the plant safely and produce electricity at a high availability factor over its design life. The design life of the early light water reactors was not originally based on neutron irradiation effects but was more to do with other design features such as fatigue usage factors [4].

The ability of a pressure vessel to withstand the normal pressure and thermal stresses during the heating and cooling part of cycles associated with plant start up and shut down together with the assessment of the stresses associated with postulated accidents are considered at the design stage. For these reasons it is important that the pressure vessel is made from appropriate materials and manufactured to a high quality standard and also meets the demands placed upon it from the design and operational requirements [5]. Pressure vessels are subject to 'allowable' pressure-temperature limits during normal operation and operational test conditions. Greater sensitivity to neutron irradiation increases the restrictions on the operating 'window' of the pressure-temperature relationship. Pressure-temperature operating limits reviews are performed periodically through life to assess the adequacy of operating parameters and to ensure against brittle fracture. Establishing quantitatively the safety margins of a nuclear pressure vessel, during normal operation and under fault conditions requires that reliable mechanical property information be available [5].

However nuclear power plants have been built and operated for many years. (Slide3) The relevance of this particular point is that in the late 1960s the role of the impurity and residual elements copper and phosphorus were identified [6][7] as increasing the sensitivity of 'model' pressure vessel steels to irradiation embrittlement. The application of the results from this, and other early work, was truly remarkable in its impact on PWRPV technology and the economics of power plant operation. Benefits have been derived in terms of plant life assurance and also in extending the life of modern plant by the removal of the major component lifetime limiting feature.

Subsequently, nickel and many other elements [8][9][10] were also found to be important in their effect on the mechanical properties of PV steels and welds on irradiation. There are anecdotes of earlier steelmaking practice which utilised automobile scrap still containing the electrical copper wire for the manufacture of pressure vessel base metal and also of using copper coated electrodes for welding [11]. The copper content of PWRPV welds and base metal made after about 1972 was substantially reduced but there remains a significant number of operating vessels with higher levels of copper in their welds. Additionally, some early pressure vessels were produced by welding practices employing a flux which resulted in a low toughness in the ductile temperature regime [12].
Thus the reactor pressure vessels typifying this earlier generation of nuclear power plant were seen as the plant lifetime limiting components and the effect of neutron irradiation on the materials in the beltline region of RPVs was an area of concern and much study. The newer generation of pressure vessels, which have benefited from the improvements in pressure vessel technology and improved understanding of irradiation effects, have a much reduced sensitivity to neutron irradiation and therefore are not restricted in life from this cause. However the annealing of the Novovoronezh 3 WWER pressure vessel to mitigate the effects of neutron irradiation, as recently as 1987, is an indication of the relatively continuous nature of the impact of this particular problem.

1.2 IRRADIATION EFFECTS ON MECHANICAL PROPERTIES

Material composition, method of manufacture, metallurgical condition, design and operating temperature can all influence the mechanical strength of the Reactor Pressure Vessel (RPV) steels and welds during neutron irradiation. All these are features to be considered in assessing the behaviour of a particular pressure vessel. There is an increase in yield strength, an increase in tensile strength and a decrease in strain hardening capability of steels and welds. A possibility which also has to be considered is for there to be also a non-hardening component of embrittlement caused by irradiation enhanced temper embrittlement or transmutation and precipitation effects during long term operation.

The main neutron irradiation effects are illustrated by the Charpy test where the amount of energy required to break specimens made from samples of the steel is measured. The higher the energy required, the less brittle or more ductile is the specimen being tested. A pendulum is swung, with a known energy, which is established by how far the pendulum swings in a free but unloaded state. When a notched specimen of standard shape is inserted in the path of the swinging pendulum then the distance now swung is a measure of the energy absorbed in fracturing the specimen. Steels and welds used for RPVs demonstrate a transitional behaviour as a function of test temperature, (Slide 4), where the material exhibits brittleness at low temperature and ductile behaviour at higher temperatures. In general terms the effect of neutron irradiation is to shift this transitional behaviour to higher temperatures and to reduce the energy absorbed for fracture in the ductile region. There may also be an increase in the temperature range of the transition region. This shift in Ductile- Brittle Transition Temperature (ADBTT) is frequently defined at the reference absorbed energy level of 41J (30ft.lbs.) which generally correlates with the transition temperature defined as the Reference Nil Ductility Transition Temperature (RTNDT) from a separate drop weight test, ADBTT is therefore to be considered to
be equivalent to the shift in RTNDT (ΔRTNDT) which is commonly employed to ‘reference’ the fracture toughness of the material.

Δ DBTT varies with the neutron dose (fluence) experienced by the RPV. (Slide 5) The energetic neutrons encompass a range of energies. The fluence is usually specified in terms of energetic neutrons above a particular neutron energy level, for example, greater than 0.1, 0.5 or, more usually, greater than 1.0 Million electron Volts, (MeV). Neutron fluence is now usually expressed in units of neutrons (n) per square metre but, much data, many workers and formulations still use neutrons per square centimetre so both will be used here. There is also a current trend to express neutron effects in terms of the damage produced by incident neutrons in the matrix iron crystal lattice. Collisions between the more energetic neutrons and the lattice atoms produce primary knock-on atoms (PKAs), which in turn, lose their energy by interacting with the lattice atoms to produce, under the appropriate conditions, vacancies and interstitials. At higher energies, where collisions occur every lattice spacing, collision or displacement cascades are produced in which the final configuration is generally thought to be a vacancy rich region surrounded by interstitial atoms which may be clustered. The calculated number of point defects produced by PKAs provides a measure of neutron exposure expressed as displacements per atom (dpa).

1.3 NEUTRON FLUENCE

Knowledge of the neutron exposure of a pressure vessel is necessary in order to analyse changes in mechanical properties [5]. The aspect of reporting neutron fluence is a particularly important feature in normalising data derived from different irradiation positions or different reactors and then relating them to a particular location in an actual RPV, where the neutron flux and energy spectrum are usually substantially different. Information on irradiation effects on the mechanical properties of RPV steels has also been gained from accelerated irradiations carried out in Materials Test Reactors where, again, the neutron flux could be significantly higher and the neutron energy spectrum could be significantly different from that of an actual location in an RPV where the data is to be applied.

Surveillance capsules are located inside the RPV of power reactors, where the neutron flux and energy spectrum is also usually higher than the actual RPV. The method of reporting neutron fluence continues to be important and for example, earlier workers [13] found that “...neutrons with energies 1MeV would account for over 75%....but neutrons with energies greater than 0.1MeV would account for 94% of the transition shift temperature increase observed for virtually every spectrum”. (Slide 6) Indeed some of the uncertainties associated with calibrating the fluence can also be removed by the use of a ‘standard’ reference material, (i.e. one
having a 'known' response in its change of mechanical properties to neutron irradiation), in surveillance programmes. Such a material is that designated as JRQ in the IAEA Coordinated Research Programme, Phase 3.

1.4 IRRADIATION EFFECT TRENDS

The effects of irradiation on mechanical properties are described in reference [5] The effect of irradiation on the mechanical properties can be followed by irradiating specimens taken from representative archive samples of an operational RPV (or research samples or candidate materials for future RPVs) under representative neutron fluences, and irradiation temperatures. This is accomplished by loading specimens in special assemblies in materials test reactors or in power producing reactors (surveillance assemblies) [5] and testing them after irradiation to establish the change in mechanical property. For example, the transition temperature increase at a particular neutron fluence can then be derived from knowledge of the equivalent unirradiated curve and this gives one point on the ΔDBTT-neutron fluence curve (trend curve): The curves shown in Slide 5 [14] show the increase in ΔDBTT with increasing neutron fluence and are for steels and weldments of particular compositions. As we have mentioned earlier, different compositions, particularly with respect to impurity and residual element concentrations, can have a significant effect on the sensitivity of the steel to neutron irradiation.

The significance of having appropriate and flexible surveillance programmes has continued to increase over the years. The data obtained from tests on surveillance specimens are used to set operating pressures and temperatures of RPVs; the data is used to assess vessel integrity for actual plant transients; the technique can be used to evaluate the effect of re-irradiation of samples from annealed vessels; a rescheduling of the surveillance programme can meet the needs of a plant life extension programme; the characterised facilities can be used for the irradiation of samples from other reactors which may be temporarily shut down or unavailable, or samples of generic or candidate materials. Indeed for the Spanish reactors (32) problems are emerging of rescheduling their surveillance programmes against a background of surveillance results which show a low irradiation sensitivity and the potential for a much extended PV life.

1.5 EMPIRICAL MODELLING OF IRRADIATION EFFECTS

Earlier empirical models describing irradiation effects on mechanical properties for guidance for Regulatory purposes led to the preparation and use of United States Nuclear Regulatory Commission (USNRC) Regulatory Guide 1.99 [15][Slide 7]. This merely took the copper and phosphorus content of the steel and related them to the neutron fluence to give the trend of the shift
DBIT for that particular steel. This was later modified and refined into a Revision 2 [16] in order to take into account the increasing base of surveillance data, to recognise the importance of nickel in irradiation sensitivity and to treat the weld data as a separate family. Phosphorus content was not included as a variable in the USNRC Reg. Guide Rev.2 because the role of phosphorus could not be distinguished from the data base, which included results for a restricted range of phosphorus and, perhaps more importantly, for the higher copper content of the US steels where the effect of phosphorus is not so marked. It is 'bulk' copper that is used to describe neutron irradiation sensitivity. For a schematic illustration of the effect of copper see Slide 8.

But, copper is present in these steels and weldments in many forms. A significant observation was the discovery [33][35] of 'Digenite', copper sulphide inclusions, in these steels. Copper can also be present as coarse particles, precipitated or undissolved, after stress relief heat treatment of the steels and weldments. Copper is also present as precipitates during irradiation. However it is the copper in solution that provides the major potential for future irradiation embrittlement. Thus, depending upon the quantitative and relative amounts of these forms of copper then it may be that the best descriptor of 'effective' copper is the unirradiated level of 'dissolved' copper (Slide 9)[after McElroy 37]. But for a fuller description of irradiation effects there would appear to be a need for this level of knowledge on other elements. However, for the range of steels being used for RPVs elsewhere in the world phosphorus, as well as the major effect of copper, is seen as an important variable in describing hardening and non-hardening changes in mechanical properties. For example, French [17], Japanese [18] and Russian [19] empirical models, deriving from their own national steels' irradiation effects data bases include phosphorus, and, in some cases, other elements also as variables. (Slide 10) There is therefore an intent, because of the shortcomings of some of this large number of Guides which may be based on limited or somehow unique data, for an IAEA data base to be established which will encompass a large amount and a greater variety of the international irradiation data and which will reflect the large variety of materials in use. It is now generally thought that phosphorus is also a significant element in promoting irradiation sensitivity. Annealing studies [19][34] suggest that phosphorus behaves in two ways as an embrittling agent. Firstly as ultra-fine phosphide precipitates (Slide 11), in the same way as copper, and also as a grain boundary segregate to produce temper embrittlement(Slide12). Clearly there is still scope for further work to describe the the role and behaviour of phosphorus.

The empirical models and Guides serve a variety of purposes and usually describe or predict the mechanical properties after the irradiation of RPV steels and weld specimens. These models are produced in the context of a national programme and are usually, as stated in the previous paragraph, within the constraints of the data base used to generate those models. They could therefore be
subject to modification or further refinement as new data outside the existing range becomes available. In some cases no surveillance data base exists for a particular plant and also there may be no archive material available for irradiation. The materials may belong to a particular ‘family of steels’ and these empirical models can be used to predict behaviour in a ‘generic’ way but confidence in the results can always be enhanced with additional information on the materials such as chemical composition from samples taken from irradiated vessels or by generating more data by irradiating equivalent materials [5].

1.6 MECHANISTIC MODELLING

More sophisticated comprehensive models based on the underlying mechanisms of neutron irradiation effects on pressure vessel steels and welds have and continue to be developed [5][20][21][37]. The detailed underlying irradiation effects and the resulting metallurgical structures which lead to changes in mechanical properties of steels and welds continue to be the subject of detailed investigation. New techniques are available which permit a more detailed description of metallographic structures [5]. The potential of these new developments is being realised to meet technological requirements of operating plants. A full description of the detailed metallography of the pressure vessel materials of a specific plant will augment and underpin the formulation of empirical and mechanistically based relationships between neutron fluence and mechanical property change and also possibly explain the role and significance of other alloying, residual and impurity elements. Increasingly, as these refinements continue to be developed, a detailed metallographic description will become available to help characterise the condition of real pressure vessels and will be of direct value in independently characterising their irradiation and mechanical condition.

Neutron irradiation surveillance results have recently been reviewed and discussed from a consideration of mechanistic models [11]. (Slide 13). The mechanical property changes are attributed to the interaction of dislocations with the following features resulting from neutron irradiation:

- Defect production (i.e. vacancies, interstitials, dislocation loops, vacancy clusters, caused by neutron displacement cascades

- Formation of ultra-fine copper - rich, coherent precipitates (age hardening)

- Ultra-fine phosphide formation

- Ultra-fine carbide formation

- Temper embrittlement caused by phosphorus segregation
It is now generally thought that the first two mechanisms are the most significant. Phosphorus is thought to behave in the same way as copper, that is, by hardening the matrix by precipitation as phosphides. However there is also the possibility for phosphorus to contribute to a non-hardening but embrittling mechanism by segregation to grain boundaries. Nevertheless, irradiation effects in model (simulation), and actual pressure vessel steels involve complicated processes and there may be differences between the two sets of materials, these continue to be the subject of much investigation. This overall situation will continue to be the case whilst predictions are required outside the boundaries of existing data bases or where further refinement of data is required for interpolation of data.

1.7 EXTENSION OF DATA TO NEW OPERATIONAL LIMITS

It is to be stressed that models and guides are constrained within the limits of the data bases used for their formulation and knowledge of the underlying controlling mechanisms. The enlargement of these limits to areas outside current operational requirements can produce data which is not readily explicable with present mechanisms. By way of an example (Slide 14) results from the Kola 3 surveillance data showed no saturation effect at fluence levels exceeding end-of-life (EOL) values. There was, as can be seen in the slide an enhancement of irradiation response. The curve shows remarkable similarity to that shown in Slide 15 which is taken from the Fisher and Buswell paper [20]. This is an example which illustrates the value of modelling irradiation behaviour for 'real' RPV materials.

For the future the improvement in irradiation response of steels and weldments is likely to provide a greater lifetime for pressure vessels and the behaviour of pressure vessels will need to be assessed against those operational requirements.

1.8 CURRENT ‘STATE OF THE ART’

Thus neutron irradiation effects a change in the mechanical properties of PV steels and welds and the changes derive from a multiplicity of factors. These factors include the method of manufacture (plates, forgings and welds), fabrication (including variations in heat treatment), chemical composition (the degree of change being mainly dependant on the copper content) and metallurgical structure, neutron fluence, neutron energy spectrum neutron flux, irradiation temperature and time of irradiation. The changes can be predicted by using data derived from materials test reactor irradiations or from lower fluence rates from surveillance irradiations. Predictions can be obtained from the use of empirical guides together with additional confidence from the added knowledge of generic data, the composition of the steel or and metallurgical structure. However, amelioration or mitigation of these irradiation effects can contribute to the extension of
operational life of some older plants. For nuclear power plants with ‘modern’ pressure vessels the problems of irradiation effects have been largely overcome and new problems of plant lifetime limiting features need to be assessed.

The stresses associated with a postulated transient which causes a rapid cooldown of the pressure vessel at high or increasing system pressure, Pressurised Thermal Shock (PTS), in a region-sensitive to neutron irradiation containing flaws have been evaluated in the USA. This led to ‘PTS screening criteria values’ which, in turn, have led to flux reduction programmes or other measures in several plants to ensure that the screening values will not be exceeded during current (licence) life. However, the USNRC is planning [31] to change (update) the current PTS rules with the result that many US PWR PV will exceed the PTS screening criteria before the end of (licenced) life.

1.9 MITIGATION OF IRRADIATION EFFECTS

Recovery of mechanical properties by annealing irradiation damage has been the subject of many investigations [5][22][19]. Significant recovery of unirradiated mechanical properties, both the shift in ductile brittle transition temperature and upper shelf fracture strength, could be obtained under relatively modest conditions of temperature and time. Interestingly the kinetics of upper shelf recovery is different from the shift in transition temperature indicating that different mechanisms may be operating in these different regimes.

The US Army SM-1A reactor was annealed in 1967 [23] and the BR-3 vessel in 1984 [24]. The feasibility and economics of annealing vessels has been considered in the USA [25][26] and a recommended guide for in-service annealing has been drafted by the ASTM [27]. However, no ‘commercial’ PWRPV has yet been annealed outside the former USSR or former Comecon countries.

After surveillance capsules were withdrawn from the WWER Loviisa plant in Finland, and some of the former USSR plants, and the samples tested and analysed was it realised that the degradation in mechanical properties was greater than expected [Slide16][28][29]. The enhanced degradation in mechanical properties was ascribed to a combination of high copper and phosphorus and a high neutron fluence. From the subsequent evaluation of the WWER plants, in the light of these results, mitigating measures were proposed (Slide 17) and implemented to a greater or lesser extent on a plant specific basis. Amongst these measures were included a modification to the pressure-temperature limits, replacement of outer fuel assemblies by dummy elements which acted as neutron shields, use of highly depleted fuel assemblies at the core periphery and annealing to recover the unirradiated mechanical properties.

31
One of the major developments, and achievements, of the WWER programme [30] has been the annealing and recovery of a large number of actual irradiated pressure vessels (fourteen by the end of May 1993) (Slide 18) starting with the Novovoronezh 3 reactor pressure vessel, after 16 years service, in May 1987. This was carried out at a temperature of 420°C for 150 hours after extensive studies on irradiation and annealing. The recovery was only partial. Higher degrees of recovery have been achieved in subsequent annealing of the other vessels by raising the annealing temperature to 475°C. From the underlying studies [19] the residual embrittlement after annealing was found to be independent of neutron fluence (Slide 19) but significant dependence on phosphorus content was found as we saw earlier in Slide 11. Residual embrittlement was inversely proportional to annealing temperature.

1.10 ANNEALING AND RE-IRRADIATION OF 'ACTUAL' PRESSURE VESSELS (Slides 20 and 21)

The purpose of annealing pressure vessels is to allow their continued use and for plant life assurance/plant life extension. The efficient annealing of pressure vessels is therefore associated with the requirements of knowledge of the re-embrittlement rates and the period for continued operation. This in turn could require surveillance irradiations of annealed material and detailed metallography for a full evaluation of the irradiation response. The techniques used for the mechanical property evaluation may utilise sub-standard specimens and the testing of 'recovered' material using sub-standard sized specimens which have been calibrated against standard sized specimens of reference or archive material.

1.11 INTERNATIONAL PROGRAMMES (Slide 22)

Studies of neutron irradiation effects on pressure vessel steels and weldments continues to absorb much effort worldwide. The emphasis changes from the needs of the older generation of vessels to those associated with the longer anticipated life of the newer vessels.

The IAEA continues to be a focus for international activities in the field of neutron irradiation effects on pressure vessel steels and weldments (Appendix 1). The activities were coordinated in the Division of Nuclear Power under the aegis of the International Working Group on the Reliability of Reactor Pressurised Components (IWG - RRPC), which now operates under as the International Working Group on Nuclear Power Plant Life Management (IWG - NPPLM). There are three main activities in this field. The first is through regularly held Specialist Meetings at different venues (e.g. Balatonfüred, Hungary in 1990) and this present meeting held as a joint meeting with the CSNI Working Group 3. from the OECD. The second area of international activity has been through the Coordinated Research Programme on irradiation effects [5]. This long standing, but highly successful,
venture has now reached the end of Phase 3 and the work to date is described in another paper to be presented at this meeting. The third area of IAEA activity is in the generation of a more comprehensive international data base on irradiation effects which will cover a greater range of materials and conditions. This work has now been initiated and will be coordinated by the IAEA.

The International Group on Radiation Damage Mechanisms (IG-RDM) was founded in 1987 with USNRC sponsorship, to bring together in a 'workshop' atmosphere scientists and engineers involved directly with RPV embrittlement issues. Involvement with the Group has led to many inter-laboratory comparison exercises. This aspect of the international activity has resulted in greater understanding of damage mechanisms because of the obvious advantages of applying a variety of techniques to the same material.

The European Action Group on RPV Materials Irradiation Effects and Studies (AMES) (Appendix 2) is one of a number of network groups being set up in Europe. There are a large number of objectives and while essentially European it is hoped to include a larger international membership at a later stage.

Indeed with the maturing and rationalisation of nuclear technology worldwide there is a perceived increase in cooperative activity in this area. By these means there is a 'sharing' of advanced equipment and techniques and participation in extra-national activities.

1.12 REFERENCES


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35. BUSWELL, J.T., Where he found Digenite in the submerged arc weldment. 'Examination of materials by electron microscopy'. In report of the UK contribution to the Phase 2. IAEA Co-ordinated programme on the analysis of the behaviour of advanced reactor pressure vessel steels under neutron irradiation, ed. L M Davies UKAEA 1983.


APPENDIX 1

IAEA IWG-LMNPP ACTIVITIES SO FAR (IN 1992/93)

1 SPECIALISTS MEETINGS


• 'Fracture mechanics verification by large scale testing', held as joint meeting with OECD/CSNI PWG3, Oak Ridge, USA, 26-29 October 1992. Full proceedings are in the course of publication.

2 COORDINATED RESEARCH PROGRAMMES

• 'Optimisation of RPV surveillance programmes and their analysis', Balatonfured, Hungary, 29 September - 2 October 1992. Meeting of National Chief Scientific Investigators and observers. Essentially complete but some results will become available this year because of delays in some irradiations. Final meeting to incorporate delayed data, complete the CRP and finalise the report - being considered for November 1993.

• 'Coordinated research programme on the management of ageing of an RPV primary circuit nozzle', project launched with objectives to exchange information on the state-of-the-art in assessing the remaining life of RPV nozzles and effects of ageing and perform a collaborative study. Continuing.

3 DATA BASE

• The project is 'to develop a wide international data base on the results from RPV surveillance'. Initiated by the publication of the document 'International data base on ageing management and life extension - data base specification'. Continuing.

4 PUBLICATIONS

• Outstanding proceedings, other than those mentioned above, published. (e.g. ASTM -STP 1170).

• Report in the Technical Report Series on 'Neutron irradiation effects in pressure vessel steels and weldments'. Continuing - in course of publication. Fourteen Chapters by separate authors on different aspects of the subject with one Chapter giving different 'National perspectives' of current national studies in the field.
IWG-LMNPP FUTURE ACTIVITIES

Besides those activity items identified as 'continuing' in Appendix 1 and those not yet agreed authorised and approved the list below.


- Specialists Meeting on "Non-Destructive Examination, Practices and Results", joint meeting IAEA/NEA/CEC Petten, 8-10 March, 1994.


PROGRAMME for 1995 and 1996

To be considered at the IWG-LMNPP in early 1994.
Objectives

- Validation and establishment of safe limits for mitigation and amelioration measures (annealing, etc.);
- Formulation of a microstructurally based model capable of predicting the effect of annealing and re-irradiation;
- Validation of the applications of novel techniques, including reconstructing specimens, miniature and in-situ mechanical test procedures and also advanced microstructural techniques to RPV condition assessment for use in the longer term.
- Maintenance of a European capability expertise and resources for RPV condition assessment and remedial action;
- Participation in collaborative programmes with organisation in the former Soviet Union and Eastern Europe;
- Advice to regulatory bodies and provision of a base for development of common European Standards.

Major Tasks

The network will cover the following range of activities on material studies and expertises:

- Review the capabilities within its member organisation together with the existing knowledge base from previous work programmes.
- Studies on other components than the Reactor Pressure Vessel e.g. internals.
- Assess the availability of stocks of irradiated and unirradiated materials that might be made available for work programmes as well as material that might be recovered from operating or decommissioned reactors.
- Studies on model alloys to improve the understanding of the underlying effects for irradiation damage, thermal ageing and annealing.
- Annealing validation and re-irradiation studies on materials of current interest for LWR (Light Water Reactor) systems in Europe and the former Soviet Union.
- Development of microstructural models of irradiation damage, thermal ageing and annealing.
- Studies on other new materials than the only steels used in the old power plants.
- Studies on irradiation and thermal degradation of materials for a new generation of reactors.
- Survey of national Regulatory Requirements and identification of existing, planned and required Standards at European level relevant to material damage and mitigation methods
Those will be allocated to the following major Tasks:

Task1: Evaluation of Mitigation Methods of Irradiation Damage

Task2: Survey of National Regulatory Requirements

Task3: Identification of Existing, Planned and Required Standards at a European Level (relevant to irradiation damage and mitigation methods)

Task4: Harmonisation of Rules for Defining RPV Material Condition (with the aim of reducing safety margins where possible)

Steering Committee (December 1992)

AEA Technology, UK; TRACTEBEL and CEN/SCK, Belgium; MPA Stuttgart and SIEMENS, FRG; CEA/CEN, France; VTT, Finland; TECNATOM, Spain; JRC/IAM, DG XVII, DGXI of CEC.

Funding

- Various sources: national programmes, regulatory bodies, utilities, CEC programmes and JRC/IAM Support.
- Contribution in kind of all task members.

Operating Agent

JRC, Institute for Advanced Materials of CEC in principle with the help of national institutions of excellence as required by the tasks: VTT, MPA Stuttgart, TRACTEBEL, CEA/CEREM.

Officers

- Chairman of the Steering Committee: M. Davies UK
- Vice Chairman of the S.C.: to be nominated UK
- Network Scientific Advisor: C. English JRC/IAM CEC
- Network Manager: S. Crutzen JRC/IAM CEC
- Network Secretary: U. von Estorff JRC/IAM CEC
- Task Chairmen:
  - Task1: K. Törrönen VTT
  - Task2: R. Gerard TRACTEBEL
  - Task3: J. Foehl MPA
  - Task4: P. Petrequin CEA/CEREM

Cooperation with non-European Institutions

NRC, USA (probable)
World's reactors

At the end of 1992 there were 496 power reactors of about 400,000 MWe(net) capacity in operation or under construction
Number of reactors by age (end 1992)

Charpy curves (ferritic steels)

Notes:
(i) Irradiation reduces upper shelf
(ii) Increases DBTT
(iii) Decreases slope of curve in transition region
IAEA CRP Phase2 results

Note. 03 refers to reference plate HSST O3

IAEA TRS 265, 1986, Fig.18

Neutron flux and fluence

>0.1 MeV
>0.5 MeV
>1.0 MeV
dpa

Questions:
(i) Should there be mixed mode reporting of results?
(ii) Should reference material be included in surveillance and test irradiations as a matter of course?
Guides for describing irradiation response

USNRC Rev.1-took copper and phosphorus to generate trend curves.

USNRC Rev.2-took copper and nickel to generate trend curves.

But besides these there are many national trend curves!

Questions?

(i) What of the adequacy (statistics and range of data) and self consistency of the data base?

(ii) What of the future?

Schematic effect of copper and dose rate

Questions:

(i) What about real materials?

(ii) Does effect saturate with dose for real materials?

(iii) What about phosphorus—does it behave in the same or in a 'mixed' way?
Embrittlement effects

\[ \Delta R T_{NDT} = \Delta R T_{\text{matrix}} + \Delta R T_{\text{ppt}} + (\Delta R T_{\text{seg}}) \]

- point defect clusters
  - microvoids
  - dislocation loops or clusters (stabilized by nitrogen)
  - carbon
  - dose dependence (fluence)\(^{1,2}\)
  - nitrogen effect (low dose/low temp)
  - dose rate effect at \(<300\) C
  - etc., etc., etc., etc., etc., etc., etc., etc.

\[ C_{\text{bulk}} = C_{\text{heai}} + C_{\text{ppt}} + C_{\text{Cu}_{1.5}S} + C_{\text{matrix}} \]

Questions:
- What about welds, forgings?
- What about annealing, re-irradiation?
- What about the upper shelf?

Data bases

- Needed for those cases where there is inadequate data or where comparison with other data is sought.
- But they have to be large for statistical reasons and also encompass a wide range of variables of materials and irradiation conditions to meet possible needs.
- Input data may have to be further refined to allow other interpretations.
- Above all, data has to be consistently treated in an agreed way to be comparable.
Phosphorus (annealing)

Notes:
(i) Recovery increases with annealing temperature
(ii) Rate of recovery decreases with phosphorus content at given temperature
(iii) Residual embrittlement related to %P and annealing temperature

Phosphorus

IAEA CRP Phase III pilot HAZ study. SHIFT 69C on irradiation.

Hardness tests show recovery on annealing at 475C for one week, to be complete

BUT CHARPY TESTS SHOW SHIFT OF 155C!

Ref. C A English et al, this meeting.
Modelling and mechanisms

- defect production from displacement cascades (vacancies, interstitials, dislocation loops, vacancy clusters, vacancy - interstitial pairs)
- formation of ultra fine copper rich precipitates
- ultra fine phosphide formation
- ultra fine carbide formation
- temper embrittlement

It is generally considered that the first two processes are the most important for irradiation embrittlement

REF: Pavinich et al, ASTM STP 1170, 1993

Irradiation beyond EOL fluence

Kola-3 surveillance specimens

Only after surveillance specimens were withdrawn and evaluated from the Loviisa, Kola and Rovensk plants was significant deviation seen in embrittlement from that expected.
Mitigation measures

- modification of operation pressure temperature limits
- replacement of outer fuel assemblies with shielding dummies
- use of highly depleted fuel assemblies at the core periphery
- temperature increase in the boric acid injection tanks
- replacement of the injection pipes from the cold to the hot leg of main circulation loop
- installation of fast closing valves in the main steam system
- recovery annealing: performed on nine units, another three planned

degree of implementation is plant specific

REF. IAEA TECDOC-659

Annealed WWER-440/230 units

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REFERENCE: IAEA-draftTECDOC
Residual embrittlement on annealing

(Ref. Amajev et al ASTM STP 1170,1993)

Notes:
(i) Annealing conditions were 420C for 150h
(ii) Sv10ChMFT, 0.028%P, 0.18% Cu.
(iii) E > 0.5 Mev (?)
Annealing and re-embrittlement behaviour

Armenia NPP surveillance specimens

Ref. Amaev et al. Joint
Soviet-American Seminar 1990

International activities

- IAEA
- AMES
- IGRDM
OVERVIEW OF FRENCH ACTIVITIES ON NEUTRON RADIATION EMBRITTLEMENT OF PRESSURE VESSEL STEEL

BY

C. BRILLAUD* - F. de KEROLAS** - C. PICHON*** - A. TEISSIER****

ABSTRACT

This paper describes recent developments in France's pressure vessel surveillance program, in particular aimed at assessing the irradiation-caused embrittlem ent of EDF's pressurized water reactors. The first part presents surveillance program results for base metal, weld metal and heat-affected zones for 74 capsules removed from 34 units. Fluence ranges from $3.10^{19}\text{n.cm}^{-2}$ to $5.5.10^{19}\text{n.cm}^{-2}$. The second part considers research and development activities in this area. These include the metallurgical structure effects of segregated bands on mechanical properties and the embrittlement rate under irradiation, as well as the effect of irradiation parameters such as flux and neutron spectrum on irradiation embrittlement, in particular to obtain the best damage assessment.

INTRODUCTION

Electricité de France (EDF) started monitoring and studying the effects of irradiation on pressurized water reactor vessels some 25 years ago, with the start-up of its first PWR reactor, Chooz A. However, this activity actually reached full speed starting in 1977 as France implemented its nuclear power plant construction program, which now counts some 54 reactors in operation. Each vessel is individually monitored for embrittlement by neutron irradiation. Through this effort, EDF seeks to guarantee a high level of installation safety, as specified by law, and provide optimum management of the design life of its nuclear system.

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The pressure vessel plays a very specific role in the system, and any damage it experiences through neutron irradiation may limit its lifetime, and therefore the lifetime of the facility itself, since there are no plans for replacement of this component.

Given this background, EDF is conducting a major neutronics and metallurgy-oriented R&D effort with Framatome and the Commissariat à l'Energie Atomique (French atomic energy agency) concerning damage caused by neutron irradiation.

**THE SURVEILLANCE PROGRAM**

**Organization**

Since the organization set up to monitor the irradiation embrittlement of reactor vessel steel has been covered in several papers [2, 4, 5, 9], we will only review the basic points here.

Each vessel is subject to an individual surveillance program, involving from 6 to 8 sampling capsules attached to the outer surface of the thermal shield. The methodology used to monitor the embrittlement of reactor vessel steel was to a large degree inspired by American regulatory practices. It meets the requirements of the decree and memo of February 26, 1974. The collected "Règles de Surveillance en Exploitation des Matériels mécaniques des îlots nucléaires REP" (RSEM), or "Rules for the operational surveillance of mechanical equipment on PWR nuclear islands," describes the practical measures chosen to survey the effects of irradiation on reactor vessel steel.

This surveillance applies to the different constituent materials in the beltline region of the reactor vessel: shell base metal, weld metal for the welding of shells and the welding heat-affected zone. The materials chosen for the surveillance program are those that involve the highest risk of brittle fracture at end-of-life, and therefore the lowest degree of toughness. Toughness is evaluated on the basis of a reference curve that is indexed on the reference temperature nil ductility transition (RT$_{ndb}$). Materials are chosen based on the RT$_{ndb}$ and an evaluation of their irradiation shift using prediction equations.

Mechanical testing specimens are taken from the materials selected for the surveillance program and placed in capsules to be irradiated in the reactor. The irradiation locations are on the outer surface of the thermal shield. The azimuth position was chosen according to the damage lead factor sought in relation to the vessel. This lead factor is defined as the ratio between the flux of neutron energy greater than 1 MeV received by the capsule at the reference point, and that of the vessel at the most highly irradiated point. Legally, this lead factor should not exceed 3.

Each capsule also includes instruments designed to measure the dose of neutrons absorbed by the specimens (fissile and activation dosimetry) and the maximum
irradiation temperature (alloys with low melting points). The embrittlement of materials by neutron irradiation is measured by the increase in transition temperature that it causes ($\Delta T_{cv}$). The temperature $T_{cv}$ determination is based on the CHARPY V transition curve, as the maximum value between $TK_7$ (temperature at an energy level of 56 joules) and $TK_{0.9}$ (temperature at lateral expansion of 0.9 mm). The shift, $\Delta T_{cv}$, is assumed to represent that of the toughness curve, the temperature scale of which is based on $RT_{ndt}$, and therefore:

$$\Delta RT_{ndt} = \Delta T_{cv}$$

Using experience feedback, the surveillance program was reorganized in terms of the dosimetry and the type and number of specimens used.

The surveillance program is operated by EDF’s hot laboratory at the Irradiated Materials Laboratory on the Chinon site. CEA/SAPR in Grenoble is in charge of the dosimetry.

**Operation of the surveillance program**

To date, some 75 capsules from 34 units have been analyzed. They show low content of embrittling elements (copper, phosphorous, nickel). The following table give the minimum, maximum and mean values for all 900-MW PWR units.

<table>
<thead>
<tr>
<th></th>
<th>COPPER</th>
<th>PHOSPHORUS</th>
<th>NICKEL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base</td>
<td>0.04 - 0.08</td>
<td>0.005 - 0.13</td>
<td>0.64 - 0.84</td>
</tr>
<tr>
<td>m</td>
<td>0.061</td>
<td>m = 0.007</td>
<td>m = 0.71</td>
</tr>
<tr>
<td>Weld</td>
<td>0.09 - 0.13</td>
<td>0.012 - 0.019</td>
<td>0.07 - 0.51</td>
</tr>
<tr>
<td>(CPO)</td>
<td>m = 0.11</td>
<td>m = 0.016</td>
<td>m = 0.22</td>
</tr>
<tr>
<td>Weld</td>
<td>0.03 - 0.04</td>
<td>0.003 - 0.015</td>
<td>0.52 - 0.78</td>
</tr>
<tr>
<td>(CPY)</td>
<td>m = 0.033</td>
<td>m = 0.008</td>
<td>m = 0.65</td>
</tr>
</tbody>
</table>

Maximum, minimum and mean contents of embrittling elements (%)  
CPO = Fessenheim and Bugey plants  
CPY = All other 900-MW plants

Copper and phosphorous contents are low, expect for the 6 units of the CPO series, and nearly always below the thresholds currently used in the embrittlement prediction equations. The embrittlement results, as a function of the absorbed neutron dose, are shown in the graphs on the next page for the different materials monitored by the surveillance program: base metal, weld metal and heat-affected zone. Neutron doses range from $3.10^{19}$ to $5.5.10^{19}\text{n} \cdot \text{cm}^{-2}$ for neutron energies greater than 1 MeV. For 900-MW reactor vessels, the highest dose corresponds to an operating lifetime of approximately 30 years. Dosimetry results show the very consistent operation of all plants. The ratio of measured flux to predicted flux ranges from 0.97 to 1.08, depending on the irradiation locations, with a standard deviation of 0.04 at 1 sigma.

The embrittlement of different materials remains moderate, although there is a significant dispersion in the results. The greatest embrittlement, 72°C, is seen for a
dose of $3.54 \times 10^{19}$ n.cm$^{-2}$ on the weld metal on unit 3 at Gravelines, for copper and phosphorous contents of 0.04% and 0.014 weight %, respectively.

**Shifts $RT_{ndt}$ as a function of neutron fluence ($E > 1$ MeV) for the base metal, weld and heat-affected zones.**

For base metals, this dispersion, which may reach 60°C for a given fluence, can not be correlated to the differences in the contents of embrittling elements. As shown above, the copper and phosphorous contents are generally very homogeneous. In several cases, intergranular fractures were observed on the Charpy specimens located in the segregation zones. The specimen fractures are not examined systematically, but only when a test result is significantly different from other results. Note that the embrittlement of 64°C for a fluence of $1.75 \times 10^{19}$ n.cm$^{-2}$ corresponds to the presence of an intergranular fracture on several specimens.

The dispersion of results is equally great for the welds, at approximately 40°C. The
The highest values of shift $RT_{ndt}$ generally correspond to high phosphorus content, but the opposite has not been verified; i.e., the highest phosphorous contents do not always correspond to high embrittlement figures.

Each embrittlement result obtained by the surveillance program is compared to a damage estimate based on an empirical formula, FIS, developed by Framatome for products used in France.

$$FIS \ (°C) = 8 + [24 + 1537(P-0.008) + 238(Cu-0.08) + 191Ni^2 \ Cu][Ω/10^{19}]^{0.35}$$

- $F$ : fluence in n.cm$^{-2}$ ($E>1$MeV) and $10^{18} < F < 6 \times 10^{19}$
- $Cu - 0.08 = 0$ if $Cu \leq 0.08\%$
- $P - 0.008 = 0$ if $P \leq 0.08\%$
- $275°C \leq T$ irradiation $\leq 300°C$.

Out of 75 and 64 embrittlement results obtained for the base metal and weld metal, respectively, only 10 values exceed the estimates made using the FIS formula. What is more, in 9 cases the values were exceeded by no more than $10°C$; in the tenth case (base metal), it exceeded it by $21°C$ (see graphs below).

**Comparison of measured shifts with shifts calculated by using the FIS prediction formula for base metal and weld metal.**

**RESEARCH AND DEVELOPMENT**

Within the scope of the "Life duration project", participants reflected how to highlight the problems caused by the operational degradation of components for which operating surveillance does not provide a solution [3-13]. Various specialized units of EDF, CEA and Framatome defined and implemented a certain number of R&D actions, needed to increase knowledge of degradation phenomenon, in support
of the vessel irradiation monitoring program. For reactor vessels, the hardening of materials by neutron irradiation is considered the type of damage that carried the heaviest lifetime penalty. Irradiation damage is due to rapid neutron flux and the metallurgical properties of the materials exposed to this flux. Actions are therefore designed to acquire better knowledge of the irradiation conditions in the reactor, on one hand, and to better understand the role of certain metallurgical factors on the changes in mechanical properties of the irradiated materials, on the other.

Irradiation conditions

Assessing irradiation damage on reactor vessels requires knowing, to the greatest degree of certainty possible, the dose of neutrons received by the steel constituting the core shell and by the specimens representing it. Based on the measured activity of the dosimeters irradiated with the specimens, and the knowledge of neutron sources, the fluence received by the specimens is calculated and then deduce the fluence received by the vessel. Using a correlation between the irradiation damage and the absorbed neutron dose calculated from the surveillance specimens, we then determine the damage undergone by the vessel for the neutron dose actually absorbed.

Work in this area is designed to find the best damage indicator, and to more precisely assess the neutron spectrums used to calculate the optimum cross-sections required to process capsule dosimetry. It is also designed to enhance methodologies, based on experience feedback, and to study the impact of new fuel management modes on flux levels and neutron spectrums. The main actions implemented to date are as follows:

* Calculation of neutron transport by the CEA/SERMA, using the Tripoli 2 3D code. These computations are used to determine the flux and neutron spectrum at different points of the reactor, characterizing each irradiation location for the specimens. In particular, a fine description of the geometry has enabled researchers to discover the flux disturbance caused by the internal structures (stiffeners) [7].

* An experimental dosimetry program, both inside and outside the vessel, on the Saint Laurent B1 reactor, studies the influence of MOX (mixed oxides) fuel on the flux and neutron spectrum. This program involves an instrumented capsule, comprising 80 dosimeters of 8 different types distributed over 23 measurement points. Complementing this instrumentation are 5 temperature sensors covering the range from 263°C to 304°C. Dosimetry has also been set up in the cavity, comprising 79 dosimeters of 7 different types, including 3 solid-state track recorders (SSTR). Initial results have not shown any significant change in irradiation parameters [12].

* The CEA has launched a second experimental program on its Siloe and Osiris reactors to study the influence of the neutron spectrum and find the best damage indicator. Two materials, representing core shell weld metals for high and low contents of embrittling elements, are irradiated at the same damage fluence, but in very different neutron environments in terms of spectrums. In one case,
irradiation conditions comparable to those of a PWR are sought, and in the other
the spectrum is "degraded" in order to obtain a ratio on the order of 2 between
the rapid fluences (E > 1 MeV) of the two irradiations.

* An analysis of the influence of current new fuel management modes on neutron
flux, flux and fluence factors on the vessel at end-of-life. In addition, EDF's
Research and Development Group has developed a simplified program to
calculate neutron transport, which enables counting the fluence for each vessel
over its whole lifetime, taking into account the different fuel loading plans that
are successively implemented [10].

* Improvement of dosimetric instrumentation and processing methods, based on
experience feedback [1-6], focusing on:
  - the specification for maximum impurity contents for the dosimeters and
    housings;
  - using titanium to replace nickel and stainless steel in the fissile dosimeter
    housings;
  - improving the efficiency of fissile dosimeter filters, in terms of thermal and
    epithermal neutrons; the cadmium was replaced by boron nitride;
  - using new activation dosimeters (niobium);
  - modifying the location of the dosimeters in the capsules;
  - highlighting and taking into account disturbances to the neutron flux caused
    by the presence of the internal structures (stiffeners);
  - refining the neutron spectrum calculated for dosimetry processing by using a
    statistical optimization method;
  - assessing the uncertainties linked to the computation of flux factors and
    measurement of flux and fluence.

Metallurgical studies

These actions and measures are designed to lead to a better knowledge of the
materials' mechanical characteristics, and their changes over time due to irradiation.
Work carried out covers fundamental studies aimed at a better understanding of
irradiation damage mechanisms, as well as experimental work using an atomic pile
or reactor. Key actions under way include the following:

* The study of the influence of metallurgical heterogeneities in the vessel base
  metal on toughness, due to the presence of segregated zones, and their behaviour
  under irradiation. This action, jointly conducted by EDF, Framatome and the
  CEA, aims at better specifying the mechanical properties of the irradiated
  materials that must be taken into account to analyze fast fracture risk margins.
  This program involves the mechanical characterization of piping cut-outs, as
  well as a scrapped vessel shell. It should enable verifying the conservative
  assumption of the toughness curve taken as a reference for calculating the break
  margins.

* The study of the effect of irradiation on heat-affected zones (HAZ) by cladding
  welding. In the case of shell weld heat-affected zones, the mechanical
properties taken into account are those of the base metal, and the conservative nature of this approach is confirmed by the surveillance program. On the other hand, since we do not have the same elements for heat-affected zones under the cladding, this study is designed to collect data on the ageing of these zones under irradiation.

* EDF has started working with Framatome and Creusot Loire to provide a more precise and complete description of the metallurgical and mechanical properties of the core shells now in operation. The manufacturer has developed a model for the manufacture of forged parts, based on experience feedback. This model enables products to be characterized, by analyzing all available elements of the manufacturing process. This program is designed to establish the position of the segregated zones over the whole of the shell, and to appraise the extent to which the surveillance specimens are metallurgically representative of the shell from which they are taken.

* EDF's Research and Development Group is currently carrying out fundamental research into irradiation damage mechanisms. Both the material of the vessel in the surveillance program and synthetic alloys irradiated in an atomic pile are subject to microstructural examination [8-14].

* A study of the uncertainties involved in establishing mechanical properties, and how they evolve under irradiation. The purpose of this study is a finer processing of the surveillance results, while taking into account uncertainties in the risk analysis of fast fractures on vessels.

* Lastly, we should also mention the upcoming major expert assessment program for the Chooz A vessel, scheduled for 1994-95. One of this program's prime objectives is to validate the surveillance approach by comparing the results obtained on the surveillance program with the results of samples taken directly from the vessel material. Significant work was already carried out in 1987 within the scope of the reassessment of fast fracture margins for this plant, in particularly involving the CEA/SAPR's development of a dosimetry technique based on material samples taken directly from the irradiated material [11]. This technique will once again be implemented for the new expert assessment program, in order to determine the dose of neutrons received by the various samples taken from the vessel.

OUTLOOK

Results to date confirm the real capability of vessels to provide a service life of 40 years, under good safety conditions. To ensure this lifetime, the challenge we face is to manage this key asset in two main areas:

- Monitor and limit fluence on the vessels.
- Pursue R&D actions already underway in order to minimize the uncertainties weighing on the results of the operational surveillance programs, and to better understand the various ageing factors involved.
REFERENCES


Russian Research Center "Kurchatov Institute"

A. Amaev, A. Kryukov, V. levit, P. Platonov, M. Sokolov

RADIATION STABILITY AND RECOVERY OF WWER-440 MATERIALS

The main results of a complex investigation of radiation embrittlement of WWER-440 reactor vessel (RV) materials, carried out in Russia are presented. The object of the investigation was surveillance specimen (SS) evaluations of RV materials. It has been found that at the irradiation temperature of 270°C neither the base metal (steel 15Kh2VFA) nor weld metal exhibits saturation of radiation embrittlement in irradiation of specimens up to neutron fluences of $7 \times 10^{20}$ n/cm$^2$ (E$0.5$ Mev). Regularities in the influence of impurity elements (copper and phosphorous) on radiation embrittlement of RV materials have been investigated. It is shown that radiation embrittlement of the weld metal is determined by at least four processes associated with the individual effects of copper and phosphorous, their joint effect and the mechanism of direct build-up of radiation defect in the metal. The effect of dependence of the radiation embrittlement characteristics of the WWER-440 materials on the neutron flux has been found. A tendency has been observed for the values of the radiation embrittlement coefficients $A_1$, characterizing sensitivity of the steel to radiation embrittlement, to reduce with an increase of neutron fluence in irradiation specimens with flux of $4 \times 10^{11}$ n/cm$^2$s.

The main results obtained from studying recovery of radiation embrittlement of reactor vessel steels are presented. The effect of the annealing temperature and annealing time, neutron fluence, and phosphorous and copper impurity contents on recovery of the ductile-to-brittle transition temperature was studied. Recovery of the transition temperature depends mainly on the annealing temperature. At an annealing temperature 420 and 460°C, residual post-annealing embrittlement does not depend on neutron fluence.

Experimental results are presented from a study of changes in ductile-to-brittle transition temperature under cyclic irradiation and recovery. Specimen irradiation was carried out in a commercial reactor in three irradiation and recovery cycles.

Annealing of specimens was performed at 340, 420, 460°C. Change in the transition temperature as a result of annealing depends both on the intermediate annealing temperature and on the chemical composition of the metal, in particular, on the phosphorous content. It has been found that the greatest shift during re-irradiation corresponds to the highest temperature of annealing.
Features of the first generation reactor vessels.

1. Transportability by railway of ready-made reactor vessels.

2. Vessel welds are only circumferential; one of the welds (Number 4.) is situated in region nearby maximum neutron flux.

3. Some vessels have welds with a high content of impurities (P, Cu).

4. A number of vessels was produced without stainless steel plating.

5. Some of the vessels were produced without surveillance samples.

6. Neutron flux is by an order of magnitude greater in places where surveillance samples are irradiated than on vessel wall.
Table 1

<table>
<thead>
<tr>
<th>Grade of vessel steel</th>
<th>Mass content of elements, %</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C</td>
<td>Si</td>
<td>Mn</td>
<td>S</td>
<td>P</td>
</tr>
<tr>
<td>15KH2MFA</td>
<td>0.11-0.22</td>
<td>0.17-0.37</td>
<td>0.3-0.6</td>
<td>0.025</td>
<td>0.025</td>
</tr>
<tr>
<td>15KH2MFAA</td>
<td>0.11-0.16</td>
<td>0.17-0.37</td>
<td>0.3-0.6</td>
<td>0.015</td>
<td>0.012</td>
</tr>
<tr>
<td>Sv-10KHMF</td>
<td>0.07-0.12</td>
<td>0.15-0.35</td>
<td>0.4-0.7</td>
<td>0.030</td>
<td>0.030</td>
</tr>
<tr>
<td>Sv-10KHMF TU</td>
<td>0.07-0.12</td>
<td>0.15-0.35</td>
<td>0.4-0.7</td>
<td>0.030</td>
<td>0.030</td>
</tr>
<tr>
<td>15KH2NMFA</td>
<td>0.13-0.18</td>
<td>0.17-0.37</td>
<td>0.3-0.6</td>
<td>0.020</td>
<td>0.020</td>
</tr>
<tr>
<td>15KH2NMFAA</td>
<td>0.13-0.18</td>
<td>0.17-0.37</td>
<td>0.3-0.6</td>
<td>0.012</td>
<td>0.012</td>
</tr>
<tr>
<td>Sv-10KHGNM AA</td>
<td>0.08-0.14</td>
<td>0.05-0.20</td>
<td>0.9-1.2</td>
<td>0.012</td>
<td>0.010</td>
</tr>
<tr>
<td>Sv-08KHGNM TA</td>
<td>0.05-0.10</td>
<td>0.22-0.37</td>
<td>0.7-1.0</td>
<td>0.012</td>
<td>0.010</td>
</tr>
</tbody>
</table>

Table 1 continued

<table>
<thead>
<tr>
<th>Grade of vessel steel</th>
<th>Mass content of elements, %</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cr</td>
<td>Ni</td>
<td>Mo</td>
<td>V</td>
<td>Ti</td>
</tr>
<tr>
<td>15KH2MFA</td>
<td>2.0-3.0</td>
<td>0.4</td>
<td>0.6-0.8</td>
<td>0.25-0.35</td>
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<tr>
<td>15KH2MFAA</td>
<td>2.0-2.5</td>
<td>0.4</td>
<td>0.6-0.8</td>
<td>0.25-0.35</td>
<td>-</td>
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<tr>
<td>Sv-10KHMF</td>
<td>1.4-1.8</td>
<td>0.3</td>
<td>0.4-0.6</td>
<td>0.20-0.35</td>
<td>0.05-0.10</td>
</tr>
<tr>
<td>Sv-10KHMF TU</td>
<td>1.4-1.8</td>
<td>0.3</td>
<td>0.4-0.6</td>
<td>0.20-0.35</td>
<td>0.05-0.12</td>
</tr>
<tr>
<td>15KH2NMFA</td>
<td>1.8-2.3</td>
<td>1.0-1.5</td>
<td>0.5-0.7</td>
<td>0.1-0.12</td>
<td>-</td>
</tr>
<tr>
<td>15KH2NMFAA</td>
<td>1.8-2.3</td>
<td>1.0-1.5</td>
<td>0.5-0.7</td>
<td>0.1-0.12</td>
<td>-</td>
</tr>
<tr>
<td>Sv-10KHGNM AA</td>
<td>1.8-2.1</td>
<td>1.6-1.9</td>
<td>0.55-0.7</td>
<td>0.03</td>
<td>-</td>
</tr>
<tr>
<td>Sv-08KHGNM TA</td>
<td>1.55-1.85</td>
<td>1.1-1.4</td>
<td>0.5-0.7</td>
<td>0.03</td>
<td>0.03-0.10</td>
</tr>
</tbody>
</table>

Note: For fabrication of VVER-1000 RV the steel is additionally alloyed with nickel in order to improve characteristics of strength and to reach the lower T ko.

Table 3

Limits of content of some impurity and alloying elements and the range of ductile-to-brittle transition temperature in vessel materials of VVER-440 reactors of first generation (production before 1980)

<table>
<thead>
<tr>
<th>Steel of grade 15KH2MFA</th>
<th>Weld metal of wire Sv-10KHMF</th>
</tr>
</thead>
<tbody>
<tr>
<td>P, %</td>
<td>0.010 - 0.019</td>
</tr>
<tr>
<td>Cu, %</td>
<td>0.09 - 0.15</td>
</tr>
<tr>
<td>C, %</td>
<td>0.13 - 0.17</td>
</tr>
<tr>
<td>Ni, %</td>
<td>0.13 - 0.31</td>
</tr>
<tr>
<td>Mn, %</td>
<td>0.38 - 0.69</td>
</tr>
</tbody>
</table>

max. 0 +60
Tk. °C min. -50 -13
Fig. 3 Dependence of shift in transition temperature as a function of fast neutron fluence for surveillance specimens of base and weld metal.
1 - base metal
2 - weld metal
Fig. 4 Coefficient of irradiation embrittlement $A_f$ as function of content of phosphorus and copper in base metal (o) and its weld metal (●) at irradiation temperature 270°C.
Fig. 5 Dependence of shift in transition temperature at irradiation temperature 270°C of base metal (○) and of weld metal (●) as function of content of phosphorus and copper, irradiation in Rovno Unit 1.
F=1.1×10^{19} \text{n/cm}^2 > 0.5 \text{Mev}.

\[ \Delta T_F(270^\circ C) = [609(P+0.1\text{ Cu}) - 2] (F/F_o)^{1/3} \]
Fig. 7 Dependence of shift in transition temperature at irradiation temperature 270°C of base metal and of weld metal as function of fast neutron fluence as characterised by different values of K.
Fig. 13 Radiation embrittlement of base metal at irradiation temperature of 270°C.

- Fe = 0.020%; Cu = 0.11%
- \( \varphi = 1.54 \times 10^{19} \text{ cm}^{-2} \cdot \text{cm}^{-2} \cdot \text{s}^{-1} \)
- \( \varphi = 2.8 \times 10^{17} \text{ cm}^{-2} \cdot \text{s}^{-1} \)
Fig. 14 Radiation embrittlement of weld metal at irradiation temperature of 270°C.
Fig. 17 Radiation embrittlement of Novo-Voronezh unit 1 reactor vessel material (●, ■) and surveillance specimens (○, □) at irradiation temperature 250°C.
Fig. 18 Change of the transition temperature of steel 15Kh2MFA under irradiation in shield (○) and without shield (●).
Fig. 25 $\Delta T_F$ recovery of irradiated weld metal vs. anneal temperature. Annealing duration is 150 h.
Fig. 26 Degree of $\Delta T_r$ recovery of reactor pressure vessel materials vs. annealing duration.
Fig. 27 Impact energy of weld metal steel 15Kh2MFA at the different condition.

- $F = 1 \times 10^{19}$ cm$^{-2}$
- $F = 1 \times 10^{20}$ cm$^{-2}$
- $F = 4 \times 10^{20}$ cm$^{-2}$
- $F = 4.9 \times 10^{20}$ cm$^{-2}$

- $\Delta$, $\circ$, $\square$ - specimens annealed at 420°C, 150 h
- $\bigcirc$ - unirradiated material
Fig. 28 Dependence of the $\Delta T_{res}$ after annealing at 420°C for 150 h of the pressure vessel steel 15Kh2MFA on their phosphorus content.
Fig. 29 Dependence of the $\Delta T_{\text{res}}$ after annealing at different annealing temperature (150 h) on phosphorus content in steel 15Kh2MFA.

- $\circ$ - $T_{\text{ann}} = 340^\circ\text{C}$
- $\square$ - $T_{\text{ann}} = 420^\circ\text{C}$
- $\triangle$ - $T_{\text{ann}} = 460^\circ\text{C}$
Fig. 31 Radiation embrittlement and post-irradiation annealing recovery of transition temperature vs. fast neutrons fluence at different annealing temperature.
SESSION B

NEUTRON IRRADIATION EFFECTS
IAEA Specialist Meeting on 
Irradiation Embrittlement and Optimization of Annealing 
Paris - FRANCE 20-23 Sept 1993

The irradiation embrittlement of two pressure vessel steels - Contribution of local approach

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ABSTRACT

Within the Phase 3 of the IAEA Coordinated Research Programme on "Optimizing of Reactor Pressure Vessel Surveillance Programmes and their Analyses", the French participation has been focused on the contribution of the local approach on the determination of sensitivity to radiation embrittlement of two different pressure vessel steels.

For that, a low sensitive French forging steel (FFA) and a high sensitive "monitor" Japanese plate steel (JRX) were irradiated to a fluence of $3.10^{19}$ n/cm$^2$ at 290$^\circ$C.

The irradiation embrittlement of the two steels measured by the shift of Charpy V transition curves was in good agreement with the estimated shifts given by theoretical prediction.

The fracture toughness properties were carried out at low temperature with brittle fracture and at service temperature, 290$^\circ$C, with ductile tearing.

The values of $K_{1C}$ or $K_{JC}$ for the brittle fracture and $J_{1C}$ for the ductile rupture are compared with predictions established using the local approach of cleavage fracture (Weibull analysis) and the critical rate of void growth ($R/R_0C$ respectively.

INTRODUCTION

In order to make a first evaluation of the ability of the local approach to estimate, from a probabilistic point of view, the embrittlement of pressure vessel steels, two different materials were irradiated at $3.10^{19}$ n/cm$^2$ and 288$^\circ$C under nominally identical conditions.

One of these materials is a low sensitive forging steel (referred FFA) and the other a special "radiation sensitive" correlation monitor steel (referred JRX) tailored to have a great scale of behaviour in the transition region and in the ductile area.
• MATERIALS AND SPECIMENS

The French forging steel FFA is a part of a cutout H2 of the nozzle course Q17 of a thickness of 275 mm (305 mm when quenched). This was manufactured by Creusot-Loire and provided by FRAMATOME.

The chemical composition, the heat treatments and the mechanical properties are summarized as determined at the acceptance of the shell course. table 1a, b and c.

The mechanical specimens were taken from the three quarter-thickness (3/4T) locations, where the steel is homogeneous, in the transverse (TL) orientation.

The Japanese rolled plate is a monitor steel of a thickness of 225 mm manufactured by Kawasaki Steel Corporation.

The chemical composition, the heat treatment and the mechanical properties are summarized in table 2a, b and c.

The steel was planned to be tested in the TL orientation. However, CT specimens, aimed at determining fracture toughness before and after irradiation, were inadvertently machined in the LT orientation. So some complementary testing was done with the longitudinal and the transverse orientation in order to get some insight into the anisotropy of the plate. The specimens were machined from the 1/4T and 3/4T locations.

• IRRADIATION CONDITIONS AND RESULTS

The irradiation was performed in the OSIRIS Material Test Reactor with a SIAT capsule (fig.1). The rig is rotated 180° at the midtime of the radiation exposure to correct the irradiation gradient through the thickness.

The temperature was measured with twelve thermocouples placed on different levels in the rig. As shown schematically on Fig.1, there was a thermal gradient between 277 and 297°C and thermal fluctuations in the range of ± 10°C during irradiation. The mean temperature was 285.3°C. The two thermal neutron fluence detectors based on the 59 Co (n,γ) 60 Co reaction, indicated that the thermal fluence was 4.4 \times 10^{19} \text{n.cm}^{-2}. The fast fluences were measured from detectors using the 63 Cu(n,α) 60 Co reaction, with fast fluences amount to from 2.4 to 3.16 \times 10^{19} \text{n.cm}^{-2} (E > 1 \text{MeV}). These results are illustrated in Fig.1 and summarized in table 3.

The irradiation induced temperature shift indexed at the 8.5 da J1cm^{-2}, Charpy V impact results for the two steels are illustrated in figures 2a and 2b. 46°C for FFA and 100°C for JRQ steel, are in good accordance with FIS formula (1) employed in France, but are underestimated by the R.G. 1-99 rev.2 (2) and the DAS formula (3) for JRQ. This is probably because these formulae do not take into account the high phosphorus content for R.G. 1-99 and is established only for lower residual elements for DAS formula (table 4.1).

The irradiation produces no change in the upper shelf energy for FFA steel but a significant decrease for the sensitive JRQ steel and consequently an expected lowered toughness.
Tensile testing was done at a rate of $3.1 \times 10^{-4}$ s$^{-1}$. The results are illustrated in Fig. 3a and b. For FFA steel, it appears that the two results at -80°C for the irradiated state are very scattered. Otherwise, in the temperature range from -150°C to 0°C, the hardening $\Delta T_f$ due to irradiation is approximately of 20 MPa.

For the JRQ steel (Fig 5b), the paucity of results for the irradiated state is due to the damaging of specimens during the removal from the capsule. We have therefore constructed a curve relating irradiated yield stress and temperature from a single unirradiated test and the unirradiated data. A hardening of 104 MPa is assumed at all temperatures.

It is obvious that the previously established empirical relationship (3) $\Delta T = 0.5 \Delta T_f$, gives a temperature transition shift of 10°C for FFA steel and 52°C for JRQ steel much lower than the observed values of 46°C and 100°C respectively. These results seem to show that the embrittlement is more pronounced than the hardening for these two steels. This may be due to an evolution of the mechanisms of rupture after irradiation with a possible participation of intergranular failure.

- FRACTURE MECHANICS PROPERTIES

Fracture Toughness in Cleavage - Steel FFA.

The fracture toughness measurements for the unirradiated state were carried out at low temperatures between -120 and -80°C and at -80°C after irradiation. For these temperatures, cleavage toughness was taken as $K_{IC}$ when elastic behaviour prevail and as $K_{IC}$ deduced from $J_C$ when brittle fracture occurring after plastic deformation of the specimen. The results are given in Table 5 and compared with Finnish results (4) in LT orientation on Fig.4. For this comparison a thickness correction was made with the formula.

$$K_J(a_{25}) = K_{JC}(12.5)(12.5/25)^{1/4}$$

The toughness in the transverse orientation is lower than in longitudinal orientation, but more importantly is the great scatter, confirmed by the VTT results with a transition temperature at 100 MPavm estimated at -80°C, higher than the temperature of -105°C in the longitudinal orientation.

Steel JRQ

In this case also, as for the FFA steel, the fracture toughness for the unirradiated state were carried out at low temperature, but for two orientations, longitudinal and transverse, and only at 0°C in longitudinal orientation after irradiation. The results are given in Table 5 and illustrated in Figures 5 and 6. In the Fig.5, the toughnesses obtained in longitudinal orientation are compared with VTT results. They indicate good agreement and a transition temperature at 100 MPavm of -55°C. In the transverse direction, as illustrated Fig.6, the same transition temperature of -55°C is estimated. For this steel, the scatter of toughness values is less pronounced than for the FFA steel, despite some pop-in at low temperature tests.
Fracture Toughness for Ductile Rupture
Steel FFA

At 290°C the crack resistance JR curves for unirradiated and irradiated states were obtained for TL orientation by testing with the single specimen unloading compliance methodology. J1C parameters were evaluated from the experimental points with \( \Delta a < 0.15 \) (W-a) and \( J < (W-a) \Delta \tau / 15 \) J1C is the intersection of the linear regression fitted line for the J values lying between the 0.15 and 1.5 mm exclusion lines with the blunting line expressed as \( J = 4 \sigma_Y \Delta a \). The results are given in table 7. These results, with a mean value of 309 KJ/m², are higher than the mean value of VTT of 250 J/m² in the LT orientation at 250°C. After irradiation a decrease of 55 J/m² is observed but with only one result, to compare with the unaffected Upper Shelf Energy determined with Charpy V specimens.

Steel JRQ

The crack resistance JR curves determined for LT orientation give for J1C a mean value of 300 KJ/m², the same toughness that for steel FFA. This high value is justified, as for steel FFA, by a low sulphur content of 0.04 %.

After irradiation a decrease of 50 KJ/m² is observed, is this case, in good accordance with a lowering of the Upper-Shelf of 17 % (table 7).

- CONTRIBUTION OF LOCAL APPROACH

- Application of a local criterion of cleavage rupture.

The determination of valid K1C values requires the use of large size specimens (CT 100 for example) which is not compatible with the space in irradiated rig. The random aspect of cleavage is another difficulty and it is necessary to employ for analysis pessimistic values of K1C as they are proposed by K1R curve.

From the classical fracture mechanics point of view, it is difficult to evaluate "safety margins" effectively and introduce them in a probabilistic analysis. In order, to quantify these margins, it is proposed to use a local approach based on the Weibull statistics, as developed by MUDRY (5) and BEREMIN (6).

At low temperature in the brittleness temperature range, local approach was used to determine the microscopic cleavage characteristics of the steel and their evolution under irradiation. The axisymmetric notched specimens were tested at low temperature to determine average stress and strain at rupture.

These tests on notched specimens were carried out in order to determine the Weibull stress \( \sigma_W \) of the cumulative probability to failure expressed by the formula

\[
PR = 1 - \exp\left[-\left(\frac{\sigma}{\sigma_u}\right)^n\right]
\]

These parameters were determined for the two steels in the two states, before and after irradiation, with a limited number of specimens type AE 4-6 of 6 mm diameter with a notch root of 2.4 mm.
For steel FFA tests were conducted at -90°C before irradiation with 12 specimens and at -80°C after irradiation with only 7 specimens. The representation of the failure probability Fig.7 gave the determination of the Weibull parameters which are:

\[ \sigma_u = 1901 \text{ MPa and } m = 65 \text{ in unirradiated state} \]
\[ \sigma_u = 1981 \text{ MPa and } m = 62 \text{ in irradiated state.} \]

The values of \( m \) representative of the scatter of results are very near each of other and very high, not in agreement with the large scatter of results obtained with CT specimens. A possible reason is the ductile area in the center of the specimen, preceding the brittle rupture, leading to a unification of the conditions of rupture. For the steel JRQ tests were conducted at -160°C before irradiation with 7 specimens and at -20°C after irradiation with the same limited number of specimens. The representation of the failure probability illustrated Fig.8 gave the determination of Weibull parameters which are:

\[ \sigma_U = 2060 \text{ MPa and } m = 30 \text{ in unirradiated state} \]
\[ \sigma_U = 1991 \text{ MPa and } m = 65 \text{ in irradiated state.} \]

In the unirradiated condition, the value of \( m \) is quite low, but by suppressing one point with the highest value of \( \sigma_w \), a new value of 77 was found \( m \). It is obvious that the number of points is unfortunately not sufficient for further analysis.

- **APPLICATION OF STATISTICAL THEORY OF CLEAVAGE TO FRACTURE TOUGHNESS TEST RESULTS.**

The application of the statistical cleavage fracture theory with the stress distribution given in the HRR model leads to:

\[ \ln \left( \frac{1}{[1-P_R]} \right) = (\sigma y m^{-4} K^2 C^4 BC_m)/V_{0au}^m \]

where \( C_m \) is a numerical coefficient, \( V_0 \) a characteristic volume generally taken as equal to (50 \( \mu \)m\(^3\)) and \( B \) the thickness of the specimen.

For the FFA steel, an evolution of \( \sigma y \) as illustrated by Fig.9 was considered to determine the fracture toughness probability, compare with unirradiated (Fig.10) and irradiated results (Fig.11). It is apparent that with a \( m \) value of 65, the scatter of toughness results is not taken into account. In another view, the shift of 30°C is reasonable, comparing with the shift of 46°C obtained with Charpy V tests, but it depends essentially of the hardening and the change of \( \sigma y \) with temperature which is not accurately determined.

Due to the difficulties to interpret accurately the probability of failure with these results with an insufficient number of notched specimens and an evolution of \( \sigma y \) not sufficiently documented, the pressure vessel steel properties determined by Beremin, were applied to the two steels FFA and JRQ, as precedentely made for an irradiated steel (7) with a value of \( m = 22, \sigma y = 2560 \) and \( C_m = 1.310^6 \). For that, the values of \( \sigma y \) used are illustrated in figures. 5a and b.
The probabilities of failure are illustrated on figures 12, 13, 14 and 15 for steels FFA and JRQ in the two states. In these cases, we have a better appreciation of the scatter of toughness results but shifts due to irradiation are widely underestimated: 8°C for FFA steel at 100 MPaVm and 60°C for JRQ steel at 60 MPaVm.

Considering the results summarized in Table 8, these shifts are in very good accordance with the shifts deduced from the hardening.

**APPLICATION OF LOCAL APPROACH IN THE DUCTILE RANGE**

At high temperature, in the ductile range, the local approach was used to determined the microscopic ductile rupture characteristics, i.e., the critical rate of void growth at rupture \((R/R_0)_C\) and the estimated initiation toughness \(J_{1C}\). These parameters are calculated with the results of the testing of axysymmetric specimens at 290°C according to the following formula established for AE4 specimen at 100°C by MUDRY [5].

\[
\ln \left( \frac{R}{R_0} \right)_C = 1.186 \varepsilon_R + 0.01 \\
\text{and} \quad J_{1C} = 4.5 \sigma_y s \Delta a_0 \ln \left( \frac{R}{R_0} \right)_C
\]

with \(\Delta a_0\) chosen as 0.2 mm. The results are given in Table 9 for FFA and JRQ steels before and after irradiation.

The results obtained in the ductile range are summarized in Table 10. The first observation is the difference between measured and calculated values. The reason is the value of \(\Delta a_0\) which is an adjustable parameter, dependant of the distance between nonmetallic inclusions. In a precedent work with a high sulfur materials this value of 0.2 mm overestimated calculated \(J\) values [8]. Considering the evolution due to irradiation, there is a good agreement between the decrease observed with USE results calculated \(J_{1C}\), and measured \(J_{1C}\), for JRQ steel but not for FFA steel.

**CONCLUSION**

The ductile to brittle transition temperature shifts were obtained from CV testing for the pressure vessel steels FFA and JRQ irradiated at 288°C to \(3.10^{19}\) n.cm-2 (E<1MeV).

No upper shelf reduction was observed for the FFA steel, but the reduction is substantial for the JRQ steel. This behaviour were well predicted by local approach with the evolution of the critical growth rate \((R/R_0)_C\) under irradiation.

Tensile testing was done and the hardening by irradiation appeared too low to be able to explain the shifts.

Local approach methodology in cleavage was tentatively used to understand the shift and to have a determination of probabilistic properties of toughness. Some difficulties are due to a loss of definition of the tensile properties and the Weibull parameters \(\sigma_u\) and \(m\). Nevertheless satisfactory representation was obtained for a fixed value of \(m\) to 22, but with shifts which underestimate the Charpy V results, are in good accordance with the measured hardening.
References


**TABLE 1a - CHEMICAL COMPOSITION OF STEEL FFA, % BY WEIGHT**

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>S</th>
<th>P</th>
<th>Si</th>
<th>Mn</th>
<th>Ni</th>
<th>Cr</th>
<th>Ti</th>
<th>Mo</th>
<th>Cu</th>
<th>Co</th>
<th>V</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>LADLE</td>
<td>0.17</td>
<td>0.004</td>
<td>0.007</td>
<td>0.22</td>
<td>1.33</td>
<td>0.73</td>
<td>0.18</td>
<td>0.48</td>
<td>0.07</td>
<td>0.01</td>
<td>0.008</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td>SHELL</td>
<td>0.17</td>
<td>0.004</td>
<td>0.006</td>
<td>0.19</td>
<td>1.28</td>
<td>0.71</td>
<td>0.16</td>
<td>0.47</td>
<td>0.08</td>
<td>0.01</td>
<td>0.005</td>
<td>0.01</td>
<td></td>
</tr>
</tbody>
</table>

**TABLE 1b - HEAT TREATMENTS**

Austenitize 665°C-900°C, 4 h 30'; Water quench; Tempering 630°C-860°C, 8 h 55'; Air cool
+ Shell - Heat from 315°C to 615°C, 30°C/C, h⁻¹; Stress relieving at 615°C, 7 h; Furnace cool from 615°C to 315°C, 30°C/C, h⁻¹; Air cool
+ Acceptance specimens - Heat from 315°C to 615°C, 55°C/C, h⁻¹; Stress relieving at 615°C, 16 h; Furnace cool from 615°C to 110°C, 30°C/C, h⁻¹; Air cool

**TABLE 1c - MECHANICAL PROPERTIES OF THE ACCEPTANCE SPECIMENS**

<table>
<thead>
<tr>
<th>TEMPERATURE, °C</th>
<th>YIELD STRESS, MPa</th>
<th>ULTIMATE STRESS, MPa</th>
<th>ELONGATION, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>475</td>
<td>616</td>
<td>72</td>
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<tr>
<td>343</td>
<td>407</td>
<td>564</td>
<td>73</td>
</tr>
</tbody>
</table>

**TABLE 1c - MECHANICAL PROPERTIES - LT orientation - 1/4 thickness**

<table>
<thead>
<tr>
<th>TEMPERATURE, °C</th>
<th>IMPACT ENERGY, J/cm²</th>
<th>SHEAR, %</th>
<th>LATERAL EXP. mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>189</td>
<td>70</td>
<td>2.0</td>
</tr>
<tr>
<td>0</td>
<td>189</td>
<td>75</td>
<td>2.1</td>
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<tr>
<td>-20</td>
<td>128</td>
<td>45</td>
<td>1.5</td>
</tr>
<tr>
<td>-20</td>
<td>155</td>
<td>55</td>
<td>1.9</td>
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<td>-20</td>
<td>108</td>
<td>40</td>
<td>1.3</td>
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**TABLE 2a - CHEMICAL COMPOSITION OF STEEL JRQ, % BY WEIGHT**

<table>
<thead>
<tr>
<th>LOCATION</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>Cu</th>
<th>Co</th>
<th>V</th>
<th>Sol Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ladle</td>
<td>0.18</td>
<td>0.24</td>
<td>1.42</td>
<td>0.017</td>
<td>0.004</td>
<td>0.14</td>
<td>0.84</td>
<td>0.12</td>
<td>0.51</td>
<td>0.002</td>
<td>0.014</td>
<td></td>
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<tr>
<td>Plate Top 1/4t</td>
<td>0.19</td>
<td>0.25</td>
<td>1.41</td>
<td>0.019</td>
<td>0.004</td>
<td>0.14</td>
<td>0.84</td>
<td>0.12</td>
<td>0.50</td>
<td>0.003</td>
<td>0.012</td>
<td></td>
</tr>
<tr>
<td>A Bottom 1/4t</td>
<td>0.18</td>
<td>0.25</td>
<td>1.39</td>
<td>0.017</td>
<td>0.003</td>
<td>0.14</td>
<td>0.83</td>
<td>0.12</td>
<td>0.50</td>
<td>0.003</td>
<td>0.012</td>
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**TABLE 2b - HEATS TREATMENTS AFTER ROLLING**

Normalize 900°C; Quench 880°C; Tempering 665°C, 12 h; Stress relieving at 620°C, 40 h

**TABLE 2c - MECHANICAL PROPERTIES**

**Tensile**

<table>
<thead>
<tr>
<th>LOCATION</th>
<th>YIELD STRENGTH (MPa)</th>
<th>TENSILE STRENGTH (MPa)</th>
<th>ELONGATION (%)</th>
<th>RED. OF AREA (%)</th>
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<tbody>
<tr>
<td>Top 1/4t</td>
<td>487</td>
<td>635</td>
<td>25</td>
<td>77</td>
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<tr>
<td>Bottom 1/4t</td>
<td>467</td>
<td>624</td>
<td>27</td>
<td>76</td>
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**Charpy V**

<table>
<thead>
<tr>
<th>ORIENTATION</th>
<th>LOCATION</th>
<th>TRANS. TEMP. °C</th>
<th>60% S. TEMP. °C</th>
<th>50% S. FATT. °C</th>
<th>ABSORBED ENERGY AT -12°C J</th>
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</thead>
<tbody>
<tr>
<td>Longitudinal</td>
<td>Top 1/4t</td>
<td>-28</td>
<td>-15</td>
<td>2</td>
<td>75</td>
</tr>
<tr>
<td>Transverse</td>
<td>Top 1/4t</td>
<td>-23</td>
<td>-13</td>
<td>2</td>
<td>69</td>
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**Drop weight**

NDT = -15°C
### TABLE 3: FLUX AND FLUENCE OF THE IRRADIATION FO 842

<table>
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<tr>
<th>Irradiation</th>
<th>Thermal 10^{12} n{ cm^{-2}¥ s^{-1}}</th>
<th>&gt;0.1MeV 10^{12} n{ cm^{-2}¥ s^{-1}}</th>
<th>&gt;1MeV 10^{10} dpa s^{-1}</th>
<th>dpa</th>
<th>Thermal 10^{18} n{ cm^{-2}}</th>
<th>&gt;0.1MeV 10^{18} n{ cm^{-2}}</th>
<th>&gt;1MeV 10^{18} n{ cm^{-2}}</th>
<th>dpa</th>
</tr>
</thead>
<tbody>
<tr>
<td>FO 842</td>
<td>1.49</td>
<td>2.45</td>
<td>9.35</td>
<td>1.5</td>
<td>4.5</td>
<td>7.41</td>
<td>2.83</td>
<td>45.3</td>
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### STEEL

<table>
<thead>
<tr>
<th>Steel Type</th>
<th>Temperature °C</th>
<th>Flux (E&gt;1MeV) 10^{10} n{ cm^{-2}}</th>
<th>T(5) °C</th>
<th>T(8.5) °C</th>
<th>T(9) °C</th>
<th>T(50%) °C</th>
<th>ΔT(85) J/cm²</th>
<th>ΔT(85) °C</th>
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<tbody>
<tr>
<td>JRQ (TL)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Unirradiated</td>
<td>-24</td>
<td>-7</td>
<td>-13</td>
<td>+20</td>
<td>226</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>290</td>
<td>3.05</td>
<td>71</td>
<td>93</td>
<td>+77</td>
<td>188</td>
<td>100</td>
<td></td>
<td></td>
</tr>
<tr>
<td>JRQ (TL)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Unirradiated</td>
<td>-47</td>
<td>-40</td>
<td>-47</td>
<td>-19</td>
<td>223</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>290</td>
<td>3.0</td>
<td>-8</td>
<td>+6</td>
<td>-3</td>
<td>213</td>
<td>46</td>
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### TABLE 4a: RESULTS OF THE STANDARD CHARPY V IMPACT TESTING

<table>
<thead>
<tr>
<th>Reference of material</th>
<th>ΔTT measured at 85 J/cm²</th>
<th>ATT Calculated</th>
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<tbody>
<tr>
<td>JRQ</td>
<td>100</td>
<td>79</td>
</tr>
<tr>
<td>JRQ</td>
<td>100</td>
<td>85±25</td>
</tr>
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</table>

### TABLE 4b: COMPARISON OF MEASURED AND CALCULATED VALUES OF TEMPERATURE TRANSITION SHIFT

<table>
<thead>
<tr>
<th>Material</th>
<th>Irradiation conditions</th>
<th>Specimen number</th>
<th>Orientation and location</th>
<th>W mm</th>
<th>B mm</th>
<th>Test temperatur. °C</th>
<th>K_c MPa m</th>
<th>K_JC MPa m</th>
<th>K corrected MPa m</th>
</tr>
</thead>
<tbody>
<tr>
<td>FFA</td>
<td>Unirradiated</td>
<td>23</td>
<td>TL</td>
<td>25</td>
<td>12.5</td>
<td>-120</td>
<td>51.7</td>
<td>-</td>
<td>43.4</td>
</tr>
<tr>
<td>JRQ</td>
<td>Unirradiated</td>
<td>291</td>
<td>LT-1/4T</td>
<td>25</td>
<td>12.5</td>
<td>-100</td>
<td>77.7</td>
<td>88.7</td>
<td>74.4</td>
</tr>
</tbody>
</table>

### TABLE 5: FRACTURE TOUGHNESS FOR CLEAVAGE OF STEELS FFA AND JRQ.
<table>
<thead>
<tr>
<th>Specimen number</th>
<th>Orientation and location</th>
<th>W (mm)</th>
<th>B (mm)</th>
<th>Test temperature (°C)</th>
<th>KIC (MPa.m)</th>
<th>KJC (MPa.m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>169</td>
<td>T</td>
<td>25</td>
<td>12.5</td>
<td>-166</td>
<td>20.2</td>
<td>-</td>
</tr>
<tr>
<td>171</td>
<td>T</td>
<td>25</td>
<td>12.5</td>
<td>-160</td>
<td>25.2</td>
<td>-</td>
</tr>
<tr>
<td>167</td>
<td>T</td>
<td>25</td>
<td>12.5</td>
<td>-160</td>
<td>40.5</td>
<td>-</td>
</tr>
<tr>
<td>154</td>
<td>T</td>
<td>25</td>
<td>12.5</td>
<td>-160</td>
<td>36.7</td>
<td>-</td>
</tr>
<tr>
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* Pop-in

**TABLE 6** - FRAC TURE TOUGHNESS FOR CLEAVAGE OF THE STEEL JRO IN THE UNIRRADIATED STATE.

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<th>Material</th>
<th>Irradiation conditions</th>
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<th>W (mm)</th>
<th>B (mm)</th>
<th>Test temperature (°C)</th>
<th>Δapp (mm)</th>
<th>ΔF (kJ.m⁻²)</th>
<th>ΔJC (kJ.m⁻²)</th>
<th>dJ/ds (MPa)</th>
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**TABLE 7** - FRAC TURE TOUGHNESS FOR DUCTILE RUPTURE OF STEELS FFA AND JRO.

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<th>Δσ (MPa)</th>
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**TABLE 8** - SUMMARIZED RESULTS FOR THE BRITTLE BEHAVIOUR OF STEELS FFA AND JRO.

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*Table 9*- RESULTS OF FRACTURE TESTS ON AXISYMMETRIC NOTCHED SPECIMENS OF STEELS FFA AND JRO.
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*: TL orientation, **: LT orientation, ***: Estimated value.

**TABLE 10 - RESULTS OBTAINED IN THE DUCTILE RANGE.**
**Figure 6** - Fracture toughness of the JRQ steel in the transverse direction and in the unirradiated state as a function of temperature.

**Figure 7** - Determination of Weibull parameters for FFA steel.

**Figure 8** - Determination of Weibull parameters for JRQ steel.
**FIGURE 12** - PROBABILITY OF FAILURE FOR UNIRRADIATED FFA STEEL. COMPARISON WITH TESTS RESULTS.

**FIGURE 13** - PROBABILITY OF FAILURE FOR IRRADIATED FFA STEEL. COMPARISON WITH TESTS RESULTS.

**FIGURE 14** - PROBABILITY OF FAILURE FOR UNIRRADIATED JRQ STEEL. COMPARISON WITH TESTS RESULTS.

**FIGURE 15** - PROBABILITY OF FAILURE FOR IRRADIATED JRQ STEEL. COMPARISON WITH TESTS RESULTS.
Investigation of the Templet cut out of the Kozloduy Unit 2 Reactor Pressure Vessel

Kryukov, A. "Kurchatov Institute"
Klausnitzer, E., Leitz, C. "Siemens KWU"
Rieg, C.Y. "EDF"

ABSTRACT

In the framework of the 6 mont WANO programme small templets were cut out of the inside of the Kozloduy NPP Unit 2 reactor pressure vessel to assess the actual state of the RPV material before and after annealing.

The actual values of the weld metal characteristics required for estimates of the radiation-limited lifetime - the ductile-to-brittle transition temperature in the initial state ($T_{10}$), the phosphorous and copper contents that determine the radiation stability of steel were not determined during manufacture. The Kozloduy Unit 2 pressure vessel had no surveillance specimens programme and its radiation stability was evaluated on the basis of dependencies obtained from the results of investigations of surveillance specimens from other WWER-440 reactor. For this reason, the determination of the actual values of the pressure vessel characteristics and their changes in the course of reactor operation, as well as the comparison of the experimental data with those, obtained from calculation were set forth as the principle objectives of the study.

Instrumented impact tests were carried out on sub-size specimens of base and weld metal. Using correlation dependancies, values of ductile-to-brittle transition temperature were determined for base metal and weld metal corresponding to standard specimen tests (in accordance with Russian standards):

- base metal before annealing 40°C
  after annealing 16°C
- weld metal before annealing 212°C
  after annealing 70°C

The estimated value of $T_k$, corresponding to the initial, unirradiated, state of metal, was 50°C.

The experimental results were compared to a prediction of the extent of radiation-induced embrittlement of Kozloduy Unit 2 pressure vessel materials. It was confirmed that radiation-induced embrittlement of the base metal does not impose any limitation on the radiation-limited lifetime of the pressure vessel.

The predicted increase of the ductile-to-brittle transition temperature of weld metal as a result of irradiation (about 165°C) is practically equal to the experimental result (162°C). However, the absolute value of $T_r$ before annealing (212°C), obtained from tests is about 40°C higher than the estimated value, that is, the calculation does not produce a conservative estimate. This was explained by an understated value of $T_{10}$ (+10°C), which had been produced by calculation on the basis of the data from chemical analysis of the
weld metal, performed by the manufacturer. The results of investigations on the
templates, however, gave an estimated value of $T_{ko} = 50^\circ C$.

The effectiveness of annealing as a means of recovery of the mechanical properties
of irradiated WWER-440 reactor pressure vessels was confirmed. The ductile-to-brittle
transition temperature of the weld metal was recovered by no less than 85%.
PRE-PRELIMINARY RESULTS FROM THE PHASE III OF THE IAEA CRP "OPTIMIZING OF REACTOR PRESSURE VESSEL SURVEILLANCE PROGRAMMES AND THEIR ANALYSIS"

M. BRUMOVSKÝ
F. GILLEMOT
A. KRYUKOV
V. LEVIT

to be presented
at the

Joint IAEA/NEA Specialists meeting on

Irradiation Embrittlement and Optimization of Annealing

Paris, France, 20-23 September, 1993
Preliminary results from the Phase III of the IAEA CRP "Optimizing of reactor pressure vessel surveillance programmes and their analysis"

by

Milan BRUMOVSKÝ et al.
Nuclear Research Institute Řež plc
250 68 Řež, Czech Republic

ABSTRACT

Paper gives preliminary results and some conclusions from Phase III of the IAEA Coordinated Research Programme "Optimizing of Reactor Pressure Vessel Surveillance Programmes and their Analysis" which was realized during last seven years in fifteen member states.

First analysis has been aimed to the following directions:
- comparison of results from initial, unirradiated materials condition,
- comparison of transition temperature shifts (from notch toughness testing) with respect to:
  - content of residual (P, Cu) and alloying (Ni) elements,
  - type of material - base metal and weld metal
  - irradiation temperature - 288 and 265 °C
  - type of fluence dependance

Special effort has been taken to the analysis of the behaviour of a chosen reference steel - JRQ : its homogeneity, scatter of results and resistance against irradiation embrittlement.
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<td>0.71</td>
</tr>
<tr>
<td>BW</td>
<td>0.012</td>
<td>0.22</td>
<td>1.58</td>
</tr>
<tr>
<td>GFB</td>
<td>0.006</td>
<td>0.05</td>
<td>0.85</td>
</tr>
<tr>
<td>JF</td>
<td>0.016</td>
<td>0.04</td>
<td>0.60</td>
</tr>
<tr>
<td>GWA</td>
<td>0.013</td>
<td>0.04</td>
<td>0.93</td>
</tr>
<tr>
<td>JFL</td>
<td>0.004</td>
<td>0.01</td>
<td>0.74</td>
</tr>
<tr>
<td>JFM</td>
<td>0.014</td>
<td>0.15</td>
<td>0.79</td>
</tr>
<tr>
<td>JPA</td>
<td>0.018</td>
<td>0.33</td>
<td>0.82</td>
</tr>
<tr>
<td>JPB</td>
<td>0.017</td>
<td>0.01</td>
<td>0.83</td>
</tr>
<tr>
<td>JPC</td>
<td>0.007</td>
<td>0.01</td>
<td>0.81</td>
</tr>
<tr>
<td>JPD</td>
<td>0.006</td>
<td>0.16</td>
<td>0.10</td>
</tr>
<tr>
<td>JPE</td>
<td>0.006</td>
<td>0.16</td>
<td>0.39</td>
</tr>
<tr>
<td>JPF</td>
<td>0.020</td>
<td>0.16</td>
<td>0.62</td>
</tr>
<tr>
<td>JPG</td>
<td>0.017</td>
<td>0.16</td>
<td>0.82</td>
</tr>
<tr>
<td>JPH</td>
<td>0.006</td>
<td>0.17</td>
<td>1.18</td>
</tr>
<tr>
<td>JPI</td>
<td>0.006</td>
<td>0.01</td>
<td>0.69</td>
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<tr>
<td>JPJ</td>
<td>0.005</td>
<td>0.05</td>
<td>0.63</td>
</tr>
<tr>
<td>JRQ</td>
<td>0.019</td>
<td>0.14</td>
<td>0.83</td>
</tr>
<tr>
<td>JWN</td>
<td>0.008</td>
<td>0.02</td>
<td>0.87</td>
</tr>
<tr>
<td>JWO</td>
<td>0.005</td>
<td>0.03</td>
<td>0.78</td>
</tr>
<tr>
<td>JWP</td>
<td>0.009</td>
<td>0.03</td>
<td>0.90</td>
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<td>JWQ</td>
<td>0.026</td>
<td>0.26</td>
<td>1.10</td>
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COMPARISON OF INITIAL VALUES OF JRQ MATERIAL

<table>
<thead>
<tr>
<th>CODE</th>
<th>DEPTH</th>
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<th>$T_{411J}$</th>
<th>USE</th>
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<tbody>
<tr>
<td>BE</td>
<td>55</td>
<td>-</td>
<td>-38.3</td>
<td>180</td>
</tr>
<tr>
<td>JA</td>
<td>10</td>
<td>T-L</td>
<td>-81.8</td>
<td>164</td>
</tr>
<tr>
<td>JA</td>
<td>10</td>
<td>L-T</td>
<td>-</td>
<td>230</td>
</tr>
<tr>
<td>CS</td>
<td>35</td>
<td>L-T</td>
<td>-33.0</td>
<td>225</td>
</tr>
<tr>
<td>JA</td>
<td>55</td>
<td>T-L</td>
<td>-27.0</td>
<td>170</td>
</tr>
<tr>
<td>RG</td>
<td>55</td>
<td>T-L</td>
<td>-10.5</td>
<td>191</td>
</tr>
<tr>
<td>GD</td>
<td>53</td>
<td>T-L</td>
<td>-20.0</td>
<td>195</td>
</tr>
<tr>
<td>JA</td>
<td>55</td>
<td>L-T</td>
<td>-27.0</td>
<td>230</td>
</tr>
<tr>
<td>AR</td>
<td>48/5</td>
<td>L-T</td>
<td>-75.7</td>
<td>191</td>
</tr>
<tr>
<td>UK</td>
<td>55</td>
<td>L-T</td>
<td>-21.8</td>
<td>200</td>
</tr>
<tr>
<td>CS</td>
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<td>L-T</td>
<td>-14.8</td>
<td>225</td>
</tr>
<tr>
<td>HU</td>
<td>50</td>
<td>L-T</td>
<td>-27.9</td>
<td>130</td>
</tr>
<tr>
<td>RU</td>
<td>65</td>
<td>T-L</td>
<td>-4.4</td>
<td>178</td>
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<td>US</td>
<td>65</td>
<td>T-L</td>
<td>-11.3</td>
<td>202</td>
</tr>
<tr>
<td>CH</td>
<td>65</td>
<td>L-T</td>
<td>-28.7</td>
<td>201</td>
</tr>
<tr>
<td>ES</td>
<td>68/80</td>
<td>T-L</td>
<td>-11.3</td>
<td>199</td>
</tr>
<tr>
<td>SF</td>
<td>55</td>
<td>L-T</td>
<td>-21.8</td>
<td>212</td>
</tr>
<tr>
<td>GD</td>
<td>107</td>
<td>T-L</td>
<td>-5.2</td>
<td>181</td>
</tr>
<tr>
<td>JA</td>
<td>112/159</td>
<td>T-L</td>
<td>-27.0</td>
<td>197</td>
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<tr>
<td>JA</td>
<td>112</td>
<td>L-T</td>
<td>-26.1</td>
<td>230</td>
</tr>
</tbody>
</table>

$T_{411J}(T-L,55)$ = -14.1 +/- 7.3
$T_{411J}(L-T,55)$ = -25.0 +/- 5.5
$T_{411J}(55)$ = -21.9 +/- 9.0

USE (T-L,55) = 189.2 +/- 11.5
USE (L-T,55) = 212.0 +/- 14.0
USE (55) = 201.5 +/- 17.2
### COMPARISON OF INITIAL TENSILE PROPERTIES OF JRQ MATERIAL

<table>
<thead>
<tr>
<th>CODE</th>
<th>DEPTH</th>
<th>ORIENT</th>
<th>DIA</th>
<th>Rp</th>
<th>Rm</th>
<th>Z</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>CS</td>
<td>35</td>
<td>T</td>
<td>4</td>
<td>564</td>
<td>675</td>
<td>71.6</td>
<td>27.0</td>
</tr>
<tr>
<td>ES</td>
<td>56</td>
<td>T</td>
<td>4</td>
<td>494</td>
<td>627</td>
<td>70.0</td>
<td>24.7</td>
</tr>
<tr>
<td>FR</td>
<td></td>
<td></td>
<td></td>
<td>460</td>
<td>602</td>
<td></td>
<td></td>
</tr>
<tr>
<td>RG</td>
<td>56</td>
<td>T</td>
<td>4</td>
<td>485</td>
<td>627</td>
<td>73.0</td>
<td>25.6</td>
</tr>
<tr>
<td>JA</td>
<td>66</td>
<td>T</td>
<td>4</td>
<td>479</td>
<td>616</td>
<td>73.5</td>
<td>22.6</td>
</tr>
<tr>
<td>UK</td>
<td></td>
<td></td>
<td></td>
<td>481</td>
<td>624</td>
<td>73.7</td>
<td>25.9</td>
</tr>
<tr>
<td>US</td>
<td>65</td>
<td>T</td>
<td>4</td>
<td>484</td>
<td>628</td>
<td>70.4</td>
<td>25.5</td>
</tr>
<tr>
<td></td>
<td>55/65</td>
<td>T</td>
<td>4</td>
<td>482.3</td>
<td>626.5</td>
<td>74.2</td>
<td>25.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>+/- 7.2</td>
<td>7.0</td>
<td>2.6</td>
<td>0.7</td>
</tr>
</tbody>
</table>

| JA | 5 | T | 12.5 | 564.0 | 686.5 | 81.5 | 25.8 |
|    |   |   |      |       |       |     |     |
|    |   |   |      |       |       |     |     |
|    |   |   |      |       |       |     |     |

\[ \Delta R_p = 88.87 \ (F.10^{-23})^{0.52} \]

\[ \Delta R_m = 92.4 \ (F.10^{-23})^{0.315} \]
RESULTS FROM IRRADIATION OF JRQ MATERIAL

\[ T_{\text{IRRADIATION}} = 290 \, \text{C} \]

\[ \Delta T_{41J} (T-L) = 65.8 \left( F.10^{-23}\right)^{0.362} \]

\[ \Delta T_{41J} (T-L) = 51.2 \left( F.10^{-23}\right)^{0.334} \]

\[ \Delta T_{41J} (T-L) = 58.7 \left( F.10^{-23}\right)^{0.34} \]

\[ T_{\text{IRRADIATION}} = 270 \, \text{C} \]

\[ \Delta T_{41J} = 86.6 \left( F.10^{-23}\right)^{0.30} \]
CRP PHASE III
MODELS

MODEL 1 :

REG.GUIDE 1.99, Rev.1 

\[ \Delta T_{41J} = \frac{5}{9} [40 + 1000(Cu-0.08) + 500(P-0.008)]F^{0.5} \]

BM + WM :
\[ \Delta T_{41J} = \frac{5}{9} [54.3 + 353(Cu-0.08) + 3250(P-0.008)]F^{0.453} \]
\[ \sigma = 22.0 \text{ C}, R^2 = 0.772, R_{xy} = 0.878 \]

BM :
\[ \Delta T_{41J} = \frac{5}{9} [57.7 + 266(Cu-0.08) + 3500(P-0.008)]F^{0.403} \]
\[ \sigma = 21.0 \text{ C}, R^2 = 0.671, R_{xy} = 0.819 \]

WM :
\[ \Delta T_{41J} = \frac{5}{9} [57 + 441(Cu-0.08) + 4630(P-0.008)]F^{0.365} \]
\[ \sigma = 23.5 \text{ C}, R^2 = 0.90, R_{xy} = 0.949 \]

MODEL 2 :

Guthrie and McElroy :
\[ \Delta T_{41J} = \frac{5}{9} [-10.4 + 472.Cu + 352.Cu.Ni]F^{0.272} \]

BM + WM :
\[ \Delta T_{41J} = \frac{5}{9} [22.5 + 256.Cu + 357.Cu.Ni]F^{0.466} \]
\[ \sigma = 22.8 \text{ C}, R^2 = 0.756, R_{xy} = 0.87 \]

BM :
\[ \Delta T_{41J} = \frac{5}{9} [29.3 + 202.Cu + 360.Cu.Ni]F^{0.47} \]
\[ \sigma = 22.5 \text{ C}, R^2 = 0.621, R_{xy} = 0.789 \]

WM :
\[ \Delta T_{41J} = \frac{5}{9} [24.7 -1502.Cu + 2230.Cu.Ni]F^{0.361} \]
\[ \sigma = 23.9 \text{ C}, R^2 = 0.90, R_{xy} = 0.949 \]
MODEL 3

Randall (for Ni > 0.5 %)
\[ \Delta T_{41J} = \frac{5}{9} [30 + 1000(\text{Cu}-0.05)].F^{0.35} \]

BM + WM :
\[ \Delta T_{41J} = \frac{5}{9} [20.4 + 353.\text{Cu}].F^{0.523} \]
\[ \sigma = 24.0 \text{ C}, R^2 = 0.729 \]

BM :
\[ \Delta T_{41J} = \frac{5}{9} [29.1 + 471.\text{Cu}].F^{0.492} \]
\[ \sigma = 23.73 \text{ C}, R^2 = 0.58 \]

WM :
\[ \Delta T_{41J} = \frac{5}{9} [15.8 + 763.\text{Cu}].F^{0.365} \]
\[ \sigma = 23.78 \text{ C}, R^2 = 0.898 \]

MODEL 4 :

Japan :
\[ \Delta T_{41J} = \frac{5}{9} [15.25 + 432.\text{Cu.P}].F^{0.332} \]

BM + WM :
\[ \Delta T_{41J} = [18.38 + 16300.\text{Cu.P}].F^{0.403} \]
\[ \sigma = 21.0 \text{ C}, R^2 = 0.792, R_{xy} = 0.890 \]

BM :
\[ \Delta T_{41J} = [19.65 + 15740.\text{Cu.P}].F^{0.414} \]
\[ \sigma = 20.54 \text{ C}, R^2 = 0.685, R_{xy} = 0.828 \]

WM :
\[ \Delta T_{41J} = [16.79 + 17850.\text{Cu.P}].F^{0.358} \]
\[ \sigma = 23.68 \text{ C}, R^2 = 0.899, R_{xy} = 0.948 \]

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IRRADIATION EMBRITTLEMENT OF SOME 15Kh2MFA PRESSURE VESSEL STEELS UNDER VARYING NEUTRON FLUENCE RATES

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** IVO International Ltd, FIN-01019 IVO, Finland

ABSTRACT. Irradiation exposure parameters in the surveillance positions of Loviisa power units have varied considerably due to installation of dummy fuel elements and to the fact that some specimens were loaded in a gradient fluence rate position. The applied fluence values range from $0.1 \times 10^{19}$ n/cm$^2$s, E > 1 MeV, the fluence rate values from $0.1 \times 10^{11}$ n/cm$^2$s, and the irradiation times from 1 to 9 reactor cycles. Material response to irradiation was studied with Charpy-V and with Charpy-size fracture mechanics specimens. The derived trend curves are within the limits given in the current Russian Norm. Significant fluence rate effects or time at irradiation temperature effects on embrittlement sensitivity were not found in this forging material. The results are relevant to VVER-440 reactors which have resorted to mitigating measures like fluence rate reduction or annealing.

KEYWORDS: pressure vessel steel, embrittlement, fluence, fluence rate, Charpy-V, fracture toughness, cleavage fracture

Irradiation embrittlement of reactor materials is not only a function of the total neutron fluence but also of parameters like irradiation temperature and neutron fluence rate. For some materials thermal ageing or thermal ageing combined with low fluence rate irradiation might give an additional component to the total embrittlement.

The uncertainty due to a possible fluence rate effect is taken into account by setting requirements on the applied lead factor in surveillance irradiations. However, in cases when NPPs have resorted to fluence rate mitigation methods like installation of dummy elements, a simple definition of a lead factor is not any more available. In these cases material behaviour should be studied by irradiations resembling the real irradiation history of the pressure vessel. The complicated irradiation history due to core reduction measures applied in Loviisa NPP and the simultaneous material irradiations are described and the base metal material data analysed in this study.
DESCRIPTION OF MATERIALS, SPECIMENS AND TESTING

The material under study is the Russian 15Kh2MFA pressure vessel steel used for fabricating the vessels for VVER-440 reactors. This material notation is used for forgings. The material originates from authentic forgings which have undergone the relevant metallurgical heat treatments including quenching, tempering and stress relief heat treatments of totally 70 hours' duration in five stages. Chemical compositions of the materials are given in Table I. The two materials have almost identical chemical composition.

Table I. Chemical composition of the materials in atomic percentage.

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>S</th>
<th>P</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>V</th>
<th>As</th>
<th>Co</th>
<th>Cu</th>
</tr>
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<tbody>
<tr>
<td>Base-1</td>
<td>0.15</td>
<td>0.30</td>
<td>0.49</td>
<td>0.017</td>
<td>0.010</td>
<td>2.56</td>
<td>0.15</td>
<td>0.67</td>
<td>0.34</td>
<td>0.01</td>
<td>0.002</td>
<td>0.16</td>
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<tr>
<td>Base-2</td>
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<td>0.25</td>
<td>0.40</td>
<td>0.015</td>
<td>0.012</td>
<td>2.75</td>
<td>0.16</td>
<td>0.69</td>
<td>0.30</td>
<td>0.01</td>
<td>0.009</td>
<td>0.12</td>
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</table>

ISO Charpy-V (CVN) and Charpy-size pre-cracked (CVN_{pc}) specimens in LT-orientation were used in the study. Charpy-V specimens were tested with an instrumented impact hammer using 5.4 m/s impact velocity. The precracked specimens were used for static fracture mechanics tests performed with a servohydraulic testing machine. Crack growth was measured with partial unloading compliance method the compliance being calculated from crack mouth opening versus load compliance. The precracked specimen is shown in Figure 1. The specimens were manufactured and precracked in another laboratory. Before testing 10% side grooves were machined on both sides of the specimen.

Figure 1. Precracked Charpy specimen.

Only Charpy-V impact energy values are included in the analyses even if lateral expansion and fracture appearance are measured for each specimen.

Only fracture toughness values corresponding to the onset of brittle fracture are analysed in the study. The critical J-integral value measured in the experiment is transformed to $K_{JC}$ for analyses.
IRRADIATIONS

Specimens were irradiated in the vertical surveillance positions on the outer surface of the core barrel in Loviisa power plants. One irradiation set consists of a chain of small capsules. The walls of the capsules were able to withstand the pressure load, and hence thermal contact between the specimens and the cooling water is guaranteed only by fabrication tolerances between the specimens, aluminium spacers and the capsule walls. Specimen temperature in the irradiation position was measured during one reactor cycle with a mock-up irradiation capsule instrumented with thermocouples [1]. During this cycle the measured specimen temperature followed the cooling water temperature within ±2 °C margin. The irradiation temperature of the specimens was 265 °C.

The installation of dummy fuel elements into the cores of Loviisa-1 and Loviisa-2 units during specimen irradiations lead to a varying irradiation fluence, fluence rate and time history. Some of the capsules were exposed to low fluence rate only. The irradiation times and fluence and fluence rate ranges of Charpy-V specimens and precracked specimens are given in Table II.

Table II. Irradiation history and irradiation parameters of Charpy-V and $CVN_{pc}$ specimens. Lo13 denotes base-1 material irradiated in capsule number 3, etc.

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Cycles</th>
<th>Irradiation time</th>
<th>Fluence $10^{19}$</th>
<th>Fluence rate $10^{11}$</th>
<th>Fluence $10^{19}$</th>
<th>Fluence rate $10^{11}$</th>
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<tr>
<td></td>
<td></td>
<td>FC [FPD] RC [FPD] CVN</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>L11</td>
<td>1</td>
<td>285</td>
<td>4.1</td>
<td>16.6</td>
<td>0.2 - 3.6</td>
<td>0.8 - 14.6</td>
</tr>
<tr>
<td>L12</td>
<td>1 - 3</td>
<td>925</td>
<td>13.8</td>
<td>17.2</td>
<td>0.8 - 13.7</td>
<td>1.0 - 17.2</td>
</tr>
<tr>
<td>L13</td>
<td>1 - 3</td>
<td>925</td>
<td>13.6</td>
<td>17.0</td>
<td>0.8 - 13.7</td>
<td>1.0 - 17.2</td>
</tr>
<tr>
<td>L14</td>
<td>1 - 3</td>
<td>925</td>
<td>13.5</td>
<td>17.0</td>
<td>0.8 - 13.7</td>
<td>1.0 - 17.2</td>
</tr>
<tr>
<td></td>
<td>4 - 9</td>
<td>1759</td>
<td>3.7</td>
<td>2.4</td>
<td>0.3 - 3.4</td>
<td>0.2 - 2.2</td>
</tr>
<tr>
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<td>1 - 3</td>
<td>925</td>
<td>13.5</td>
<td>17.0</td>
<td>0.8 - 13.7</td>
<td>1.0 - 17.2</td>
</tr>
<tr>
<td>L16</td>
<td>1 - 3</td>
<td>925</td>
<td>13.5</td>
<td>17.0</td>
<td>0.8 - 13.7</td>
<td>1.0 - 17.2</td>
</tr>
<tr>
<td></td>
<td>3 - 4</td>
<td>277</td>
<td>1.0</td>
<td>2.4</td>
<td>0.3 - 0.5</td>
<td>0.1 - 2.1</td>
</tr>
<tr>
<td>L21</td>
<td>1</td>
<td>294</td>
<td>4.1</td>
<td>16.1</td>
<td>0.2 - 4.1</td>
<td>0.9 - 16.3</td>
</tr>
<tr>
<td></td>
<td>2 - 7</td>
<td>1783</td>
<td>3.6</td>
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<td>0.2 - 2.4</td>
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<tr>
<td>L22</td>
<td>1</td>
<td>294</td>
<td>4.1</td>
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<td>0.2 - 3.7</td>
<td>0.8 - 14.7</td>
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<tr>
<td>L23</td>
<td>4 - 8</td>
<td>1427</td>
<td>3.1</td>
<td>2.5</td>
<td>0.3 - 2.8</td>
<td>0.2 - 2.3</td>
</tr>
<tr>
<td>L24</td>
<td>2 - 7</td>
<td>1783</td>
<td>3.4</td>
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<td>0.3 - 2.9</td>
<td>0.2 - 1.9</td>
</tr>
<tr>
<td>L25</td>
<td>2 - 4</td>
<td>855</td>
<td>1.7</td>
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<td>0.1 - 1.5</td>
<td>0.2 - 2.0</td>
</tr>
<tr>
<td>L26</td>
<td>2 - 5</td>
<td>1158</td>
<td>2.3</td>
<td>2.3</td>
<td>0.2 - 2.2</td>
<td>0.2 - 2.2</td>
</tr>
</tbody>
</table>

FPD - Reactor full power days  FC - Full core  RC - Reduced core
Fluence parameters in units n/cm², E < 1 MeV

The change from full core loading to reduced core loading decreased the specimen irradiation fluence rate by a factor of 7 - 9 depending on whether transition core or the final low leakage core is referred to.

The irradiation chains were longer than the core height, and hence on the upper and lower ends of the chains a considerable gradient in the fluence rate was found.
Precracked Charpy specimens were located in this area. The measured fluence rate distribution along the irradiation channel is shown in Figure 2. Fluence of Charpy-V specimens in one irradiation set is rather homogeneous. The fluence of the precracked Charpy specimens on the contrary varies almost linearly in one irradiation chain.

![Figure 2. Fluence rate distribution along the irradiation channel. The loading positions of CVN and CVN_{pc} specimens in the chain are pointed out.](image)

The irradiation fluence versus fluence rate parameters of Charpy-V specimens are shown in Figure 3. The same irradiation parameter ranges for base-1 CVN_{pc} specimens are shown in Figure 4 and for base-2 CVN_{pc} specimens in Figure 5.

![Figure 3. Irradiation parameter range of Charpy-V specimens. Closed points indicate a constant irradiation condition. The open points are given in pairs, the higher fluence rate irradiations were applied first, the lower rate irradiations thereafter. Total fluence values are given in the figure. One irradiation set consists of 12 specimens.](image)
Figure 4. The range of irradiation parameters for base-1 CVN\textsubscript{pc} specimens. One line shows the parametric range of one set of specimens in the gradient irradiation position. The dashed lines (L14 and L16) indicate the two subsequent irradiation periods, the higher fluence irradiation was performed first. n is the number of specimens in one irradiation set. L11 (R) specimens were reconstituted from broken Charpy-V specimens halves.

Figure 5. The range of irradiation parameters for base-2 CVN\textsubscript{pc} specimens. One line shows the range of parameters in one set of specimens in the gradient irradiation position. The dashed lines L21a, L21b show the subsequent irradiation periods given for L21 specimens, the higher fluence irradiation was performed first. n is the number of specimens in one irradiation set.
NEUTRON DOSIMETRY

The complicated irradiation history of the specimens requires careful evaluation of neutron fluences. Neutron fluence rate within one surveillance chain varies greatly in the axial direction as shown in Figure 2, and also slightly in the radial direction because the rotation and the loading angles of the two side by side laying specimens inside the vertical chain is not known. The relative neutron fluence of all individual specimens was measured with iron samples taken from specimen corners by means of the well known reaction $^{54}$Fe(n,p)$^{54}$Mn. The assessment of the shape of the neutron energy spectrum was based on spectral calculations performed with computer code ANISN and on experimental measurements using Nb, Fe, Ni, Ti, Cu, $^{238}$U and Co activation detectors. The gradual change from the initial fuel loading pattern to low-leakage core was found to have only minor influence on the neutron spectrum. The accuracy of the specific activity measurements is estimated to be 1 - 3 %. Larger systematic uncertainties to the fluence values arise from cross-section data, approx. 7 - 15 %, and from the shape of the neutron spectrum, 5 - 20 %. However, the systematic uncertainties do not bias the conclusion because all the specimens were irradiated in identical surveillance positions. The variation of fluence rate during one irradiation cycle was estimated with the computer code PREVIEW. A correction of -5 % to fluence values at surveillance chain end positions was attributed.

DATA ANALYSING METHODS

Charpy-V data

Charpy-V transition curves are approximated by a hyperbolic tangent function

$$E = 1/2 \times B + 1/2 \times \text{TANH} \left( \frac{(T-T_o)}{C} \right)$$

(1)

where

E  impact energy
T  test temperature
B  upper shelf energy
$T_o$ and C  fitting parameters

Lower shelf energy is always approximated to zero and only the impact energy values are analysed in this paper. In addition to the least square fitting criterion also orthogonal fitting was made for some transition curves. In the orthogonal fitting the sum of the squares of the orthogonal distances between the measured points and the fitted curve is minimised. In the later fitting the same weight was used for the energy value given in joules as for the temperature value given in degrees centigrade.

Fracture toughness data

J-integral values are calculated according to ASTM E 813-81. Hence the crack growth correction given in this standard is included in the measured values. The critical J values
corresponding to the onset of cleavage fracture are transformed to K-parameter values according to (2) irrespective of temperature.

\[ K_{Jc} = 15 \times \text{SQR}(J_c) \]  
(2)

All brittle fracture values are included in the data irrespective of the amount of ductile crack growth preceding the brittle fracture.

Fracture toughness data are analysed by using a functional form proved to be universal for a wide range of steels [2]. The mean value of \( K_{Jc} \) as a function of temperature for Charpy-size specimens is given by (3) and the 5% and 95% probability curves by equations (4) and (5). The parameter \( T_0 \) gives the transition temperature at 100 MPa\(\sqrt{m} \) toughness level for 25 mm specimen size.

\[ K_{i}(T) = 88.5 \times \exp[0.019 \times (T-T_0)] + 33 \]  
(3)
\[ K_{5\%}(T) = 46 \times \exp[0.019 \times (T-T_0)] + 26.7 \]  
(4)
\[ K_{95\%}(T) = 127.6 \times \exp[0.019 \times (T-T_0)] + 38.4 \]  
(5)

Fluence dependence of the transition temperature is described by

\[ T_0 = T_{0,\text{unirr}} + A \times [\text{fluence} \times 10^{-18}]^m \]  
(6)

where

- \( T_{0,\text{unirr}} \): \( T_0 \) parameter defined by unirradiated specimens
- \( A, m \): parameters describing irradiation embrittlement

All the measured \( K_{Jc} \) values are drawn on a single transition curve. The measured values for irradiated specimens are transformed to zero fluence by relation (6).

The following fitting criteria are applied when the parameters describing the mean transition curve (3) are defined. Criteria (7) is applied when one parameter has to be assessed, and criteria (8) in case of two parameters. The scatter of the mean curve is proportional to the term given in the denominator of the formulas.

\[ \sum_{i} \frac{K_i(T_i) - K_i(T_0)}{|K_i(T_i) - K_{\text{min}}|} = 0 \]  
(7)
\[ \sum_{i} \frac{|K_i(T_i) - K_i(T_0)|}{|K_i(T_i) - K_{\text{min}}|} = \text{minimum} \]  
(8)
RESULTS

Charpy-V tests

Base-1 specimens were irradiated first with high fluence rate to a medium or to a high fluence value. The irradiation of two high fluence capsules was further continued one or six years in low fluence rate. The upper shelf energies of base-1 specimens are shown in Figure 6. The function given in Figure 6 is used for defining the upper shelf energy for individual irradiation sets. Figure 7 shows the fitted transition curves for each irradiation set with free variation of parameters $T_0$ and $C$. Transition curve parameters are given in Table III. The $T_{42}$-transition temperature as a function of fluence is shown in Figure 8.

![Base-1 Energy vs Fluence](image)

$\Delta E = A\left[\phi/10^{18}\right]^n$

$n = 0.252 \quad A = -15.71$

Figure 6. Upper shelf impact energy of base-1 specimens. Test temperatures varied from 45 °C to 160 °C.
Figure 7. Charpy-V transition curves for material base-1.

Table III. Charpy-V transition curve parameters for base-1 material. The number of specimens in each transition curve is 12.

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Total fluence</th>
<th>$T_{42F}$ °C</th>
<th>B</th>
<th>C °C</th>
<th>$T_0$ °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref</td>
<td>-</td>
<td>-38</td>
<td>200</td>
<td>40</td>
<td>-12</td>
</tr>
<tr>
<td>L11</td>
<td>4.08</td>
<td>30</td>
<td>160</td>
<td>32</td>
<td>47</td>
</tr>
<tr>
<td>L12</td>
<td>13.8</td>
<td>74</td>
<td>145</td>
<td>35</td>
<td>90</td>
</tr>
<tr>
<td>L13</td>
<td>13.6</td>
<td>65</td>
<td>145.5</td>
<td>37.6</td>
<td>82</td>
</tr>
<tr>
<td>L14</td>
<td>17.2</td>
<td>101</td>
<td>142</td>
<td>30</td>
<td>115</td>
</tr>
<tr>
<td>L15</td>
<td>13.5</td>
<td>77</td>
<td>145.5</td>
<td>22.6</td>
<td>88</td>
</tr>
<tr>
<td>L16</td>
<td>14.5</td>
<td>66</td>
<td>144.6</td>
<td>43.6</td>
<td>86</td>
</tr>
</tbody>
</table>
Figure 8. Charpy-V transition temperature as a function of fluence for base-1 material.

Base-2 specimens were exposed to a moderate fluence and hence the measured transition temperature shifts are also relatively small. At the same time the scatter in the irradiated specimen values is somewhat higher than with base-1 material, which leads to a less accurate transition temperature estimation. The following procedure was followed when estimating the transition temperatures. Base-1 upper shelf energy values are scaled to base-2 upper shelf energy values with reference specimens. Upper shelf energy dependence on fluence is estimated by all the upper shelf energy values as shown in Figure 9. This function is used for calculating upper shelf energy parameter B for each set of specimens. Two kinds of curve fittings are made for the irradiation sets. First the fitting parameter C is assumed to be constant and secondly an orthogonal fitting is made. Transition curves from the first estimation with the measured impact values are shown in Figure 10. Transition curve parameters are given in Table IV. Fluence dependence of transition temperatures from the first estimation are shown in Figure 11 and from the second estimation in Figure 12.
Figure 9. Upper shelf energy of base-2 material as a function of fluence.

Figure 10. Charpy-V transition curves for base-2 material. Parameter C is fixed.
Table IV. Charpy-V transition curve parameters for base-2 material.

<table>
<thead>
<tr>
<th>Capsule</th>
<th>Total fluence (10^{19} \text{n/cm}^2, \text{E &gt; 1 MeV})</th>
<th>BJ</th>
<th>Conventional fitting</th>
<th>Orthogonal fitting</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>(T_{421}) (^\circ\text{C})</td>
<td>(C) (^\circ\text{C})</td>
</tr>
<tr>
<td>Ref</td>
<td>-</td>
<td>190</td>
<td>-36</td>
<td>24.5</td>
</tr>
<tr>
<td>L21</td>
<td>7.7</td>
<td>146</td>
<td>31</td>
<td>24.8</td>
</tr>
<tr>
<td>L22</td>
<td>4.1</td>
<td>155</td>
<td>2</td>
<td>24.8</td>
</tr>
<tr>
<td>L23</td>
<td>3.1</td>
<td>159</td>
<td>14</td>
<td>24.8</td>
</tr>
<tr>
<td>L24</td>
<td>3.4</td>
<td>158</td>
<td>4</td>
<td>24.8</td>
</tr>
<tr>
<td>L25</td>
<td>1.7</td>
<td>165</td>
<td>-2</td>
<td>24.8</td>
</tr>
<tr>
<td>L26</td>
<td>2.3</td>
<td>162</td>
<td>18</td>
<td>24.8</td>
</tr>
</tbody>
</table>

![Graph showing Charpy-V transition temperature as a function of fluence. Parameter C is fixed.](image)

\[ \Delta T_{42J} = A \cdot \left( \frac{\phi}{10^{18}} \right)^n \]

\(n = 0.342, A = 14.1\)

Figure 11. Charpy-V transition temperature as a function of fluence. Parameter C is fixed.
Figure 12. Charpy-V transition temperature as a function of fluence when orthogonal fitting is applied.

Fracture Toughness Tests

Base-1

Base-1 test results can be divided into subgroups by different criteria. In Figure 13 the data are grouped according to specimen preparation history. The first group, surveillance specimens, were tested but not prepared in our laboratory. The second group, the reconstituted specimens, were both prepared from broken Charpy-V specimen halves and tested by us. In Figure 14 the test values are divided into rather homogeneous groups according to irradiation history and fluence.

Figure 13. Test results of base-1 material graded according to specimen history, original surveillance specimens and afterwards reconstituted specimens.
Figure 14. Test results of base-1 material graded according to irradiation history and fluence.

Base-2

Precracked base-2 specimens fall into three groups on the basis of the irradiation history. Irradiation capsules L23, 24, 25 and 26 have only experienced low fluence rate irradiation and the total fluence of the specimens remains low. Capsule L22 was irradiated one year in high fluence rate only. The set L21 was exposed to high fluence rate one year and to subsequent low fluence rate 6 years. All the specimens irradiated in a lower fluence rate than $5 \times 10^{11}$ n/cm$^2$s, $E > 1$ MeV, were included in the curve fitting. The base-2 test results graded as described above are shown in Figure 15.

Figure 15. Fracture toughness transition curve for Charpy-size pre-cracked base-2 specimens. Only specimens irradiated in a lower fluence rate than $5 \times 10^{11}$ n/cm$^2$s, $E > 1$ MeV are used in fitting. Other points are only transferred to the estimated curve.
Irradiation embrittlement as a function of fluence

Irradiation embrittlement trend curves from the mean curve fittings are compared to the current Russian Norm [3]. Base-1 trend curves are given in Figure 16 and base-2 trend curves in Figure 17.

Figure 16. Base-1 mean trend curves and the norm trend curve.

Figure 17. Base-2 mean trend curves and the norm curve.
DISCUSSION

The effect of fluence rate on irradiation embrittlement is not straightforward. Besides the embrittling mechanism itself the response depends also on the detailed material characteristics especially on the impurity content and dislocation structure. Basically, diffusing vacancies and interstitial atoms create the microstructural changes in the material but effective density of these elements will be a complicated function of the source strength i.e. fluence rate and of recombination or annihilation strengths in different impurity structures.

In the western pressure vessel steels copper precipitation has been identified as one of the main embrittling mechanisms. General tendency in these steels is that in low fluence rate in low fluences the embrittlement sensitivity is higher and in high fluence rate in high fluences lower than with medium fluence rate and fluence values. This tendency has been found in mechanistic studies using microhardness and tensile tests in wide variety of model alloys and pressure vessel steels [4]. Also some studies using Charpy-V impact tests and fracture mechanics tests showing the same tendency are available [5]. However, the medium values of fluence rate and fluence parameters, where no fluence rate dependence is observed, depend on the material conditions.

The embrittling mechanism in the Russian CrMoV-pressure vessel steels is connected to copper and phosphorus contents, and due to often high phosphorus content in weld metal the main response comes from phosphorus. However, the micromechanism in these steels has not been identified properly. The fluence rate has been reported to have a considerable effect on embrittlement sensitivity in a weld metal [6]. Charpy-V tests were used in the experiments and the fluence rate values are approximately the same as in this study, when the different cut-off energy used in fluence estimation is taken into account.

In the present study different fluence and fluence rate combination were applied in the irradiations. In Charpy-V tests performed with base-1 material the effect of a subsequent low fluence rate irradiation after a high fluence rate irradiation to a fluence value of approx. 14 x 10¹⁹ n/cm², E > 1 MeV is found out. The irradiation times in the subsequent low fluence rate irradiations were one (L16) and six years (L14) reactor cycles. The upper shelf energy, Figure 6, and transition temperature shift, Figure 8, of L14 specimens fit very well to the high fluence rate trend curve. The upper shelf energy for L14 specimens is lower and the transition temperature shift higher than the trend curve behaviour suggesting that a small additional embrittling component is caused by a long time low fluence irradiation.

Charpy-V specimens of base-2 material have irradiation parameter combinations, where only fluence rate is changed and one combination of high fluence rate followed by low fluence irradiation. The trend curve given in Figure 12 does not suggest any fluence rate dependent effects. Due to the higher scatter in the impact values and to the small transition temperature shifts the uncertainty in the trend curve is higher than for base-1 material.
The data of fracture mechanical tests performed with base-1 material are shown in Figures 13 and 14. It is clear that the specimen history is a strong variable and it is further discussed in [7]. Fluence versus fluence rate parameter range, described in Figure 4, is divided into four groups in Figure 14. These four groups do not show any bias according to fluence of fluence rate. The conclusion is that the large scatter in surveillance specimens does not depend on specimen fluence or fluence rate.

Fracture mechanics specimen of base-2 material have experienced three kinds of irradiation histories, low fluence rate only, high fluence rate only, and high fluence rate followed by 6 years in low fluence rate. In Figure 15 the low fluence rate specimens and high fluence rate specimens form a rather homogeneous group. Data from the third group, when only high fluence values are included, fall below the mean curve even if the points are well within the total scatter bands. This suggests that a small additional component to embrittlement is caused by long time low fluence irradiation.

CONCLUSIONS

Irradiation sensitivity of two forging materials was measured with Charpy-V and fracture mechanics tests.

Irradiation sensitivity of the materials was found to be less or equal to the current Russian standard. Only at high fluence the value given in the standard was slightly exceeded. For material base-1 the behaviour was measured up to a fluence value of $17 \times 10^{19}$ n/cm$^2$, $E > 1$ MeV. With this material Charpy-V impact test gives the same irradiation sensitivity as fracture mechanics specimens.

For material base-2 the measured irradiation sensitivities were lower than given in the standard. Charpy-V test gives somewhat lower irradiation sensitivity than fracture mechanics specimens.

The data was analysed in order to find out any fluence rate dependencies in the irradiation sensitivity. Irradiation sensitivity is well described by the fluence parameter only. A slight additional effect on embrittlement from a long term low fluence irradiation is noticed but even this effect is within the total scatter band of the data.

Many of the VVER-440 power units have resorted to fluence embrittlement mitigation by fluence rate reduction measures. These measures can lead to complicated fluence versus fluence rate histories in the pressure vessel. However, in the parameter range applied in the present study the fluence parameter only is needed to describe embrittlement sensitivity of the forging material. This is in contradiction to some data presented for VVER-440 weld material.
REFERENCES


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SESSION C

SURVEILLANCE
ANALYSIS AND RESULTS FROM STANDARD SURVEILLANCE PROGRAMMES
OF WVER 440/V-213C REACTOR PRESSURE VESSELS

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ANALYSIS AND RESULTS FROM STANDARD SURVEILLANCE PROGRAMMES
OF WWER 440/V-213C REACTOR PRESSURE VESSELS

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ABSTRACT

In Czech and Slovak republics there are six units of WWER 440/C type of reactors (PWR type) that are completed by surveillance specimens programmes. The sets of specimens are determined for static tensile testing, impact notch toughness testing and fracture toughness determination. They are supplemented by necessary sets of neutron fluence indicators and irradiation temperature monitors.

Results of mechanical properties of these specimens after irradiation in interval between one and five years of operation are summarized and discussed with respect to the effect of:
- individual heats and welding joints,
- radiation embrittlement,
- annealing recovery.

KEY WORDS: pressure vessel steels, radiation embrittlement, transition temperature shifts, WWER 440/V-213C, surveillance programmes, annealing recovery
INTRODUCTION

Surveillance specimens programmes from the reactor pressure vessels are one of the most important parts of the in-service quality assurance programmes that are necessary for realistic and reliable assessment of reactor pressure vessel lifetime.

Structure, volume, type of specimens, withdrawal schedule and testing procedures depend first of all on time where the reactor was designed and also on volume and other possibilities of reactor pressure vessel construction.

In the case of WWER 440/V-213c reactors, six sets of surveillance specimens are placed into each pressure vessel. The set contains the specimens for study of radiation embrittlement after from one to five years of operation as well as study of thermal ageing and recovery annealing.

DESCRIPTION OF SURVEILLANCE PROGRAMMES

Test materials

Reactor pressure vessels for WWER 440/C reactors have been manufactured in ŠKODA Concern, Power Machinery Plant from the following materials:
- base metals: Steel 15Kh2MFA (Cr-Mo-V steel)
  Steel 15Kh2MFAA (Cr-Mo-V type with supplementary requirement on content of P,Cu,As,Sb,Sn) is used for active core cylindrical rings
- weld metals: submerged-arc weld wire Sv 10KhMFTA (Cr-Mo-V type)

Whole pressure vessels are manufactured from forgings-rings and from forged plates used for covers. Rings are welded together into sections that are stress relieved intermediary. At the end the whole pressure vessel is stress relieved by final heat treatment regime.

Test specimens from base materials are manufactured from upper part of cylindrical ring that is used for the centre part of pressure vessel in front of active core. Test specimens from weld metal and welding joints are made from surveillance welding joint that is manufactured by the same technology and using the same welding wires and flux as for the welding joint situated in the lower part of active core.
Test specimens

Static tensile (3 mm, gauge length 30mm), Charpy V-notch impact and COD (10x10x55 mm) specimens for fracture toughness determination (10 mm thick, fatigue precracked with side grooves) are provided for unirradiated and irradiated tests.

For the studies, the sets of 12 specimens are designed for determination of one curve (transition temperature dependence) of Charpy impact and fracture toughness specimens. For static tensile tests six specimens are included (three for room temperature and three for tests at operation temperature-265 °C).

The specimens were cut from the centre part of material thickness not closer than 1/4 of thickness to pressure vessel surface.

Capsule assemblies and removal schedule

The test specimens are placed into capsules made from austenitic stainless steel that prevents corrosion effects on specimens. Each capsule contains 6 tensile specimens or two Charpy-impact or COD type specimens. Some of them also contain neutron flux and irradiation temperature monitors. Capsules are situated into chains - each contain 19 or 20 capsules in active core position. Two chains also include another 13 capsules that are located in upper part of reactor pressure vessel, out of neutron field. These specimens are determined for thermal ageing effect study.

Two chains represent one set of the specimens - six sets of chains are placed in each pressure vessel. Removal schedule is given by operation schedule of reactor, i.e. by fuel elements changing. Interval between these changes is approximately one year:
- one year of operation - SET 1
- two years of operation - SET 6
- three years of operation - SET 3
- five years of operation - SET 5
- six years of operation - SET 1 (for recovery annealing study)

Neutron dosimetry

All six reactors are practically identical from the point of view of their neutron fluxes on surveillance positions as well as on reactor pressure vessel surfaces (inner and outer). Thus, results form individual reactors can be compared and also put into larger set of results.

Relatively high lead factor has been observed and approved for this type of reactors. It means, one year of operation provides such an irradiation damage in surveillance specimens as approximately 10 years of operation on inner pressure vessel
surface. After five years of operation, results from surveillance specimens represent full designed lifetime of reactor (40 years). One additional set of specimens that is removed after 6 years of operation is determined for annealing recovery experiments.

Design of chains (length of chain) of capsules has been shown as not practical because only central part of chains is located in neutron constant field. Other parts received quite different fluences that do not enable to make test from one chain, especially in the case of specimens for fracture toughness determination. The specimens in one chain received fluences that are different more than 20-times.

**SPECIMENS TESTING**

Specimens testing is carried out in Nuclear Research Institute in hot and semi-hot sell laboratories Tensile. Charpy impact and fracture toughness test are carried out following Czech material standards (CSN) that are very close to ASTM codes.

Tensile and fracture toughness tests are performed by INSTRON type servo-hydraulic testing machine.

Charpy impact energy levels are measured using instrumented universal impact tester Tinius Olsen Model 74 with velocity of 5.6 m.s⁻¹ within the temperature range -160 to +200 °C.

Fracture toughness tests are carried out over the temperature range -190 to +200 °C. After pre-cracking, all of the TPB specimens were sidegrooved to the depth of 0.1 net thickness. Unloading compliance single specimen method is used in the whole temperature range. Load point displacement is measured using LVDT gauges.

Results from Charpy impact tests have been statistically evaluated using the mean square method and following equation:

\[ KCV = A + B \cdot \tanh \left( \frac{T - T_o}{C} \right) \]

In the case of fracture toughness (in transition region):

\[ K_C = A + B \cdot \exp \left( C \cdot T \right) \]
Transition temperatures were determined according to USSR Codes [1] and were named as Tko - criterion for this temperature is as follows:
- KCV = 49 Jcm⁻² at transition temperature,
- KCV = 74 Jcm⁻² at temperature Tko+30 °C,
- brittle fracture appearance is lower than 50% at temperature Tko+30 °C.

Transition temperature shifts are evaluated following [1], using the formula:

$$\Delta T_F = A_F \cdot (F \cdot 10^{-22})^{1/3}$$

where

$\Delta T_F$...transition temperature shift.

$A_F$...irradiation embrittlement coefficient.

$F$...neutron fluence with energies higher than 0.5 MeV.
EXPERIMENTAL RESULTS

Radiation embrittlement

Six reactor pressure vessels are characterized by six different heats of steel 15Kh2MFAA type. For four welding joints in four pressure vessels the same welding materials were used, thus only three different welding materials characterized six pressure vessels.

The chemical composition of materials is given in Table 1 and mean mechanical properties in unirradiated state in Table 2. There are not any large differences between individual heats of base as well as welding metals, especially between different welds made by the same welding material.

In Fig. 1 all results of Charpy impact transition shifts from base materials are given and statistically evaluated. Dashed line represent results that can be connected with heats that contain approximately 0.014 % P and 0.08 % Cu. Practically all values lie very close to this line but its exponent is equal to 0.49. For comparison maximum allowable results following [ 1 ], i.e. with \( \Delta F = 18 \), are also given (full line). All fluences are shown with energies higher than 0.5 MeV.

In Fig. 2 similar results for weld metals are given. The open marks represent welding metal No. 76 303 and its mean line has exponent equal to 0.59 and lies much lower than other ones. All other two welds can be taken as one set of results, its mean line is shifted to higher values (full marks). For comparison maximum allowable values according to [ 1 ], i.e. with \( \Delta F = 15 \), are also shown.

In both diagrams practical all results, with exclusion of few ones for higher fluences, are situated below these maximum allowable trend lines.

In Figures 3 and 4, typical transition curves are summarized for unirradiated and irradiated state of base metal (Fig. 3) and weld metal (Fig. 4).

Post-irradiation annealing

Two chains (one set) of the surveillance specimens have been removed after six years of operation. A standard part of these chains is determined for study of annealing recovery effect.

The specimens have been annealed under following conditions:

Annealing temperature : 475 – 480 °C
Time : 168 hours.
In Fig. 6.7.8 and 9, recovery of tensile properties (yield stress, tensile strength, ductility and contraction) after annealing are given for three heats of reactor pressure vessel steels. The changes of tensile test results after irradiation (5 years of operation) are compared with results obtained after annealing recovery. In the case of yield stress and tensile strength it has been obtained partial recovery of the properties. In the case of plastic properties, i.e. ductility and contraction, it can be observed nearly full recovery.

Recovery of transition temperature shifts and upper shelf energy (KCV_{max}) measured by Charpy impact tests is shown in Fig.10 and 11 for three heats of RPV steel. Results of Charpy impact tests after irradiation (5 years of operation) are compared with post-irradiation annealing results. It is clear from Fig.10, the temperature transition shifts has decreased after annealing recovery but it has not been reached full recovery. Plastic properties (KCV_{max}) have nearly full recovered.

DISCUSSION OF RESULTS

Transition temperature shifts

Analysis of results of transition temperature shifts of both materials shows to the fact that this type of steel is characterized by relatively high resistance against irradiation embrittlement. Even for very high fluences (2.5x10^{24}n.m^{-2}, E>0.5MeV) at irradiation temperature 265 °C, maximum temperature shift is about -100 °C. This shift represents situation at the end of design lifetime. In the case of results obtained after five years of operation with fluences about 4.5x10^{24}n.m^{-2} (E>0.5MeV), the transition temperature shifts represent situation at more than full reactor pressure vessel lifetime.

Comparing all results with proposed trend lines according to the USSR Code [1], all results are well below this line for both materials (with exclusion of a few for very high fluences - especially after 5 years of operation). But their fluence dependences are quite different. While [1] supposed that irradiation embrittlement law is characterized by exponent 1/3, experimental results show to somewhat larger value - for base as well as veld material between 0.49 and 0.59. The sets of results, especially of veld material, are not too large to be able to solve problem of this exponent value. But from Fig.1 and 2, it is clearly seen that exponent is closer to 1/2 than 1/3.
Post-irradiation annealing

In Fig. 6 to 11 there are summarized the results from annealing recovery studies. It is clear that tensile and Charpy impact properties have recovered better in the case of plastic properties (ductility, contraction, upper shelf energy) than for yield stress, tensile strength and transition temperature shifts.

CONCLUSIONS

Results from surveillance specimens programmes from reactor pressure vessels of six WWER 440/c units operated in Czech and Slovak republics have been shown and discussed. The most important conclusions that have been received are:
- there is a high gradient of neutron flux along surveillance chains,
- high lead factor between surveillance position and inner reactor pressure vessel surface (10.9 for E=0.5MeV) - five years of operation results represent more than full reactor pressure vessel lifetime,
- transition temperature shifts are well below trend lines proposed by USSR Code for base and weld material with exclusion of few results for high fluences that correspond to more than full designed lifetime of reactor pressure vessel,
- fluence dependence of transition temperature shifts is better illustrated by exponent of 1/2 than by 1/3, given in The USSR Code,
- after post-irradiation annealing, plastic properties have been nearly full recovered. In the case of tensile strength, yield stress and transition temperature shifts, it has been only reached partial recovery.
LITERATURE


LIST OF ILLUSTRATIONS

Table 1: Chemical composition of surveillance specimens (mass %)

Table 2: Room temperature mechanical properties of surveillance specimens in unirradiated state

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Fig. 2: Transition temperature shifts for weld metals

Fig. 3: Typical transition curves from impact testing of base material (Heat No. 41 230)

Fig. 4: Typical transition curves from impact testing of weld material (Heat No. 76 303)

Fig. 6: Recovery of yield stress for base metals of three reactor pressure vessels at 265 °C

Fig. 7: Recovery of tensile strength for base metals of three reactor pressure vessels at 265 °C

Fig. 8: Recovery of ductility A for base metals of three reactor pressure vessels at 265 °C

Fig. 9: Recovery of contraction Z for base metals of three reactor pressure vessels at 265 °C

Fig. 10: Recovery of transition temperature shifts for base metals of three reactor pressure vessels

Fig. 11: Recovery of KCV_{max} (upper shelf measured by Charpy impact test) for base materials of three reactor pressure vessels
Table 2: Room temperature properties of surveillance specimens

<table>
<thead>
<tr>
<th>Unit</th>
<th>Heat No.</th>
<th>( R_{p0.2} ) [MPa]</th>
<th>( R_m ) [MPa]</th>
<th>( A_s ) [%]</th>
<th>( Z ) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>BASE METALS</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3 EBO</td>
<td>27407</td>
<td>516</td>
<td>631</td>
<td>23.4</td>
<td>77.0</td>
</tr>
<tr>
<td>4 EBO</td>
<td>37191</td>
<td>555</td>
<td>668</td>
<td>21.0</td>
<td>74.0</td>
</tr>
<tr>
<td>1 EDU</td>
<td>28313</td>
<td>538</td>
<td>638</td>
<td>26.6</td>
<td>74.0</td>
</tr>
<tr>
<td>2 EDU</td>
<td>34589</td>
<td>564</td>
<td>675</td>
<td>20.0</td>
<td>74.5</td>
</tr>
<tr>
<td>3 EDU</td>
<td>41230</td>
<td>535</td>
<td>647</td>
<td>21.5</td>
<td>75.0</td>
</tr>
<tr>
<td>4 EDU</td>
<td>39655</td>
<td>504</td>
<td>637</td>
<td>22.3</td>
<td>76.0</td>
</tr>
<tr>
<td>WELD METALS</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3 EBO</td>
<td>80532</td>
<td>478</td>
<td>617</td>
<td>24.9</td>
<td>69.0</td>
</tr>
<tr>
<td>4 EBO</td>
<td>76306</td>
<td>448</td>
<td>614</td>
<td>24.7</td>
<td>67.0</td>
</tr>
<tr>
<td>1 EDU</td>
<td>81797</td>
<td>468</td>
<td>624</td>
<td>25.8</td>
<td>69.8</td>
</tr>
<tr>
<td>2 EDU</td>
<td>76306</td>
<td>468</td>
<td>607</td>
<td>24.2</td>
<td>66.6</td>
</tr>
<tr>
<td>3 EDU</td>
<td>76306</td>
<td>451</td>
<td>604</td>
<td>23.0</td>
<td>67.0</td>
</tr>
<tr>
<td>4 EDU</td>
<td>76306</td>
<td>451</td>
<td>604</td>
<td>25.3</td>
<td>68.0</td>
</tr>
</tbody>
</table>
Transition shifts $T_k$ [deg.C]

Fig. 1: Transition temperature shifts

Base metals:

Fluence $[10^{22} \text{ n.m}^{-2}, E > 0.5 \text{ MeV}]$

$n = 0.486$

$0.014P + 0.08Cu$

$A_f = 18$
Fig. 2: Transition temperature shifts

Weld Metals:

Fluence ($10^{17}$ n$_{eq}$/cm$^2$, $E < 0.5$ MeV)
Fig. 3: Transition curves from impact testing of base metal (Heat No. 41230)

- Unirradiated
- After 1 year
- After 3 years
- After 6 years

Fig. 4: Transition curves from impact testing of weld metal (Heat No. 76306)

- Unirradiated
- After 1 year
- After 3 years
- After 6 years
Fig. 6: Recovery of yield stress for base metals at 265 deg.C
- Irradiated 5 years
- Annealing recovery

Delta Rp0.2 [MPa]

Heat No. 27407  Heat No. 37191  Heat No. 28313

Fig. 7: Recovery of tensile strength for base metals at 265 deg.C
- Irradiated 5 years
- Annealing recovery

Delta Rm [MPa]

Heat No. 27407  Heat No. 37191  Heat No. 28313
Fig. 8: Recovery of ductility \( A \)
for base metals at 265 deg.C
- Irradiated
- 5 years
- Annealing recovery

Delta \( A \) [%]

Heat No. 27407  Heat No. 37191  Heat No. 28313

Fig. 9: Recovery of contraction \( Z \)
for base metals at 265 deg.C
- Irradiated
- 5 years
- Annealing recovery

Delta \( Z \) [%]

Heat No. 27407  Heat No. 37191  Heat No. 28313
THE RESULTS OF THE SURVEILLANCE SPECIMEN PROGRAM PERFORMED IN THE RPVs NPP V-2 IN JASLOVSKÉ BOHUNICE

by
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2Division of Nondestructive Testing, Nuclear Power Plant (EBO) Jaslovenské Bohunice, 91931 Jaslovenské Bohunice, Slovak Republic
3Slovak Energetic Enterprise, Hranicná 12, 827 36 Bratislava, Slovak Republic

SUMMARY

The RPVs VVER-440 V-213 type are equipped with the surveillance specimen programme, which is used for the Reactor Vessel Material Embrittlement evaluation. Because of high "lead factor" the fluence of the irradiated specimens is in present time on the planned end of life both RPVs.

The topic of the paper are:
- the basic description of the present status of the Bohunice NPP Unit 3 and 4 RPVs embrittlement assessment
- the surveillance specimen program results
- neutron exposure measurements
- RPV lifetime evaluation.

Except items mentioned above in the paper are discussed the possibilities of improving existing surveillance specimen programme, too.

Key words: irradiation embrittlement, reactor pressure vessel, surveillance specimen, lifetime evaluation, chemical composition, 15CH2MFA steel, neutron fluence
2. MATERIAL SPECIFICATIONS

2.1. THE MECHANICAL PROPERTIES OF THE RPVs MATERIAL

For the reactor pressure vessel of VVER 440/213 type manufacturing was chromium-molybdenium-vanadium steel 15CH2MFA used as a base metal and for the weld joints was 10CHMFT steel used. The welding procedure under flux was used, the type of the flux was AN-42A. The basic mechanical properties are presented in the table 1 for base and weld metal.

Table # 1. Mechanical properties of the RPVs base and weld metal

<table>
<thead>
<tr>
<th>Prop.</th>
<th>test temp. [°C]</th>
<th>Mean values were obtained from the four testing samples series</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Unit 3 BM</td>
</tr>
<tr>
<td>Rp0.2</td>
<td>20</td>
<td>516</td>
</tr>
<tr>
<td>Rm</td>
<td>20</td>
<td>631</td>
</tr>
<tr>
<td>A5</td>
<td>20</td>
<td>23.4</td>
</tr>
<tr>
<td>Z</td>
<td>20</td>
<td>76.5</td>
</tr>
<tr>
<td>KCV</td>
<td>20</td>
<td>191</td>
</tr>
<tr>
<td>Rp0.2</td>
<td>350</td>
<td>464</td>
</tr>
<tr>
<td>Rm</td>
<td>350</td>
<td>527</td>
</tr>
<tr>
<td>A5</td>
<td>350</td>
<td>17.1</td>
</tr>
<tr>
<td>Z</td>
<td>350</td>
<td>74.2</td>
</tr>
<tr>
<td>TK0</td>
<td>-50±±70</td>
<td>54.5</td>
</tr>
</tbody>
</table>

* - (50J/cm²) criterion

2.2. CHEMICAL COMPOSITION OF THE BASE AND WELD METAL

The chemical composition has an important role in the mechanical properties changes of the RPV steel due to the irradiation. Especially, Cu and P contents must be limited as low as possible.

In the table 2 are presented the chemical composition of the base and weld metal of unit 3 and 4 RPVs.
Table # 2. Chemical composition of the RPVs base and weld metal

<table>
<thead>
<tr>
<th>UNIT#</th>
<th>METAL</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>S</th>
<th>P</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>V</th>
<th>As</th>
<th>Co</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>base</td>
<td>0.14</td>
<td>0.21</td>
<td>0.49</td>
<td>0.016</td>
<td>0.013</td>
<td>2.81</td>
<td>0.06</td>
<td>0.6</td>
<td>0.33</td>
<td>0.007</td>
<td>0.009</td>
<td>0.07</td>
</tr>
<tr>
<td></td>
<td>weld</td>
<td>0.044</td>
<td>0.61</td>
<td>1.16</td>
<td>0.016</td>
<td>0.017</td>
<td>1.44</td>
<td>-</td>
<td>0.46</td>
<td>0.26</td>
<td>-</td>
<td>0.004</td>
<td>0.10</td>
</tr>
<tr>
<td>4</td>
<td>base</td>
<td>0.16</td>
<td>0.24</td>
<td>0.41</td>
<td>0.016</td>
<td>0.015</td>
<td>2.86</td>
<td>0.06</td>
<td>0.70</td>
<td>0.32</td>
<td>0.008</td>
<td>0.008</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>weld</td>
<td>0.031</td>
<td>0.6</td>
<td>1.13</td>
<td>0.015</td>
<td>0.010</td>
<td>1.31</td>
<td>0.05</td>
<td>0.52</td>
<td>0.19</td>
<td>-</td>
<td>0.006</td>
<td>0.06</td>
</tr>
</tbody>
</table>

3. MEASUREMENTS AND CALCULATIONS

3.1. NEUTRON FLUX MONITORING AND CALCULATIONS

For the neutron fluence monitoring are used Fe, Cu, Nb and Co foils detectors placed in the irradiation capsules. The neutron fluence was calculated for neutron energy $E_n > 0.5$ MeV and $E_n > 0.1$ MeV. The neutron detectors are located in capsules #1, 3, 7, 11, 15 and 19 along the core axis in both reactors. The value of the neutron fluence depends on the position of the detectors in the reactor, as is shown on the figure 1.

For the neutron fluence estimation of unit 3 and 4 RPVs are the neutron flux values from unit 1 used and then re-calculated according to effective days (in unit 1 are at outer RPV wall activation detectors used). This estimation gave us good results due to the same core design for all units (before dummy elements installation in the unit 1 and 2). The estimated neutron fluence values of unit 3 and 4 are shown in the table #3.
Table #3 Neutron fluence values of unit 3 and 4

<table>
<thead>
<tr>
<th>Campaign #</th>
<th>UNIT 3</th>
<th>UNIT 4</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Te</td>
<td>( \phi )</td>
</tr>
<tr>
<td>1</td>
<td>265.61</td>
<td>0.786</td>
</tr>
<tr>
<td>2</td>
<td>236.67</td>
<td>0.701</td>
</tr>
<tr>
<td>3</td>
<td>288.10</td>
<td>0.853</td>
</tr>
<tr>
<td>4</td>
<td>259.62</td>
<td>0.768</td>
</tr>
<tr>
<td>5</td>
<td>292.33</td>
<td>0.865</td>
</tr>
<tr>
<td>6</td>
<td>301.94</td>
<td>0.894</td>
</tr>
<tr>
<td>7</td>
<td>294.22</td>
<td>0.871</td>
</tr>
<tr>
<td>8</td>
<td>330.13</td>
<td>0.977</td>
</tr>
</tbody>
</table>

* accumulated values, for \( E > 0.5 \text{ MeV} \times 10^{23} 

3.2. DETECTOR ACCURACY

From the fig. 1 follows the high dependence of the neutron fluence distribution, according to the position of the activation foils in capsules along the core axis. It is caused by the core design. The accuracy of the neutron fluence estimation is \( \leq 20\% \). It is recommended to perform and independent neutron fluence monitoring in both units.

3.3. IRRADIATION TEMPERATURE MONITORING

An irradiation temperature is very important parameter for the irradiation embrittlement assessment. The irradiation temperature is determined by the diamond powder lattice parameters changes which is irradiated together with samples in irradiation capsules. But the results of these measurements were not satisfactory. It is because why are implemented the corrections
for "the real irradiation temperature" according to the neutron fluence by following formula [4]:

$$T_{IRR} = T_{MEASURED} - 8T$$

where the correction factor depends on the neutron fluence by formula:

$$8T = 9.3 \times 10^6 \cdot \phi^{-1/3}$$

where $\phi = \text{neutron fluence in \,[10^{16} \text{ nm}^{-2}]}$

The mean values of irradiation temperatures obtained from all temperature indicators are following:

unit 3 : $T_{irr} = 288 \pm 18 \, ^\circ C$
unit 4 : $T_{irr} = 272 \pm 19 \, ^\circ C$

3.4. FUEL LOADING CALCULATION OF REACTOR CORE

The fuel loading calculations of the unit 3 and 4 reactor cores are in Nuclear Power Plants Research Institute (VÚJE) performed according to LINDA, TOPER and BIPR-5 codes.

The calculations are performed in three stages:
- out-core fuel management
- in-core fuel management
- nuclear design, i.e. neutron - physics calculation of core.

The fuel sortiment is calculated for 5 campaigns ahead according to numbers, to optimize costs and fuel enrichment of the peripheral zone.

One month before planned outages the analysis of core design are performed prior to the end of the campaign.

Using by TOPER code, the fuel assembly distribution is optimized for next campaign core design. Usually, 24 assemblies after 3 years of operation are in peripheral zone used.

Neutron - physics characteristics are re-calculated using by a BIPR-5 code.
3.5. INFLUENCE OF FUEL LOADING ON THE VALUE OF RPV NEUTRON FLUENCE EXPOSURE

The neutron flux density on RPV wall depends on the assemblies power in the peripheral zone.

The fuel loadings which are used nowadays at unit 3 and 4 caused decreasing of neutron flux density on RPV wall about 25% with compare to a standard fuel loading.

4. SURVEILLANCE SPECIMEN PROGRAMME PERFORMANCE

4.1. MATERIAL OF THE SURVEILLANCE SPECIMENS

The testing samples for the surveillance specimen programme were manufactured by the RPVs manufacturer ŠKODA Plzeň. Surveillance specimens were prepared from the base metal (steel 15Kh2MFA) core region forging for unit 3 RPV from melt # 27408M and for unit 4 RPV from melt # 37191.

Samples of weld metal and heat affected zone were made from so called R-weld, which was prepared from the control plates of the same chemical composition steel as core region ring and using by the same technology and welding procedure of 0.1.4 (5/6) RPV weld. As a weld wire was used steel 10CHMFT. The melt numbers of unit 3 RPV are # 80532 and for unit 4 RPV # 76306.

4.2. THE NUMBER AND TYPES OF SURVEILLANCE SPECIMENS

The type and number of surveillance specimens is shown in the table # 4. Each RPV is furnished by six "twin chains" which consists from the irradiation capsules arranged in the core axis direction. The upper part of the chains # 1 and 4, are located in the position of lower neutron flux (in the RPV nozzles region). The chains # 2, 3, 5 and 6, are located in the core region. Except these samples, there was prepared the set of samples for the initial conditions properties testing - so called zero condition material properties (table # 5). The type and number of these testing samples is sufficient for the monitoring the changes of material properties after irradiation.
### Table # 4. The types and number of surveillance specimens

<table>
<thead>
<tr>
<th>Chain number</th>
<th>Base metal CH COD ten.</th>
<th>Weld metal CH COD ten.</th>
<th>HAZ CH COD ten.</th>
<th>Indicators T n-flux</th>
</tr>
</thead>
<tbody>
<tr>
<td>1G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>1G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 9</td>
</tr>
<tr>
<td>2G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 9</td>
</tr>
<tr>
<td>2G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 9</td>
</tr>
<tr>
<td>3G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 9</td>
</tr>
<tr>
<td>3G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 9</td>
</tr>
<tr>
<td>4G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>4G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>5G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>5G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>6G1</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td>6G2</td>
<td>12 12 6</td>
<td>6 6 6</td>
<td>12 12 6</td>
<td>3 6</td>
</tr>
<tr>
<td><strong>Summary</strong></td>
<td><strong>72 72 36</strong></td>
<td><strong>72 72 36</strong></td>
<td><strong>72 72 36</strong></td>
<td><strong>36 60</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Specimens located above the reactor core region</th>
</tr>
</thead>
<tbody>
<tr>
<td>1G1</td>
</tr>
<tr>
<td>1G2</td>
</tr>
<tr>
<td>4G1</td>
</tr>
<tr>
<td>4G2</td>
</tr>
<tr>
<td><strong>All</strong></td>
</tr>
<tr>
<td><strong>Summary</strong></td>
</tr>
</tbody>
</table>

### Table # 5. The types and number of surveillance specimens used for the initial conditions properties testing

<table>
<thead>
<tr>
<th>Specimen type</th>
<th>Material</th>
<th>Number of spec.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charpy tension</td>
<td>BM</td>
<td>2 x 18</td>
</tr>
<tr>
<td>COD</td>
<td>BM</td>
<td>2 x 15</td>
</tr>
<tr>
<td></td>
<td>BM</td>
<td>2 x 6</td>
</tr>
<tr>
<td>Charpy tension</td>
<td>VM</td>
<td>2 x 18</td>
</tr>
<tr>
<td>COD</td>
<td>VM</td>
<td>2 x 15</td>
</tr>
<tr>
<td></td>
<td>VM</td>
<td>2 x 6</td>
</tr>
<tr>
<td>Charpy tension</td>
<td>HAZ</td>
<td>2 x 18</td>
</tr>
<tr>
<td>COD</td>
<td>HAZ</td>
<td>2 x 15</td>
</tr>
<tr>
<td></td>
<td>HAZ</td>
<td>2 x 6</td>
</tr>
</tbody>
</table>

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4.3. THE SYSTEM OF SURVEILLANCE SPECIMEN TESTING

According to the quality assurance programme of the RPV, there was prescribed the schedule for the surveillance specimens withdrawal. The irradiated chains withdrawal schedule is shown in the table # 6.

Table # 6. The chains withdrawal schedule

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>Start</td>
<td>2G1</td>
<td>6G1</td>
<td>3G1</td>
<td>-</td>
<td>5G1</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>irradi</td>
<td>1G2</td>
<td>6G2</td>
<td>3G2</td>
<td></td>
<td>5G2</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>-</td>
<td>Start</td>
<td>2G1</td>
<td>6G1</td>
<td>3G1</td>
<td>-</td>
<td>5G1</td>
</tr>
<tr>
<td></td>
<td>irradi</td>
<td>2G2</td>
<td>6G2</td>
<td>3G2</td>
<td></td>
<td>5G2</td>
<td></td>
</tr>
</tbody>
</table>

4.4. COMPARISON WITH DESIGNED LIFETIME

According to the results of the surveillance specimen programme after 5 years of irradiation, there are not limits for the operation during RPVs designed lifetime.

4.5. RESULTS OF THE SURVEILLANCE PROGRAMME PERFORMANCE

At the both units were completed material test results after five years of irradiation. The changes in $R_{p0,2}$ are shown in figures # 2 and 3. The shift of $T_{KF}$ values for the unit 3 and 4 is presented in figures 4 to 11. $T_{KF}$ shift was calculated according to the energy criterion (50 Jcm$^{-2}$).

From these values the trends of $T_{KF}$ shift were predicted for postulated time (years) of operation for both units.

As it is shown in figures #12 and 13, the $T_{KF}$ shift for both units for base and weld metals, is acceptable for designed lifetime.
5. CONCLUSIONS

From the calculations, and experimental results follows:
- the chemical composition of the both RPVs V-213 type is from the point of view the Cu and P contents satisfactory
- the neutron fluence monitoring and calculations needs some improvements because of the problems with the irradiation capsules orientations to the reactor core
- the irradiation temperature monitoring is not satisfactory and needs substantial improvement based on the new system of temperature measurement with the melting monitors or thermocouples
- the improved fuel loading scheme give us benefit through decreasing of the neutron fluence impinging on the RPV wall
- from the surveillance specimen programmes for the both RPV V-213 follows, that there are not expected the limits for the operation during planned operation lifetime.

6. REFERENCES

[1] Beňová H., Beňo P., Kupča L.: Comparison of the surveillance specimen programme results from the RPVs Unit-3 and 4 NPP EBO after five years of irradiation, Report VÚJE #360/34/92, december 1992
[4] NTD Interatomenergo: The material properties testing of the base metals, weld metals and austenitic cladding, Moscow, November 1986

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Fig. #5: Shift of $T_{kf}$ for Unit EBO4  
After 5 Years of Irradiation

Fig. #6: Shift of $T_{kf(BM)}$ for RPV EBO3  
KCV Criterion 5 Years of Irradiation
Fig. #7: Shift of Tkf(WM) for RPV EBO3
KCV Criterion 5 Years of Irradiation

Fig. #8: Shift of Tkf(HAZ) for RPV EBO3
KCV Criterion 5 Years of Irradiation
Fig. #11: Shift of Tkf(HAZ) for RPV EBO4
KCV Criterion 5 Years of Irradiation

Fig. #12: Tkf-BM trends for Units 3 & 4
Inner Surface in Maximum n-Flux
Non Typical Results of the first and second Irradiation Set of the Surveillance Programme of a Boiling Water Reactor in Switzerland

by

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Surveillance Programme, Automatic Weldment, Copper Content

In the surveillance programme for the pressure vessel of the nuclear power station Mühlberg, Switzerland, tensile- and impact tests on the base material (ASTM A 508 Cl 2) and two weldments (main and automatic, respectively) as well as the heat affected zone between base material and weld material was performed.

Examinations of the first irradiation set with a neutron fluence of about $5.5 \cdot 10^{17}$ nvt ($E > 1$ MeV) revealed shifts of $\Delta NDT \leq 33$ K at an energy level of 50 ft lbs.

The automatic weld (with a copper content of about 0.3 wt %) however exhibited a shift of 81 K.

Within the second irradiation set, with a neutron fluence of about $1.1 \cdot 10^{18}$ nvt ($E > 1$ MeV), the shifts are increased to values of $\Delta NDT \leq 44$ K. An exception was found in the automatic weld without any further shift as compared to the first irradiation set.

The possibilities of this discrepancy is outlined within this contribution and is topic of discussion.
1. Introduction

The surveillance program for the determination of the mechanical properties of the reactor pressure vessel material as a function of the neutron dose is provided for impact- and tensile tests.

According to the specification for the performance of the post irradiation investigations the tests had to be undertaken on

- the base material
- the automatic weldment (weld material)
- the automatic weldment / heat affected zone
- the hand weldment (filler metal)
- the hand weldment / heat affected zone.

The NDT-temperature obtained on these types of materials at an energy level of 30 ft-lbs should under consideration of the maximum shift deliver the operator the data for the underpressure installation temperature according to

\[ T_U = NDT + 33 \, K. \]

Due to the recommendations of the Swiss licence authority for atomic power plants, for the judgement of the material additionally the 50 ft-lbs shift had to be determined.

2. Experimental Procedure

Under investigation is the modified RPV-material ASTM A 508 Cl 2, the chemical composition of which is given in Table 1. The deposited weld metal was the "Molytherme G.S." due to Sulzer brothers with 0.3 wt % Cu maximum.

To determine the fast neutron fluence, Ni, Fe and Cu activation detectors were irradiated along with the charpy specimens. The fluence was determined from the measured monitor activities.

2.1 Impact Tests

The tests were performed according to DIN 50115 with an impact tester according to DIN 51222. An automatic centering device with a cooling chamber and a resistance furnace was adapted for use on the impact machine. For the temperature regime \( 210 \leq T/K \leq 283 \) (\(-60 \leq T/^\circ C \leq 10\)) a mixture of pure ethanol with 2 % ketone was used in combinations with a refrigerator and heaters for cooling and heating, respectively. The temperatures were measured using calibrated Chromel-Alumel thermocouples. Approximately 900 s were necessary for the low temperature regime in order to obtain temperature equilibrium for the specimens under investigation.

The specimen was pushed to the anvil of the impact machine with the centering device and 4 seconds later impact occurred. The respective cooling down or heating up rate during these 4 second was previously determined using calibration curves and to obtain the precise temperature at impact a corresponding supercooling or an overheating was selected.

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For the evaluation the following values had to be taken

- impact energy
- deformed part of the fracture surface due to ASTM-A-370
- In addition to DIN 50115 the lateral expansion (LE) and the root notch contraction at the broken specimens is determined.

2.2 Tensile Tests

For the first irradiation set a hydraulically driven universal test machine of the supplier Mohr & Federhaff, Mannheim, with a maximum region of 400 kN was used.
During the investigations of the second irradiation set a servo hydraulic 250 kN-test machine, delivered by MTS, Berlin, of the type MTS 810.13 was applied.
The determination of the characteristic values was due to the DIN 50145.
The yield strength $R_{p0.2}$ was obtained by means of an electronical high sensitive strain gauge of the type AW15-50 of the supplier Mohr & Federhaff with a magnification of 1000, which brought an accuracy of 1%.

The gauge length of 25 mm was taken by ringtyped knife-edges, which were adapted by a mounting bed, which was especially developed for the hot-cells.
The normal load-displacement diagram was taken with a magnification of 10.

3. Test results

The results of the tensile tests are not shown here, because no significant changes were observed. From that point of view they will briefly be discussed later.
The impact values are presented after the first irradiation ($5.4 \cdot 10^{17}$ nvt) for the base material and the weldments in figs. 1 -2. Fig. 3 reveals the impact values for the automatic weldment for the second irradiation set ($1.1 \cdot 10^{18}$ nvt).
Fig. 4 shows the position plan, which exhibits the position where the different specimens were taken from. Fig. 5 shows the impact values for the automatic weldment again, which will be taken as a point of discussion.

4. Discussion

4.1 Tensile Test Results

In principal remains the following statement with respect to the values. In all material states an evident increase of about 10 pct for the yield strength was observed for the second irradiation set, which was not found for the first irradiation set with the exception of the automatic weldment.

The automatic weldment revealed for the second irradiation set an increase of the yield strength of about 20 pct, i.e. e.g. at room temperature
\[ R_{p0.2} = 502 \text{ MPa} \quad \text{unirradiated} \quad \text{to} \quad R_{p0.2} = 601 \text{ MPa} \quad \text{irradiated}. \]

Because the tensile strength increases at room temperature as well as at operational temperature (288°C) in the same scale, i.e. the ratio

\[
\left( \frac{R_{p0.2}}{R_{m}} \right)_{\text{unirradiated}} \sim \left( \frac{R_{p0.2}}{R_{m}} \right)_{\text{irradiated}} \sim 0.82
\]

remains constant, reveals the material state of the automatic weldment with at the percentage elongations after fracture of 20.4 and 18.4 pct at room temperature as well as 15.4 pct at operational temperature at remaining unchanged reductions of area after fracture Z of 58 to 60 pct an excellent material behaviour.

4.2 Impact Test Results

The impact energy versus temperature for the different material states is presented in Figs. 1 to 3. For the determination of the NDT-Temperature of the several material states at 520 kJ/m² and 860 kJ/m² (30 ft-lb and 50 ft-lb, resp.) the toughness vs. temperature diagrams served as the basis. For each temperature the averaged value of the measured toughness values are taken as the real value. In such cases, where possibly "phase jumps" in the toughness-temperature curve could lead to unreasonably high toughness values, for reasons of a steady slope the lower part of the scatter band is applied.

The shift \( \Delta \text{NDT} \) for the different treatments are summarized in Table 2.

The shifts \( \Delta T_{50} \) for different material states (Table 2), as evaluated from the graphs, reveal the weakest part important for safety considerations. This part is the automatic weldment with a shift of \( \Delta T_{50} = 79°C \). This apparent insensitivity of the automatic weldment can be explained as follows:

In the first irradiation set, the outer layer of the weldment is compared with unirradiated specimens, which were taken from the mid-position, Fig. 4. If the outer layer is compared with the outer layer, a shift of \( \Delta T_{50} = 60°C \) is observed, Fig. 5.

According to Fig. 6, which reveals the change of the toughness values over the wall thickness, for the first irradiation set for 1/4 T - 1/2 T (T: Thickness) a correction of \( \Delta T_{50} = 60 - 66°C \) can be derived. In the 2. irradiation set only the mid-position is available. From that the observed shift of 79°C proceeded from an extreme case

\[
\Delta T_{50}^{\text{total}} = \Delta T_{1}^{50} (60°C) + \Delta T_{2}^{50} (19°C) = 79°C
\]

and for the other extreme case

\[
\Delta T_{50}^{\text{total}} = \Delta T_{1}^{50} (66°C) + \Delta T_{2}^{50} (13°C) = 79°C
\]

The change of the shift \( \Delta T_{50} \) in the meantime of the first to the second irradiation set is at least 13°C and no more than 19°C. The justification of this argument can be taken from the tensile tests.
5. Conclusions

Within the surveillance program for the nuclear power plant discussed here the following conclusions can be derived from the test results:

- From the tensile tests no drastic changes can be derived with respect to the material behaviour due to irradiation. The result are within the usual changes.

- The shifts $\Delta T_{50}$ and $\Delta T_{30}$, which were taken from the impact energies as a function of the temperature exhibit only small changes within the time of irradiation. The large shift of the automatic weldment was explainable by different positions within the weldment, where the specimen were taken from. The copper content plays a minor role. In principle this weld can be handled like a forged and/or cast material.

Acknowledgement: Mr. A. Christen performed these experiments in the hot cell.
### Table 1: Chemical Composition of the Base Material A 508 Cl 2 and specific Analysis for Cu, Mn, Si of the Welds

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>Ni</th>
<th>Mo</th>
<th>Si</th>
<th>Mn</th>
<th>Cr</th>
<th>Cu</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hand</td>
<td>.18</td>
<td>.86</td>
<td>.60</td>
<td>.32</td>
<td>.69</td>
<td>.35</td>
<td>.147</td>
<td>.018</td>
</tr>
<tr>
<td>Automatic</td>
<td>.30</td>
<td>1.05</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>.20</td>
<td>1.61</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 2: Transition Temperatures $T_{50}$ for 50 lb-fts for unirradiated and irradiated States of the Material with neutron fluences of $5.5 \cdot 10^{17}$ ntv (2) and $1.1 \cdot 10^{18}$ ntv (3) unirradiated (1):

<table>
<thead>
<tr>
<th>$T(1)_{50}{^\circ C}$</th>
<th>$T(2)_{50}{^\circ C}$</th>
<th>$T(3)_{50}{^\circ C}$</th>
<th>$\Delta T_{50(2-1)}{^\circ C}$</th>
<th>$\Delta T_{50(3-1)}{^\circ C}$</th>
<th>Base material</th>
</tr>
</thead>
<tbody>
<tr>
<td>-21</td>
<td>-9</td>
<td>-4</td>
<td>12</td>
<td>17</td>
<td>Basematerial</td>
</tr>
<tr>
<td>-25</td>
<td>+56</td>
<td>+54</td>
<td>81</td>
<td>79</td>
<td>Automatic weld</td>
</tr>
<tr>
<td>-20</td>
<td>-10</td>
<td>0</td>
<td>10</td>
<td>20</td>
<td>Hand weld</td>
</tr>
</tbody>
</table>
Fig. 1: Impact energy as a function of temperature for base material A 508 Cl 2.
Fig. 2: Impact energy as a function of temperature for the hand weldment.
Fig. 3: Impact energy as a function of temperature for the automatic weldment.

- unirradiated
- irradiated $1.1 \times 10^{18}$ nvt (E > 1MeV)
Fig. 4: Detail for the fabrication and surveillance test program for the automatic weldment-position of the specimens within the weldment.
Fig. 5: Impact energy as a function of temperature for the automatic weldment under consideration of different position.
Fig. 6: Impact energy as a function of the position within the automatic weldment.
Surveillance Extension Experience
at WWER-440 Type Reactors.

by

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to be presented at the joint IAEA/NEA Specialist's meeting on
"Irradiation Embrittlement and Optimization of Annealing"

to be held in
Paris
France
ABSTRACT

SURVEILLANCE EXTENSION EXPERIENCE at WWER-440 TYPE REACTORS.

by

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ABSTRACT.

In WWER-440 reactors the surveillance specimens are located in accelerated irradiation positions. After five years all specimens are withdrawn and the operational changes are not monitored. At Paks NPP a new surveillance program extension is started to eliminate of this disadvantage of the original program, and obtaining further data for plant lifetime management.
The paper is includes:
- Research performed to prepare the surveillance extension programme.
- The elaborated surveillance extension programme
- The evaluation method prepared for the surveillance extension programme.
- The first results.

Keywords: surveillance, radiation embrittlement, RPV integrity.

Prepared for the Specialist's Meeting
on Irradiation Embrittlement and Optimization of Annealing.
Paris 1993 September

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1. Introduction

The second generation of the WWER-440-s, the so-called V-213 type, has a uniq surveillance programme. Such units are operated in Hungary at Paks site. The task of the surveillance testing is to verify the operational safety, to supply data for PTS analysis and for evaluation of the exact lifetime. To fulfil these requirements the original surveillance program of the V-213 reactors had to be extended.

2. The original surveillance program of the WWER-440-s

To understand the extended surveillance program a short review of the original surveillance is necessary.

Specimen sets

Each WWER-440 V213 unit has 6 sets of specimens for surveillance testing. Each set contains specimen series made from base material, HAZ, and the weld. Each series consists of 12 Charpy V specimens, 12 Charpy size TPB specimens and 6 tensile specimens. This means a total of 90 irradiated specimens in each set. Two sets also contain specimens for studying thermal embrittlement. Every set has Fe, Ni, Co, and Cb foils for dosimetry.

The specimens are located between the core and the vessel wall in pipes welded to the outer side of the core barrel. The specimens are encapsulated in stainless steel capsules. FIG. 1. The Charpy size specimens are encapsulated two by two, and the tensile specimens are encapsulated six by six, and 19 or 20 capsules are connected together as a chain. Two chains form a complete set of specimens. Since the chains are quite long, the first and last capsules are out of the maximum flux. The Charpy specimens are located in the middle of the sets, the tensile specimens are in one capsule, and give reliable results, but the COD specimens are irradiated by different fluences. Since the small ligaments of these specimens also give confusing results generally only the results obtained on weld material can be accepted as valid fracture toughness values.

FIG.1. Specimen capsules in the WWER-440-s

The specimens are cut from below 1/4 depth of the wall thickness, from four or in some cases five different layers.

The layers are named D, G, I, E, K as it is shown in FIG. 2.
FIG. 2. Specimen cut plan of WWER-440-s

Dosimetry

The accuracy of dosimetric measurements and calculations is strongly influenced by the facts that flux and fluence change with the burning of the fuel and with refuelling; great fluences cause saturation effect, and some foils (e.g. Cb) may be activated by gamma radiation which does not cause embrittlement in the wall. The distribution of the flux changes along the active zone, too. Figure 3. shows the results of the measurements performed in the first unit of the Paks Nuclear Power Station.

FIG. 3. Fluence distribution in Paks unit 3
The Charpy (ISO V) specimens, which are the source of the most important information for the surveillance, are situated in neighbouring capsules facing the centre of the active zone and thus the divergence of the flux is small. A more significant divergence is caused as there are two Charpy specimens in one capsule and – depending on their position as compared to the active zone – they shield each other or themselves. The divergence of fluence between the farther surfaces of the two specimens may reach 40%. Because of the irregularities of the fluence the specimens are used in random order during testing.

Withdrawal schedules

Since the surveillance specimens in WWER-440 V-213 units are relatively far from the vessel wall the lead factor is about 11.8 for the base material and about 18 for the weld metal as the welds are below or above the core level. The high lead factors imply that the specimens are withdrawn in the first 5 years. The withdrawal schedules applied in Hungary are shown in TABLE 1.

**TABLE 1. Surveillance itinerary at Paks**

<table>
<thead>
<tr>
<th>Unit</th>
<th>Capacity[M We]</th>
<th>Supplier</th>
<th>First operation</th>
<th>Specimen set withdrawn year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paks1</td>
<td>440</td>
<td>Skoda</td>
<td>1982</td>
<td>84, 85, 86(2), 87</td>
</tr>
<tr>
<td>Paks2</td>
<td>440</td>
<td>Skoda</td>
<td>1984</td>
<td>85, 86, 87(2), 88</td>
</tr>
<tr>
<td>Paks3</td>
<td>470</td>
<td>Skoda</td>
<td>1986</td>
<td>87, 88(2), 89, 90</td>
</tr>
<tr>
<td>Paks4</td>
<td>460</td>
<td>Skoda</td>
<td>1987</td>
<td>88, 89(2), 90, 91</td>
</tr>
</tbody>
</table>

One set of specimens in every unit stays for a long period (minimum 10 years) to study the thermal embrittlement.

3. The extended surveillance program

The surveillance testing of WWER-s is different from that prescribed in the ASTM E-185 standard. Difficulties in the evaluation of this type of surveillance are caused by the high lead factor, by the fact that there is no direct thermocouple for temperature measurement of the capsules (diamond powder is used as temperature monitor) and by the self-shielding of the specimens. The small size COD specimens can only be used to obtain valid fracture toughness values at a very low temperature. On the other hand the large number of the specimens allows statistical evaluation, even the low temperature fracture toughness values give extra information on the material performance and because the results are obtained in an early stage the utility has an extended period for plant lifetime management.
To avoid the disadvantages and to satisfy fully the requirements of the ASTM E-185 standard an extended surveillance program has been elaborated in Hungary and it has been accepted by the Hungarian authority. The new specimens are placed into the empty surveillance channels.

The new specimen sets

The new specimen sets consist of three different types of forged (base) material. The materials are: a special heat of the 15H2MFA material, the IAEA reference material JRQ, and the original archive material of every unit.

Every specimen set consists of 12-20 Charpy and 6 tensile specimens of each of the above mentioned materials. The extended surveillance specimen sets are shown in FIG.4.

FIG.4. Specimen sets for extended surveillance program at NPP Paks
F= relative fluence (max=1), 15h2m=15H2MFA reference steel, archiv=reconstituted Charpy specimens made from archive material (remnants of 0 level testing), JRQ=IAEA reference steel, CV= Charpy V notched specimen, T= round tensile specimen (2 smooth and 4 notched in 1 capsule).

For a forty year operation period every unit requires about 150 uniform Charpy and 60 uniform tensile specimens plus specimens for zero level testing from every material. All of the specimens are located against the middle of the core to be exposed with the same irradiation fluence, with the exception of 4 Charpy specimens in every set, which are located in low flux positions. These specimens will be collected from the four units, and they will be used to evaluate the flux rate effect, which may affect the surveillance results.
Production of the specimens

In case of reference materials the requirement is to use specimens cut from the same specific layer of the material, since the material properties especially in case of the IAEA reference material JRQ are changing in the function of distance from the surface (depth).

Due to the limited availability of archive materials (practically only the remnants of specimens used for zero level testing are available) reconstituted type specimens are used. Electron beam or laser welding is used for the reconstitution. Instead of individual welding the remnants of the archive Charpy specimens are welded together with two metal blocks, called "combs" due to their shape. See Fig. 5. The thickness of the combs is a bit more than the thickness of the Charpy remnants, so there is no crater at the edge of the welding. Eight-twelve specimens and two combs are welded together.

FIG. 5 Charpy remnants and "comb" cut for welding

Before the welding the pieces are placed into a special rig. See Fig. 6. The rig is made from copper to assist heat removal. To avoid the forming of martensitic structures in the weld or in the HAZ, preheating suitable to the material type is necessary. The welding parameters are carefully selected to ensure defectless seams and a small heat affected zone at the same time. A computer model to calculate the heat distribution during welding in the function of the technology parameters has been elaborated. Fig. 7 shows the calculated sizes of the differently heat affected zones made by electron-beam welding.
FIG. 7. Calculated size of the heat affected zone during EB welding

The model has been verified by thermocouple measurements during welding and by metallography (FIG 8. shows a metallography picture on an electron-beam welded specimen), by hardness testing on the welded pieces.

FIG. 8. Cross section of an electron beam welded Charpy specimen.

Finally, careful machining and control of the specimen size are the last steps of the specimen production.

According to the tests performed until now these specimens gave the same results as the original ones. For further verification we participate in the ASTM Round-Robin program on reconstituted Charpy specimen production.

Every specimen got a specific number engraved on both ends by laser beam. This technology gives good visibility without destroying any part of the specimens.

Dosimetry

In the original surveillance program the dosimetry foils are located in an eccentric position. Since the specimen holder chain can turn around during location into the irradiation channels, the self-shielding increases the scatter of the results. In the extended surveillance program centrally located foil holders are used.

The shielding effect of the wall of the foil holders is equivalent to the shielding effect of a half thickness of a Charpy specimen. See FIG. 9. The foil holders are in the head of the selected capsules. There are 6-10 dosimetry foil sets are in every extended surveillance set. The foils used in the extended surveillance programme are: Cb; Fe54; Cu, Co.

FIG. 9. The new foil holder
d. Encapsulation

The specimens are encapsulated into stainless steel capsules, which are the same design as the original surveillance capsules. The materials for capsule production are carefully selected, they have quality guarantee by the producer. The sealing of the capsules is made by TIG welding in argon gas. The weld technology and quality satisfy the requirements for nuclear pressurised components. Every capsule is leak tested. The geometry and flexibility of the complete sets are also checked.

Every set of specimens including material properties, cut plan, specimen geometry, location in the chains, location, mass, material of the dosimetry foils are carefully documented. The national authority checks the documentation and permits the utility to reload the specimens.

Preparation of the surveillance extension program

Four sets of specimens were used to prepare this surveillance extension program. These sets are similar to the sets which will be used in the future, but they contain more dosimetry foils. In the first two sets JFL, JRQ, JWP, and JWO materials were also irradiated for the IAEA co-ordinated irradiation research program.

In the research sets irradiated for 1 year Ti, and U238 dosimetry foils are also used. This extended foil set gives a better estimation on spectra of Paks unit 2, meaning an increase of the accuracy of dosimetry results and EOL calculation.

These research sets had been placed into the empty surveillance channels of Paks unit 2, for 1, 1, 2, and 3 years of irradiation. This made it possible to develop the standard technology of the production of the new specimen sets, and permits better comparison with the previous surveillance results.

Irradiation schedule

Every specimen set will be irradiated for four years in the reactor. Table 2. shows the schedule for the extended surveillance sets at NPP Paks. Until this year 4 preparation (research) sets and 3 extended surveillance sets have been reloaded.

**TABLE 2. Surveillance extension reloading itinerary at Paks**

<table>
<thead>
<tr>
<th>Unit</th>
<th>Capacity [MWe]</th>
<th>First operation</th>
<th>Extended specimen set reloading year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paks1</td>
<td>440</td>
<td>1982</td>
<td>94, 98, 2002, 06, 10, 14, 18</td>
</tr>
<tr>
<td>Paks3</td>
<td>470</td>
<td>1986</td>
<td>92, 96, 2000, 04, 08, 12, 16, 20, 24</td>
</tr>
<tr>
<td>Paks4</td>
<td>460</td>
<td>1987</td>
<td>93, 97, 2001, 05, 09, 13, 17, 21, 25</td>
</tr>
</tbody>
</table>

* Remark: preparation sets for research purpose
The bold numbers indicate the sets already reloaded.
Results obtained from the first two research sets

The first two research sets have already been withdrawn and tested.

The purposes of installing the research sets were:
- to get experience in designing, production and reloading of specimen sets and to test the manipulating technology.
- to check the maximum temperature of the specimens during irradiation.
- to get informations on the radiation embrittlement behaviour of the materials used in the extended surveillance program.
- to get further dosimetry results.

No difficulties arose during the production, reloading and withdrawal of the first research set. During irradiation 3 capsules became inermetic, and the surface of the specimens slightly corroded.

In WWER-s the maximum capsule temperature is determined by the measurement of the lattice parameter of diamond powder irradiated together with the specimens. Due to the high flux the gamma heating is a considerable factor affecting the specimen temperature. In the original surveillance program the diamond powder is located away from the specimens, which means it tests the temperature of itself. Also there is some uncertainty during the measurement and evaluation of the lattice parameter. In the research specimen set small holes were drilled into the end of some specimens and into some of the aluminium heat conduction elements contained in every capsule, and melting temperature monitors were inserted into the holes. See Fig. 13.

**FIG. 13. Cavity for melting temperature monitor in Charpy specimen.**

Only two monitors with melting a temperature of max 290 °C were melted. (FIG. 14.). Since the melting monitor alloys contain several elements (like lead, cadmium...) having very high cross section values a high quantity of gamma heat is generated in them. This means that the specimen temperature remained below 290 °C in every case, in good agreement with the thermocouple measurement results performed by VTT (Finnland) in similar type capsules.

**FIG.14. Melted temperature monitor in Charpy specimen**

The Charpy results of materials tested in Paks unit 2 are shown on Fig.15. Table 3. and 4. shows the hardness testing and tensile testing results.
FIG. 15. Charpy results obtained on JRQ, JWQ, JWP and materials. (Zero level and 1 year irradiated specimens. Fluence $3 \times 10^{19}$ n/cm$^2$ E$>1$ MeV)

**TABLE 3. Tensile results on JFL and JRQ material.**

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>JRQ01</td>
<td>20</td>
<td>698</td>
<td>794</td>
<td>1423</td>
<td>62</td>
<td>1026</td>
</tr>
<tr>
<td>JRQ02</td>
<td>20</td>
<td>702</td>
<td>803</td>
<td>1345</td>
<td>58</td>
<td>887</td>
</tr>
<tr>
<td>JRQ03</td>
<td>100</td>
<td>691</td>
<td>787</td>
<td>1154</td>
<td>51</td>
<td>654</td>
</tr>
<tr>
<td>JRQ04</td>
<td>100</td>
<td>675</td>
<td>751</td>
<td>1097</td>
<td>44</td>
<td>517</td>
</tr>
<tr>
<td>JRQ05</td>
<td>300</td>
<td>623</td>
<td>746</td>
<td>952</td>
<td>32</td>
<td>301</td>
</tr>
<tr>
<td>JRQ06</td>
<td>300</td>
<td>626</td>
<td>744</td>
<td>1247</td>
<td>58</td>
<td>803</td>
</tr>
<tr>
<td>JFL01</td>
<td>20</td>
<td>587</td>
<td>707</td>
<td>1087</td>
<td>60</td>
<td>774</td>
</tr>
<tr>
<td>JFL02</td>
<td>20</td>
<td>558</td>
<td>674</td>
<td>1023</td>
<td>64</td>
<td>799</td>
</tr>
<tr>
<td>JFL03</td>
<td>100</td>
<td>530</td>
<td>644</td>
<td>915</td>
<td>53</td>
<td>547</td>
</tr>
<tr>
<td>JFL04</td>
<td>100</td>
<td>513</td>
<td>616</td>
<td>928</td>
<td>62</td>
<td>705</td>
</tr>
<tr>
<td>JFL05</td>
<td>300</td>
<td>505</td>
<td>653</td>
<td>738</td>
<td>42</td>
<td>334</td>
</tr>
<tr>
<td>JFL06</td>
<td>300</td>
<td>500</td>
<td>636</td>
<td>697</td>
<td>53</td>
<td>547</td>
</tr>
</tbody>
</table>
Evaluation

After every four year period the results of every unit will be compared with the original surveillance results. If the new results differ greatly from the original ones, the EOL calculated from the original surveillance program results will be modified. The modification will be performed by the use of the following formula.1

\[ E_m = E_f - C - \sum_{i=1}^{n} 4 \times K \times \frac{\Delta T_{i[n]}}{\Delta T_{i[1]}} \]

where \( E_m \) is the remaining lifetime, \( E_f \) is the full lifetime, calculated from the original surveillance program, \( C \) is the number of operational years until the loading of the first new surveillance set, \( n \) is the serial number of the new sets in the unit, \( K \) is a correlation factor characterizing the relationship between the original and the first extended surveillance period finally \( \Delta T_{i[0]} \) is the transition temperature shift (in K) measured on the \( n \) sets of new specimens. \( K \) is evaluated from the operational history, its value generally is 1. If in case of any extended set the value of: \( \frac{\Delta T_{i[n]}}{\Delta T_{i[1]}} > 1.5 \) the utility must study why the embrittlement rate increased.

5. Summary

The surveillance program extension at NPP Paks will complete the original surveillance program and eliminate all its disadvantages. The surveillance programs with the extensions at the Paks units satisfy the requirements of ASTM E-185 standard, supports life time management of the PWR-s, and will give more data for PTS calculations.

6. Acknowledgement

This surveillance extension program was elaborated by the sponsorship of NPP Paks, in the form of the government supported national research program on Nuclear Safety under the Gouverment contract no 91-97-42-0339.

The following scientists and their institutes participated in the work beside the authors: Dr. J. Rittinger (ERÖKAR), Dr. A. Fehérváry (VASKUT), Dr. É. Zsolnay (Technical Univ. of Budapest)

7. References.


4.) F. Gillemot; Survey of Irradiation Embrittlement Effects of the Mechanical Properties of Alloyed Steels. 


5.) P. Trampus, F. Gillemot; Paks Reactor Pressure Vessels Meet the Requirements. Science and Technology in Hungary. 1992 Augustus, Budapest

THE PRELIMINARY RESULTS OF THE THERMAL ANNEALING PROCESSES PERFORMED ON THE RPVs NPP V-1 IN JASLOVSKÉ BOHUNICE
by
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ABSTRACT

The annealing procedure was planned and prepared many years before not for the first two units V-230 type in NPP V-1 only, but well before in another countries which are operating these type of former Soviet technology. After all it was 13-th and 14-th procedure realized till this time, but first two annealings without Russian know-how and technology.

The samples of weld and base metal of both RPVs NPP V-1 were prepared by special apparatus in the narrow gap between the outside surface of the RPV and the reactor thermal shielding in January and April 1993, from the critical circumferential weld joint #4. The chemical composition of the samples was analyzed in Nuclear Power Plants Research Institute (VÚJE) laboratories. Except results achieved from analysis of the irradiated samples are presented the results of the hardness measurements of reactor pressure vessel material across the weld #4 too. All this results will serve as input data for the irradiation embrittlement recovery evaluation of the both RPV NPP V-1 in Jaslovenské Bohunice.

Keywords: WVER, reactor, 15CH2MFA steel, annealing, irradiation embrittlement recovery, chemical analysis, hardness measurement,

INTRODUCTION

The annealing procedure was prescribed in the "Regulation Act #5" of the former Czechoslovak Atomic Energy Commission (ČSKAEB) issued in 1990, concerning the higher reliability and safe operation of the PVR VVER-440 type with Reactor Pressure Vessels (RPV) V-230 type [1]. On the base of this decision were performed:
- the sampling of the RPV material both units from the weld #4, and from the base material above and below this weld
- the hardness measurement across the weld #4, on the external surface of these RPVs, in the very narrow gap between the RPV and thermal shielding in reactor cavity before and after annealing
- thermal annealing procedure

Annealing procedure was performed:

Unit # 1: 20-th till 27-th April 1993
Unit # 2: 30-th January till 6-th February 1993
The sampling was performed:

- on the unit #2:
  - from January 14-th till 20 - before annealing
  - from February 12-th till 14-th - after annealing.
- on the unit #1:
  - from April 5-th till 9-th - before annealing

The hardness measurement (Brinnel) was performed:

- in the same time both of base and weld metal across the critical weld #4 in the regions of maximum and minimum neutron flux from the azimuthal crosssection point of view (fig.#1).

The samples prepared by milling were used:

- for the chemical analysis, both the weld and base metal, to achieve more precise and reliable informations as are available in original Soviet documentation
- for the dosimetry measurements to analyze the positive effects of the dummy elements, which were inserted to the both reactor cores on the base of recommendations of OKB Gidropress [2], which were accepted by former Nuclear Regulatory Body.
- for the electron-positron annihilation measurements to analyze the regeneration procedure efficiency

The hardness measurement using the ball indentor through the stripe from Ni-foil was performed:

- to identify the real position of the weld #4
- to compare the results of the hardness before and after annealing to analyze the regeneration procedure efficiency.

THE HARDNESS MEASUREMENT ON RPVs ACROSS THE WELD #4

Before the beginning of the hardness measuring both RPVs were performed these operations:

a) the determination of real weld positions on RPV by three independent methods (chemical etching of weld #2, determination of weld #3 position by chemical etching and measurement, identification of the real position of critical weld #4 by the measurements of altitudes based on the data from the ultrasonic control operation performed inside the RPV, and using the Eddy current probe).

b) the grinding of RPV outside surface around the weld #4 in the maximum and minimum positions of neutron flux in azimuthal plane of the reactor core [3].

c) the milling of samples from the weld metal #4 and the base metal above and under this weld for the chemical composition determination (fig.#2 and #3).

The hardness measurement was performed by following parameters:

- Brinnel method with the ball indentor, pressed on the grounded RPV surface through the Ni-stripe
- diameter of the indentor is 2,5 mm
- dimensions of the Ni-stripe: 9 mm wide and 250 mm long
- force 1840 N.
By the hardness measurement evaluation are the diameters of recorded prints measured using the optical microscope and image analyser. The results of these measurements are summarized in fig. #4 and #5 [4].

THE CHEMICAL COMPOSITION DETERMINATION

Total ten samples were prepared for the chemical analysis of base and weld metals from Unit-1, and nineteen samples from the Unit-2, because of lack of precise and reliable informations about the composition of this critical part both RPVs. For the determination of elements: C, Mn, Cr, Ni, Mo, P, Si, Co, V, S, and Cu were used the methods: coulombimetric titration, spectroscopy and AAS. The chemical composition is presented in table #1 [12,13]. The sampling coordinates are measured from the axis of weld #2. The samples were taken in minimum and maximum of the neutron flux (300 from axis #III, or 920 mm on the outer diameter RPV, and 300 mm left from the axis #III). From the known chemical compositions it is possible to calculate the irradiation embrittlement coefficients using Yermakov and Dragunov [5] so called "chemical relations" (C.R.) for base metal and weld metal:

\[
\begin{align*}
A_{F-BM270} &= 1100.\% P - 2 \\
A_{F-VM} &= 800 \ (\%P + 0.07\%Cu)
\end{align*}
\]

and the transition temperature by relation:

\[
T_{K0-VM} = 101.6 - 171.\%\text{Mn}.\%\text{Mo} + 151.8.\%\text{Mn}.\%\text{V} + 8224.\%\text{Cu}.\%P - 42139 .\%S .\%P - 163 .\%\text{Cu}.\%\text{Mo} - 2726 .\%P
\]

The shift of the transition temperature is possible to calculate, if the fluence on RPV is known by formula:

\[
\delta T_{KF} = A_F \cdot (F\cdot10^{-22})^{1/3}
\]

Table #1: Chemical composition of materials of RPV’s Unit #1 and #2

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>V</th>
<th>S</th>
<th>P</th>
<th>Co</th>
<th>Cu</th>
</tr>
</thead>
<tbody>
<tr>
<td>BH Unit1</td>
<td>0.140</td>
<td>0.31</td>
<td>0.37</td>
<td>2.64</td>
<td>0.20</td>
<td>0.58</td>
<td>0.27</td>
<td>0.017</td>
<td>0.014</td>
<td>0.019</td>
<td>0.091</td>
</tr>
<tr>
<td>VM Unit1</td>
<td>0.048</td>
<td>0.37</td>
<td>1.11</td>
<td>1.00</td>
<td>0.12</td>
<td>0.39</td>
<td>0.13</td>
<td>0.013</td>
<td>0.043</td>
<td>0.024</td>
<td>0.103</td>
</tr>
<tr>
<td>BH Unit2</td>
<td>0.132</td>
<td>0.25</td>
<td>0.37</td>
<td>1.225</td>
<td>0.17</td>
<td>0.55</td>
<td>0.25</td>
<td>0.018</td>
<td>0.010</td>
<td>0.017</td>
<td>0.082</td>
</tr>
<tr>
<td>VM Unit2</td>
<td>0.066</td>
<td>0.26</td>
<td>0.88</td>
<td>1.110</td>
<td>0.14</td>
<td>0.34</td>
<td>0.19</td>
<td>0.013</td>
<td>0.026</td>
<td>0.019</td>
<td>0.109</td>
</tr>
</tbody>
</table>
THE RESULTS OF THE ANALYSIS

FROM THE DOSIMETRY MEASUREMENTS:
- the activity measurements of the RPV samples show that dummy elements caused the shift of local maximum of the neutron flux in the azimuthal crosssection
- the high energy neutron fluence is 2.6 times lower and in the thermal energy region is the shielding factor higher than 4.5
- from these measurements is possible estimate the relative contents of some elements present in the samples too

FROM THE CHEMICAL COMPOSITION ANALYSIS:
- the impurities contents of Cu, and S is significantly lower than the producer data and comparing with the analysis results obtained from the sampling in 1989. This fact is possible to explain by the higher precision of the chemical analysis methods, and better statistics from more samples which were sampled in two layers
- the P contents is comparable with the producer data

FROM THE HARDNESS MEASUREMENT:
- the precision of the hardness measurement was improved by the correlations measurement of the hardness standards measurements in laboratory conditions and by the image analysis system application to the 10% value
- the recovery of hardness after annealing is highest in weld metal (up to 48 HB), and 33 HB in base metal
- the values of the hardness after anealning operation are for both RPVs similar
- we do not find the annealing temperature gradient influence on the hardness recovery due to the annealing procedure
- recalculation of the hardness values to the Rm, give us the unirradiated values of the RPV steel

FROM THE ANNEALING PROCEDURE EFFICIENCY:
The annealing procedure efficiency was estimated in following way:
- according the data based on the experiences of the producer, the recovery of the RPV material properties depends on the difference between temperature of annealing and temperature of irradiation in our case for more than 200°C is the recovery efficiency in range 95 - 100%
- the calculations of the strengths values from hardness value before and after annealing by ČSN 420379 give us 100% recovery efficiency
- the Mössbauer spectroscopy and electron-positron annihilation methods applications give us the high degree of ordered structure after annealing too
- LAST BUT NOT LEAST THE COMPARISON OF DATA MEASURED IN LOWER PART OF RPV, WHICH IS NOT INFLUENCED WITH HIGH NEUTRON FLUENCE, GIVE US THE SAME INFORMATIONS TOO.
REFERENCES

[1] ČSKAE: "Regulation Act No.5", Prague, December 1990


[5] Kupča L. et al.: The results of hardness measurements in the weld #4 region, chemical analysis and dosimetry of the samples taken from RPV EBO Unit #1, Report VÚJE #360/15/92


Fig. No. 1: Neutron flux distribution on RPV Unit 1
Fig. No. 2: Positions of hardness measurement on RPV Unit 1 before annealing
Axis of max. "n"-flux

WELD 0.1.4

Position No.5

Fig. No. 3: Positions of hardness measurement on RPV Unit 1 after annealing

WELD 0.1.2

Position No. 6

12 prints

16 prints

18 prints

10 prints

12 prints

Axis of min."n"-flux
Fracture mechanics investigations within the swiss surveillance programme for the pressure vessel of modern nuclear power plants

Fracture toughness, stress intensity, ductile fracture

by

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Paul Scherrer Institut, LWV, Würenlingen u. Villigen, CH-5303 Villigen PSI

In the frame of surveillance programmes of swiss nuclear power plants (Gösgen, Leibstadt) irradiations were performed on tensile-, impact-, and wedge opening load specimens as well as on three point bend type specimens as on precracked charpy impact specimens. The three point bend type specimens are used for J-integral investigations, the precracked charpy impact specimens are applied for dynamical stress intensities $K_{JD}$. From the swiss nuclear authority two different methods were asked for the determination of crack initiation. The Paul Scherrer Institute is responsible for the surveillance programmes. The method is divided into two parts; an experimental method (potential drop technique) and a mathematical procedure, which is described in detail by the "application of analytical functions to obtain crack-initiation and fracture toughness $K_{IC}$ (ASTM E 399) in the ductile region". The agreement of these both methods was found to be excellent. The mapping of the both methods to fatigue precracked small specimens (3 PB and charpy) is possible. The applicability of the analytical method to dynamical tests is also possible. Both methods are not part of any regulatory guide. The purpose of this contribution is to stimulate further discussions.
List of Abbreviations

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>P</td>
<td>Load, in Newton (N)</td>
</tr>
<tr>
<td>B</td>
<td>Thickness of a specimen, in (mm) metric units</td>
</tr>
<tr>
<td>a</td>
<td>Length of a crack, in (mm) metric units</td>
</tr>
<tr>
<td>W</td>
<td>Width of a specimen, in (mm) metric units</td>
</tr>
<tr>
<td>E</td>
<td>Young's modulus, GPa, N/mm²</td>
</tr>
<tr>
<td>ν</td>
<td>Poisson's number</td>
</tr>
<tr>
<td>σₚ</td>
<td>Yield Strength (MPa, N/mm²)</td>
</tr>
<tr>
<td>K</td>
<td>Stress intensity factor, (MPa • m₁/², N/mm³/²) conversional factor: 31.623</td>
</tr>
<tr>
<td>KᵢC</td>
<td>Critical stress intensity factor for the stress modulus I (pure tensile)</td>
</tr>
<tr>
<td>J</td>
<td>J-Integral value which describes the plastic fracture, kJ/m², N/mm</td>
</tr>
<tr>
<td>A</td>
<td>Energy derived from the integration of the load vs load point displacement diagram up to the initiation load Pᵢ</td>
</tr>
<tr>
<td>B (W-a)</td>
<td>Magnitude of the remaining ligament</td>
</tr>
<tr>
<td>η'</td>
<td>A factor, which is 2 for three point bend type specimens and ( \eta' = 2 + 0.522 \frac{W-a}{W} ) for the compact tension specimens</td>
</tr>
<tr>
<td>ε (t)</td>
<td>total strain as a response</td>
</tr>
<tr>
<td>σ (t)</td>
<td>time dependent stress</td>
</tr>
<tr>
<td>σ₀</td>
<td>stress at the very beginning, i.e. t = 0⁺</td>
</tr>
<tr>
<td>σ∞</td>
<td>stress level after relaxation</td>
</tr>
<tr>
<td>Mᵤ</td>
<td>unrelaxed modulus</td>
</tr>
<tr>
<td>Mᵣ</td>
<td>relaxed modulus</td>
</tr>
<tr>
<td>( \bar{\tau}_σ )</td>
<td>average retardation times at constant stress</td>
</tr>
<tr>
<td>( \bar{\tau}_ε )</td>
<td>average relaxation times at constant strain</td>
</tr>
<tr>
<td>T</td>
<td>absolute temperatures (K)</td>
</tr>
</tbody>
</table>
\( \delta \) : Phase angle
\( \omega \) : Angular velocity (s\(^{-1}\))
\( R_T \) : real resistivity (Ohm's resistor)
\( C \) : capacity
\( L \) : Inductivity
\( U \) : Electrical potential
\( I \) : Current
\( \gamma \) : the ratio of \( \sigma_w / \sigma_0 \)
\( w \) : a complex number
\( z, \bar{z} \) : complex numbers, \( \bar{z} \) is the conjugate of the complex number \( z \).
\( f (\frac{Z}{W}) \) : Geometrical correction factor for different types of specimens
Introduction and outline of the problem

According to the ASTM E 399 standard (1988), the stress intensity $K$ in N mm$^{-3/2}$ ($N =$ Newton, mm = millimeter) is calculated for the 1$^{st}$ CT (Compact Tension)-test piece using

$$K = \frac{P}{B\sqrt{W}} f \left( \frac{a}{W} \right) \quad (1.1)$$

Here $P$ is the linear elastic peak load, to cause the growth of crack $a$; $W$ and $B$ are the sample width and thickness (figure 1) and $f (a/W)$ the geometry factor which can be found in the literature of fracture mechanics.

From an engineering point of view following current procedures conservative limiting cases are determined. In the case of a plane-strain condition with $E' = E (1 - v^2)$ ($v =$ Poisson's Number)

$K \rightarrow K_{IC}$ applies; for the plane stress condition with $E' = E$, $K \rightarrow K_C$.

The derived empirical relationship for ferritic materials

$$B, a \geq 2.5 \left( \frac{K_{IC}}{\sigma_s} \right)^2 = g \cdot \left( \frac{K_{IC}}{\sigma_s} \right)^2 \quad (1.2)$$

with $\sigma_s =$ yield strength (N/mm$^2$) determines the thickness $B$ for brittle fracture criteria.

Since $K_{IC}/\sigma_s$ increases with temperature, $B$ and $a$ ($B = a$ for $a/W = 0.5$) must be selected to be correspondingly greater.

E.T. Wessel (1970) achieved a valid crack toughness value $K_{IC}$ of approximately 4900 N mm$^{-3/2}$ for a ferritic steel 22MnMoNi55 (~ A533 B1) with a yield strength of $\sigma_s \approx 500$ N/mm$^2$ at nearly room temperature and a sample thickness $B = 300$ mm (weight of the sample was approx. 11800 N).

Using a pressure vessel material 22NiMoCr37 Kussmaul K. (1990) achieved a value of $K_{IC} \sim 6100$ N/mm$^{3/2}$ with a sample thickness of $B = 500$ mm following ASTM E 399.

Both these large and expensive tests show, that for a poor $K_{IC}/\sigma_s$ relationship a valid fracture toughness value can only be obtained with a cheap and sensible size of test piece at low and mainly uninteresting temperatures.

A solution to this dilemma of achieving valid $K_{IC}$ values for small test pieces at higher temperatures was proposed in ASTM E 813 (1988). From the measured load-displacement curve one uses a suitable method (e.g. a potential drop technique) to determine the start of the stable crack growth (point $V^0$ in figure 5) and calculates the $J$ value from

$$J = \frac{\eta' \cdot A}{B (W-a)} \quad (1.3)$$

Here $A$ is the energy under the load-strain curve up to the crack-initiation load $P_1$, $B (W-a)$ the ligament and $\eta'$ a factor. For 3-point bending samples $\eta' = 2$, for CT samples $\eta' = 2 + 0.522 \cdot (W-a)/W$. The original idea of deriving the $K$ value with the help of the $J$ value using $K^2 = JE$ has not prevented the discussion over this standard from continuing. (Question of the blunting line, type of regression, method of measuring the initiation, $J$ converted to $K$ too high).

The following suggests a method of solution using the conformal mapping, which is applied with success in aero-, hydro-, and electrodynamics.
2 Theoretical Basis

2.1 Transformation of the elastic-plastic behaviour to an elastic one

Brittle fracture occurs when in a test piece of a given size the relationship of the volume in a plastic state to that in the elastic state is zero or at least very small.

In the case that the effective energy from the applied load cannot be absorbed in plastic deformations then the whole of the loading energy can be applied to the fracture. Such "brittle conditions" occur not only at low temperatures but also during rapid loading conditions. The task is considered to be solved, if between the constructed elastic energy-level $\sigma_0$ and the plastic energy level $\sigma_-$ a correlation can be derived. This situation can be described mathematically by means of the theory of the linear relaxation processes, combined with the introduction of an invariant of the inelastic strain, which is valid for infinitesimally small to an infinitely large amount.

2.2 Thermodynamical theory of the linear relaxation analogy of the mechanical, magnetical and electrical fields

C.L. Maxwell's equations of the electrodynamics are based on a relaxation model which can be transformed to mechanics by a serial connection of the spring and a Newtonian dashpot (fig. 2a).

In addition a parallel connection of a spring and a dashpot exists which were introduced by Kelvin-Voigt and can be appointed as retardational model, fig. 2b. In Appendix A it is shown in detail (G. Ullrich 1978), that the parallelogram $\overrightarrow{OP}$ in fig. 3 with

$$\alpha = \frac{\tan \vartheta'}{\tan \vartheta''} = \frac{\tau_a}{\tau_e}, \quad \beta = \frac{\tan \alpha}{\tan \beta} = \frac{M_r}{M_u}, \quad \gamma = \frac{\sigma_-}{\sigma_0}$$

can be derived using the generalised Maxwell-model with a continuous spectrum of the relaxation times $\tau_e$ and the generalised Kelvin-Voigt model with a continuous spectrum of the retardation times $\tau_e$ by overlapping by means of the Boltzmann superposition principle as a folding integral as follows

$$\varepsilon(t) = \int_{-\infty}^{t} \frac{\partial}{\partial t'} (\sigma(t')) \Lambda(t-t') \, dt' , \quad (2.2.1)$$

with $\varepsilon(t)$ as the total strain as response and $\sigma(t)$ the stress relaxation from the level $\sigma_0$ to the level $\sigma_-$ acc:

$$\sigma(t) = \sigma_- + \sigma_0 \exp \left( -\frac{t}{\tau_e} \right) \int_{0}^{t} \overrightarrow{R} \left( \tau_e \right) \, d\tau_e , \quad (2.2.2)$$

and the retardation function
\[ \Lambda(t) = \frac{1}{M_u} + \frac{1}{M_r} \left\{ \frac{1 - \exp \left( -\frac{t}{\tau_\sigma} \right)}{\tau_\sigma} \right\} \int_0^\infty F_1(\tau_\sigma) \, d\tau_\sigma \, , \]  

(2.2.3)

with \( M_u \) as an unrelaxed and \( M_r \) as a relaxed modulus. Introducing the normalising conditions of the distribution function in equation (2.2.2) and (2.2.3) according to

\[ \int_0^\infty F_2(\tau_\sigma) \, d\tau_\sigma = \int_0^\infty F_1(\tau_\epsilon) \, d\tau_\epsilon = 1 - \frac{M_r}{M_u} \, . \]  

(2.2.4)

One obtains formally after differentiation of equ. 2.2.1 over the time the well known dynamical state equation following Cl. Zener [1948]

\[ \sigma + \frac{\tau_\sigma}{M_u} \, \dot{\sigma} = M_r \left\{ \epsilon + \frac{\tau_\epsilon}{M_u} \, \dot{\epsilon} \right\} \]  

(2.2.5)

for

\[ \frac{\tau_\sigma}{M_u} \cdot \frac{M_r}{M_u} = \frac{\tau_\epsilon}{M_u} \]  

(2.2.6)

Equation (2.2.5) is the basis for the description of irreversible processes by diffusion processes of atoms or electrons. This dynamical state equation can be appointed as one of the most important equation of the whole physics of matter [U. Dehlinger 1955] because of its large variety of modifications.

Because of the central significance, J. Meixner 1954 undertook a derivation universally beyond thermodynamics and obtained equation (2.2.5), (see appendix B).

If a load or a strain in equation (2.2.5) occurs very rapidly on the values \( \sigma_0 \) resp. \( \epsilon_0 \), then the Newtonian dashpot plays no role (inertia) in the equivalent standard linear solid (fig. 2c). The complete elastic case is reached

\[ \tau_\epsilon \frac{d\sigma}{dt} = M_r \tau_\sigma \frac{d\epsilon}{dt} \]  

(2.2.7)

or

\[ \sigma = M_r \frac{\tau_\sigma}{\tau_\epsilon} \, \epsilon = M_u \, \epsilon \, . \]  

(2.2.8)

Therefore \( M_r \frac{\tau_\sigma}{\tau_\epsilon} \) is the valid unrelaxed modulus \( M_u \) following rapid loading. During rapid loading procedures the plasticity at a given temperature \( T_1 \) is suppressed. During slow procedures the velocity dependent term vanishes in equation (2.2.5) and therefore

\[ \sigma = M_r \, \epsilon \]  

(2.2.9)

with \( M_r \) as the measurable relaxed modulus.

Cl. Zener investigated equation (2.2.5) with success in a study of mechanical relaxation spectra under the introduction of oscillating stresses and strains according to
\[
\sigma = \sigma_0 \exp(i\omega) \quad , \quad \varepsilon = \varepsilon_0 \exp(i\omega)
\]  \hspace{1cm} (2.2.10)

For the loss or phase angle \(\delta\), for which the strain is shifted behind the stress, the following expression is found

\[
\tan \delta = \frac{\omega (\tau_0 - \tau_\varepsilon)}{1 + \omega^2 \tau_\sigma \tau_\varepsilon}
\]  \hspace{1cm} (2.2.11)

Introducing the geometrical average

\[
\overline{\tau} = \sqrt{\overline{\tau_\sigma} \overline{\tau_\varepsilon}}
\]  \hspace{1cm} (2.2.12)

in equation (2.2.11) and the consideration of equation (2.2.8) the following is obtained

\[
\tan \delta = \frac{M_u - M_r}{\sqrt{M_u \cdot M_r}} \cdot \frac{\omega \overline{\tau}}{1 + (\omega \overline{\tau})^2}
\]  \hspace{1cm} (2.2.13)

For the attenuation maximum

\[
\frac{d (\tan \delta)}{d (\omega \overline{\tau})} = 0
\]  \hspace{1cm} (2.2.14)

follows

\[
\omega = \frac{1}{\tau} = \frac{1}{\sqrt{\tau_\sigma \tau_\varepsilon}}
\]  \hspace{1cm} (2.2.15)

If the increase or the decrease of an electrical field \(E\) with the relaxation time \(\tau_I = RC\) (I: current, R: resistivity, C: capacity) is coupled with the decrease or the increase of a magnetic field \(H\) with the relaxation time \(\tau_U = \frac{L}{R}\) (L: inductivity, U: electrical potential analogue to equus. (2.2.2) and (2.2.3) [R.W. Pohl 1967]), so this leads to

\[
U + \tau_I \frac{dU}{dt} = R_r \left( I + \tau_U \frac{dI}{dt} \right)
\]  \hspace{1cm} (2.2.16)

with \(R_r\) as the relaxed measurable resistivity. Introducing the alternating electrical potential and current due to

\[
U = U_0 \exp(i\omega) \quad , \quad I = I_0 \exp(i\omega)
\]  \hspace{1cm} (2.2.17)

on the place of equation (2.2.15) is found

\[
\omega = \frac{1}{\overline{\tau}} = \frac{1}{\sqrt{\tau_U \cdot \tau_I}} = \frac{1}{\sqrt{LC}}
\]  \hspace{1cm} (2.2.18)

the well known Thomson equation for the resonance case of free oscillation within an electrical oscillation circuit with a parallel capacity and inductivity.

If the system is not attenuated (no Joule's heat) so follows with equ. (2.2.8)
\[
\frac{M_u}{M_t} = \frac{\bar{\tau}_a}{\bar{\tau}_e} = 1
\]

(2.2.19)

or

\[
\bar{\tau}_a = \bar{\tau}_e.
\]

(2.2.19a)

For the electrodynamics is analogue

\[
\tau_u = \tau_t \text{ and from that } RC = \frac{L}{R}
\]

or

\[
R_u = R_r = \frac{L}{\sqrt{C}}
\]

(2.2.20)

as an unrelaxed resistivity, special an electrical wave resistance if the propagation of electromagnetic waves without phaseshift between current I and potential U in parallel "Lecher-current-wires" are considered [Chr. Gerthsen, H.O. Kneser 1971]. The geodetic lines of the orthogonal cutting circles of a coupled electrical and magnetic field (fig. 4) can be calculated with the extremals of a problem of variation acc. L. Euler [B. Baule 1961]. Within this noneuclidean hyperbolic geometry based on C.F. Gauss the circumference \(\tilde{u}\) of a geodetic circle can be calculated acc

\[
\tilde{u} = 2\pi \, sh \, R_0
\]

(2.2.21)

with

\[
R_0 = \ln \tan \left(\frac{\alpha}{2} + \frac{\beta}{2}\right)
\]

(2.2.22)

as the geodetic radius.

Equation (2.2.22) is the condition for the angle conservation using conformal mapping.

Thus the same results can be obtained in a simpler manner by means of the analytical functions \(w(z)\) with the complex argument

\[
z = x + iy \; ; \; i = \sqrt{-1}
\]

(2.2.23)

All the mappings resulting from analytical functions are conform, i.e. conservation of angles and circles.

By means of conformal mapping the mechanics of deformable solid state bodies can be handled with the laws of fluid dynamics in an universal way, as already applied in aerodynamics, hydro-dynamics, and electrodynamics, respectively.

The starting point is the logarithmic potential

\[
w = -\ln z.
\]

(2.2.24)

The derivation is
\[ \frac{dw}{dz} = \omega' = -\frac{1}{z} \]  \hspace{1cm} (2.2.25)

and represents the velocity field of the function in equation (2.2.24) as well as a complex mirroring at the unit circle. It represents a mapping, which turns the internal of a unit circle outside and vice versa.

It is assumed that the point \( V^0 (= \sigma_i) \) on the elastic-plastic curve [fig. 5] as an equilibrium value at the \( \sigma_m \)-level is considered via a stress relaxation from the \( \sigma_0 \)-level as a pseudo stress (fig. 5). Neither \( \sigma_0 \) nor \( \sigma_m \) is known. The ratio will be called

\[ \gamma = \frac{\sigma_m}{\sigma_0} \]  \hspace{1cm} (2.2.26)

\( \sigma_0 \) and \( \sigma_m \) being considered as imaginary values \( i\sigma_0 = iy_0 \) and \( i\sigma_m = iy_m \) \( (i = \sqrt{-1}) \), then equ. (2.2.26) is not changed. If any point at the \( \sigma_0 \)-level is introduced as a complex quantity, the conformal (mirror) mapping at the unit circle around \( O \) (0, 0) with the radius of unity \( \sigma_{\max} = 1 \) acc

\[ W = -\frac{1}{Z} = \frac{\bar{Z}}{|Z|^2} = \frac{-x + iy_0}{x^2 + y_0^2} = u (x, y_0) + iv (x, y_0) \]  \hspace{1cm} (2.2.27)

represents a transformation by reciprocal radii with the abscissa \( x \) as the real axis of symmetry. Equ. 2.2.27 follows the equations of Cauchy-Riemann (e.g. Goodstein R.L. 1965).

Assuming the vector \( \overline{O_\sigma \overline{P}} = Z \) (fig. 5) with the coordinates

\[ Z = x + i \sigma_0 \]  \hspace{1cm} (2.2.28)

one obtains for \( \overline{P} (x = +1/iy_0 = i\sigma_0) \) in fig. 5\(^{10} \) following equ. 2.2.27 the vector

\[ -\frac{1}{Z} = \overline{O R}^* \]  \hspace{1cm} (2.2.29)

in the interior of the unit circle with \( |\overline{O} R^*| = \gamma \).

The same operation can be made with the vector \( Z^* = \overline{O_\sigma O}^* = -1 + iy_0 \). The circles around \( O^* \) or \( \overline{P} \) with the radius \( r = \sigma_0 \) cut the unit circle around \( \overline{O} \) orthogonally in the point \( O \) or \( O^* \). The both vectors \( Z \) and \( Z^* \) passes the unit circle on the \( \sigma_m \)-level. The line \( VR \) passes the experimental elastic-plastic curve through the point \( V^0 \) (= \( P_1 \)).

In fig. 5 the following relationships are valid:

\(^{10} \) In the rectangle \( \overline{O O P O^*} \) for the invariant \( O \overline{P} \) in fig. S equ. 17 in appendix A is also fullfilled with \( \tan \theta'/\tan \theta^* = \tan \frac{x}{P O' O_1} / \tan \frac{x}{P O^* R} = \alpha = \gamma \), and thus \( x = 1 \).
\[
\frac{V}{O^{\prime\prime}} - \frac{V}{O} = \sqrt{1 - \gamma^2} = -\gamma - 1 = \frac{\sigma}{\sigma_0}
\]

\[
\frac{R}{O^*} = \sqrt{1 - \gamma^2} = \frac{\sigma}{\sigma_0} = +\gamma + 1
\]

or

\[
O \cdot O^* = O^* \cdot P = \frac{\sqrt{1 - \gamma^2}}{\gamma} = \sigma_0
\]

and with respect to the experimental curve it is received

\[
\sigma_0 = \frac{\sigma_{\text{max}} \sqrt{1 - \gamma^2}}{\gamma} = \frac{\sigma_{\text{max}} (= \sigma_i)}{\gamma}
\]

Further informations are to be found inside the unit circle as the hyperbolic plane (fig. 6) now with respect to the \(O\) as new origin of a coordinate system. The line \(OR^*\) orthogonal to \(\overline{OR}^*\) passes the ordinate \(i\sigma\) through the point \(M\), as the center of the circle with the radius

\[
\overline{OM} = \frac{\gamma}{\sqrt{1 - \gamma^2}} = \frac{1}{\sigma_0}.
\]

This circle passes the line \(OR^*\) through the points \(P^*\) and \(P^{**}\) also with the abscissa \(x = -\gamma\) and \(= +\gamma\). The most important result of this complex inversion delivers the orthogonality

\[
OP^* \cdot OP^{**} = 1
\]

with

\[
OP^* = \text{ch} R_0 - \text{sh} R_0 = \frac{1 - \gamma}{\sqrt{1 - \gamma^2}} = \frac{1 - \gamma}{\sqrt{1 + \gamma}}
\]

\[
OP^{**} = \text{ch} R_0 + \text{sh} R_0 = \frac{1 + \gamma}{\sqrt{1 - \gamma^2}} = \frac{1 + \gamma}{\sqrt{1 - \gamma}}
\]

whereby

\[
\text{ch} R_0 = \cos (iR_0) = \frac{1}{\cos \varphi} = \frac{1}{\sqrt{1 - \gamma^2}} = O \cdot M
\]

\[
\text{sh} R_0 = -i \sin (iR_0) = \tan \varphi = \frac{\gamma}{\sqrt{1 - \gamma^2}} = \overline{OM}
\]

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\[ \text{th} R_0 = -i \text{tg} (iR_0) = \sin \varphi = \gamma = \overline{O} R^* \] (2.2.40)

The equations 2.2.36 - 2.2.40 confirm that the real hyperbolic functions can be described by trigonometric functions with an imaginary argument, whereby

\[ R_0 = \text{arctg} \gamma = \ln \frac{1 + \gamma}{\sqrt{1 - \gamma}} \] (2.2.41)

\[ e^{+R_0} = \frac{1 + \gamma}{\sqrt{1 - \gamma}} ; e^{-R_0} = \frac{1 - \gamma}{\sqrt{1 + \gamma}} \] (2.2.42)

With the help of the equation 2.2.42 the ordinates from the points P** and P* are received:

\[ \psi_2 = \gamma \cdot e^{+R_0} = \frac{\gamma(1+\gamma)}{\sqrt{1 - \gamma^2}} \] (2.2.43)

\[ \psi_1 = \gamma \cdot e^{-R_0} = \frac{\gamma(1 + \gamma)}{\sqrt{1 - \gamma^2}} \]

as points of the strophoide \( OP^* \overline{OP}^{**} \), which is inverse to itself and symmetric to the real abscissa \( x \).

From the point of view of the metric of length the equations 2.2.36 and 2.2.37 are from the type of the well known Lorentz transformation in electrodynamics. With \( R_0 \) as the radius of the geodetic circle in equation 2.2.22 acc:

\[ R_o = \ln \tan \left( \frac{\varphi}{4} + \frac{\varphi}{2} \right) \] (2.2.45)

whereby

\[ \text{sh} R_o = \frac{\gamma}{\sqrt{1 - \gamma^2}} = \frac{1}{\sigma_o} \] (2.2.46)

In fig. 7 at the first glance one can see in the triangle O\( \overline{OD}M \) with the height \( \overline{OR}^* \) a graphical illustration of the trigonometric and hyperbolic functions.

From the point of view of the metric of angle the equations 2.2.36 and 2.2.37 can also be derived from the origin \( \overline{O} \) in fig. 6

\[ \times P^* \overline{O} O = \frac{\pi}{4} - \frac{\varphi}{2} \; ; \; \times P^{**} \overline{OR} = \frac{\pi}{4} + \frac{\varphi}{2} \]

\[ \tan \left( \frac{\pi}{4} - \frac{\varphi}{2} \right) = \frac{1 - \gamma}{\sqrt{1 - \gamma^2}} \; ; \; \tan \left( \frac{\pi}{4} + \frac{\varphi}{2} \right) = \frac{1 + \gamma}{\sqrt{1 - \gamma^2}} \]

see also appendix C.
3 Determination of the stress intensity $K_{IC}$ for small CT-samples (ASTM E 399) instead of large ones. Relationship to the 3 point bend specimen

Measuring the point $V^0$ (= $P_I$ as load) (fig. 5) on the elastic-plastic curve as initiation of the stable crack growth with the help of the potential drop technique, the value $\gamma$ is received following the numerator in equation 2.2.33 acc.

$$\gamma = \sqrt{1 - \left( \frac{P_I}{P_{max}} \right)^2}$$  \hspace{1cm} (3.1)

The stress intensity $K_0$ acc. equ. 1.1 is given by equation 2.2.33 following

$$K_0 = \frac{P_0 f (a/w)}{B \sqrt{W}} = \frac{P_{max} \sqrt{1 - \gamma^2} \cdot f (a/w)}{\gamma \cdot B \cdot \sqrt{W}}$$  \hspace{1cm} (3.2)

In equ. 3.2 one has to differentiate between the 3 point bend (3PB)-specimen test with a compressive (negative) stress antiparallel to the crack propagation and the CT-specimen-test with a positive tensile stress field orthogonal to the crack propagation. Because of this orthogonality in case of the CT-specimen the equation (3.2) has to be transformed with the Lorentz-Transformations.

$$K'_0 = K_0 \cdot \frac{1 - \gamma}{\sqrt{1 - \gamma^2}}$$  \hspace{1cm} (3.3)

Introducing $E' = E / (1 - v^2)$ ($E = 206010$ N/mm²) one obtains

$$\frac{(K'_0)^2 (1 - v^2)}{E} = \frac{K_{IC}^2}{E} = J_{IC} / N/mm$$  \hspace{1cm} (3.4)

In equation 3.4 the Poisson-number $v_{eff}$ is needed. It can be derived from this experiment (see appendix C).

4 Discussions

The evaluation of the experiments on CT-samples at ≥ room temperature can be seen in table 1.

As an example experiment no 1 in table 1 is calculated with $P_{max} = 75000$ Newton and the initiation point $P_I = 69220$ Newton. From equation 3.1 one obtains $\gamma = 0.385$. Then equ. 3.2 delivers $K_0 = 8819$ N mm⁻³/². After transformation acc. equ. 3.3 the value $K'_0 = 5873$ N mm⁻³/² is received. With $v_{eff} = 0.298$ from equ. 17 appendix C one obtains $K_{IC} = 5606$ N mm⁻³/² acc. equ. 3.4 and $K_{IC}^2 / E = J_{IC} = 153$ N/mm. This result delivers acc. equ. 1.2 with the
factor $g = 2.5$ a specimen thickness $B = 366$ mm at room temperature for the material AS33 B1 with a yield-strength $\sigma_y = 463$ N/mm$^2$ in agreement with the experiments from E.T. Wessel et al (fig. 8).

If one would take the value $J_{IC} = \eta U/B(W-a) = 2.279 \cdot 71141 / 25 \cdot 26.8 = 242$ N/mm acc equ. 1.3 (ASTM E 813, table 1) one should receive the value $K_{IC} = 7060$ and a specimen thickness $B = 581$ mm.

A profound discussion of this transformation as used here in mechanics, which is beyond the scope of this work, lead us to the analogous phenomena best known in physics as transversal Doppler-effect of second order using the Lorentz-transformations. [R.W. Pohl 1967]

It is obvious to be seen that in the case of the movement of crack propagation perpendicularly to the isoforcelines in a force field a further analogous aspect in mechanics and electrodynamics is demonstrated. Thus for the 3 point bend specimen with the crack propagation antiparallel to the field of force this transformation is not required because the bending-experiment itself is inverse to that of CT-specimen-experiment. Equ. (3.2) can be used immediately.

Therefore for a 3 point bend specimen with the dimensions acc. ASTM E 399 ($W = 50$ mm $B = 25$ mm) with the same ratio $a/W = 0.464$ and the same value $\gamma = 0.385$ it is predicted a load following the inversion acc. equ. 2.2.35

$$P_{\text{max}}^{3PB} = P_{\text{max}}^{CT} \cdot \frac{OP^*}{OP^{**}} = P_{\text{max}}^{CT} \cdot \frac{1-\gamma}{1+\gamma} = 33303 N \quad (4.1)$$

This load can be turned by 90° and the bend specimen can principally be treated as a CT-specimen after having transformed acc. equ. 3.3. Taking the predicted value $P_{\text{max}}^{3PB} = 33303$ Newton the value $K_{IC} = 3738$ N mm$^{-3/2}$ acc. equ. 3.2 and 3.4 and $J_{IC} = 68$ N/mm is calculated using the boundary factor $f_{CT}(a/W) = 8.67$.

If it is intended to change from the 3 point bend specimen to the CT-specimen with aims of the transformation following equ. 2.2.37 acc.

$$K_{IC}^{CT} = K_{IC}^{3PB} \cdot \frac{1+\gamma}{\sqrt{1-\gamma^2}} = 3738 \cdot 1.5 = 5606 \text{ N mm}^{-3/2} \quad (4.2)$$

one obtains the above mentioned value $K_{IC}^{CT}$.

An example will reveal this behaviour, using immediately equ. 3.2 for a 3 point bend specimen.

An experimental curve for a specimen $25 \times 25 \times 100$ mm with a smaller ligament $B(W-a)$, $(B=W$, not quite due to ASTM E 399) published elsewhere (Krompholz K., Ulrich G., 1986) with respect to the determination of the J-Integral acc. to ASTM E 813 has delivered the following data:

Material: A 533 B1, $a/W = 0.53$, $P_{\text{max}} = 20600$ Newton, $P_i = 19140$ Newton, $P_f/P_{\text{max}} = 0.929$, $\gamma = 0.37$, $v_{\text{eff}} = 0.351$, $f_{CT}(a/W) = 10.63$. Using immediately equ. 3.2 and equ. 3.4 it is
obtained $K_{IC}^{3P} = 4117 \, N \, mm^{-3/2}$ and $J_{IC} = 82 \, N/mm$. If it is wanted to calculate the value for the equivalent CT-specimen with the width $W = 25 \, mm$ one obtains the value $K_{IC}^{CT} = 6071 \, N \, mm^{-3/2}$ acc. equ. 4.2.

From this it can be pointed out that the 3 point bend specimen delivers a more conservative value $K_{IC}$ acc. to ASTM E 399.

5 Detection of the crack initiation without any auxiliary equipment

The relevant point $P^*$ and $P^{**}$ of the strophoide in fig. 6 can be obtained mathematically by setting:

$$\frac{1}{E} \int_0^K dK = M_r \int_0^\varepsilon d\varepsilon$$

$$\frac{K_0^2}{E} = J = M_r \{1\}^2 = \frac{\sigma_{\infty}}{\{1\}} \{1\}^2 = \sigma_{\infty} \{1\} \left[ N / mm \right] \quad (5.1)$$

Hereby $K_0$ is the stress intensity, $E$ is the Young's modulus and

$$\frac{\sigma_{\infty}}{\{1\}} = M_r = \tan \angle V'O\overline{D} = \frac{\sigma_{\max} \sqrt{1 - \gamma^2}}{\{1\}}$$

whereby $\{1\} = O^n\overline{P}$ in fig. 6 is the invariable. Introducing the ligament according to

$$\sigma_{\infty} = \frac{P_{\infty}}{B(w-a)} = \frac{P_{\max} \sqrt{1 - \gamma^2}}{B(w-a)}$$

on the right side of equ. (5.1) one obtains

$$\frac{P_{\max}^2 (1-\gamma^2)}{\gamma^2 B^2} f^2 \left( \frac{a}{w} \right) = \frac{P_{\max} \sqrt{1 - \gamma^2}}{B(w-a)} \{1\} \quad (5.2)$$

The numerical value of the invariable $\{1\} = O^n\overline{P}$ for the three point bend type specimen is

$$\{1\} = \{f\} \frac{1 - \gamma}{\gamma} = \{f\} O^n\overline{P} \quad (5.3)$$

where $O^nP$ in fig. 6 is the intersection of the straight line $OV$ with the horizontal line $O^nO'$ in the point $P$. Correspondingly for the CT-specimen one finds

$$\{1\} = \{f\} \frac{1 + \gamma}{\gamma} = \{f\} O^n\overline{C}, \quad (5.4)$$

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where $O^\prime C$ is the intersection of the straight line OR in fig.6 with the horizontal line $O^\prime O$ in C. The point C is to be found outside of the diagram.

The value $\{f\}$ in equ. (5.3) and (5.4) is denoted with a length unit and the respective value $\frac{1 \pm \gamma}{\gamma}$ gets the corresponding numerical magnitude without dimension. Therefore from equ. (5.2) the fourth order polynomial in $\gamma$ is found due to

$$\gamma^4 + (\psi^2 - 1)\gamma^2 \pm 2\psi^2\gamma + \psi^2 = 0, \quad (5.5)$$

For

$$\psi = \frac{1}{\{f\}} \frac{P_{\text{max}} f^2 (a) (W-a)}{B \cdot w \cdot E} \quad (5.6)$$

With $\gamma$ and $\psi$ as values without dimensions fig. 6 forms a nomogram.

Using the explicit solution of equ. (5.5) the following is obtained

$$\psi_{1,2}^2 = \frac{\gamma^2(1 - \gamma)}{(1 \pm \gamma)^2} \quad (5.7)$$

With

$$\psi_2 = \pm \frac{\gamma(1 + \gamma)}{\sqrt{1 - \gamma^2}} \quad (5.8)$$

the positive and negative values for the CT-specimens are obtained as shown in equs (2.2.43 and 2.2.44). The positive sign (tension) is valid. With

$$\psi_1 = \pm \frac{\gamma(1 - \gamma)}{\sqrt{1 - \gamma^2}} \quad (5.9)$$

the positive and negative values for the three point bend test pieces are found. The negative sign is valid (compression).

Equation (5.8) is connected with equation (5.9) according to

$$\psi_1 = \psi_2 \frac{1 - \gamma}{1 + \gamma} \quad (5.10)$$

The following example will confirm the theory. Taking for example the first value from Table 1 for the CT-specimen the following can be calculated

$$\psi_2 = \frac{75000 \cdot 8.67^2 \cdot 26.8}{25.50 \cdot 206010} = 0.586$$
From fig. 6 or from equation (5.7), which can now be used as a working diagram one finds $\gamma \sim 0.39$ in best agreement with the potential drop technique.

Taking the values for the above mentioned three point bend type specimen with a span $s = 4 \, W$ with the boundary correction factor $f^{3PB}(\varepsilon) = 2.94$ (ASTM E 399) one obtains:

$$\psi_1 = \frac{20600 \cdot 11.75 \cdot 2.94^2 \cdot 4^2}{25.25 \cdot 206010} = 0.259$$

and from fig. 6 $\gamma \sim 0.381$, also in good agreement with the potential drop technique.

Thus this method of calculation is suitable to examine the values from the potential drop technique.

6 Final remarks

With the above mentioned solution for calculation $K_{IC}$ it will be possible to determine the temperature dependent stress intensity $K_{IC}$ on small samples $B = 25 \, \text{mm}$ (e.g. after neutron irradiation) without having to refer back to large specimens:

Without proof is should be mentioned that the $g$-factor in equ. 1.2 is found experimentally acc

$$g_{CT} = \frac{d^V (1 + \gamma)}{\{f\} \sqrt{1 - \gamma^2}}$$

Whereby $d^V$ is the path up to crack initiation and $\{f\}$ the unity of length. For room-temperature with $d^V = 1.7 \, \text{mm}$ and $\gamma$ from table 1 it is found $g_{CT} = 2.5$.

For elevated temperature $g_{CT}$ is increasing and even $B$ in spite of slightly decreasing $K_{IC}$ values above room-temperature (fig. 8).
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<td>130</td>
<td>1.95</td>
<td>403</td>
<td>2.9</td>
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<td>-</td>
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<tr>
<td>200</td>
<td>0.470</td>
<td>65500</td>
<td>61111</td>
<td>0.933</td>
<td>0.36</td>
<td>8.83</td>
<td>0.388</td>
<td>5356</td>
<td>118</td>
<td>2.1</td>
<td>414</td>
<td>3.15</td>
<td>-</td>
<td>-</td>
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<td>200</td>
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<td>67000</td>
<td>61693</td>
<td>0.920</td>
<td>0.39</td>
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<td>5012</td>
<td>134</td>
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<td>427</td>
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<td>-</td>
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<td>60633</td>
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<td>0.395</td>
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<td>0.265</td>
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<td>130</td>
<td>2.4</td>
<td>532</td>
<td>3.64</td>
<td>-</td>
<td>-</td>
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</table>

Table 1 Fracture mechanics data for CT-specimens (ASTM E 399, $B = 25$ mm) for the maerial A 533 B1 $\geq$ room temperature. $f_{CT}(a/W)$ is a boundary correction factor acc. ASTM E 399.
Appendix A

Derivation of an inelastic strain invariant of arbitrary quantity

An integration by parts of the Boltzmann's superposition principle equation (2.2.1) delivers

\[ \varepsilon (t) = \sigma (t') \Lambda (t-t') \bigg|_{t'=-\infty}^{t'=-\infty} - \int_{-\infty}^{t'} \sigma (t') \frac{\partial}{\partial t'} \Lambda (t-t') \, dt' \]  

(1)

If in equation (1) the following substitution is undertaken

\[ \sigma (t') := \sigma_\infty + (\sigma_0 - \sigma_\infty) \exp \left( - \frac{t'}{\tau_e} \right) \cos \omega t' \]  

(2)

and

\[ \Lambda(t-t') := \frac{1}{M_u} + \frac{\Delta}{M_r} \left\{ 1 - \exp \left( \frac{t-t'}{\tau_\sigma} \right) \right\} ; \]  

(3)

\[ \Delta = 1 - \frac{M_r}{M_u} \]

than by evaluation of equation (1), if the integrals are handled as improper integrals, the following will be found

\[ \varepsilon (t) = \frac{\sigma_0 - \sigma_\infty}{M_u} \exp \left( - \frac{t}{\tau_e} \right) \cos \omega t + \]

\[ \frac{(\sigma_0 - \sigma_\infty) \Delta}{M_r} \exp \left( - \frac{t}{\tau_e} \right) \left\{ \frac{\tau_\sigma^{-1} - \tau_e^{-1}}{\tau_\sigma \left[ \left( \tau_\sigma^{-1} - \tau_e^{-1} \right)^2 + \omega^2 \right]} \right\} \cos \omega t + \]

\[ \frac{(\sigma_0 - \sigma_\infty) \Delta}{M_r} \exp \left( - \frac{t}{\tau_e} \right) \left\{ \frac{\omega}{\tau_\sigma \left[ \left( \tau_\sigma^{-1} - \tau_e^{-1} \right)^2 + \omega^2 \right]} \right\} \sin \omega t. \]  

(4)

If the ratio \[ \frac{\sin \omega t}{\cos \omega t} = \tan \omega t = \tan \delta \] is formed, than for the term which contains \( M_r \), the loss angle in formally way, which was also derived by C. M. Zener, is obtained for \( \tau_e > \tau_\sigma \).

For the most general case is found

\[ \tau_\sigma > \tau_e \]  

(5)
If the deformation step is formulated within the boundaries $t = 0$ up to $t = +\infty$ for the relaxation and retardation under the condition $\omega \to 0$ than one obtains

$$
\left. \Delta \varepsilon_B \right|_{t = \infty} = - \frac{\sigma_0}{M_u} + \frac{\sigma_\infty}{M_u} - \frac{(\sigma_0 - \sigma_\infty) \Delta}{M_r \left[ 1 - \frac{\tau}{t_c} \right]},
$$

where the index $B$ stands for Boltzmann.

Moreover, by definition is $\sigma(t) = 0$ for the semiopen interval $t \in \left[ -\infty, 0 \right]$, and $\sigma(t) = \sigma_0$ for $t \in \left] 0, \infty \right]$. 

Now the generalised function $H(t)$ is introduced, which is called Heaviside's jump-function. This function is defined in the following way [A1]

$$
f(t) H(t) := \frac{1}{2} \left( |f(t)| + |f(t)| \, \text{sgn}(t) \right),
$$

where $\text{sgn}(t)$ is an unsteady function, the sign function, with

$$
\text{sgn}(t) := \begin{cases} 
1 \text{ for } t \in \left] 0, \infty \right[, \\
-1 \text{ for } t \in \left[ -\infty, 0 \right].
\end{cases}
$$

where $\left] \ldots \right]$ are semiopen intervals. The "derivation" is

$$
\frac{d \text{sgn}(t)}{dt} = 2 \, \delta(t),
$$

where $\delta(t)$ is the "Dirac-jump function".

If equations (8) and (9) are applied, one finds with $f(t) = 1$

$$
H(t) = \begin{cases} 
1 \text{ for } t \in \left] 0, \infty \right[, \\
0 \text{ for } t \in \left[ -\infty, 0 \right].
\end{cases}
$$

and

$$
\frac{d H(t)}{dt} = \frac{1}{2} \cdot \frac{d \text{sgn}(t)}{dt} = \delta(t).
$$

Drawing this into consideration, it possible to state: Because of the requirement of a conservation theorem for the strain, equation (6) must be equal to such an equation, which is obtained by the application of the former considerations.

$$
\Delta \varepsilon_D(t) = \sigma_0 \int_{-\infty}^t \frac{\partial H}{\partial t'} \Lambda(t - t') \, dt' = \sigma_0 \Lambda(t) \left| \begin{array}{c} t = \infty \\
t = 0
\end{array} \right. = \frac{\sigma_0 \Delta}{M_r}
$$

where the index $D$ stands for Dirac. Formulating the theorem of conversation

$$
\Delta \varepsilon_B + \Delta \varepsilon_D = 0
$$

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leads with \( \Delta = 1 - \frac{M_r}{M_u} \) to

\[
\frac{\sigma_0}{M_r} - 2 \frac{\sigma_0}{M_u} + \frac{\sigma_\infty}{M_u} = \frac{1 - \frac{M_r}{M_u}}{M_r} \cdot \frac{\left( \sigma_0 - \sigma_\infty \right)}{1 - \alpha}
\]

(14)

for

\[ \alpha : = \frac{\tau^{\sigma}}{\tau_e} \]

Considering Fig. 3 one finds

\[
\overline{\sigma} \overline{P} + \overline{P} \overline{P}'' = \frac{\overline{P}'' \overline{P}}{1 - \alpha}
\]

(15)

where

\[
\overline{\sigma} \overline{P} + \overline{P} \overline{P}'' = \overline{\sigma} \overline{P}
\]

\( \overline{\sigma} \overline{P} \) is introduced as the measured contribution and \( \overline{P} \overline{P}'' \) is introduced as the stored contribution. The amount \( \overline{P}'' \overline{P} \) in equation (14) is not included. Therefore a loss contribution is introduced.

By formulation of the following equation it is to be seen in Fig. 3 that

\[
\overline{\sigma} \overline{P} + \overline{P} \overline{P}'' + \overline{P}'' \overline{P} = \overline{\sigma} \overline{P}
\]

or

\[
\frac{1 - \beta}{M_r} \cdot \frac{\left( \sigma_0 - \sigma_\infty \right)}{1 - \alpha} + \frac{1 - \beta}{M_r} \left( \sigma_0 - \sigma_\infty \right) + \frac{1 - \beta}{M_r} \left( \sigma_\infty - \sigma_0 \alpha \right) = \frac{\sigma_0 \left( 1 - \beta \right)}{M_r},
\]

where \[ \beta = \frac{M_r}{M_u} \]

By dividing with \( \frac{\sigma_0 \left( 1 - \beta \right)}{M_r} \) and by definition \( \gamma : = \frac{\sigma_\infty}{\sigma_0} \), the following result is obtained.

\[
\frac{(1 - \gamma) \alpha}{1 - \alpha} + (1 - \gamma) + \frac{\gamma - \alpha}{1 - \alpha} = 1
\]

(17)

Since \( \beta \) gives no longer a contribution to this equation, the invariable covers all conditions from the smallest infinity up to the largest, i.e. in principle from microphysics to macrophysics without restriction of universality.

The difference between anelasticity (i.e. small deformation processes) and plasticity is only gradual.

On the other hand from equation (14) the relationship
\[
\frac{1}{\gamma} = \frac{1 - \alpha\beta}{\alpha - 2\alpha\beta + \beta}
\] (18)

can be derived. If the assumption is in mind that

\[
1 - \alpha\beta = 0,
\]

for

\[
\sigma_0 = 0 \ (\gamma = \infty)
\]

the relationship (2.2.6) as a boundary case is obtained.

For \( \beta = 0 \) in equation (18) with \( M_u \rightarrow \infty \) the following is observed

\[
\gamma = \alpha
\] (19)

and the distribution function, as given in equation (2.4), becomes the normalised distribution function in the classical manner.

Equation (19) delivers the condition of the rectangle

\[
\overline{OO'PQ}
\]

In Fig. 6 the invariable in equation (18) delivers

\[
\frac{(1 - \gamma)}{1 - \gamma} + (1 - \gamma) = 1, \ i.e.
\] (20)

\[
1 = 1
\]
Appendix B

Thermodynamical Consideration of Relaxation Processes

Very often "reversible processes" are combined with diffusion-like mechanisms, in which atoms or electrons have to overwhelm energy barriers under the influence of thermal movement. These processes are irreversible under normal conditions. Therefore the previously reversible processes are damped and retarded, a process which is called relaxation.

In order to describe such processes, an equation of state is needed, which exists and by means of material constants correlates the volume \( V \), the temperature \( T \) or instead the entropy \( S \).

This normal equation of state is now to replace by an extended equation, which contains a parameter \( \xi \), which is the denominator of a coupled irreversible reaction.

Let \( \xi = 0 \) for a given equilibrium state, from which also the change of pressure \( p \) and volume \( V \) should be counted. In principle \( \xi \) could be any arbitrary parameter.

For a given Temperature \( T \) for a given function with the variables \( p, \xi \) it can be written

\[
V = V(p, \xi).
\]

For the velocity of the irreversible reaction the following rate equation holds

\[
\dot{\xi} = \frac{d\xi}{dt} = -a \left( \frac{\partial f}{\partial \xi} \right)_{p,V,T} = -a \cdot A
\]  

(1)

where \( f = f(p, V, T, \xi) \) is the free energy of the system. For the equilibrium state is

\[
\frac{df}{d\xi} = A \equiv 0.
\]

The term \( a \) depends also on the variables of state, but near the equilibrium it changes only slowly so that \( a \) is regarded as "quasiconstant". \( A \) is regarded as linearly dependent of the state variables so that this can be written \( a \),

\[
A = A(p, V, T, \xi) \text{ with } A_0 = 0 \text{ and }
\]

\[
\Delta A = \left( \frac{\partial A}{\partial \xi} \right)_{V,T} \Delta \xi + \left( \frac{\partial A}{\partial V} \right)_{\xi,T} \Delta V + \left( \frac{\partial A}{\partial T} \right)_{\xi,V} \Delta T,
\]

(2)

or

\[
\Delta A = \left( \frac{\partial A}{\partial \xi} \right)_{P,T} \Delta \xi + \left( \frac{\partial A}{\partial p} \right)_{\xi,T} \Delta p + \left( \frac{\partial A}{\partial T} \right)_{P,\xi} \Delta T.
\]

(3)

Because \( p \) and \( V \) are interdependent, only \( p \) or \( V \) is used. Be \( \Delta V = 0 \) and \( \Delta T = 0 \). Than also \( \Delta p = 0 \) and equation (1) becomes
\[ \frac{d\xi}{dt} = -a(A) = -a \left( \frac{\partial A}{\partial \xi} \right)_{V,T} \Delta \xi \]  

(4)

respective

\[ \frac{d\xi}{dt} = -a \left( \frac{\partial A}{\partial \xi} \right)_{p,T} \Delta \xi \]  

(5)

The separation of the variables delivers

\[ \frac{d\xi}{\Delta \xi} = -a \left( \frac{\partial A}{\partial \xi} \right)_{x,y} dt \]  

(6)

Let

\[ \tau_i := -\frac{1}{a} \left( \frac{\partial \xi}{\partial A} \right)_{x,y} \]  

(7)

than the following hold

\[ \ln \Delta \xi = \frac{t}{\tau_i} + \ln C \text{ or} \]

\[ \Delta \xi(t) = C \cdot \exp \left( \frac{t}{\tau_i} \right). \]  

(8)

Here is

\[ \tau_i = \tau_v = -\frac{1}{a} \left( \frac{\partial \xi}{\partial A} \right)_{T,V} \]

\[ \tau_i = \tau_p = -\frac{1}{a} \left( \frac{\partial \xi}{\partial A} \right)_{T,p} \]  

(9)

\( \tau_v \) is the relaxation time at a constant volume and \( \tau_p \) is the relaxation time at a constant pressure. Analogue to the specific heat it can be shown that \( \tau_v \leq \tau_p \).

Let \( \Delta T = 0 \) and

\[ \Delta \xi(p, V, T) = \left( \frac{\partial \xi}{\partial p} \right)_{V,T} \Delta p + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} \Delta V + \left( \frac{\partial \xi}{\partial T} \right)_{p,V} \Delta T. \]

For an isothermal procedure, i.e. \( \Delta T = 0 \) the following holds

\[ \Delta \xi(p, V, T) = \left( \frac{\partial \xi}{\partial p} \right)_{V,T} \Delta p + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} \Delta V. \]  

(10)
The integral delivers
\[ \xi(p, V, T) = \left( \frac{\partial \xi}{\partial p} \right)_{V,T} p + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} V \]
and from that
\[ \frac{d\xi}{dt} = \left( \frac{\partial \xi}{\partial p} \right)_{V,T} \frac{dp}{dt} + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} \frac{dV}{dt} = -a \left( \frac{\partial A}{\partial \xi} \right) \Delta \xi. \]

With equation (10) the following holds
\[ \left( \frac{\partial \xi}{\partial p} \right)_{V,T} \frac{dp}{dt} + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} \frac{dV}{dt} = -a \left( \frac{\partial A}{\partial \xi} \right) \left\{ \left( \frac{\partial \xi}{\partial p} \right)_{V,T} \Delta p + \left( \frac{\partial \xi}{\partial V} \right)_{p,T} \Delta V \right\} \]
\[ = -a \left\{ \left( \frac{\partial A}{\partial p} \right)_{V,T} \Delta p + \left( \frac{\partial A}{\partial V} \right)_{T,A} \Delta V \right\} \]

Because \( A = A(p, V) \), so at a constant reaction rate
\[ \frac{dA}{dV} = 0 = \left( \frac{\partial A}{\partial p} \right)_{T,V} \left( \frac{\partial p}{\partial V} \right)_{T,A} + \left( \frac{\partial A}{\partial V} \right)_{T,P}. \]

Then one finds
\[ \left( \frac{\partial p}{\partial V} \right)_{T,A} = - \frac{\left( \frac{\partial A}{\partial V} \right)_{T,P}}{\left( \frac{\partial A}{\partial p} \right)_{T,V}} \]

\[ (12) \]
A substitution into (11) delivers

$$
\left( \frac{\partial \xi}{\partial p} \right)_{V,T} \dot{p} + \left( \frac{\partial \xi}{\partial V} \right)_{P,T} \dot{V} = -a \left( \frac{\partial A}{\partial p} \right)_{V,T} \left\{ \Delta p + \frac{\left( \frac{\partial A}{\partial V} \right)_{T,P}}{\left( \frac{\partial A}{\partial p} \right)_{T,V}} \Delta V \right\}
$$

$$
= -a \left( \frac{\partial A}{\partial p} \right)_{V,T} \left\{ \Delta p - \left( \frac{\partial p}{\partial V} \right)_{A,T} \Delta V \right\}
$$

$$
\dot{p} + \left( \frac{\partial p}{\partial \xi} \right)_{V,T} \left( \frac{\partial \xi}{\partial V} \right)_{P,T} \dot{V} = -a \left( \frac{\partial A}{\partial p} \right)_{V,T} \left( \frac{\partial p}{\partial \xi} \right)_{V,T} \left\{ \Delta p - \left( \frac{\partial p}{\partial V} \right)_{T,A} \Delta V \right\}
$$

$$
\dot{p} + \left( \frac{\partial p}{\partial \xi} \right)_{T,A} \dot{V} = -a \left( \frac{\partial A}{\partial p} \right) \left\{ \Delta p - \left( \frac{\partial p}{\partial V} \right) \Delta V \right\}
$$

$$
\dot{p} + a \left( \frac{\partial A}{\partial \xi} \right) \Delta p = a \left( \frac{\partial p}{\partial V} \right)_{T,A} \left( \frac{\partial A}{\partial \xi} \right) \Delta V - \left( \frac{\partial p}{\partial V} \right)_{T,A} \dot{V}
$$

and from that

$$
\frac{1}{a} \left( \frac{\partial \xi}{\partial A} \right) \dot{p} + \Delta p = \left( \frac{\partial p}{\partial V} \right)_{T,A} \left\{ \Delta V + \tau_V \dot{V} \right\}
$$

(13)

or

$$
\frac{1}{a} \left( \frac{\partial \xi}{\partial A} \right) \dot{p} + \Delta p = V \left( \frac{\partial p}{\partial V} \right)_{T,A} \left\{ \frac{\Delta V}{V} + \tau_V \left( \frac{\Delta V}{V} \right) \right\}
$$

(14)

with

$$
V \left( \frac{\partial p}{dV} \right)_{T,A} = K_r
$$

(15)

as the modulus of compression.

Analogue to equation (14) is the dynamic equation

$$
\tau_e \dot{\sigma} + \Delta \sigma = M_r \left( \Delta \varepsilon + \tau_\sigma \Delta \dot{\varepsilon} \right)
$$

(16)

according to C.M. Zener in equation 2.2.5.

The unrelaxed quantity $M_u = M_r \frac{\tau_a}{\tau_e}$ or $K_u = K_r \frac{\tau_p}{\tau_V}$ are not measurable because at least one of the both time contents are internal quantities.
Appendix C

Determination of the Poisson number experimentally

A cube of a side length of one will be loaded in pure tensile in one axial direction. By this loading the cube is elongated to 1 + \( \varepsilon \) in the tensile direction, while a diminishing of the length of the sides perpendicularly to the tensile direction to 1 - \( \nu \) takes place. Here \( \nu \) is the Poisson number (see Fig. 8). The angle between the formerly crossing diagonals is changing by the gliding \( \eta \), i.e. for \( \frac{\eta}{2} \) at each side.

From that follows, that

\[
\tan \left( \frac{\pi}{4} - \frac{\eta}{2} \right) = \frac{\varepsilon}{1 + \varepsilon}.
\]  

(1)

Under the application of addition theorems the following holds

\[
\tan \left( \frac{\pi}{4} - \frac{\eta}{2} \right) = \frac{\tan \frac{\pi}{4} - \tan \frac{\eta}{2}}{1 + \tan \frac{\pi}{4} \cdot \tan \frac{\eta}{2}} = \frac{1 - \tan \frac{\eta}{2}}{1 + \tan \frac{\eta}{2}},
\]  

(2)

we receive:

\[
\frac{1 - \tan \frac{\eta}{2}}{1 + \tan \frac{\eta}{2}} = \frac{1 - \nu}{1 + \nu}.
\]  

(3)

For small deformations the following approximation holds

\[
\tan \frac{\eta}{2} \approx \frac{\eta}{2}.
\]  

(4)

and from that equation (3) becomes

\[
\frac{1 - \frac{\eta}{2}}{1 + \frac{\eta}{2}} = \frac{1 - \nu}{1 + \nu}.
\]  

(5)

If the products \( \varepsilon \cdot \eta \) are neglected for being terms of small order, than the following is obtained

\[
\eta = \varepsilon (1 + \nu).
\]  

(6)

In the region of linear elasticity (Hooke's Law) is

\[
\frac{\eta}{\mu} \text{ and } \varepsilon = \frac{\sigma}{E}.
\]  

(7)
Where \( \tau \) is the shear stress, \( \mu \) the shear modulus, \( \sigma \) the normal stress and \( E \) Young's modulus. Equation (7) substituted into equation (6) delivers

\[
\frac{\tau}{\mu} = \frac{\sigma}{E} (1 + \nu) \tag{8}
\]

At the uniaxial stress state the main shear stress \( \tau \) is equal to the half normal stress \( \sigma \) (Mohr's stress circle). From that the following is obtained

\[
\tau = \frac{1}{2} \sigma \tag{9}
\]

and using equation (8)

\[
\frac{\mu}{E} = \frac{1}{2 (1 + \nu)} \tag{10}
\]

This is the well known approximation of the relation between Young's modulus and the shear modulus, which is also obtained from the stress-strain tensor according to

\[
\sigma_{ik} = 2 \mu \varepsilon_{ik} + \lambda \theta \delta_{ik}, \tag{11}
\]

with the Lamé's constants

\[
\mu := \frac{E}{2 (1 + \nu)} \quad \lambda = \frac{E \nu}{(1 - 2 \nu) (1 + \nu)}, \tag{12}
\]

\[
\theta = \frac{1 - 2 \nu}{E} \left( \sigma_1 + \sigma_2 + \sigma_3 \right); \tag{12}
\]

\[
\delta_{ik} = \begin{cases} 1 & \text{for } i = k \\ 0 & \text{for } i \neq k \end{cases}, \text{ Kroneckersymbol},
\]

where \( \sigma_i \) (\( i = 1, 2, 3 \)) are the three main stresses.

For the extended relation,

\[
\frac{1 - \tan \frac{\alpha}{2}}{1 + \tan \frac{\alpha}{2}} = \frac{1 - \nu \varepsilon}{1 + \varepsilon} = \frac{1 - \gamma}{\sqrt{1 - \gamma^2}} \tag{13}
\]

the last term is brought into a series expansion

\[
\frac{1 - \gamma}{\sqrt{1 - \gamma^2}} = 1 - \gamma \left[ 1 + \frac{1}{2} \gamma^2 + \frac{3}{8} \gamma^4 + .... \right], \tag{14}
\]

neglecting all \( \gamma \)-terms of higher orders one obtains

\[
\frac{1 - \nu \varepsilon}{1 + \varepsilon} = 1 - \gamma. \tag{15}
\]

The introduction of the strain invariant delivers
\[
\frac{1 - v}{2} = 1 - \gamma \quad \text{or} \quad 2\gamma = 1 + v. \tag{16}
\]

If equation (14) is applied the solutions are within a unit circle. The invariable is outside. Therefore the outer region is mapped into the internal of the unit circle. For that in equation (16) \(\frac{1}{1 + v}\) has to be used instead of \(1 + v\) and form that one obtains

\[
\gamma = \frac{1}{2 (1 + v)},
\]

which is the approximation analogue to equation (10).

From that it holds

\[
\gamma = \frac{1}{E} = \frac{1}{2 (1 + v)}. \tag{17}
\]

With the determination of \(\gamma\) in the experiment the Poisson's number \(v\) is determined. The fact, that the ratio \(\frac{\tan \theta'}{\tan \theta} = \alpha = \frac{\tau_\alpha}{\tau_\epsilon}\); see Fig. 3, of the both time constants in reality is the ratio of two moduli, demands the introduction of a viscosity \(\eta^*\) in the models

\[
\frac{\tau_\alpha \cdot \eta^*}{\tau_\epsilon \cdot \eta^*} = \frac{M'}{M''} = \frac{\tan \theta'}{\tan \theta''} = \alpha \tag{18}
\]

This also is to be seen as follows:

With \(O\) and \(O''\) as origins in Fig. 6 and with the angles \(PO''V = \theta''\); \(PO'V' = \theta'\), \(POO' = \rho\) the relation is received:

\[
\frac{1}{\tan \rho} = \frac{1}{\tan \theta'} - \frac{1}{\tan \theta''} = \frac{1 - \alpha}{\sqrt{1 - \alpha^2}} = \frac{1 - \gamma}{\sqrt{1 - \gamma^2}}; \quad \tag{19}
\]

Because \(\tan \rho = M\), it follows that \(\tan \theta'\) and \(\tan \theta''\) have to be moduli too.
Fig. A  Scheme of a deformed cube-shaped body for the derivation of the Poisson-number
Fig. 1  Schematic view of the compact tension (CT)-specimen (upper part) and the three point bend type (3PB) specimen (lower part).
CT specimen ASTM E 399 with $B = w/2$ ($B$: thickness, $W$: width). the positive load $P$ is perpendicular to the crack propagation.
3PB specimen with span $s = 4w$ (ASTM E 399) with $B = W/2$. The negative force $P$ is antiparallel to the direction of crack propagation.
Fig. 2 Description of the models
(a) Serial connection of a spring and an attenuator (Newtonian dashpot)
(b) Parallel connection of a spring and an attenuator (dashpot), introduced by Kelvin-Voigt
(c) Combination of the relaxation model (a) and the retardational model. Standard linear solid.
Load versus load point displacement diagram for the calculations of \( J_0 = J_{pl} \) value according to ASTM E-813.

The diagram represents the distribution of the defined deformational shift \( OP \) and its decomposition into three parts by a projection to \( OP \). [See also Appendix A]

\( V \) denotes the point of crack initiation.
Fig. 4  Principle of the "Lecher-Wiring". The paths of the electrical and magnetical fields, indicated by \( E \) and \( H \), respectively, form geodetical lines of orthogonally cutting circles. The wires can be regarded e.g. as charge and mirror charge over a mirror plane which reveals the same potential situation.
Fig. 5  Load versus load point displacement diagram. The point \( V^o \) on the elastic-plastic curve is an equilibrium value. The conformal mapping at the unit circle around \( O \) with the radius of \( \sigma_{\text{max}} = 1 \) represents radii with the abscissa \( x \) as real axis. \( \overline{OP} = Z \), \( \overline{OR^*} = -\frac{1}{2} \) is in the interior of the unit circle with \( |\overline{OR^*}| = \gamma \). Both vectors \( Z \) and \( Z^* \) passes the unit circle in \( V \left(-\gamma, \sqrt{1-\gamma^2}\right) \) and \( R \left(\gamma, \sqrt{1-\gamma^2}\right) \) on the \( \sigma_{\infty} \) level. The line \( VR \) cuts the experimental elastic-plastic curve in \( V^o (= P^o) \).
Fig. 6  Geometric presentation of equations (2.2.34) to (2.2.46) in the hyperbolic plane inside of the unit circle around \( \bar{O} \) for \( \gamma = \frac{\pi}{2} \). The triangle \( \overline{OM\bar{O}} \) allows a clear picture between trigonometric and hyperbolic functions. The strophoid \( OP*\overline{OP**} \) cuts the geodetic circle \( \eta = \overline{OM} = \sinh \varphi \) at \( P^* (-\gamma, \sigma_1) \) and \( P^{**} (+\gamma, \sigma_2) \), \( \tan \delta' / \tan \delta'' = \gamma \).
Fig. 7  Relationship between different trigonometric and hyperbolic functions. All different functions can be taken immediately from the triangle.
Fig. 8  \( K_{IC} \)-values as a function of temperature for the material A 533 B1

\( \bigcirc \)  E.T. Wessel et al., taken from Debray et al. (1971)

\( \blacksquare \)  E.T. Wessel, taken from P. Suter, E. Macherauch (1975), (B = 300 mm).

\( \triangle \)  derived from own measurement performed on small CT-specimens (B = 25 mm).
**Experimental Procedure**

- Determination of the Point of Initiation by a selected Method
  - $\gamma = \sqrt{1 - \left( \frac{P_i}{P_{\text{max}}} \right)^2}$
  - Calculate Ko, Ko$^\prime$, (3.2), (3.3) or J due to ASTM

**Theoretical Procedure**

- Experimental Input:
  - $P_{\text{max}}$, (a directly) of the Load vs. Displacement

- $\psi = \frac{P_{\text{max}} f \left( \frac{a}{w} \right)^2 (w-a)}{B w M u \{f\}}$

- $\gamma^4 + (\psi^2 - 1)\gamma^2 \pm 2\psi^2\gamma + \psi^2 = 0$

- forms a Strophoide
  - either
  - \begin{align*}
  \psi_{1,2}^2 &= \frac{\gamma^2(1-\gamma)}{(1 \pm \gamma)^2}
  \end{align*}

- or

- Go into the Nomogram Fig. 6

- $\gamma^2 = 1 - \left( \frac{P_i}{P_{\text{max}}} \right)^2$

- $\gamma = \sqrt{1 - \left( \frac{P_i}{P_{\text{max}}} \right)^2}$
Errata:

Page 2 \[ \beta = \frac{\tan \phi''}{\tan \phi'} = \frac{M_r}{M_u} \]

\[ \sigma(t) = \sigma_\infty + (\sigma_0 - \sigma_\infty) \exp \left( -\frac{t}{\tau_c} \right)^\infty \int_0^\infty F(t) d\tau_c \]

Appendix:

Page A1: Reference

Page 8 \[ \psi_1 = \gamma e^{-R_0} = \frac{\gamma(1-\gamma)}{\sqrt{1-\gamma^2}} \quad (2.2.43) \]
SESSION D

VARIATIONS IN IRRADIATION CONDITIONS AND MECHANISMS-MODELLING
Radiation damage structure in irradiated and annealed 440 WWER reactor pressure vessel steels

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to be presented

at the

Joint IAEA/NEA Specialists' Meeting on Irradiation Embrittlement and Optimization of Annealing

Paris, France, 20-23 September 1993
RADIATION DAMAGE STRUCTURE IN IRRADIATED AND ANNEALED 440 WWER-TYPE REACTOR PRESSURE VESSEL STEELS

by
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Nuclear Research Institute Řež plc; 250 68 Řež near Prague; Czech Republic

ABSTRACT

Conventional Transmission Electron Microscopy has clearly revealed several characteristic features of radiation damage structure in WWER-type RPV steels neutron irradiated in both, the research and the power light-water reactor. The visible part of radiation-induced defects population in power reactor surveillance specimens consists of very fine vanadium carbide precipitates, small dislocation loops and black dots. Black dots probably correspond (beside unresolvable defects like loops and precipitates) to clusters and particle embryos formed from vacancies and solute-atoms (vanadium, copper, phosphorus) and carbon associated with vanadium. The differences in damage structure of both, the surveillance and in research reactor irradiated specimens, where only hardly discernible black dots were observed, are probably associated with a low flux effect.

Radiation-induced defects are concentrated to dislocation substructure during irradiation in a power reactor, revealing the role of radiation-enhanced diffusion in damage structure forming processes. Contrary to that, the distribution of defects resulting from annealing of specimens irradiated in the research reactor is predetermined by a homogeneous distribution of radiation-induced defects prior to annealing. Increasing the number of re-irradiation and annealing cycles, the amount of dislocation loops among all defects seems to be growing. Simultaneously, the dislocation substructure recovers considerably.

Keywords: reactor pressure vessel steels, radiation damage, dislocation substructure, annealing

1. INTRODUCTION

Neutron irradiation results in significant changes in mechanical properties and the microstructure of reactor pressure vessel (RPV) steels. The degradation of mechanical properties is manifested as an upward shift in ductile to brittle transition temperature, associated with upper shelf toughness reduction, and an increase in the tensile yield strength, accompanied by a decrease in ductility. These effects - referred to as irradiation embrittlement - are generally attributed to the development of a fine scale radiation-induced defects which impede the dislocation motion under an applied stress. Defects are formed from vacancies and interstitials created in and surviving the collision cascades processes; point defects migrate freely through the crystal lattice at the service temperature, interact
each other, with solute atoms in matrix and also with dislocation substructure and precipitates, resulting in the formation of dislocation loops, atmospheres, clusters and precipitates. Generally, two - matrix and solute-related - components contribute to the radiation damage structure of RPV steels. Matrix component is produced by the coalescence of point defects, the solute-related component by the radiation-enhanced diffusion of solute-atoms and their precipitation from solid solution.

Dimensions of radiation-induced defects are less then ≈10 nm in RPV steels. Consequently, a wide range of techniques for microstructural characterization of irradiated RPV steels has been developed and applied to RPV steels of western provenance, like transmission electron microscopy [1,2], field ion/atom probe microscopy [3], small angle neutron scattering [4], positron annihilation spectroscopy [5], extended X-ray absorption fine structure analysis [2] and other methods, including also molecular dynamics computer simulations [2]. Inter-correlations of data from different techniques provide a complete characterization of radiation damage structures (RDS) allowing a modelling of detrimental radiation-induced processes in RPV steels [6].

Transmission electron microscopy (TEM) provides useful informations of number density, size and distribution of radiation-induced defects with diameter above the visibility limit of ≈2nm. This paper is intended to provide a review of a radiation damage structure of WWER-type RPV steels based on a conventional TEM investigations carried out at NRI Řež plc [7-9].

2. MATERIALS AND EXPERIMENTAL

The Cr-Mo-V ferritic steel of the 15Kh2MFA type is employed for the RPV of the WWER-440 light-water reactor. The smooth ring in the vicinity of the active core of the reactor is made from the steel of the grade AA of higher purity. Nominal chemical composition of both the base metal and weld metal (made from welding alloy Sv10KhMFT and flux agent AN 42) is given in Table 1 [10].

![Table 1: The nominal chemical composition of steels for RPV of WWER - 440 reactor](image)

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>P</th>
<th>S</th>
<th>Ni</th>
<th>Cr</th>
<th>Mo</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>15Kh2MFA base metal</td>
<td>.13</td>
<td>.30</td>
<td>.17</td>
<td>max.</td>
<td>max.</td>
<td>2.5</td>
<td>.60</td>
<td>.25</td>
<td></td>
</tr>
<tr>
<td></td>
<td>.18</td>
<td>.60</td>
<td>.37</td>
<td>.025</td>
<td>.025</td>
<td>.40</td>
<td>3.0</td>
<td>.80</td>
<td>.35</td>
</tr>
<tr>
<td>15Kh2MFA weld metal</td>
<td>.04</td>
<td>.60</td>
<td>.20</td>
<td>max.</td>
<td>max.</td>
<td>1.2</td>
<td>.35</td>
<td>.10</td>
<td></td>
</tr>
<tr>
<td></td>
<td>.12</td>
<td>1.3</td>
<td>.60</td>
<td>.042</td>
<td>.035</td>
<td>1.8</td>
<td>.70</td>
<td>.35</td>
<td></td>
</tr>
</tbody>
</table>

Cu content: 0.15, in AA grade materials 0.08 wt.%, (AA grade specify <0.015 S, 0.005 Sn and 0.005 Sb).
The maximum content of P and S in AN-42M flux agent is .012 nad 0.015 wt.%, respectively.
Basic heat treatment and technological annealing regimes during manufacturing of a pressure vessel are following: 1000 °C/oil + 680 to 720 °C/air and then 665 °C/31 to 90 hours/furnace cooling, resulting in the bainite-ferrite microstructures with bainite prevailing in a base metal and ferrite in weld metal, respectively [9].

The samples were irradiated in an inert gas-filled capsules located either in the Chouca-rig on the LVR-15 research reactor at Řež or at position for surveillance specimens in vertical channels welded to the outer surface of the core barrel of the WWER-440 reactor on the nuclear power plant. The lead factor is high, amounting the value \( \approx 10 \), in the later case because of the high flux gradient in WWER-440 reactors. The irradiation temperature \( \approx 275 \) °C was considered to be \( \approx 10 \) °C above that of the inlet water; in research reactor irradiations, the irradiation temperature was measured by thermocouples attached to the rig. Irradiation conditions are briefly summarized in Table 2.

**Table 2:** Irradiation conditions

<table>
<thead>
<tr>
<th>Reactor</th>
<th>WWER-440</th>
<th>LWR-15</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron flux (E &gt; 1 MeV), ([\text{m}^{-2}\text{s}^{-1}])</td>
<td>(5 \times 10^{14}-1.9 \times 10^{16})</td>
<td>(\approx 1.8 \times 10^{17})</td>
</tr>
<tr>
<td>Neutron fluence * / ([\text{m}^{-2}])</td>
<td>(2 \times 10^{22}-2.5 \times 10^{24})</td>
<td>(7 \times 10^{22}-2.6 \times 10^{24})</td>
</tr>
<tr>
<td>Irradiation temperature ([\degree\text{C}])</td>
<td>(\approx 275)</td>
<td>(\approx 300)</td>
</tr>
<tr>
<td>Irradiation time</td>
<td>1 - 5 years</td>
<td>2 - 3 weeks</td>
</tr>
</tbody>
</table>

*S*Smallest fluences correspond to the periphery of surveillance specimen chains

Note: Coefficients allowing convert the neutron fluxes and fluences for E > 0.5 and E > 0.1 MeV are 1.8 and 3, respectively.

Thin foils for TEM were prepared from samples cut from fractured Charpy-V specimens using Struers twin-jet electropolisher modified to low-temperature application. The conventional transmission electron microscope Tesla BS-540 operated at 120 kV was used for investigations, allowing a resolution of 2 - 3 nm for ferromagnetic materials.

3. RESULTS

3.1. EFFECT OF NEUTRON FLUENCE AND FLUX

There are differences in RDSs investigated by TEM in specimens irradiated at surveillance positions in a WWER-440 NPP and those irradiated in the research reactor at
Řež. Long-term expositions in the power reactor provide the diffusional processes to facilitate radiation-induced defects to grow to a visible scale.

The threshold neutron fluence allowing defects to be observed directly by TEM depends on both, the neutron fluence and the neutron flux. Thus, the conventional TEM failed to reveal radiation-induced defects in specimens irradiated in the LVR-15 reactor with the neutron flux of \(1.8 \times 10^{17} \text{m}^{-2}\text{s}^{-1}\) to a neutron fluence of \(\approx 1 \times 10^{23} \text{m}^{-2}\) (\(E > 1 \text{ MeV}\)) [7] probably because they are too small to be readily seen.

In surveillance specimens irradiated in the power reactor by the very low neutron flux (\(\approx 5 \times 10^{14} \text{m}^{-2}\text{s}^{-1}\), \(E > 1 \text{ MeV}\)) to a fluence of \(\approx 2 \times 10^{22} \text{m}^{-2}\) very small defects just become visible [9], after irradiation to a neutron fluence of \(\approx 8 \times 10^{22} \text{m}^{-2}\) a few clearly discernible defects are seen in the bainite (base metal), Fig. 1, and an abundant defect population is found in the ferrite (weld metal), Fig.2. This damage may represent a half of anticipated RPV end-of-life damage structure approximately. The irradiation with a neutron flux \(\approx 1.6 \times 10^{16} \text{m}^{-2}\text{s}^{-1}\) revealed a well developed defect structure since the first year of irradiation (fluence \(4.3 \times 10^{23} \text{m}^{-2}\)) [7-9], Fig. 3.

3.2. POWER REACTOR IRRADIATION

Most of radiation-induced defects appear as "black-dots", < 8 nm in size, representing point defect clusters, unresolvably small dislocation loops and very fine precipitates. At higher fluences the defects > 10 nm in size, can be resolved as small dislocation loops or fine precipitates, the total amount of defects is increasing function of the neutron fluence, Figs. 3 - 6. Particularly, defect populations involve the increased amount of dislocation loops after irradiation to higher fluences. However, a unique identification of the defect nature is very complicated and only rarely successful due to superposition of numerous disturbing experimental and material factors. It seems that both, the clusters and dislocation loops may act as new particles nucleates. No cavities or microvoids were ever found (even at favourable diffraction conditions).

In addition to visible defects, a vacancy-rich component non-visible by TEM was confirmed by positron annihilation spectroscopy [11].

Arrangement of visible defects appear to be heterogeneous. Defects concentrate first to dislocations, in particular to dislocation segments within well recovered (polygonized) areas, and also to the original precipitates; very often but not always defects nucleate from one side of a dislocation line only, Figs. 3 and 4.

Similarly, low‐angle lath boundaries are in abundance decorated by radiation‐induced defects. On the other hand the high‐angle boundaries (prior austenite, bainite packets) which are free of visible defects seem to be less favourable centers for an extended defect nucleation.

High density defect clusters, \(\approx 100 \text{ nm} \) in size, are seen at fluences higher than \(1 \times 10^{24} \text{m}^{-2}\), Fig. 5. Defects appear also within precipitate free zones observed in some ferritic grains of weld metal prior to irradiation.

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Number density and average diameter of radiation-induced defects slowly increase with increasing neutron fluence, amounting to $4 \times 10^{21}$ m$^{-3}$ and $10$ nm, respectively, at a neutron fluences above $1 \times 10^{24}$ m$^{-2}$, see Table 3.

Table 3: Quantitative characteristics of radiation-induces defects * in surveillance specimens ($N_v$ - number density, d - diameter)

<table>
<thead>
<tr>
<th>Neutron fluence (E&gt; 1 MeV) [m$^{-2}$]</th>
<th>$N_v$ [m$^{-3}$]</th>
<th>d [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2 \times 10^{22}$</td>
<td>?</td>
<td>&lt; 4</td>
</tr>
<tr>
<td>$5 \times 10^{23}$</td>
<td>$2.7 \times 10^{21}$</td>
<td>4.8</td>
</tr>
<tr>
<td>$9 \times 10^{23}$</td>
<td>$3 \times 10^{21}$</td>
<td>5.0</td>
</tr>
<tr>
<td>$1.2 \times 10^{24}$</td>
<td>$3.6 \times 10^{21}$</td>
<td>9.4</td>
</tr>
<tr>
<td>$2.5 \times 10^{24}$</td>
<td>$3.9 \times 10^{21}$</td>
<td>11.7</td>
</tr>
</tbody>
</table>

* Values measured in bainitic structure have only limited validity due to substantial experimental scatter caused by heterogeneously distributed defects, defect contrast and visibility, unknown foil thickness, etc.

A weak recovery of the dislocation substructure observed in irradiated specimens is revealed by a small dislocation density decrease (from $3 \times 10^{14}$ m$^{-2}$ prior irradiation to $1 \times 10^{14}$ m$^{-2}$ after irradiation), by formation of dislocation cells and by higher degree of a dislocation network perfection [7].

As the dislocation substructure in ferritic grains of the weld metal is more simple than in the bainite of the base metal, the population of radiation-induced defects reveal more clearly in the weld metal irradiated equally to the base metal, Figs. 2, 7 and 8.

Along with microstructural changes, changes of microchemistry may occur. For example, a refinement of VC precipitates in ferritic grains of the weld metal was found in [7], and the fringe contrast suggestive of stacking faults or thin planar precipitates was observed within some ferrite grains in the base metal [9]. It cannot be excluded, that the appearance of high density defect clusters, mentioned above, can be associated with the disintegration of original carbide particles within a bainitic microstructure.

3.3. RESEARCH REACTOR IRRADIATION

The neutron fluences, energy spectrum and irradiation temperature are supposed to be the same, but the neutron fluxes differ by the range of magnitude in favour of research reactor.
No clearly discernible radiation-induced defects were observed in both, the base metal and the weld metal up to the neutron fluence $= 1 \times 10^{24} \text{ m}^{-2}$ is reached. Defects become unlikely to be visible even at neutron fluence $= 5 \times 10^{23} \text{ m}^{-2}$ corresponding to the projected RPV end-of-life fluence.

3.4. ANNEALING AND REIRRADIATION

After post-irradiation annealing 475 °C/168 h (of the weld metal) the defects resembling radiation-induced defects appear, Fig. 9. However, these defects probably create from radiation-induced defects with diameter below the visibility limit, homogeneously distributed after the irradiation.

Resolvable radiation defects appear in specimens re-irradiated after the annealing. Increasing the number of irradiation and annealing cycles up to three, the amount of dislocation loops among all defects seems to be growing, Fig. 10. Simultaneously, the dislocation substructure recovers considerably, Fig. 11.

4. DISCUSSION

The 15Kh2MFA steel employed in 440 WWER nuclear reactors differs from the western steels mainly in its nickel, chromium and vanadium contents; last two elements are strong carbide-forming agents. The nature of radiation-induced defects in WWER steel is not yet clear enough, since only limited amount of knowledge based on TEM, and indirect methods of positron spectroscopy [12] and small-angle neutron scattering is available [13]; atom probe field-ion microscopy is missing entirely.

Similarly to the western types of RPV steels, the radiation damage structure of WWER steels consists of broad spectrum of radiation-induced defects, from small to ultra-fine in size. It is believed that in TEM seen black-dots represent (beside unresolved contrast of small dislocation loops and vanadium carbides particles) also vanadium and carbon rich clusters or vanadium carbide precipitate embryos in addition to copper precipitates and phosphorus clusters. This interpretation is not in contradiction with positron annihilation spectroscopy and small angle neutron scattering results [11-13]. Thus, it is likely to incorporate the vanadium carbide-type features (precipitates, clusters, atmospheres) separately into the model of radiation embrittlement for WWER-type steels. However, some part of the radiation-induced defect population is still less than the resolution limit of the electron microscope ($\approx 2 \text{ nm}$). Nevertheless, the combination of techniques, including field ion microscopy has to be employed to obtain a complete characterization of radiation-induced population of defects in WWER steels.

Increasing values of the defect number density with increasing neutron fluence coincide with the observed yield stress shifts [8]. However, the measured values of the defect number density are lower than the density required to explain the observed $\approx 70 \text{ MPa}$ increase in yield stress. Thus, the ultra-fine defects non-accessible to TEM have to be also
taken into account considering the radiation hardening phenomena.

A heterogeneous distribution of defects indicates the key role of a long-range diffusion in a radiation-induced evolution of the microstructure in specimens irradiated in a power reactor. The long irradiation time assists at diffusional processes and annealing of defects at temperature \( \approx 275^\circ C \), allowing them to grow to dimensions accessible to TEM (in contrast to the accelerated irradiation conditions). Other two factors, neutron flux and neutron energy spectrum, can also contribute to the growth of defects [14]. The both, lower displacement rate and reasonably softened neutron spectrum (due to water reflector) in comparison to the research reactor conditions favour survival of a higher fraction of point defects capable at a later stage to form extended defects visible by TEM.

Similarly, defects which begins to occur in the course of annealing after short-term irradiation in the research reactor corroborate the role of diffusion processes in the RDS evolution. The homogeneous distribution of defects formed in this case is predetermined by homogeneously distributed defect nucleae created during previous irradiation.

The radiation response of both ferrite and bainite microstructures is generally the same. However, some differences appear within the zones attached to ferrite grain boundaries in base metal and weld metal. In addition to that some segregation/precipitation phenomena, manifested by a fringe contrast appearance may occur in ferrite.

5. CONCLUSIONS

Conventional TEM results have clearly revealed several characteristic features of radiation damage structure in 440 WWER steels neutron irradiated in both, the research and the power nuclear reactor. The visible part of radiation-induced defects population consists of small dislocation loops, very fine vanadium carbide precipitates, and black dots. Black dots probably correspond (beside unresolvable defects like loops and precipitates) to clusters and particle embryos created from vacancies and solute-atoms (vanadium, copper, phosphorus) and carbon associated with vanadium. Defects are concentrated predominantly to dislocation lines and low-angle boundaries.

Radiation damage structure arising in the RPV steel at the reactor operating temperature depends on both, the neutron fluence and the neutron flux. The differences in damage structures of specimens irradiated in the power as well as the research reactor are associated with different fluxes. The lower is the neutron flux, the more clearly revealed is the role of radiation-enhanced diffusion in damage structure forming processes.

The distribution of defects resulting from annealing of specimens irradiated in the research reactor is predeterminte by a homogeneous distribution of radiation-induced defects prior to annealing.

REFERENCES


CAPTIONS FOR FIGURES:

Fig.1. Base metal, power reactor irradiated with a neutron flux ≈ 6×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 8×10^23 m⁻².

Fig.2. Weld metal, power reactor irradiated with a neutron flux ≈ 6×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 8×10^23 m⁻².

Fig.3. Base metal, power reactor irradiated with a neutron flux ≈ 1.3×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 4×10^23 m⁻².

Fig.4. Base metal, power reactor irradiated with a neutron flux ≈ 1.5×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 1×10^24 m⁻².

Fig.5. Base metal, power reactor irradiated with a neutron flux ≈ 1.7×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 1.3×10^24 m⁻².

Fig.6. Base metal, power reactor irradiated with a neutron flux ≈ 1.9×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 2.5×10^24 m⁻².

Fig.7. Weld metal, power reactor irradiated with a neutron flux ≈ 1.6×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 7.3×10^24 m⁻².

Fig.8. Weld metal, power reactor irradiated with a neutron flux ≈ 1.9×10^14 m⁻² s⁻¹ to a neutron fluence of ≈ 2.5×10^24 m⁻².

Fig.9. Weld metal, research reactor irradiated with a neutron flux ≈ 1.8×10^15 m⁻² s⁻¹ to a neutron fluence of ≈ 2.1×10^25 m⁻² and annealed 475 °C/168 hours.

Fig.10. Weld metal, three cycles of research reactor irradiation (with a neutron flux ≈ 1.8×10^17 m⁻² s⁻¹ to a neutron fluence of ≈ 2.1×10^25 m⁻²) and annealing (475 °C/168 hours).

Fig.11. Weld metal, three cycles of research reactor irradiation (with a neutron flux ≈ 1.8×10^17 m⁻² s⁻¹ to a neutron fluence of ≈ 2.1×10^25 m⁻²) and annealing (475 °C/168 hours).
MICROSTRUCTURAL CHARACTERIZATION OF ATOM CLUSTERS IN IRRADIATED PRESSURE VESSEL STEELS AND MODEL ALLOYS

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ABSTRACT. In order to characterize the microstructural evolution of the iron solid solution under irradiation, two pressure vessel steels irradiated in service conditions and, for comparison, low copper model alloys irradiated with neutrons and electrons have been studied. The characterization has been carried out mainly thanks to small angle neutron scattering and atom probe experiments. Both techniques lead to the conclusion that clusters develop with irradiations. In Fe-Cu model alloys, copper clusters are formed containing uncertain proportions of iron. In the low copper industrial steels, the feature is more complex. Solute atoms like Ni, Mn and Si, sometimes associated with Cu, segregate as "clouds" more or less condensed in the iron solid solution. These silicides, or at least Si, Ni, Mn association, may facilitate the copper segregation although the initial iron matrix contains a low copper concentration.

Keywords: pressure vessel steel, FeCu alloys, neutron and electron irradiations, microstructural characterization, small angle neutron scattering, atom probe, copper precipitates, copper-iron clusters, Si-Ni-Mn-Cu rich clouds.

I INTRODUCTION

Pressure vessel steels used in pressurized water reactors are low alloyed ferritic steels. They may be prone to hardening and embrittlement under neutron irradiation. The changes in mechanical properties are generally supposed to result from the formation of point defects, dislocation loops, voids and/or copper rich clusters. However, the real nature of the irradiation induced-damage in these steels has not been clearly identified yet.

In order to improve our vision of this damage, we have characterized the microstructure of steels irradiated in the French surveillance programme, i.e. materials having a chemical composition, a structure and irradiation conditions very similar to those of the vessel core zone. We present here the results obtained on two steels irradiated in CHOOZ A and DAMPIERRE 2.

Since copper is known to play an important role in the irradiation embrittlement, we have also studied some Fe-Cu model alloys irradiated with neutrons or with electrons. For comparison, a model alloy has also been characterized after thermal ageing.
The microstructural characterization has been carried out by Transmission Electron Microscopy (TEM), Positron Annihilation (PA), Small Angle Neutron Scattering (SANS) and Atom Probe Field Ion Microscopy (APFIM).

II STUDIED MATERIALS

a) Steels from surveillance programmes

Both steels from surveillance programmes are 16 MND 5 type; they are representative of the C shells of the two reactors. Their chemical compositions are given in table 1.

The steel from CHOOZ has a ferrito-bainitic structure. The samples used in the study were irradiated during up to 13 years with a fluence of $1.4 \times 10^{20}$ n.cm$^{-2}$ (all the fluences are given for neutrons with energy higher than 1 MeV), which corresponds to a dose of about 0.2 dpa. The irradiation temperature was 255°C until 1970 and then 265°C (1).

The steel from DAMPIERRE is entirely bainitic. It has been irradiated during 9 years to a fluence of $4.6 \times 10^{19}$ n.cm$^{-2}$ ($\approx 0.08$ dpa) at a temperature of 290°C.

Table 1. Chemical compositions (wt%) of the CHOOZ A and of the DAMPIERRE 2 pressure vessel steels

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>S</th>
<th>P</th>
<th>Si</th>
<th>Cr</th>
<th>Mo</th>
<th>Mn</th>
<th>Ni</th>
<th>V</th>
<th>Al</th>
<th>Co</th>
<th>Cu</th>
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<tbody>
<tr>
<td>CHOOZ</td>
<td>0.16</td>
<td>0.006</td>
<td>0.012</td>
<td>0.32</td>
<td>0.16</td>
<td>0.39</td>
<td>1.26</td>
<td>0.57</td>
<td>0.020</td>
<td>0.024</td>
<td>0.02</td>
<td>0.09</td>
</tr>
<tr>
<td>DAMPIERRE</td>
<td>0.16</td>
<td>0.008</td>
<td>0.008</td>
<td>0.19</td>
<td>0.24</td>
<td>0.55</td>
<td>1.25</td>
<td>0.74</td>
<td>-</td>
<td>0.009</td>
<td>0.01</td>
<td>0.07</td>
</tr>
</tbody>
</table>

It is noticeable that the concentrations of reputed embrittling elements (P, Ni, Cu) are rather low in both steels. Nevertheless, the CHOOZ A steel is subject to an increase in the Charpy transition temperature as high as 145°C for a fluence of about $10 \times 10^{19}$ n.cm$^{-2}$. With a fluence $4.6 \times 10^{19}$ n.cm$^{-2}$, the same parameter rises of only 40°C for the DAMPIERRE 2 steel.

b) Binary Fe-Cu alloys

Three Fe-Cu alloys have been prepared with the following copper contents: 0.1 ; 0.7 ; 1.4 wt%. They have been irradiated in the OSIRIS pool test reactor of CEA/Saclay. The fluence received in the centre of the specimens is about $5.5 \times 10^{19}$ n.cm$^{-2}$ ($\approx 0.1$ dpa) with a rather high flux of $3 \times 10^{13}$ n.cm$^{-2}$. s$^{-1}$. The irradiation temperature was close to 290°C.

The binary alloys were also irradiated with electrons in a Van de Graaff accelerator (3 MeV). The maximal received fluence was $2 \times 10^{19}$ e$^{-}.\text{cm}^{-2}$ ($\approx 1.8 \times 10^{-3}$ dpa) at a temperature of 290°C. Electrons induce the formation of isolated point defects (interstitials and vacancies) and so can favour the clustering of solutes, as neutrons are supposed to do. However, after electron irradiation the materials are not radio-active, which makes the studies easier.
For both kinds of irradiations (electron and neutron), the induced-damage was characterized by Transmission Electron Microscopy (TEM) at EDF, Positron Annihilation (PA) at CEA/Grenoble, Small Angle Neutron Scattering (SANS) at Brookhaven National Laboratory or Laue Langevin Institute (in Grenoble) and Atom Probe Field Ion Microscopy (APFIM) with a energy-compensated instrument at University of Rouen (2).

The effects of electron and neutron irradiations on the mechanical properties were studied by hardness tests with a load of 5 N. In order to make some comparisons, hardness tests and APFIM experiments have also been performed on the Fe-0.7 wt%Cu alloy thermally aged 70 hours at 500°C.

III RESULTS

3.1 Steels from surveillance programmes

a) CHOOZ A pressure vessel steel

The results of TEM, PA and SANS experiments carried out on samples from CHOOZ have already been published elsewhere (3, 4, 5, 6) and can be summarized as follows: the steel has a ferrito-bainitic structure; some regions are fully bainitic, whereas some others contain about 60% of bainite and 40% of ferrite. The observed carbides are of M3C or M2C types. Conventional TEM has not allowed to detect any irradiation induced-defect such as atom clusters, cavities or dislocation loops. However, the dislocation density seems to be slightly smaller after irradiation (3). Positron annihilation has shown that neutron irradiation had not induced the formation of microvoids containing more than 50 vacancies (4).

SANS experiments (3, 5) have revealed the presence of irradiation induced defects. It was observed that their volume fraction increases as the fluence rises; however their radius (Guinier analysis) remains nearly constant (R = 1.3 nm), at least for fluences ranging between 2.4 and 14 \(10^{19}\) n.cm\(^{-2}\). Complementary SANS measurements with magnetic field have shown that the ratio of the intensities scattered parallelly and perpendicularly to the applied field (so called A ratio) is about 2 (± 1). Hence, it can be ruled out that the irradiation induced defects are pure copper clusters (A = 11). The unambiguous determination of the composition of the scattering centres required complementary studies by APFIM.

The APFIM investigations were carried out on the steel from CHOOZ in non irradiated and irradiated states (fluence : \(1.10^{20}\) n.cm\(^{-2}\)). The first experimental evidence is the absence of particular contrast on FIM micrographs, even for the irradiated sample. For this reason, only atom probe analysis from random areas has been performed.

It was observed that the irradiation had induced the formation of a high density (\(1\ 10^{18}\) cm\(^{-3}\)) of local enrichments of the iron solid solution in solutes like Si, Mn, Ni and Cu. The apparent size of these clusters ranges from 3 to 8 nm. Their solute concentrations are very low and they can be regarded as "clouds" of solute atoms more or less condensed in the iron solid solution (7, 8). A mean chemical composition of the observed clusters is given in table 2. If nickel, manganese and silicon seem to be systematically associated all together in the
clusters with a quasi $M_2Si$ stoichiometry ($M = Mn + Ni$)(7), copper has not always been observed. Nevertheless, analysis of the matrix reveals that 60% of the total copper atoms have segregated.

| Table 2: Mean concentrations of Ni, Mn, Si, Cu (at% ± 2 σ) in clusters and enrichment ratios (compared to the nominal composition of the ferrite) |
|---|---|---|---|---|
| Ni | Mn | Si | Cu |
| average at% | 3.6 ± 0.6 | 3.8 ± 0.7 | 4.8 ± 0.7 | 0.9 ± 0.3 |
| enrichment ratio | up to 12 | up to 6 | up to 12 | up to 45 |
| Fe | balance |

The internal constitution of one cluster has been investigated thanks to a plane by plane evaporation process (figure 1). It has been shown that the core of the cluster is mainly constituted of Ni, Mn and Si atom enrichment, whereas copper atoms are systematically gathered on one border. Rather than a unique cluster, this suggests the existence of two types of symbiotic ones.

From the chemical composition given in table 1, it is possible to calculate the A ratio that such defects would give in SANS experiments. Assuming that the magnetic contrast in the clusters is proportional to their iron content (which is possible for high iron contents) we find an A ratio of 2.1. This value is consistent with the experimental one (cf 3.1.a), which indicates that the defects observed by APFIM could be the same as those revealed by SANS experiments. It is likely that only the core of the clusters is dense enough in solutes to induce a neutron scattering. This may explain why the radius of the defects appear smaller after SANS measurements than after APFIM ones.

Figure 1.
Composition profiles of Fe, Si, Ni, Mn and Cu obtained during a (100) plane by plane evaporation sequence of an irradiation induced "cloud". 30 successive atomic planes have been evaporated. The identified atoms have been positioned in their order of arrival, therefore, their actual disposition inside each plane is unknown. The core of the "cloud" is Si, Ni, Mn rich, whereas copper is mainly gathered on the top border.
The results obtained by APFIM in the steel from CHOOZ give a picture of the irradiation induced-damage which is slightly different of the usually proposed scenarios (9-16). For this low copper highly irradiated steel, not only copper but also Ni, Mn and Si are concerned in the solid solution demixing process.

b) DAMPIERRE 2 pressure vessel steel

The DAMPIERRE 2 pressure vessel steel is slightly different from the previous one. More precisely, its copper and silicon concentrations are lower, conversely the nickel content is higher than for the CHOOZ A steel (table 1). The irradiation conditions are different too: the DAMPIERRE 2 steel has been submitted to a lower fluence with a weaker flux, but it has been irradiated at a higher temperature.

**SANS examination** (6) of the non irradiated and irradiated samples exhibit almost the same feature. In both cases, no apparent neutron scattering is observed.

**Atom probe random analyses** (8) of irradiated samples give no evidence of solute peaks on concentration profiles. However, small differences exist between solute distribution in irradiated and non irradiated samples. These differences can be revealed thanks to statistical tests like contingency tables (17) which allow to check the first stages of clustering. These tests show that after irradiation, solute atoms are no longer completely randomly distributed in the iron solid solution. There appears a trend to the formation of Si-Ni, Si-Mn or Si-Ni-Mn very small clusters of a few atoms of each species which do not exist in the non irradiated condition. The eventuality of copper segregating to these clusters is uncertain because of the high statistical error bar due to the tiny number of detected copper atoms.

3.2 *Irradiated Fe-Cu MODEL ALLOYS*

a) Results of hardness tests

The fluences of the neutron and electron irradiations are high enough for the hardness of each alloy to reach a plateau. For both irradiations, the evolution of the maximum hardness increase ($\Delta H_v$ for electrons and $\Delta H_v$ for neutrons) is plotted in figure 2 as a function of the initial copper content in the solid solutions. $\Delta H_v$ and $\Delta H_v$ evolve with the copper content according to parabolic laws. Whatever the copper content, it is noticeable that the hardening is higher after neutron than electron irradiation. This can be due to an effect of flux or to the difference of the primary damage induced by the particles: Frenkel pairs and cascades for neutrons and only Frenkel pairs for electrons.

b) Microstructural examinations

**SANS experiments** have been carried out on the alloys with 0.7 and 1.4% of copper after neutron irradiation (18). It was noticed that the intensity scattered at small angles clearly increases with the alloy copper content. The measured Guinier radii of the scattering centres are respectively 1.2 and 1.5 nm; their A ratios are 2.2 and 4.1. These values show that the defects are not pure copper clusters ($A = 11$).
Figure 2. Maximum hardness increase after neutron irradiation, electron irradiation or thermal ageing as a function of initial solid solution copper content.

On the APFIM micrographs of neutron or electron irradiated specimens, no well defined contrast is detected. This precludes selected area analyses to be carried out. Thus, with the hope of a sufficient number density of particles, random analyses have been performed.

Random Atom Probe analyses of the neutron irradiated samples with 0.7 and 1.4 wt% copper contents, revealed copper rich clusters having a non unique distribution of apparent sizes, which can suggest the existence of several populations or, more probably, the simultaneous presence of different stages of development of the same population. 20% of the smallest clusters are particularly copper rich and considering the large uncertainties, they could be pure copper particles.

In the electron irradiated Fe-0.1%Cu, no special event was detected. In the Fe-0.7%Cu, electron irradiation has induced the formation of small copper clusters. Their density appears much lower than after neutron irradiation.

For all irradiation conditions and copper contents, it appears that the iron solid solution is copper depleted down to about 0.1 at%. This concentration seems to be a limit under which irradiation (neutron or electron) cannot induce the clustering of copper atoms. This is likely why the Fe-0.1%Cu sample does not decompose after electron irradiation.

c) Analysis of the measurements

In order to study the effects of the copper content and to compare the two kinds of irradiations, we have treated all the experimental results through a same geometrical model (19) which assumes a random distribution of identical spherical precipitates.
Taking into account the uncertainties of the experimental measurements, this analysis has led to determine possible intervals of values for the copper content, the size, the density and the volume fraction of the "mean cluster" in each irradiated sample (8). The results are given in Table 2.

Table 2. Atom probe results on thermally aged and neutron or electron irradiated Fe-Cu model alloys.

<table>
<thead>
<tr>
<th>Material</th>
<th>State</th>
<th>Cu in the matrix (at%)</th>
<th>Cu in clusters (at%)</th>
<th>Diameter of clusters (nm)</th>
<th>Density of clusters (10^17 cm^-3)</th>
<th>Volume fraction of clusters (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe-0.7wt%Cu</td>
<td>Thermally aged</td>
<td>0.14±0.02</td>
<td>100</td>
<td>7±1</td>
<td>0.30-0.35</td>
<td>0.5-0.6</td>
</tr>
<tr>
<td></td>
<td>Electron irradiated</td>
<td>0.10±0.02</td>
<td>20-60</td>
<td>4.5-11.0</td>
<td>0.3-0.9</td>
<td>0.8-2.5</td>
</tr>
<tr>
<td></td>
<td>Neutron irradiated</td>
<td>0.08±0.02</td>
<td>15-30</td>
<td>4-6</td>
<td>3-7</td>
<td>1.8-3.6</td>
</tr>
<tr>
<td>Fe-0.1wt%Cu</td>
<td>Electron irradiated</td>
<td>0.09±0.02</td>
<td>*</td>
<td>*</td>
<td>*</td>
<td>*</td>
</tr>
<tr>
<td>Fe-1.4wt%Cu</td>
<td>Neutron irradiated</td>
<td>0.08±0.02</td>
<td>20-40</td>
<td>4-6</td>
<td>4-9</td>
<td>3.0-5.8</td>
</tr>
</tbody>
</table>

* No defect has been observed

From this analysis, it appears that the "mean clusters" may have rather high iron contents. We notice also that their volume fraction seems to increase with the copper content of the alloys.

The uncertainties are too high to observe any influence of the kind of irradiation (electron or neutron) on the chemical composition and size of the "mean clusters". However, the density and volume fraction of these latter appear higher after electron than neutron irradiations.

3.3 Thermally aged Fe-0.7 wt%Cu model alloy

In order to compare the clusters induced by electron or neutron irradiations with those due to thermal ageing, some analysis have been carried out on the Fe-0.7%Cu alloys aged at 500°C. At this temperature, copper has an extremely low solubility in iron and precipitates via a diffusion controlled mechanism. The samples were treated during 70 h so as to reach the maximum increase in hardness (ΔHV_th). The latter is plotted in figure 2 with two other values of peak hardness measured in iron copper alloys and given in the literature (21, 24). ΔHV_th also follows a parabolic law according to the Russel - Brown model (22). Whatever the copper content, we notice (18) that ΔHV_th < ΔHV_e < ΔHV_n.

Field Ion Microscopy (FIM) reveals the existence of the precipitates induced by the thermal treatment. They appear as well defined but scarce dark contrasts in the bright pole figure of the iron solid solution (figure 3). They have been analysed by the selected area analysis method. These techniques allow a higher precision on shape and chemical measurements than the random analyses carried out on the irradiated samples. The analysis on
the selected areas has been done with a constant lateral resolution of 0.5 nm. The results are given in table 2.

In this case, the precipitates are unambiguously composed of pure copper. Like after irradiation, the solid solution is copper depleted down to about 0.1 at%. By evaporating atomic planes one after one, we noted that the precipitates have a spherical or slightly ellipsoidal shape with a mean diameter of 7 nm and a number density of $3 \times 10^{16}$ cm$^{-3}$ (6). Thus, they are slightly larger and less numerous than the defects observed in the same alloys after electron or neutron irradiation.

Figure 3. Video recording field ion micrograph of the thermally aged Fe-0.7 wt%Cu sample. The dark area near the (132) pole of the bcc structure of the brightly imaged iron matrix is the image of the copper precipitate.

IV DISCUSSION

The results obtained in the steels irradiated in CHOOZ and DAMPIERRE are consistent. For these low copper steels, it appears that neutron irradiation induces a clustering of elements like Ni, Mn and Si (maybe accompanied by Cu?). The features detected in the steel from DAMPIERRE may be considered as the first stage of this process and the "clouds" encountered in the highly irradiated CHOOZ A steel can be regarded as a more developed one. New experiments on DAMPIERRE 2 samples have to be carried out in order to increase the statistical confidence. In addition, a kinetics study is in progress on CHOOZ A samples irradiated with different fluences in order to establish the successive stages of the clustering process.
In the irradiated Fe-Cu model alloys, we have shown that the irradiation-induced clusters present a non-unique distribution of size. We have noticed that the smallest ones were copper rich, but we were not able to decide if they were pure copper or not. However, the following results tend to show that most of the clusters (at least the largest ones) are not made of pure copper:

- the A ratio measured by SANS (A = 2.2 and 4.1) is lower than the value expected for pure copper defects (A = 11);
- the clusters do not exhibit any contrast on the FIM micrographs while pure copper precipitates obtained by thermal ageing have one;
- a geometrical analysis of the AP measurements revealed that the "mean cluster" in each sample contains an uncertain iron content.

This study shows that most of the particles induced by irradiation in a ferritic matrix (at least with damage lower than 0.2 dpa) look like clouds of solutes rather than real precipitates. The presence of defects composed of iron, copper atoms and vacancies has already been envisaged (for example ref. 24) in order to justify the values of A ratios obtained with SANS experiments on FeCu alloys.

It still has to be determined if the hardening and the embrittlement of steels under irradiation is mainly due to the presence of such clouds or if there is an other component due to point defects. An unambiguous answer to this question will need further heavy work. In order to test the hypothesis of vacancy clusters, eventually associated with copper, SANS measurements with an applied magnetic field, as well as PA experiments are in progress.

V CONCLUSION

We have shown that in ferritic industrial or model alloys most of the irradiation induced-particles look more like clouds of solutes than real precipitates. These clouds can be more or less condensed. In low copper steels irradiated in service conditions, they are composed of Ni, Mn, Si, and sometimes Cu. The segregation of this latter may be facilitated by Si, Ni, Mn associations formed in early stages of the irradiation.

The presence of vacancies or microvoids isolated or related to these atom clusters cannot be ruled out. In order to conclude on the existence of these features, SANS measurements with an applied magnetic field as well as positron annihilation experiments are in progress.

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RPV STEEL EMBRITTLEMENT: DAMAGE MODELING AND MICROMECHANICS IN AN ENGINEERING PERSPECTIVE

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ABSTRACT

A new, consolidated strategy for improved Light Water Reactor (LWR) pressure vessel surveillance is proposed. The methodology incorporates statistical fracture mechanics and damage modeling, while taking maximum advantage of the data generated by conventional surveillance practices. Available reconstitution and miniaturisation techniques allow to implement such strategy with minimum material inventory.

KEY WORDS: Radiation effects, pressure vessel steel, safety and regulation, surveillance program, impact and tensile tests, fracture toughness, reconstitution techniques.
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Figures 1 to 27
ABSTRACT

A new, consolidated strategy for improved Light Water Reactor (LWR) pressure vessel surveillance is proposed. The methodology incorporates statistical fracture mechanics and damage modeling, while taking maximum advantage of the data generated by conventional surveillance practices. Available reconstitution and miniaturisation techniques allow to implement such strategy with minimum material inventory.

1. INTRODUCTION AND EXECUTIVE SUMMARY

Regulatory requirements to insure adequate fracture toughness of Light Water nuclear Reactor Pressure Vessels (RPV) throughout their anticipated life rely on the \( C_n \) notch impact test of specimens aimed at being representative of the beltline materials. The temperature at which 41J is absorbed (TT41) is used to 'index' the crack initiation (\( K_{ic} \)) and arrest (\( K_{ia} \)) fracture toughness, two exponential functions of temperature \( T \). These functions are assumed to be universal for RPV steels, depending only on T-RTNDT, where RTNDT is a reference temperature characterizing the ductile-brittle transition temperature (DBTT). It is furthermore assumed that, during service, RTNDT increases as TT41. This methodology of US vintage [1][2], and adopted by many countries including Belgium, is summarized by Figure 1.

Regulation also refers to predictive correlations, by which the effect of service exposure on TT41 (thus on RTNDT) is described in function of neutron fluence and the steel composition - namely, its content of copper, nickel (USNRC Regulatory Guide 1.99 Revision 2 [3]) and phosphorus (See [4] for a review of phosphorus and tin influence). Whenever surveillance results exceed the upper bound of such correlations, the steel is considered an 'outlier': this may entail severe penalties when defining the margins to be applied for consistency with today's prevailing empiricism.

The Research and Development (R&D) efforts described here are primarily born from the need to address steel 'outlier' behaviour in a more scientific manner than outlined above. For the cases analyzed so far, it is found that the major underlying cause of apparent anomaly is the inadequacy of the 41J- \( C_n \) fix for toughness indexation. More generally, it is concluded that this indexation does introduce distortions and unwarranted scatter into present engineering correlations of embrittlement.

This central contention results from consistent application of current knowledge in the fields of "Damage modeling" and "Micromechanics". Fundamentally, the steel DBTT and brittle crack initiation fracture toughness are governed by the uniaxial flow properties, the microscopic fracture stresses and the size and spatial distribution of cracked particles. The influence of these properties can be assessed using the load-time traces detected by the instrumented \( C_n \) impact test, in combination with the conventional tensile test and with \( K_{ia} \) measurements at (minimum) one selected temperature (using small, i.e. surveillance-size specimens).
Recently, we have made use of C, traces and tensile data for a re-examination of damage mechanisms - applying the established Fisher et al. model [5] [6] to account for the role of irradiation-enhanced copper precipitation. This leads to confirm the anneal activation energies, the kinetics and the saturated intensity of three distinct hardening processes, previously suggested for the A302-B reference plate and its 'outlier' companion, the Yankee surveillance plate [7]. The same parameters fully reproduce the observations for a A533-B plate and two B&W Linde-80 welds irradiated in support of the BR3 vessel anneal: the anomalies plaguing the corresponding 41J shift data are entirely eliminated by the present, physically-grounded DBTT approach. This work will be reported separately.

Micromechanics has been successfully applied also to the reconstitution of 10x10x10mm remnants from broken C, specimens, and to the miniaturisation of the C, impact test. The results are found to be independently supportive of the preceding considerations.

2. GENERAL PHILOSOPHY OF BELGIAN SURVEILLANCE R&D PROGRAM

Micromechanics-based Indexation of Fracture Toughness.

Structural integrity evaluations require to know the fracture toughness of the material. Current reactor pressure vessel steel surveillance programs do not directly determine this property under service exposure, but rely on the C, notch impact test for its estimation. This is done through an indexation scheme (Fig.1) [1][2] based on unirradiated data (for relevant A302-B, A533-B, A508, ... melts): the observed invariance of the shape of the corresponding lower bound toughness-versus-temperature curves is assumed to remain applicable to irradiated steels, but there is a paucity of supportive experimental data and no consensus as to their interpretation (see for instance [8]). On another hand, as shown and explained in this paper, toughness indexation to a fixed level of absorbed energy or lateral expansion in the C, impact test may be significantly inadequate and induce erroneous conclusions, even at the licensing level, in some cases. Finally, the irradiation-induced shift of $K_{ic}$ is generally expected to exceed the corresponding shift of $K_{isc}$, which is in direct contradiction with the Regulatory implication (Fig.1) of equal shifts.

An example is given by Figure 2 for the high copper weld 73W comprehensively investigated by the USNRC/ORNL Heavy Section Steel Irradiation (HSSI) program [9] to [11]. (Here, the toughness data have been expressed as kJ/m² and the fitted lines are indicative only). The bottom part of the Figure is a preliminary illustration that the onset temperature TO of the upper shelf in the C, test does track well the irradiation effect on $K_{isc}$ and that the TO shift also is of similar magnitude as the Pellini NDT shift, while the $K_{isc}$ shift is indeed significantly larger.

A major reason for the current reliance on C, indexation is that measurements of crack initiation fracture toughness $K_{ic}$ (or $K_{isc}$, elasto-plastic tests) are expensive and cumbersome if to be considered 'valid' in the sense of existing standards [12] [13]. In principle, $K_{ic}$ should be determined by means of specimens having the full vessel thickness.
Large specimens have been irradiated only under accelerated conditions, i.e. in test reactors, at rather high neutron dose rates as compared to the ones in a PWR vessel. Also, this has been done for only a few base and weld metals [9],[14],[15].

A germinal view in the background of this program is that small test specimens, machined or reconstituted from broken surveillance remnants, can be used to measure $K_{ie}$ shifts in service, and, even more importantly, to measure irradiated $K_{ie}$ curves for one steel relative to another one.

This is so promising that a dedicated experimental project in cooperation with ORNL/USNRC and the VTT Metals Laboratory, Finland, is being initiated for demonstration.

But even so, the material inventory of implemented surveillance programs is generally too limited to derive statistically valid, size representative, complete $K_{ie}$-temperature curves from small specimens. Furthermore, for older plants, relevant baseline specimens or remnants are often not available at all. Last, but not least, it is important also to determine $K_{1a}$ not only for the analysis of accidents such as pressurized thermal shock, but also because the lower bound of $K_{1a}$ is essentially identical to the reference toughness $K_{1c}$ [1], which governs the determination of plant heat-up and cool-down restrictions.

One is thus compelled to be inventive and to try and salvage from the $C_\gamma$ test whatever information can be used in a more fundamental manner, in the fracture mechanics context. The $C_\gamma$, load-deflection diagram recorded in the instrumented version of the test allows just that, along two broad lines:

**Approach I. Transition Temperature Concept.**

The thrust here is to rely on an improved definition of the ductile-brittle transition temperature DBTT for toughness indexation along existing Regulatory lines. Thus, the only difference is that the 41J $C_\gamma$ shift is replaced by a more physically grounded concept adequate to index $K_{id}$ ($K_{1a}$) shifts. The assumption of shape invariance of the temperature dependency upon service exposure remains necessary.

The $K_{1a}$ shift is then derived by accounting for the strain rate effect estimated by comparing the uniaxial tensile properties to the $C_\gamma$ impact load diagram.

If the $K_{id}$ shift obtained in this way does significantly differ from the Regulatory, 41J $C_\gamma$ shift, it is requested that this be backed-up by 'Damage Modeling', an activity overviewed below, calling for consistency between yield strength increase, microcleavage fracture stress and DBTT shift data.

**Approach II. Micromechanics-based Fracture Toughness Indexation.**

The approach is to use advanced statistical micromechanics for the evaluation of small specimen $K_{ie}$ fracture toughness measurements, in combination with some enhancement of current, commercial surveillance test matrices. In this way, maximum advantage can thus be taken of existing data banks.
Analytical work is in progress to develop a statistical, critical stress model for slip-initiated cleavage fracture under Mode I loading, applicable to low-alloy ferritic RPV steels in the brittle-ductile transition range. The final validation and routine use will refer to a capability for finite element calculations, and encompass competition of cleavage with ductile stable crack growth. The starting point has been the well-known model by Ritchie, Knott and Rice (RKR model) [16][17]. A number of studies along similar lines can be found in the literature ([18] to [25]). The present approach is directly tailored to engineering lower bound toughness indexation, rather than being of fundamental scientific nature.

This leads to propose an improved fracture toughness surveillance strategy, whose ingredients are gathered on Fig. 3. A brief explanation is the most easily offered by referring to the original RKR model. This is done for illustration purposes only and does not aim at describing the statistical SCK-CEN micromechanical model under development.

The representation of the RKR model, adopted for this orientation outline, uses for simplicity [17] the asymptotic small scale yielding solution developed by Hutchinson and Rice and Rosengren (HRR) for the plane strain tension stress distribution around a stationary crack tip in a non-linear elastic material. Note that this solution does not account for the very significant effects of crack tip blunting. Strain hardening is approximated by the Ramberg-Osgood constitutive law, with hardening exponent N. The model is based on a local criterion for slip-initiated cleavage ahead of sharp cracks: it requires the maximum principal tensile stress $\sigma_{yy}$ to exceed a critical microscopic fracture stress $\sigma^*_f$ over a microstructurally significant distance $l^*$, or more appropriately over a suitable volume $V^*$, function of the nature and spatial distribution of "eligible" cleavage trigger particles (carbides, inclusions, ...) as well as of the spatial distribution of the stress-strain field ahead of the crack or notch of interest. This characteristic volume depends also on temperature.

This lower-transition toughness model can be synthetized by a single equation, sufficiently representative of physically meaningful ingredients for our present purposes:

\[
K_c = \sqrt{\beta(N)^{(N-1)}} \sqrt[4]{I} \frac{\sqrt{\sigma^*(N-1)}}{\sigma_y}
\]

(1)

where $K_c$: Mode I plane strain fracture toughness
$\beta(N)$: Amplitude of HRR stress singularity
$\sigma_y$: Uniaxial tensile yield strength.

In order to take advantage of the "Modified RKR model", one needs - just as one does for the original model, equation (1) - to experimentally determine the following "ingredients":

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a) **The flow properties**, i.e. true stress - true strain curves by static tensile tests at a few (5 to 6) temperatures in the range \(-200^\circ\text{C}\) to \(300^\circ\text{C}\).  

b) **The microscopic fracture stress \(\sigma_f^*\) and its scatter**  

b.1 - Using the instrumented \(C_v\) test, as shown below, some reconstituted specimens are needed to obtain more low temperature load-time traces than routinely available (this is often done at reduced hammer velocity, say 1 m/s)  

b.2 - Using four-point notched bars for static tests at a few selected temperatures (Griffiths-Owen design [26])  

c) \(K_{ic}\) and its scatter, at least at one selected temperature, by three-point slow bend of reconstituted precracked \(C_v\)'s or by static tests of axisymmetric precracked notched tensile bars. In the current development phase, extensive use of the scanning electron microscope is considered essential both to define the critical volume \(V^*\) and to identify whether the cleavage initiators belong to one or more populations.  

Referring to equation (1) and Fig. 3:  

Item a) provides \(\sigma_{yc}\) (static yield) and \(N\) in a way allowing use of a strain hardening model more precise than the Ramberg-Osgood law, whenever desired  

Item b.1 provides \(\sigma_{yd}^*\), \(\sigma_{yd}\) (dynamic yield) and the strain rate sensitivity  

Item b.2 confirms \(\sigma_f^*\), its scatter and its strain rate independence  

Item c) provides \(I^* (V^*)\), \(\sigma_f^*\) at higher (transition) temperature and a validation of the model’s statistical treatment.  

The implementation of such proposed capability is in progress and its **routine** application is believed feasible within two to three years. It is important to emphasize that Items a) and b.1) will already allow to reach two milestones, for any steel to which they are applied (such as Doel-I,-II welds [27]):  

1. Full-fledged application of the "Improved Transition Temperature Concept" (Approach I above).  
2. Insight as to which damage mechanisms govern embrittlement, i.e. in particular, does one have to suspect weakening of grain or lath boundaries, such as could be induced for instance by phosphorus segregation.  

The "Improved Transition Temperature Concept" is illustrated to some extent in this paper. It is a physically defendable representation of the ductile-brittle transition temperature DBTT. But aside of being relevant for indexation of generic fracture toughness curves, it is also the proper concept for the modeling of irradiation damage effects.

---

1 High temperature is needed to separate athermal component of flow stress (see Section 4).
Guidelines to Damage Modeling.

Damage modeling aims at understanding the sub-microstructural interactions causing the worsening of RPV steel's mechanical performance under service exposure. The focus is on the neutronic component of the irradiation field (gamma rays and electrons play a negligible role). Thermal and strain ageing are also considered a concern [27].

The property of interest for structural integrity is of course fracture toughness, but surveillance data bases are limited to Cv impact and tensile properties. This should suffice, insofar as micromechanics (see above) will allow to complete the picture.

Figure 4 summarizes the foundations of damage modeling as applied here for the transition temperature range. The simple plot on the upper right corner of the Figure is well known and often referred to in literature as the Ludwig-Davidenkov diagram (for ex., [28]); it is generalized by the lower plot. Put crudely, these diagrams view DBTT, the ductile-brittle transition temperature, as the temperature at which the yield stress intersects the microscopic fracture stress. More precise definitions are used in the Belgian R&D program.

In general, in-service steel embrittlement can be accounted for by matrix hardening alone: namely, by the increase of the flow stress through the creation of new obstacles to the mobility of dislocations; this is reflected by an increase of hardness (Vickers, Rockwell, ...) - proportional whenever work hardening is not affected by irradiation. The fracture stress does then correspond to the microcleavage fracture stress - which is generally independent of temperature, strain rate and irradiation. The resulting increase (shift) of DBTT is roughly proportional to the increase of the uniaxial yield stress, or, for that matter, to the increase of the C, general yield stress. The precise relationship is more complicated, but will have to be discussed in detail in a subsequent publication.

At PWR operation regimes, there are at least three distinct hardening mechanisms contributing to the embrittlement of RPV steels, see Figure 5; they feature anneal activation energies in the range 1.8-2.1 eV, as well as different neutron fluence and irradiation temperature sensitivities [7]. The best known of these mechanisms entails radiation-enhanced diffusion and precipitation of copper [5] [6] [29]-[31]; it is described in the Belgian program by the well-established model of Fisher & al. [6], accounting for the effective amount of copper available in solid solution [32]-[34]. The kinetics - but not the saturated contribution - of this mechanism is sensitive to both dose rate and irradiation temperature (Fig.5).

Further microstructural characterization work is needed to fully grasp the physical origin of the other mechanisms and be able to better ascertain the reliability of predictive projections of surveillance data into vessel walls. Such work is in progress in cooperation with the International Group on Radiation Damage Mechanisms (IGRDM). The specific Belgian contribution does presently focus on conventional TEM, positron annihilation spectroscopy and internal friction as its experimental tools, with emphasis on addressing the less well known damage mechanisms such as the ones labelled 2 and 2A on Figure 5 (along a classification scheme by order of increasing anneal activation energy, similar to [35]).
Service exposure can sometimes also lower interboundary cohesive energies, causing a decrease of fracture stress and resulting in easier microcrack propagation beyond the hard particles in which they form. One example is the occurrence of irradiation enhanced segregation at grain boundaries, leading to mixed mode fracture (intergranular cracking and transgranular cleavage). This 'non-hardening' embrittlement may compound with the one stemming from matrix hardening or, albeit very seldomly, be found in isolation: in such case, the yield stress, the hardness and the $C_v$ upper shelf do not change, although the $C_v$ shift can be sizeable.

This brief outline shows that the mechanical data important for damage modeling are the flow properties and the microscopic fracture stresses; these can be obtained from the load signals of the instrumented $C_v$, notch impact test, in combination with conventional, static tensile tests - a topic more extensively addressed below.


3. DUCTILE-BRITTLE TRANSITION TEMPERATURE BY USE OF INSTRUMENTED C, LOAD-TIME TRACES

Results:

In the context of PWR pressure vessel surveillance, the significance of the C, notch impact test, instrumented by strain gages, is revisited.

The load diagram - general yield, maximum, brittle fracture and arrest loads versus temperature - is the most fundamental feature of the test; it is directly correlated to the appearance (percentage shear) of the fracture surface, a feature further explored herein.

The bulk of the absorbed energy and lateral expansion stems from plastic deformation associated to ductile stable crack growth under conditions unrepresentative of the constraints and stress-strain field near the tip of a sharp crack in a pressure vessel.

It is shown that the temperature at which a fixed energy is absorbed in the test (say, 41 or 68J) cannot always trace, to acceptable accuracy, the effect of steel service exposure on the ductile-brittle transition temperature DBTT, and on cleavage fracture toughness. It is contended that this can be done reliably by using characteristic temperatures of the load diagram.

Concepts and Definitions.

From a combined dislocation dynamics and fracture mechanics perspective, the most fundamental information contained in the C, impact test is the load diagram [36]-[44], as derived from the instrumented load versus time (deflection) traces recorded by strain gages on the hammer tup. This is schematized by Fig. 6.

Considering a typical signal in the transition temperature range, left-hand part of the Figure, it is well known - and acknowledged by testing standards in preparation [45] [46] - that at least four characteristic loads can be defined:

\[ F'_y \]: General Yield Load; \[ F_m \]: Maximum Load; \[ F_u \]: Brittle Crack Initiation Load; \[ F_a \]: Crack Arrest Load.

**General Yield Load**: a point at which local yielding has spread to the entire area corresponding to the ligament under the notch (net area), while strain hardening is still negligible, overall;

**Maximum Load**: a point at which a ductile crack initiated by void nucleation (on inclusions or second phase particles), growth and coalescence, at some distance under the notch (at time marked i on Fig. 6, left), has extended into a crack front across the specimen width, collinearly to the notch;

**Brittle Crack Initiation Load**: in the transition temperature range, ductile stable crack growth is interrupted at this point by the competition of stress-controlled cleavage, initiated within "trigger" particles (such as grain boundary carbides or other "weak" spots) and propagated further (into the ferrite matrix, to the next grain, ...);

**Arrest Load** for fast (brittle) crack propagation and re-conversion to ductile crack growth, but under plane stress conditions, with formation of shear lips along slant planes at \( \approx 45^\circ \) angle to the main fracture surface.
Some energy partitioning can be associated to the $C_s$ load diagram, and if done according to the characteristic loads identified by the testing standards, this leads to four distinct fractions, labelled LSE, A, B and C. Their definition is illustrated by the left-hand side of Fig.6. In this work, all energies are corrected for elastic interaction at the load points. Figure 7 presents a typical implementation of such energy partitioning.

The energy fraction LSE, Lower Shelf Energy, is composed of elastic energy ($I_{se}$) and brittle crack propagation energy ($E_d$). It is largely insensitive to service exposure, and normally never exceeds 10 J.

In literature, $A+I_{se}$ is often called "Initiation Energy", $B+E_d$ "Propagation Energy". Actually, ductile initiation occurs at point marked i on Fig. 6, left; this should in principle be taken to define the boundary between the energy fractions A and B, corresponding to the physically distinct phenomena of "Ductile Initiation" and "Ductile Propagation under near plane strain conditions". In practice, this refinement has not been found to add much, as compared to the more straightforward partitioning of Fig. 6.

Concerning the transition temperature concept, the emphasis in this work lies generally on the characteristic temperatures marked $T_r$ and $T_e$, right-hand side of Fig. 6. To this respect, the 50% transition temperatures such as TTA, etc. on Fig. 7 are supportive informations only: for ex., TTA is useful in assessing the irradiation-induced shift of $T_r$, to which it is directly correlated. Indeed, in engineering surveillance practice, limited attention is usually placed on tests at absorbed energy levels low enough to directly assess $T_r$ to sufficient accuracy. Note finally that indexation to an energy level of $28 J$ is generally equivalent to using TTA as reference.

The onset of $C_s$ impact upper shelf regime begins at Characteristic Temperature $T_0$ (right-hand side of Fig. 6), above which the load$^2$ curves $F_s(T)$ and $F_s(T)$ coincide, i.e. there is no brittle crack initiation.

In this upper shelf regime, a "fictitious $F_0$ load" can eventually still be defined as velocity inflexion point, believed to broadly separate plane stress from nearly plane strain conditions. Such fictitious $u$ point is sometimes used in this work to assign a shelf level to the Post-Arrest Energy Fraction, labelled Fraction C on Fig. 6, along with ref.[42]. Actually, the procedure is somewhat ambiguous, especially at $C_s$, upper shelf levels exceeding $\approx 120 J$, and it is often more appropriate to extrapolate a plot of energy $C$ versus shear up to 100 %.

We can further identify:

$T_{nu}$ "Ductility Temperature"

Onset of Energy Fraction B; this literature terminology is based on the idea that ductile crack initiation occurs at maximum load, which may be true for what concerns the beginning of propagation of a whole crack front;

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$^2$ Loads or forces are denoted by the symbol $F$, with appropriate index; the corresponding stresses are denoted by the symbol $P$. 

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$T_D$ - "Ductile Crack Initiation Temperature"

Onset of actual ductility, i.e. temperature at which the shear fracture appearance in the C, test begins to exceed 0% (See below);

$T_D$ - "Brittleness Temperature"

In a nutshell, the maximum temperature of "elastic fracture". This is also the temperature relevant to the 'plastic collapse' failure analysis approach.

More accurately, $T_D$ is the temperature corresponding to the load at which plastically induced cleavage coincides with general yielding (yielding spread over the entire ligament under the notch, with associated plastic hinge, as described in textbooks [47], [48]). Most significantly, this is the characteristic temperature for the OROWAN [49] cleavage fracture criterion applied to a notched specimen:

$$\sigma_{yy}^{\text{max}} = K_{op} \sigma_{y|\xi,T} \geq \sigma^*_{f} \quad (2)$$

where:

$\sigma_{yy}^{\text{max}}$: Maximum value of maximum principal tensile stress;

$K_{op}$: Plastic constraint factor, elevating the local stress beyond the uniaxial yield stress in order to reach general yielding;

$\sigma_{y|\xi,T}$: Uniaxial yield stress at the temperature $T$ and strain rate $\xi$ of interest;

$\sigma^*_{f}$: Microcleavage fracture stress.

Note that the simple brittle fracture criterion of equation (2) is valid here only because of the relative spatial flatness of the stress-strain field ahead of the shallow C, notch.

In the older literature, $T_D$ has often been considered a good definition for DBTT, from a physically-oriented viewpoint. The present views are that $T_D$ is actually the most adequate.

A criterion similar to (2) can in any case also be applied at $T_i$; this is usually done in this work. Between $T_D$ and $T_i$, the failure load is slightly larger than, and more or less parallel to the general yield load, while strain hardening primarily occurs in the space between the net and the gross specimen sections, causing some deflection, but only minor energy increase. At high strain rates, such as in the C, test, $T_D$ is generally close to $T_i$. 

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On another hand, it has been recently shown that $T_d$ is a more universal characteristics of the materials than $T_d$ [50]-[52]; it is also rather insensitive to specimen thickness and notch angle [53] (but it depends on notch acuity and strain rate). Given furthermore that $T_d$ does correspond to the temperature of first, local ductile microcrack initiation at the higher temperature end of the cleavage range (see below), it can realistically be asserted that $T_d$ is certainly a most physically-grounded definition of the ductile-brittle transition temperature DBTT applicable to notched specimens. It is also adequate for fracture toughness indexation. Certainly, the increase $\Delta T_d$ under service conditions (thermal and strain ageing, irradiation) is a good measure of the corresponding shift of the initiation fracture toughness for the same strain rate - assuming the temperature dependency (shape) to be invariant, as also done in the Regulatory, ASME XI based framework. In other words, $\Delta T_d$ is a good index for the dynamic initiation fracture toughness $K_{id}$.

**Significance of Absorbed Energy in $C_v$ Impact Test.**

It is most useful to further appraise the actual significance of $C_v$ energy and of its partitioning. This can be done on basis of Figures 8 and 9.

Figure 8 is a "bench-mark", in the sense that the steel and test conditions have been chosen to closely match the ones adopted for a decisive finite difference investigation [54], backed-up by extensive experimental scrutiny, including stop-block impact tests [55]. Time (deflection), load and energy at ductile crack initiation obtained in this literature study agree very well with the observations made by means of the Fraunhofer Institute Magnetic Emission (ME) detector [56], as installed in the SCK-CEN hot cell.

Figure 9 compiles ME detector results on irradiated 22NiMoCr37 RPV steel, over the entire test temperature range relevant to surveillance. The Figure also summarizes the conclusions.

It is clear in particular that the bulk of absorbed $C_v$ energy is unrelated to ductile crack initiation. The $C_v$ test is primarily a propagation one, and the best use of its energy information may be in trying to derive the tearing modulus for ductile stable crack growth, as exemplified in ref. [57] [58].

**Correlation between $C_v$ Impact Load Diagram and Fracture Appearance.**

Another important lesson of the work, summarized by Figures 8 and 9, is that ductile crack initiation in the $C_v$ impact test occurs at a load such that

$$F_i = F_y + (1-k)(F_m - F_y)$$

(3)

where, generally, $k \approx 0.5$, i.e. the initiation load corresponds roughly to the average of $F_y$ and $F_m$. 

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The implication of (3) is that the fracture appearance of $C_i$ impact specimens can always be determined from the load diagram through the relationship

$$SFA(\%) = \left[ 1 - \frac{F_u - F_a}{F_m + k(F_m - F_y)} \right] \times 100$$  \hspace{1cm} (4)

This can be easily demonstrated on basis of equation (3), see Appendix.

The best value for $k$ is most accurately defined, on a case-to-case basis, by means of simultaneous fit to both the load diagram and to planimetry measurements on the fracture surfaces.

We have found that $0.4 < k < 0.6$ ; across-the-board, it seems that little error can be made by taking $k=0.5$, but significant distortion often results for $k=0$, as proposed in the DIN standard [45].

Typical application of this equation is illustrated by Fig. 10³.

**Inadequacy of Fracture Toughness Indexation to a Fixed C, Impact Absorbed Energy Level and Physically-Based C, Impact Indexation Temperatures.**

Intuitively, it is difficult to believe that the DBTT shift upon service could ever exceed the shift of either $T_i$ or $T_o$. This view is reinforced by the fact that one always observes that $\Delta T_i \approx \Delta T_o$. Yet, it may happen that the 41J shift $\Delta TT41$ significantly exceeds $\Delta T_p$, $\Delta T_i$ and $\Delta T_o$. An example is illustrated by Fig. 11. In such case, $\Delta TT41$ is certainly inadequate for fracture toughness indexation.

The non-physical behaviour of TT41 in this example is explained by Fig. 12. It is seen that the 41J temperature in the baseline is controlled by the plane strain, pre-maximum energy fraction, A+LSE, while some amount of post-arrest energy fraction C - i.e., of plane stress, shear lip formation energy - is needed to reach the same 41J level after irradiation; this represents a mismatch of totally unrelated physical phenomena, which furthermore do each shift by about the same amount upon service exposure. The 41J shift is strongly affected by the shelf levels and mid-shelf temperatures of the fractions in the steel baseline: it is NOT independent of the un-irradiated condition; for ex., when A+LSE exceeds 41J both before and after service, the 41J shift is considerably less than otherwise, and agrees with the physically grounded shift. So, two plates of same chemistry may shift quite differently at 41J, under identical environmental conditions, depending on their baseline. This guarantees that unwarranted scatter and distortion does affect empirical, engineering or Regulatory embrittlement correlations, such as the ones in [3]. Furthermore, some steels may be considered as "outliers", while they actually are not. Examples are the Yankee Rowe and BR3 plates, and some Linde-80 welds (To be published).

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³ Loads or forces are denoted by the symbol F, with appropriate index; the corresponding stresses are denoted by the symbol P.
The 50% FATT shift is often closer to the physical reality than the TT41 shift, but occasionally, it may also be too large (as on Fig. 11). Nevertheless, Cₜ, FATT has been shown to be superior to the Pellini drop weight NDT for toughness indexation of a variety of unirradiated steels [59] [60], and to first approximation, one generally finds that upon reactor exposure, ΔFATT ≈ ΔTT ≈ ΔT0.

The question must be asked: could Tₒ, the Cₜ impact upper shelf onset, provide physically grounded indexation of Kₘ on its own merit, i.e. independent of the fact that its shift is not very different from the Tᵢ shift?

By contrast to the case of Tᵢ, fundamental foundation seems to lack here to provide a "first-principle" type justification. A theoretical-type demonstration might be workable on basis of tearing modulus considerations [61], but this has not been attempted. Anyway, "hard" experimental evidence is needed, ultimately. From this point of view, Figure 2 is certainly supportive, as well as earlier Westinghouse experimental data [62]. In general, literature Kₘ data suggest that the onset of the fracture toughness upper shelf may sometimes occur at somewhat higher temperature than the Cᵢ, Tₒ value, but more often than not, the differences appear small. In any event, no firm conclusion can be drawn for the service shift, because available data are too sparse.

Early work by Kobayashi & al. [63] shows that Tₒ may correspond to the crack arrest temperature (CAT) of the Robertson test, but this evidence is certainly too scanty for generalization.

A recent study by Pachur [64] does conclude that the Pellini drop weight nil ductility temperature (NDT) does correlate with the onset of the post-arrest energy fraction C, and with its 50% temperature TTC. In particular, the NDT shift by irradiation is significantly less than the 41J Cᵢ shift, for the investigated steels, and this is so apparently for similar reasons as illustrated by Fig. 12. However, the physical meaning of the drop weight test is not fully clear. The possibility of experimental bias due to the brittle bead deposition method [65] does not seem a problem in this case, nor in a similar German study by Schmitt et. al. [66]. However, inspection of Pachur's data reveals a good correlation of ΔNDT with ΔTTB as well; and the Schmitt et.al. results can be interpreted as consistent with an indexation to TT (i.e. Tᵢ, or an absorbed energy near 28J). Actually, in some cases, NDT is close to the 'ductility temperature' Tᵢ (the unirradiated HSST-03 plate for example).

Reference [67] summarizes and evaluates a host of pre-1981 literature correlations between fracture toughness and other mechanical properties (Cᵢ, tensile, drop weight). It is beyond the scope of this paper to examine all these correlations in terms of the present views.

However, a more recent correlation by Wallin [68] must be addressed some length, as it seems to entail a significantly different conceptual background. For unirradiated steels, Wallin shows that the temperature at which 28J is absorbed in the Cᵢ test, TT28, is on average about 18°C larger than the temperature at which Kₘ reaches the 100MPaVm level. The standard deviation of this linear correlation is 15°C. No problem so far. However, the theoretical underpinning to this semi-empirical correlation may conflict with some of the ideas of the present paper. Central to Wallin's argument is that a blunt notch, as opposed
to a crack, shifts the toughness transition to lower temperature, while the strain rate effect
does just the opposite; the magnitude of both shifts is deemed to be inversely related to
the yield strength and the two shifts considered tend to cancel-out one another. Wallin
does further state that dynamic fracture toughness correlates neither with static toughness
nor with $C_n$, because the compensation effect between strain rate and notch acuity is lost.
In the present work, the notch-versus-crack and strain rate effects are micromechanically
assessed in terms of the microcleavage fracture stress and the flow properties (see outline
in next section), but of course, this does hold for approach II of section 2, and does not
apply to the improved transition temperature concept as forwarded herein. However, near
the indexation temperature $T_o$, we believe that there is enough ductility so that crack
sharpening effects are essentially eliminated; as the strain rate is adequate relative to
dynamic toughness, it is not too surprising thus that $T_o$ be adequate for $K_{th}$ indexation.,
even despite the near plane stress state (for both unirradiated and irradiated conditions).
On another hand, we concur with Wallin that a crack leads to a larger $T_i$ value than a
notch; this is primarily because the stress intensification factor in the Orowan relation (2)
is larger (see next section). Yet, upon irradiation, we always observe that $\Delta T_i \approx \Delta T_o$, i.e.
hardening does not seem to affect much the stress intensification near general yielding. By
contrast, to keep his correlation valid for irradiated steels, Wallin recommends to raise the
$C_n$ indexation level by the ratio of unirradiated to irradiated upper shelf energy. This is
similar to the Russian indexation approach [69], but at odds with the foundations of the
present approach. The cornerstone of these differences seems to entail the very definition
of $K_{th}$ and its lower bound in the transition temperature range; to this respect, we do
consider the energy associated to stable tearing irrelevant to cleavage fracture toughness
(see also references [70] and [71]).

$C_n$ reconstitution and miniaturization work summarized in a subsequent Section of this
paper may yet provide the strongest evidence against toughness indexation to fixed energy
or lateral expansion levels in the $C_n$ test, and the strongest evidence in favour of our
emphasis on the load diagram approach, with its physically meaningful characteristic
temperatures $T_i$ and $T_o$.

**Brief Statistical Considerations and Analytical Fits.**

All fits to $C_n$ data in this paper intend to characterize the *mean* behaviour of a set of
specimens corresponding to a given material condition. Bounding intervals can also be
defined.

Metallurgical scatter causes departures from the mean trends, especially for what concerns
deflections and energies; in general, the load diagram is the least affected, provided the
instrumented traces have been obtained under adequate experimental conditions.

Nevertheless, even the characteristic temperatures defined above must be understood as
best mean values; for instance, it may happen that a specimen broken at a temperature
slightly exceeding $T_o$, the mean upper shelf onset, does not display fully ductile, 100% shear response, and vice versa for $T_i$, 0% shear (Fig. 10).

It has been found that the most significant scatter in the $C_n$ test entails the deflection, thus
also the energy, of fraction B. A typical illustration is provided by Fig. 13; here, the total
absorbed energy of unirradiated plate HSST-03 (L-T) at a test temperature of 40-41°C ranges from \(\approx 108\) J to \(\approx 135\) J, i.e. the dispersion is \(\approx \pm 13\) J. This is traceable to a systematic increase of energy B across the population of 12 tests. Closer inspection reveals that what fluctuates the most widely is indeed the amount of stable crack growth between the apparition of a ductile crack front under the notch and the subsequent conversion to brittle cleavage. The competition between fracture modes is strongly affected by the inhomogeneous distribution of trigger particles and this leads to much larger energy spreads than the ones caused by the same particles under lower shelf regimes. Why? Presumably, because large deformations, thus large deflections, can be accommodated by unit crack length propagation, owing to the enhanced plasticity in the transition range. This is reminding of the amplification of \(K_{\text{sc}}\) scatter by stable crack growth prior to brittle failure in the upper transition temperature regime.

The increase of Fraction B with increasing temperature is generally the steepest one as compared to the other fractions, i.e. energy B goes from zero to its shelf value over a small temperature interval (10 to 30°C), at least compared to the interval between lower and upper shelf for the total energy (typically 120 ± 25°C). Fraction B is often the one most affected by irradiation. The small scatter associated to the C, testing of specimens subject to substantial in-pile embrittlement is due to the significant decrease of this fraction, and/or to its shift to higher temperatures (sometimes even exceeding \(T_{\text{c}}\)).

Also, it will be seen in a subsequent Section that the major difference between C, transverse orientation (T-L) and strong orientation (L-T) data in base metals stems from the reduction of energy B in the former case - case in which the scatter is correspondingly reduced. Upper shelf orientation effects are linked to the spacing, shape and distribution of inclusions (sulphides, oxides, ...), which govern microvoid coalescence and the critical strain for ductile fracture. Thus, energy B is totally unrelated whatsoever to stress-controlled brittle fracture, and to DBTT. Sometimes in this paper, fraction B is loosely designated as "Tearing Fraction", simply to remind the present considerations.

Finally, energy partitioning has been found most useful to help identify outlier specimens, an important issue in surveillance applications - given the usually limited test matrix. In particular, a fraction A or fraction C misbehaviour may not be obvious from total energy, lateral expansion and fracture appearance alone. The approach, extended to encompass deflection and expansion data, and illustrated by Fig. 14, is a very powerful evaluation tool, because it is comprehensive and physically guided; moreover, it does indisputably allow to consistently appraise the entire information contained in the C, notch impact test, starting from the fundamentally relevant load diagram.

To complete this outline, it may be mentioned that a PC-software (EXCEHL) has been written which incorporates all concepts of this paper; it is presently under debugging; the detailed equations programmed into this code must be skipped here.
4. TOWARDS AN ENHANCED SURVEILLANCE PRACTICE BY COMBINED USE OF INSTRUMENTED C, LOAD-TIME TRACES AND UNIAXIAL TENSILE TESTS

Synopsis

Consistent evaluation of C, notch impact general yield data and of static uniaxial tensile yield data allows to:

- Quantify the influence of strain rate on the ductile-brittle transition temperature;
- Ascertain the importance of hardening in the degradation of the service performance of ferritic steels.

Such evaluation is possible provided the tests cover a sufficiently broad range of temperatures. In the case of nuclear reactor pressure vessel surveillance, this requires some enhancement of current test matrices - eventually including dedicated reconstitution of broken specimens. This is also essential for future, micromechanics-based assessment of irradiated fracture toughness bounds.

The mathematical formulation is outlined of a recommended analysis procedure tailored to the RPV surveillance context.

Classical dislocation dynamics is used. The separation between short and long range obstacles to the movement of dislocations is quantified in terms of activation enthalpy. Strain rate and temperature sensitivities are considered to be governed by Palerme's type barriers to the plastic flow, at least in the un-irradiated condition; complications arising under service exposure are discussed, as well as some vital features of the correlation between brittle fracture toughness and ductile-brittle transition temperature.

Brief Reminder of Thermal Activation Theory.

Steel deformation (strain) under the influence of an applied stress is governed by the interaction of dislocations with various obstacles, both short range, thermally activable ones (for ex., lattice atomic rows), and long range, 'athermal' ones (interstitial impurities, solute atoms, carbide precipitates, ...).

Thermal energy transmitted by lattice vibrations helps the dislocations to overcome the short range energy barriers. As in atomic diffusion, this can be represented by a state equation, here between shear stress \( \tau \), strain \( \varepsilon \), strain rate \( \dot{\varepsilon} \) and absolute temperature \( T \) (K). For a single controlling deformation mechanism, this may be reduced to an Arrhenius equation for the strain rate (i.e. the rate of dislocation motion):

\[
\dot{\varepsilon} = \dot{\varepsilon}_0 e^{-\frac{\Delta G(\tau,T)}{kT}}
\]

where: \( \Delta G(\tau,T) \) = Gibbs free energy,
Energy of the barrier to dislocation movement caused by the considered obstacle

\[ \dot{\varepsilon} = \sum_i \dot{\varepsilon}_0^i e^{-\frac{\Delta G_i(\tau,T)}{kT}} \]

For mechanisms acting in series:

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\[ \dot{\varepsilon}_0 = N b L \nu \]

Intrinsic Strain Rate Sensitivity,

Measures the rate of attempts to jump the barrier, times the strain produced by a successful attempt

with  
\( \frac{N}{b} \) = density of mobile dislocations involved in the thermal fluctuation
\( b \) = Burgers vector
\( L \) = distance covered in the thermal fluctuation
\( \nu \) = frequency of jump attempts

\( \dot{\varepsilon}_0 \) is largely independent of \( \tau \) and \( T \)

\[ k = \text{Boltzmann constant} \ (1.38 \times 10^{-23} \text{ J/K}, \ 8.62 \times 10^{-5} \text{ eV/K}) \]

The free energy of activation \( \Delta G \) is not directly measurable, but is thermodynamically linked to \( \Delta H \), the experimentally derived enthalpy of activation, by

\[ \Delta G = \Delta H - T \Delta S \]

where the entropy is defined as

\[ \Delta S = -\left( \frac{\partial \Delta G}{\partial T} \right)_\tau \]

In practice, the entropy during activation \( \Delta S \) is essentially due to the temperature dependence of the shear modulus \( \mu(T) \)

\[ \mu(T) = \mu_0 (1 - \alpha T) \]

with \( \alpha = 2.8 \times 10^{-4} \text{ K}^{-1} \) for iron and steel.

It can generally be assumed that the long range, internal stress field is proportional to the shear modulus; this 'athermal', i.e. thermally un-activated component, is thus expressed here as

\[ \tau_\mu(T) = \tau_0 (1 - \alpha T) \] (6)
An effective shear stress for the thermally-activated part of the plastic flow can be defined as

\[ \tau^*(T, \dot{\varepsilon}) = \tau(T, \dot{\varepsilon}) - \tau_e(T) \]  \hspace{1cm} (7)

This is illustrated by the bottom part of Figure 15, for the uniaxial lower yield stress of Fe E 460, a normalized, German reference structural steel [72].

For any fundamental investigation of flow mechanisms, it is obviously vital to properly separate the athermal component; as shown by the Figure, this can be accomplished by tests at sufficiently elevated temperature. But even in the present engineering context, it has been found important, not only to do the separation, but also to examine the thermally-activated contribution(s) at the light of relevant defect-dislocation interaction models. Reasonable semi-empirical fits to yield stress data can be obtained with the lumped expression

\[ \tau(T, \dot{\varepsilon}) = \tau_0' \left[ kT \ln\left(\frac{\dot{\varepsilon}}{\dot{\varepsilon}_0}\right)\right]^n \]  \hspace{1cm} [73]; but this is not suitable to unravel some key, albeit generally ignored subtleties of irradiation effects on RPV steels.

An important thermodynamical quantity is the 'Activation Volume', defined as

\[ V^*_A = -\left(\frac{\partial \Delta G}{\partial \tau}\right)_T \]

It is a measure of the actual volume within which thermally-activated work is done; it is directly related to the spacing between obstacles.

For a single activated mechanism, assuming - most reasonably - the mobile dislocation density to be unaffected by the stress, it has been shown that

\[ V^*_A(\tau, T) = kT \left(\frac{\partial \ln\dot{\varepsilon}}{\partial \tau}\right)_T \]  \hspace{1cm} (8)

and under such conditions, \( V^*_A \) does generally not depend on temperature. One also has:

\[ \Delta H(\tau, T) = -T V^*_A(\tau, T) \left(\frac{\partial \tau^*}{\partial T}\right) \]  \hspace{1cm} (9)

Thus, the activation volume \( V^*_A \) and activation enthalpy \( \Delta H \) can be determined from tests at two strain rates in function of temperature. This approach is suited to the surveillance context.

The activation enthalpy and activation volume can be compared to the predictions of various models for the kinetics of dislocation motion. This allows to decide which energy barrier is applicable, and what types of obstacles are responsible for the strain rate and
temperature sensitivity of the material. The activation volume can be measured the most easily and accurately by a differential method (change the strain rate at constant temperature and observe the stress); it is the most sensitive indicator to distinguish between competing models.

Peierls Mechanism and the Dorn-Rajnak Model.

In this work, it is accepted that the Peierls mechanism [74] - eventually perturbed - is responsible for the entire thermally-activated part of the flow stress for unirradiated b.c.c. lattices, including RPV steels. We do not consider deformation at very high strain rate (>3000 s⁻¹), i.e. phonon friction, ...

The experiments are represented by a single rate-controlling mechanism [equation (5)], involving only lattice hardening, i.e. the intrinsic lattice resistance to the movement of dislocations.

In pure metals, such as α-iron, this simply entails the jumping of screw dislocations over the energy hills associated to close-packed atomic rows (TEM morphologies suggest that edge dislocations play a minor role).

Alloying generates localized strain centres (tetragonal or spherical) which perturb the Peierls barrier, and result in solid solution softening and strengthening. This is accounted for most easily in the interpretation of the Peierls' mechanism offered by the Dorn-Rajnak model [75], as adopted herein.

In this model, a straight dislocation line lying (at its lowest energy) in a potential valley parallel to the closest atom packing lines on the slip plane, can be moved forward to the next valley by applying a shear stress whose maximum value, i.e. at 0°K, is equal to - by definition - the Peierls' stress \( \tau_p \). Above 0 K, the effective stress \( \tau^* \) is less than \( \tau_p \), as the assistance of thermal fluctuations allows to more easily overcome the Peierls hill: they cause the dislocation to vibrate about its mean position; eventually, it will break into two partial kinks of energy sufficient to jump over the barrier ('Double-kink' mechanism).

Under some simplifying, albeit reasonable assumptions⁵, it is found that the effective stress \( \tau^*(T,\dot{e}) \) needed for the dislocation to surmount the stress hill by formation and motion of double kinks can be expressed as

\[
\frac{\tau^*(T,\dot{e})}{\tau_p} = \frac{U_n}{2U_k}
\]

where \( U_n \) 'saddle-point' free energy for nucleation of a pair of kinks
\( U_k \) energy of an isolated kink

---

⁵ In particular: - Mobile dislocation density is unaffected by stress  
- Effect of interstitials on kink velocity negligible  
- No initiation of twinning
The function $\Psi$ is 'universal', at least for pure b.c.c. crystals (Fe, Mo, Cr, Ta, V, W...), in the sense that it is little affected by the exact shape of the Peierls hill and depends exclusively on the physical features of atomic bonding. The difference between metals is entirely reflected by their different Peierls stress $\tau_p$ and different 'activation energy' $2U_k$ - the maximum energy achievable by the double kink process (0.5 - 0.9 eV).

Various Peierls lattice energy barriers of different shape are considered in literature: sinusoidal, parabolic, camel-hump, broken bond, ... A completely analytical solution in closed form only exists for the quasi-parabolic case [76]:

$$\frac{U_n}{2U_k} = \left( \frac{1-\tau^*}{\tau_p} \right)^2.$$

We could fit the numerical results for other cases - to extremely high precision - using a 'generic' expression

$$\frac{U_n}{2U_k} = \left[ 1 - \left( \frac{1}{\tau_p} \right)^{m'} \right]^m,$$

where $m$, $m'$: constants.

The transform, directly applicable to evaluate surveillance data:

$$\frac{\tau^*}{\tau_p} = \left[ 1 - \left( \frac{U_n}{2U_k} \right)^{m'} \right]^m,$$

has been used to compare some of the plausible shapes, Figure 16. We have added an empirical representation by Smidt [77], and re-written the equation in the previous notations i.e. as:

$$\frac{\tau^*}{\tau_p} = \left[ 1 - \left( \frac{G_c}{G_c} \right)^{m'} \right]^m \quad (11)$$

where $G_c$ denotes the activation energy.

An early linear representation introduced by Conrad [78] corresponds to $m=m'=1$.

Also shown on Fig. 16 is the barrier developed by Fleischer ([79]-[81]) for a quite distinct, yet also thermally-activated process: hardening by tetragonal lattice distortions, such as induced by irradiation (e.g. [82]), or possibly by interstitial atomic defects (solid solution hardening). This may be the proper explanation for the strength of some b.c.c. metals at low temperature (for ex., iron containing carbon). At PWR irradiation temperatures, it is generally accepted that the defects introduced by service exposure can be represented as long range obstacles causing an increase of the athermal part of the flow stress; yet, in two cases so far (a German steel and the Doel-I,II Soudotenax base metals),
we have found indication of shorter range irradiation-induced defects, tentatively
depictable by the Fleischer barrier. This could be detected only because yield strength
data covered a sufficiently broad range of temperature, at two strain rates. Work is in
progress to confirm such preliminary findings - possibly traceable to the so-called "matrix
damage".

For one unirradiated steel, the one examined on Fig. 15, the Fleischer model seems to fit
the experiments better than the Dorn-Rajnak model, for which some deviations are seen at
small effective stress $\tau^*$. This is a difficult range, where separation of the athermal
component can easily conduce to error; a range also where competing mechanisms may
play a role. These could entail the dragging force exerted by interstitial atoms on the
moving dislocations, or the recombination of sessile dissociated screw dislocations [83], or
the process of intersection of dislocations - suggested for f.c.c. metals (where the Peierls
hills are very low).

However, all the other data examined to firm-up the formulation finally adopted for the
Dorn-Rajnak model (a perturbed sinusoidal barrier corresponding to a parameter $\alpha$ [75] of
-1) are most satisfactorily represented, without a need to evoke any other mechanism than
double kink nucleation at the Peierls barrier, or transverse slip of the kinks, a competing
effect also described by (10) [replacing $2U_k$ by $U_m$, the migration energy for kink
migration]. This amounts to say that (11) is applicable to RPV steels provided $\tau_p$ and $G_c$
be changed from their values for pure iron, and treated as variables depending on the
steel's microstructure and composition. A few such fits are exemplified by Fig. 17. All
data stem from tensile tests at a variety of strain rates, except in the last case of the
Doel-4 base metal, where the high strain rate measurements were obtained by means of the
instrumented C, notch impact test. Incidentally, it can be seen that alloying does strongly
influence the athermal component of the flow, as expected; it also affects the
thermally-activated part.

It must be mentioned that another interpretation of the Peierls-Nabarro stress has been
proposed by Armstrong [84], aimed at relating the Petch "friction" stress to the thermal
activation rate equation. This approach has not been retained; it is not easily amenable to
direct validation in a dislocation dynamics perspective tailored to our purposes. We have
also examined, and abandoned the representation of Yaroshevich and Ryvkina [85], based
on the graphical differentiation of their $\tau$($\dot{\gamma}$) measurements. This had given:
\[
\frac{\partial \tau}{\partial \dot{\gamma}} = -m(\tau - \tau_p) = -m \tau^*
\]
i.e. an exponential dependency $\tau^* = \tau^0 e^{-m\tau}$ [similar to Armstrong]. This form has been
used by some authors (e.g. [86] [87]), but does not fit well some recent, careful experi-
ments; also, the activation energies corresponding to $m$ are offly low, and activation
volumes are poorly represented. We concluded that an exponential behaviour is only
approximate, and is incorrect for small $\tau^*$. 

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The final equation for the flow stress, (applied in particular on Figures 15 and 17), can now be obtained by combining equations (5), (6), (7) and (11):

\[ \tau(T, \dot{\varepsilon}) = \tau_0(1 - \alpha T) + \tau_p[1 - \left(\frac{kT}{H_c'\dot{\varepsilon}}\right)^{\frac{1}{m'}}]^{m'} \]  

(12)

Note that equation (12) assumes that \( H = G \) in (11). A more exact form of the equation is actually used, in which the entropy is accounted for by the temperature dependence of the shear modulus \( \mu(T) \).

The stress calculated by means of Equation (12) can be plotted, either relatively to the temperature, e.g. see top of Figure 15, or relatively to the activation enthalpy (energy), bottom of Figure 15 and Figure 17. The strain rate - temperature sensitivity is lumped into the activation energy (enthalpy) parameter

\[ H = kT \ln \frac{\dot{\varepsilon}_0}{\dot{\varepsilon}} \]  

(13)

which brings together into a single trend all experimental data at the various strain rates. The Peierls' component vanishes at \( H = H_c' \), the "Activation Energy". To this does correspond a critical temperature above which the measurements provide the athermal stress component.

The present work suggests that strain rate effects on fracture toughness cannot be anticipated to correlate to the material's yield strength alone, as proposed by Barsom [88], nor to \( \Delta \sigma_y/\sigma_y \) alone [89]. Also, the existence of a fundamental correlation between the intrinsic strain rate sensitivity \( \dot{\varepsilon}_0 \) and the free energy in (5) has been evidenced by our scrutiny of literature data for binary iron-based alloys; this, too, bears on the toughness strain rate sensitivity. Further elaboration of these points will be left to future papers.

**Combination of C. Impact and Static Tensile Tests.**

It is useful at this time to specify further the relationship between the general yield load \( PG_y \) and the uniaxial tensile stress \( \sigma_y \). The general yield load \( PG_y \) for a notched specimen tested in three or four point bending can be related to the uniaxial yield stress \( \sigma_y \), by considering slip-line field solutions [90] for the form of the plane strain plastic zone below the notch. A number of papers have dealt with this question [91]-[93]. The **Constraint Factor** \( \phi \) is defined as the ratio of the general yield load for the notched bar to the load causing general yielding of an un-notched specimen of same cross-sectional area (tested at the same strain rate as existing below the notch). Note that \( \phi \) refers to the applied load, hence to the applied *mean* stress required for yielding all across the notched cross section. This differs from the **Plastic Stress Concentration Factor** \( K_p \) [eq. (2)], which, by contrast, is defined as the ratio of the *maximum* tensile stress below the notch root, to the tensile stress of an un-notched specimen (again at same strain rate, ...; see also [47] p.300).
Typically, $K_{op}$ is of the order of 2.5, while $\zeta$ is about 1.3.

For an ASTM type A $C_v$ notch impact specimen (three-point bending), the constraint factor $\zeta$ depends on the tup indenter width [93]:

- $\zeta = 1.363$  ASTM E23
- $\zeta = 1.274$  DIN 50115

while for the same specimen subject to pure (four-point) bending:
- $\zeta = 1.261$ (essentially independent on the notch shape).

The bending moment $MG_y$ in three- or four- point bending is given by

$$MG_y = \frac{\zeta}{2} (W-a)^2 B \tau_y$$

and this is equal to $PG_y/2$ times the bending arm $S/2$

where

- $W =$ Specimen width
- $a =$ Notch depth
- $W-a =$ Ligament below the notch
- $B =$ Specimen thickness
- $\tau_y =$ Shear yield stress
- $S =$ Loading span (For $C_v$ testing: $S/W = 4$).

Thus, the general yield load $PG_y$ (kN) and the shear yield stress $\tau_y$ (MPa) are related as follows:

$$PG_y = \frac{2 \zeta (W-a)^2 B \tau_y}{S}$$  \hspace{1cm} (14)

and the tensile yield stress $\sigma_y$ can be inferred from $\tau_y$ using either a Tresca or Von Mises yield criterion:

- Tresca: $\sigma_y = 2 \tau_y$  \hspace{1cm} (14.a)
- Von Mises: $\sigma_y = \sqrt{3} \tau_y$  \hspace{1cm} (14.b)

It has been our practice to scale the entire $C_v$ load diagram according to (14), in order to allow its evaluation in combination with uniaxial static tensile test data. We have found that a criterion midway between the above ones provides best overall consistency. On another hand, no effort has been made to assess a constraint factor strictly applicable to the maximum load: the factor used at general yield has simply been assumed valid at larger deflections; by doing so, no difference has been found, in general, between the static ultimate tensile strength and the corresponding dynamic $C_v$ maximum stress in the temperature range governed by the athermal component of the plastic flow; of course,
differences occur in the range of strain rate sensitivity. This observation encourages one to think that the strain rate sensitivity of the work hardening coefficient (N in the Ramberg-Osgood relation) can be estimated from comparison of $C_y$. Impact to Static Tensile data; this feature is relevant to the development of our "Modified RKR Model".

Extensive attention has been devoted at scrutinizing the procedure to establish $C_y$ load diagrams to accuracies suitable for engineering, surveillance applications. Various testing installations have been directly or indirectly intercompared. A particular concern in the present context was to decide whether sufficiently reliable use could be made of data recorded with older machines, featuring maybe less fast electronics than required by some modern standards. A favourable conclusion has been reached, which should allow to reanalyze many PWR surveillance and test reactor experiments along the lines of the present approach. An example of such quality assurance effort is displayed on Figure 18. The Figure also summarizes various important features of such diagrams: small scatter, insensitivity to notch orientation and to hammer geometry, ...

**Microcleavage Fracture Stress.**

Basically, the $C_y$ load diagram also allows to define the microcleavage fracture stress; the latter is generally invariant upon all service conditions, unless non-hardening embrittlement takes place. Figure 19 provides a composite illustration to assist the subsequent discussion.

The determination of the microscopic fracture stress is important, in the advanced surveillance perspective of this report, for three reasons:

1) Definition of improved fracture toughness indexation temperature, including dynamic versus static shifts.
2) Damage modeling.
3) Micromechanically supported experimental definition of $K_{IC}$ shift and lower bound for irradiated $K_{IC}$ and $K_{ID}$ curves.

Scatter is also associated to the microscopic fracture stress - equal to the microcleavage fracture stress $\sigma_f^*$ if transgranular cleavage (by slip or twinning) is the governing brittle failure mode. It is generally accepted that $\sigma_f^*$ is independent of test temperature, strain rate and irradiation embrittlement: the usual argument is that it relates to quite large, and stable microstructural features (grain boundary carbides, bainitic lath substructure, ...).

Nevertheless, even from a microscopic viewpoint, $\sigma_f^*$ (microcleavage) is of a statistical nature; it does not characterize a single step process, like breaking the bond between two atoms, but a succession of at least two, uncorrelated events, such as initiation of a microcrack in a brittle particle, and its further propagation across a boundary - whose orientation is random, favourable or not (see for ex. [94]).

In any event, any useful measurement of $\sigma_f^*$ is macroscopic and its result has a statistical distribution, function of the microstructure and of the stress field in the vicinity of stress concentrators: a blunt notch or a sharp crack do "sample" differently the size and space distribution of fracture-initiating particles and thus will not lead to the same mean value,
as can be grasped easily by a glance at Figure 20. The stress needed to crack a particle is inversely proportional to the square root of its size, and the microcleavage fracture stress is the statistical mean of the critical tensile stress with respect to the cleavage fracture probability. The uniaxial tensile test in particular tends to reflect the strength of the largest "eligible" feature, while a sharp precrack can trigger some of the more numerous, smaller particles whose strength is matched at some point during loading. All this is well known [20]-[25].

Although the mean microscopic fracture stress defines the magnitude of the "mean" fracture toughness, its scatter does not normally govern the toughness lower bound. This is illustrated by Figure 21, using normalized, two-parameter Weibull plots. The Weibull \( m \) parameter, which describes the scatter, generally ranges from about 10 to 60 for the microscopic fracture stress, while its theoretical value is 4 for \( K_{\text{in}} \) and 2 for COD (or \( J_{\text{c}} \)). It is the initiation site location, and thus the "Critical Activated Volume" \( V^* \), which controls the scatter of the fracture toughness and its lower bound.

Returning to Fig. 19, upper insert, it might seem that \( \sigma_{T^*} \) is quite exactly defined by equation (2) applied at \( T_D \) or \( T_T \). Actually, the scatter is hidden and would appear if the equation had also been applied below \( T_D \), using the appropriate plastic concentration factor, in function of \( \sigma_{\text{max}}^{\gamma} / \sigma_{\gamma|T_D} \) (extrapolating the denominator below \( T_D \), by means of the Dorn-Rajnak model). The values of \( \sigma_{T^*} \) at \( T_T \) can also be derived from [31] [47]:

\[
E = g P G^2 Y^2 \left( \frac{\sigma_{T^*}}{K_{\text{ap}} P G Y} - 1 \right)
\]

(15)

where:  
\( E \) = total absorbed \( C_t \) energy (J)  
\( P G Y \) = general yield stress (MPa)  
\( g \) = fitting constant

This is illustrated by the lower insert of Fig. 19, and the result is in good agreement with the direct application of (2) at \( T_T \). However, insofar as no effect of irradiation, anneal, ... is detected for the considered steel, the scatter is now accounted for by consideration of an ensemble of \( C_t \) curves, rather than a single one, so that confidence in the mean value obtained for \( \sigma_{T^*} \) is significantly enhanced.

It must nevertheless be reminded that the current Belgian R&D approach does also consider a number of static tests at or below \( T_D \) to allow unravelling, through a Weibull-type plot, whether the fracture is purely transgranular and unimodal (if not, to find what value of \( \sigma_{T^*} \) should apply to each mode). An example involving admixture of intergranular fracture is described in [95]. In other instances, a small population of manganese sulfides ([96], [97]) in presence of a large population of cracked carbides may govern the lower bound. This is why SEM measurements on fracture surfaces are deemed most important in the RPV surveillance framework, even if specific initiators cannot always be identified [98],[99].
Figure 22 compiles a number of microcleavage fracture stress measurements for various steels and specimen geometries (It is fortuitous that the mean values are similar for these steels). Note that the literature data have been re-evaluated, using in particular the calculations of Xu et. al. [100] for the Griffiths-Owen [26] specimens. This figure aims at illustrating the type of approach followed in developing the "Modified RKR Model". Not only does one address strain rate effects as outlined above, but also the influence of notch acuity (i.e. notch versus precrack), and of specimen design. The objective is to "recuperate" commercial surveillance data in a more physically-grounded (damage modeling), and micromechanically-grounded, fracture mechanics perspective.
5. CONSTRAINT, SIZE AND STRAIN RATE EFFECTS FOR C, NOTCH IMPACT TEST.


The reconstitution of broken C, specimens by the stud welding technique [101] has been thoroughly and successfully qualified at SCK-CEN over the past decade (e.g. [102],[103]). Normally, this entails a central insert, 15-20mm long, of the original, "As-received" material, out of which a new sample is to be fabricated.

The surveillance programs of older plants often do exclusively encompass base metal specimens representative only of the 'strong', L-T notch orientation, well suited for normal and upset operation conditions (hoop pressure stresses governing). More recent concerns with hypothesized accidental occurrences, featuring large thermal stresses, have driven the reconstitution technology towards the challenge of "Notch Reorientation", i.e.: L-T to T-L.

Upon request and with the sponsorship of Yankee Atomic Company, a well documented demonstration experiment ([103]-[105]) has been realized using the unirradiated plate HSST-03. The maximum "As-received" volume here is a cube 10x10x10 mm, but original specimens of both orientations were available and tested, so that four conditions are represented: see Figure 23. (Some "As-received" MEA data for the same original inventory agree well with the Mol results). The energy differences among the four conditions are all traceable to deflection differences before the arrest load, Figure 24; post-arrest deflections and energies (Fraction C) are invariant. The lateral expansion is also decreased, but to less an extent, because reflecting deformation effects nearer to the crack plane. Constraints by the hard welded zones have been well assessed: the local perturbation of the plastic flow causes the reconstitution losses. On another hand, T-L to L-T differences increase with the volume fraction of inclusions and are linked to their shape and spacing - all factors which affect cavity growth and eventual coalescence with the main crack front. In other words, the four curves on Figure 23 are distinct because tearing and plastic deformation are differently influenced, either by constraint, or by microstructure; the differences tend to vanish with decreasing plastic flow capability (hardening), and decreasing inclusion content for what concerns orientation effects. Thus, one primarily deals here with upper shelf ductility differences. This should bear no direct relationship with elementary yielding and work hardening, nor with the microcleavage fracture stress: the C, load diagram, fracture appearance and the ductile-brittle transition temperature should essentially not be influenced. This is exactly what Figure 25 does confirm. This particular type of experiment has been repeated at other occasions, for other RPV steels, with similar results.

Such data are important because they point to a longstanding misconception in literature, regarding the relation or lack of relation between the C, test and fracture toughness. A basic position of this paper is that "Tearing energy (C, Fraction B)" does not belong to the realm of structural integrity analysis of a pressure vessel at transition temperature regimes (below Td).

Figure 25 furthermore implicates that plane strain conditions are essentially realized in the crack propagation plane of a C, notch specimen, up to the upper shelf onset tempera-
ture $T_p$.

If advantage is to be taken of ductile stable crack growth in the transition range, a possibly untractable mismatch between tearing and cleavage failure criteria can only be avoided by adequate plastic flow constraint - whether the specimen be notched or precracked.

Based on the above, it was also concluded that the side grooving and precracking of reconstituted 10x10x10mm "As-received" Charpy inserts should remove the interaction of the plastic flow with the hard welded zones, and possibly allow to measure the plane strain toughness with such minimal inventory of material. To prove the prediction, a modest, scoping experiment was undertaken in cooperation with B. Neale (Nuclear Electric, UK). It was decided to use plate HSST-03 specimens from the same stock as above, and to compare the static initiation fracture toughness at 260°C before (i.e. full length, normal specimen) and after reconstitution. No difference was indeed observed.

**A Preliminary Look at C, Miniaturization.**

The load diagram approach has been applied to a scoping experiment using unirradiated Doel 4 base metal. The ASTM practice was followed for the conventional-size specimens in the comparison, while miniaturization was done according to the German standard DIN 50115. The details will be reported elsewhere.

It is generally observed that such C, miniaturization shifts the curves to lower temperature [106]-[109]. Only this aspect is briefly discussed here.

It can be seen from the load diagrams on Figure 26 that strain rate does play a major role here. Once accounted for, i.e. turning to an activation energy abscissa, it is remarkable that no size effect is found insofar as $T_p$ is concerned, thus supporting its validity as toughness indexation temperature.

The measurements did not extend to sufficiently low temperature to define $T_D$ for the miniature geometry. However, there is a clear size effect on $T_p$, and it is not due to an enhanced plastic stress concentration factor in Orowan's relation (2), of course - as this factor does instead decrease with the size reduction at equal a/W ratio, ... Indeed, referring to Fig. 22, the miniature specimen would fall somewhere on the steeply ascending branch of the curves. The stress field is more flat for the small specimen as compared to the large one; therefore, its mean microscopic fracture stress is smaller. This interpretation is being verified by finite element calculations.

All in all, the largest effect is the one of strain rate, and its modeling prediction agrees well with the KWU trend band [108], as shown by Figure 27.
6. CONCLUSIONS.

Nuclear Regulation relies on the C, notch impact test for indexation of fracture toughness degradation of reactor pressure vessel steels upon service exposure.

Yet, it is well known that, from a structural integrity analysis viewpoint, this test suffers a number of limitations, among which:

1) Governance of propagation energy and arbitrary use of energy "fixes" (e.g. 41J)
2) Strain rate unrepresentative for static initiation conditions
3) Specimen unrepresentatively constrained (as compared to vessel)
4) Shallow notch (rather than pre-crack) as stress concentrator

The most critical deficiency of the current engineering "indexation" approach stems from item 1) above; this is a serious potential source of mismatch which can cause large distortions and faulty safety judgments for certain steels; furthermore, this does unduly enhance the scatter of predictive chemistry-fluence correlations and associated Regulatory guides for RPV embrittlement projection.

All these deficiencies can be accounted for along the lines developed in this paper. Reconstitution technologies allow to re-visit prior surveillance programmes and to expediently obtain additional experimental data, including $K_{IC}$ whenever needed for implementation of the proposed, consolidated surveillance strategy.

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APPENDIX

Equation (4) of the paper can be rationalized as follows.

As is well known, to first approximation, loads scale to the surface under the notch, i.e. to \((W-a)B\), where \(W-a\) = ligament, and \(B\) = thickness; thus, area is proportional to \(F\).

Load change associated to brittle crack propagation is

\[
\delta F_{\text{brittle}} = F_a - F_s
\]

Load change associated to ductile crack growth is composed of

- Propagation before max. load: \(F_m - F_1\)
- Propagation after max. load: \(F_m - F_s\)
- Propagation by lip formation: \(F_s\)

thus:

\[
\delta F_{\text{ductile}} = F_m + (F_m - F_1) - (F_a - F_s)
\]

Shear fracture area is equal to

\[
SFA(\%) = \left[ 1 - \frac{\delta F_{\text{brittle}}}{\delta F_{\text{brittle}} + \delta F_{\text{ductile}}} \right] \times 100
\]

or

\[
SFA(\%) = \left[ 1 - \frac{F_a - F_s}{F_m + (F_m - F_1)} \right] \times 100
\]

which is identical to (4) if one uses (3) to express \(F_1\).

It is obvious from (4) that \(SFA = 100\%\) only if \(F_a = F_s\), which corresponds to the temperature \(T_o\) of Fig. 6, right-hand side.

On another hand, \(SFA = 0\) requires that \(F_aF_s = F_m + (F_m - F_1)\), which is possible in all generality only if:

\[
F_s = 0
\]

and

\(F_a = F_s\) (as there is no ductile initiation at this point)

or, given (3) and the fact that \(k > 0\): \(F_f = F_m = F_s\).

This condition corresponds to \(T_r\), the ductile initiation temperature (Fig. 6).
Fig. 1  REGULATORY INDEXATION OF FRACTURE TOUGHNESS FOR RPV STEELS

- Crack Initiation and Arrest Toughness
  Taken as Unique Functions of Temperature
  Indexed to

  REFERENCE TEMPERATURE  RT_{NDT}

- Lower Bound Toughness (ASME XI Code):
  \[ K_{IC} (\text{MPaVm}) = 36.5 + 3.084 \exp[0.036x(T+56-RT_{NDT})] \]
  \[ K_{IA} (\text{MPaVm}) = 29.5 + 1.344 \exp[0.026x(T+89-RT_{NDT})] \]
  Temperatures in °C

- Un-Irradiated RT_{NDT} Equal to
  Drop Weight NIL DUCTILITY TEMPERATURE
  NDT
  Unless Charpy Impact Energy at NDT+33°C
  Lower than 68 J

- Embrittlement Shifts Toughness Curves
  According to Recipe

  \[ \Delta RT_{NDT} = \Delta TT41 \]
  i.e. Shift of Cv- Impact 41J Temperature
Fig. 2  Irradiation Shift of $K_{JC}$-, $K_{LA}$- and $Cv$- SFA

HSSI High Copper Weld 73W

---

Fracture Toughness $J_c$ (kJ/m²) vs. Test Temperature (°C)

Fracture Toughness (kJ/m²) or $Cv$- Shear (%) vs. Test Temperature (°C)
Fig. 3  INGREDIENTS OF IMPROVED RPV FRACTURE TOUGHNESS
SURVEILLANCE STRATEGY FOR BELGIAN POWER PLANTS

Enhanced "Classical" Surveillance
- Tensile Tests (True Stress/Strain) Over Extended Temperature Range
- Instrumented Cv- Impact Tests Over Extended Temperature Range
- Static Fracture Stress Tests at Selected Temperatures

KJc Tests (Selected Temperatures)
- 3PT Slow Bend Tests (Reconstituted Precracked Cv' s)
- Precracked Axisymmetric Notched Tensile Bars

Statistical Micromechanics
- Modified RKR Model
- Finite Element Backing

\[ K_{lc,d} = F [\sigma_{y,c,d}, \sigma_f^*, N, l^*] \]

- \( \sigma_{y,c,d} \): Static, Dynamic Yield Stress
- \( \sigma_f^* \): Microcleavage Fracture Stress
- \( N \): Work Hardening Exponent
- \( l^* \): Characteristic Microstructural Dimension (Volume \( V^* \))

Strain-Rate Effect on Initiation Fracture Toughness
Plate HSST-02 (Unirradiated)
Modified RKR Model

---

*Plastic Zone Size smaller than "Characteristic Microstructural Distance"
Fig. 4  DUCTILE-BRITTLE TRANSITION

**DUCTILE-BRITTLE TRANSITION TEMPERATURE (DBTT)**

- **GENERAL YIELD STRESS**
  - CLEAVAGE FRACTURE STRESS
- For Temperature > DBTT:
  - Strain Hardening Needed for Fracture
    (Plastic Deformation)

**"CLASSICAL" MATRIX HARDENING**

- Irradiation Creates Obstacles to Movement of Dislocations
  (for ex., Precipitates)
- Material Flow is Hampered
  i.e. Yield Stress Increases
- **DBTT INCREASE PROPORTIONAL TO YIELD STRESS INCREASE**

**Guidelines to the "Modeling" of Steel Embrittlement**

**NON-HARDENING EMBRITTLEMENT**

- **DECREASE OF FRACTURE STRESS**
  CAN CAUSE FURTHER DBTT INCREASE
- **SEGREGATION AT GRAIN BOUNDARIES**
  (Or at Lath Boundaries, ...)
  Intergranular Fracture
  Phosphorus, Tin, Antimony, ...
- **GAZ EMBRITTLEMENT**
  - Nitrogen, Hydrogen, Helium
  - Fracture Remains Transgranular?
Fig. 5
A302B PLATE TENSILE YIELD STRENGTH AFFECTED BY
3 DISTINCT DAMAGE MECHANISMS

STRENGTH INCREASE (MPa)

1000

100

10

ASTM REF. PLATE

YANKEE SURV. PLATE

149 C

290 C

COPPER PRECIPITATION

2A

2

FLUENCE >1MeV (cm\(^{-2}\) x 1.0E19)
Fig. 6 USE OF INSTRUMENTED CHARPY-V LOAD-TIME TRACES

Typical Signal
(Transition Temperature Range)

Typical Load Diagram and Energy Partitioning

LSE (Lower "Shelf" Energy)
Fig. 7  Charpy-V Notch Load Diagram and Energy Partitioning

In Addition to a Characteristic Temperature Matching the Load Diagram, Each Energy Fraction is Characterized by its Transition Temperature (TTA, TTB, TTC), Taken at 50% of its Shelf Energy.
Fig. 8 Ductile Crack Initiation in Charpy-V Notch Impact Test

MAGNETIC EMISSION

LOAD

Maximum

Yield

Ductile Initiation

11 J.

Unirradiated A533-B Cl.1 Plate HSST-02 (L-T), DIN Hammer, 4.9 m/s., Total Energy: 161 J.
Fig. 9  Magnetic Emission Detection of Crack Initiation for Charpy-V Notch Impact Test of RPV Steel

At Ductile Crack Initiation:

Absorbed Energy: 6 to 14 J.
Deflection : 1.0 to 1.2 mm

Bulk of Absorbed Cv-Energy Linked to:

DEFORMATION
STABLE TEARING

Good Agreement of Experiment with Finite Difference Calculation
Correlation of Fracture Appearance to Instrumented Cv- Impact Load Diagram

Doel-4 Un-Irradiated Base Metal
FATT - Shift

- Governed by Post-Arrest Response
- Can be less than 41 J - Shift

Fig. 11

Energy (J)

Temperature (°C)

Baseline

Irradiated

Shear (%)
Upon Irradiation of Considered Steel

CONTROL OF 41 J. TEMPERATURE "SHIFTS"

FROM PRE-MAXIMUM ENERGY FRACTION IN BASELINE TO POST-ARREST ENERGY FRACTION AT 5E19 cm⁻²

BIASING 41J TOUGHNESS FIX
Fig. 13  FRACTION B OF LOAD SIGNAL GOVERNS DATA SCATTER IN TRANSITION TEMPERATURE RANGE

Fractional Energy (J)

Test Temperature: 40-41 °C

Total Energy (J)

PLATE HSST-03 (Unirradiated)
Fig. 14 Cv-Notch Impact Test Evaluation Approach
Fig. 15  Temperature and Strain Rate Sensitivity of RPV Steel: Modeling- Based Evaluation Approach

Lower Yield Stress (MPa)

Strain Rate (sec⁻¹)
- .0001
- 1.0
- 100.

Temperature (K)

MODEL
- Dorn- Rajnak
- Fleischer

Activation Energy Parametrization
Data at 6 Strain Rates: .0001 to 100 sec⁻¹

Lower Yield Stress (MPa)

MODEL
- Dorn- Rajnak
- Fleischer

"Peierls" Component

Athermal Component

Steel FE E 460
Aachen University
Fig. 16  Lattice Resistance to Screw Dislocations:
CANDIDATE FLOW-CONTROLLING BARRIERS

Shear Stress/Peierls' Stress

Dorn-Rainak Model
- Sinusoidal
- Perturbed (-1)
- Quasi-Parabolic

Smidt 1969 (Empirical)

Fleischer Model

Normalized Activation Energy (H/Hc)

Generic Expression for Considered Barriers:

Shear Stress Ratio = \[1 - (H/Hc)^{1/m'}\]

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<thead>
<tr>
<th>BARRIER</th>
<th>m</th>
<th>m'</th>
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<td>Perturbed (-1)</td>
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<td>Quasi-Parabolic:</td>
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<td>Smidt 1969 (Empirical):</td>
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<tr>
<td>Fleischer:</td>
<td>2.0</td>
<td>2.0</td>
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</table>
Fig. 17 Application of Dorn-Rajnak Model to Iron and Selected Steels

- **Lower Yield Stress (MPa)**
  - Annealed Al-Killed Mild Steel (●)
  - Pure Iron (△)

- **Activation Energy (in 10E-20 J)**

- **Steel C10 (Aachen University)**
  - Strain Rate (sec⁻¹)
    - .0001 to .01 (●)
    - .1 (△)
    - .5 (△)
    - 1 (△)
    - 10 (△)
    - 100 (△)

- **Lower Yield Stress (MPa)**

- **Activation Energy (in 10E-20 J)**

- **20MnMoNi55 (Quenched & Tempered)**

- **Strain Rate (sec⁻¹)**
  - .0001 (●)
  - .01 (△)
  - 10 (△)
  - 100 (△)

- **General Yield Stress (MPa)**

- **Activation Energy (in 10E-20 J)**

- **A508 CL3 (Doel-4 Base)**

- **Strain Rate (sec⁻¹)**
  - 300 (Gy-impact) (●)
  - .0002 (Tensile) (△)
Fig. 18 Quality Assurance for Cv- Impact Load Diagram Procedure

For Forcing Ring for EGF Round Robin on 'Local Approach'

Siemens Research Plate (Balt et al. 1991)

ASTM E23  □ LHMA
DIN 50165 □ VITO
□ FhG □ STATIC TENSILE

L-T & T-L Data Undistinguishable

22NiMoCr37 Unirradiated

Reconstitution of KKW-1 Surveillance Plate

For Three Considered Steels:
Uniaxial Static Yield and Cv- Impact
General Yield: Consistent

Diagram Insensitive to
- Notch Orientation
- Hammer Geometry
- Reconstitution

Diagram Data Scatter: Usually Small

Good Interlaboratory Agreement
of Tests at LHMA, VITO
SIEMENS, FhG
Fig. 19 Brittleness Temperature $T_D$ and Microcleavage Fracture Stress for Charpy-V Impact Test

Generally, under **Surveillance Impact Conditions**

**Brittleness Temperature** $T_D = \text{Ductile Initiation Temperature} T_I$

Static and Dynamic Yields Are Consistent at High Temperature

Microcleavage Fracture Stress as Derived from Instrumented Cv- Load-Deflection Traces **Generally Invariant** upon Irradiation, Annealing and Post- Anneal Re-irradiation Conditions
Fig. 20 Stressed Volume Dependency of Microcleavage Fracture Stress

Typical Size Distribution of Potential Microcrack Initiators

Silicate Inclusions C-Mn Electroslag Weld
Cracking Strength (Relative Scale)

Maximum Principal Stress / Yield Stress

n = 0.15
C = 560 MPa

PRECRACKED
R.M. McMeeking (1977)

NOTCHED
X.X. Xu et al. (1989)
Griffiths & Owen (1971)
Fig. 21 Scatter Sources for Slip-Induced Brittle Cleavage Fracture

Failure Probability \(1 - \exp[-(X/X0)^m]\)

C-Mn Weld Tests at 70°C

\[m = 50\]
\[m = 1.7\]
\[m = 2\]

- Microcleavage Fracture Stress \(\sigma_f^*\)
- Initiation Site Distance to Crack Tip
- Initiation Fracture Toughness (COD)

Relative Value of Parameter \(X/X0\)

Data: J.H. Chen et al. (Metall. Trans. 22A, 2287, 1991)
Fig. 22  Influence of Stress-Strain Field Concentration on Microcleavage Fracture Stress

Symbols

This Work: 22NiMoCr37 Base
- ○ Notched Tensile
- ■ Smooth Tensile
- ● Cv Impact
- ○ PCCv Impact

J.H. Chen et. al. *
- △ C-Mn Base  (n= 0.243)
- ◆ C-Mn Weld  (n= 0.119)

Beremin Group
- □ A508 Cl3 Base

* Cv- and 4PT Bend Data Rescaled to FEM Calculations by X.X. Xu et. al.
DEFORMATION CONSTRAINTS BY HARD WELDED ZONES AND NOTCH ORIENTATION CAUSE SIGNIFICANT ENERGY DIFFERENCES IN Cv- IMPACT TEST

* Cv- Reconstitution: 10x10x10 MM REMNANTS, Plate HSST-03 Un-Irradiated Condition STUD WELDING TESTS PER ASTM E23
Deflection $(T) = E(T) \times 0.066 \text{ mm/J}$
Fig. 25  INVARIANCE OF Cv- LOAD DIAGRAM AND FRACTURE APPEARANCE

A533-B Plate HSST-03 (Un-Irradiated) Cv Tests with ASTM Hammer
L-T & T-L  Reconstituted Data: 10 x 10 x 10 mm Remnants

All Data Fitted by Unique Set of Parameters
Fig. 26 Load Diagram for Miniaturized Charpy-V Impact Test

Doel 4 Base Metal Un- Irradiated

Activation Energy Parametrization
Fig. 27 Strain Rate Effect on Miniaturization of Cv- Notch Impact Test

**MODELING PREDICTIONS**

Temperature Shift (°C) *

Reference Pre-Exponential Rate (sec⁻¹):
- 1.0 E8
- 1.0 E9
- 1.0 E12

Activation Energy (in 10E-20 J)

**COMPARISON TO LITERATURE**

Miniature Cv- Transition Temperature (°C)

Modeling for 3 Strain Rate Sensitivities
*i.e., Reference Pre-Exponential Rate (sec⁻¹) of:*
- 1.0 E12
- 1.0 E9
- 1.0 E8

Reference Cv- Transition Temperature (°C)

**This Work**
- 41 J.
- 50%
- 68 J.

Shear

*Reference Cv (ASTM E23) - Miniature Cv (DIN 50115) [8x4 mm]*
EFFECT OF CHEMICAL COMPOSITION ON IRRADIATION EMBRITTLEMENT
AND ANNEALING IN Ni-Cr-Mo-V REACTOR PRESSURE VESSEL STEEL

Petr NOVOSAD

Nuclear Research Institute Řež plc, 250 68 Řež near Prague,
Czech Republic

to be presented

at the

Joint IAEA/NEA Specialist's Meeting on Irradiation Embrittlement
and Optimization of Annealing

Paris, France 20-23 September 1993
EFFECT OF CHEMICAL COMPOSITION ON IRRADIATION EMBRITTLEMENT AND ANNEALING IN Ni-Cr-Mo-V REACTOR PRESSURE VESSEL STEEL

by
Petr NOVOSAD, Nuclear Research Institute Řež plc, 250 68 Řež near Prague, Czech Republic

ABSTRACT

Paper summarizes results of study of copper and phosphorus influence on radiation induced changes of Ni-Cr-Mo-V steel mechanical properties.

Correlation between different mechanical properties for steels with different chemical composition is presented. Comparision of transition temperature shifts obtained for static and dynamic fracture toughness tests and Charpy impact tests is discussed. Recovery of radiation hardening, measured by hardness test after isochronnal annealing of steels with different compositions is also shown.

Copper content affects strongly irradiation induced changes of mechanical properties, but phosphorus content in connection with variable copper content has only small effect on mechanical properties degradation by neutron irradiation.

Keywords: reactor pressure vessel steels, radiation embrittlement, chemical composition, fracture toughness tests, annealing
INTRODUCTION

The Cr-Ni-Mo-V steel is used for manufacturing of PWR VVER 1000 since 1980. Within the framework of cooperation with USSR in production of nuclear power plant pressure vessel component, appropriate czechoslovak made reactor pressure vessel steel of this type was designed.

Programme for resistance to radiation damage testing included accelerated irradiation experiments of steels with different copper and phosphorus content.

Irradiation embrittlement in terms of transition temperature shifts of Charpy impact energy level and static and dynamic fracture toughness were evaluated after irradiation in experimental reactor. Annealing experiments were carried out on the hardness tests.

MATERIALS

For this experiments were used six heats doped with copper and phosphorus, made from original 15CH2NMFA Grade AA heat. All six experiment materials have received simulated heat treatment: 920°C/2 hours/air + 650°C/10 hours/air + 620°C/25 hours followed 650°C/20 hours/furnace and up to 300°C/air.

From this materials were made Charpy-V type samples for both static and dynamic fracture toughness tests and for impact tests. Specimens for static fracture toughness testing were fatigue pre-cracked and side grooved. The microstructure of investigated materials is a mixture of tempered bainite and martenzite.

Chemical composition of each examined heats is given in Table 1.

IRRADIATION CONDITIONS

Irradiation was carried out in LWR pool type experimental reactor LVR 15, using electrically heated irradiation rig Chouca - MT located inside the core.

Neutron flux during irradiation was $1.3 \times 10^{17} \text{n.m}^{-2}\text{s}^{-1}$, E $\geq 1\text{MeV}$.

Neutron fluence was evaluated using reaction on Cu and Co with respect to measured
neutron spectrum, it was $2.6 \times 10^{23}$ n/m$^2$, $E > 1$ MeV.

Temperature during irradiation was monitored and controlled by thermocouples with computer control, it was $288^\circ \pm 7^\circ C$.

EXPERIMENTAL PROCEDURES

All tests were carried out in NRI laboratories.

For tensile and static fracture toughness tests are used computer controlled servohydraulic INSTRON machines.

Tensile properties were measured on specimens of 4 mm diam. and 20 mm length at ambient temperature by $4.2 \times 10^{-3}$ s$^{-1}$ deformation rate.

Static fracture toughness tests were performed within temperature range -190$^\circ$C to 200$^\circ$C by use of sidegrooved precracked Charpy-V notch specimens for single specimen unloading method. Displacement at load point is measured remotely using high linear LVDTs.

Dynamic fracture toughness tests were performed on instrumented, computer controlled TINIUS-OLSEN pendulum impact machine within temperature range -190$^\circ$C to 200$^\circ$C. Impact velocity was 1.5 m/s to 5.6 m/s. All Charpy-V notch specimens were precracked and had no sidegrooves.

Charpy impact energy levels were measured using standard testing methods and equipment with working capacity 294 J and tup velocity 5.6 m/s within temperature range -190$^\circ$C to +200$^\circ$C. The test temperature was measured and controlled with accuracy better than $\pm 2^\circ$C.

Annealing effect was studied by annealing of disc 8 mm diameter for hardness HV 30 measurement. Hardness was measured using BMCOTEST digital hardness tester. Annealing was carried out within temperature range from 288$^\circ$C to 600$^\circ$C isochronnaly (1 hour) in vacuum.
RESULTS

Tensile properties, measured at ambient temperature are used for $B_F$ parameter calculation, defined as:

$$\Delta R_p0.2 = B_F(F \times 10^{-22} \text{n.m}^{-2})^{1/3}$$

$B_F$ is characterising irradiation hardening and their values for all six heats are in Table. 2.

Charpy impact energy levels vs. temperature are fitted in the form by Oldfield:

$$K_{CV} = A + B \tanh \left((T - T_0)/C\right)$$

Transition temperature and transition temperature shifts are in Table. 4. From this values a parameters $A_F$, defined as:

$$\Delta TT = A_F(F \times 10^{-22} \text{n.m}^{-2})^{1/3}$$

are calculated and summarised in Table 3.

Dynamic and static fracture toughness levels vs. test temperature are fitted (in transition region) by relationship:

$$K = A + B \exp(CT)$$

Transition temperatures and transition temperature shifts for 100 MPam$^{1/2}$ level are in Table 4. Parameter $A_F$ as described above is in table 3.

Recovery of hardness HV 30 after annealing is summarised for all six heats in Fig. 1

DISCUSSION

Observed values of irradiation hardening are in expected dependence on chemical composition, value of $B_F$ increased with increased content of copper, role of the phosphorus
is not so visible.

Irradiation embrittlement measured by TT shift and $A_F$ value for static fracture toughness, dynamic fracture toughness and Charpy tests showed the same picture. Irradiation embrittlement increased with copper content, by negligible effect of phosphorus. Differences of properties for dynamic and static tests in unirradiated conditions for all heats is still not fully explained /1/. There is no direct influence of chemical composition, some explanation should be a scatter of test results in transition temperature region due small amount of test samples, precracking condition etc.

In contrary to this fact, the cross-correlations between static fracture toughness transition temperature shift, dynamic fracture toughness transition temperature shift and Charpy transition temperature shift appears to be good linear Fig. 2,3,4. It is recognised, that usually quoted correspondence 1 to 1 is only between CVN (Charpy) and $K_{ID}$ (dyn. fract. tough.) Fig.2, but for other properties correlations 1 to 1 is not fully satisfied Fig.3,4 /2,3/.

Recovery of radiation hardening, measured as hardness changes is influenced by impurities content only in first stage of annealing by value of primary irradiation hardening /4/ Fig.1.

CONCLUSION

Radiation hardening depends on amount of Cu and P, influence of P is not important in this case.

The TT shift increased with increasing content of Cu, for CVN, static and dynamic transition temperature. Influence of P is far less significant.

This shifts are of the same value for CVN and $K_{ID}$, but not for $K_{IC}$

Recovery rate is higher for higher content of Cu, no detectable effect of P.

The irradiation induced properties changes, related with Cu and P content have their origin in sub-microstructure.
LITERATURE


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Table 1: Chemical composition of examined steels
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Table 3: Irradiation embrittlement coefficient $A_F$
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Fig. 3: Correlation between dynamic fracture toughness transition temperature shifts and static fracture toughness transition temperature shifts for heats with different Cu and P content
Fig. 4: Correlation between static fracture toughness transition temperature shifts and Charpy-V impact energy transition temperature shifts for heats with different Cu and P content
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<td>2.14</td>
<td>0.58</td>
<td>0.08</td>
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Table 1. Chemical composition of examined steels.
<table>
<thead>
<tr>
<th>Cu/P</th>
<th>$\Delta R_{P,0.2}$ [MPa]</th>
<th>$B_F$</th>
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<tr>
<td>0,08/0,014</td>
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<td>34</td>
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<td>0,52/0,018</td>
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**Table 2.** Irradiation hardening coefficients $B_F$

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<tr>
<th>Cu/P</th>
<th>$\Delta T_T$</th>
<th>$\Delta T_T$</th>
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**Table 3.** Irradiation embrittlement coefficients $A_F$
<table>
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<tr>
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<td>unirr.</td>
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<tr>
<td></td>
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<td>108</td>
</tr>
<tr>
<td>0.52/0.015</td>
<td>unirr.</td>
<td>-35</td>
<td>-101</td>
</tr>
<tr>
<td></td>
<td>irr.</td>
<td>86</td>
<td>121</td>
</tr>
<tr>
<td>0.52/0.021</td>
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<td>-44</td>
<td>-110</td>
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<td></td>
<td>irr.</td>
<td>101</td>
<td>148</td>
</tr>
</tbody>
</table>

Table 4. Transition temperatures and transition temperature shifts
Fig. 1. Recovery annealing
Fig. 2. Correlation between dynamic fracture toughness transition temperature shifts and Carpy-V impact energy transition temperature shifts for heats with different Cu and P content.
Fig. 3. Correlation between dynamic fracture toughness transition temperature shifts and static fracture toughness transition temperature shifts for heats with different Cu and P content.
Fig. 4. Correlation between static fracture toughness transition temperature shifts and Carpy-V impact energy transition temperature shifts for heats with different Cu and P content
Stability Of Thermally Induced Copper Precipitates Under Neutron Irradiation.

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AEA Technology, Harwell Laboratory
Didcot, Oxfordshire, OX11 0RA, UK

Abstract

It is well known that copper precipitation plays a key role in the degradation of mechanical properties of irradiated Reactor Pressure Vessel (RPV) steels. Depending on the thermo mechanical treatment of the steel and the irradiation conditions experienced, a range of copper precipitate size and number densities can be produced. Generally, in high copper welds (>0.3wt% Cu), typical post weld heat treatments produce stable copper precipitates ~10nm diameter. The remaining copper in solution has the potential to precipitate out under irradiation, forming coherent precipitates ~2nm in diameter, similar in size to those found in thermally aged samples in the peak hardness condition. The factors controlling the nucleation, growth and stability of copper precipitates under irradiation is therefore of importance to understanding the materials behaviour.

Model Fe 1.3%Cu and Fe 1.3%Cu 1.1%Ni alloys have been thermally aged at 550°C for 2hrs (peak) and 10hrs prior to irradiation at 288°C to a dose of 5.10^5n/m². Results of a microstructural investigation using dedicated field emission gun scanning transmission electron microscopy (FE-STEM) and small angle neutron scattering (SANS) to assess precipitate stability in the binary alloy will be presented. These data are then used to predict a hardness change as a result of copper precipitation for comparison with the measured values obtained using standard 5kg Vickers hardness tests on the SANS samples. The implications of these data to the embrittlement of the RPV by subsequent copper precipitation will be discussed.

1 Introduction

This paper describes the preliminary results from an experimental programme set up to establish the stability of copper precipitates formed during post weld heat treatment or vessel annealing in plant life extension programmes. Copper is known to precipitate out of solution under thermal or irradiation conditions and the resultant precipitates form efficient barriers to dislocation motion with a resultant increase in hardness and subsequent embrittlement. Under irradiation the copper available to precipitate in this way may not necessarily be the same as the bulk level. In high copper welds, post weld heat treatment (PWHT) can remove copper from the matrix forming copper precipitates that generally are observed as ~10nm diameter 'foe' precipitates associated with dislocations. In welds, further copper can be removed as a result of copper sulphide formation. In practice this reduces to the following equation:

\[\text{Cu}_{\text{matrix}} = \text{Cu}_{\text{bulk}} - \text{Cu}_S - \text{Cu}_{\text{HT ppt}} - \text{Cu}_{\text{irrad ppt}}\]

\[\text{Cu}_S = \text{Copper associated with sulphur in the form of copper sulphide}\]

\[\text{Cu}_{\text{HT ppt}} = \text{Copper in the form of large foe copper precipitates formed during post-weld heat treatment}\]

\[\text{Cu}_{\text{irrad ppt}} = \text{Copper in the form of small bcc precipitates formed under irradiation}\]

A measure of copper precipitation therefore requires knowledge of the copper matrix value and the development of techniques to measure this critical parameter either directly or indirectly.

In plant life extension, various annealing options are being considered that will remove the hardening effect of the small copper precipitates by thermally treating to permit these precipitates to grow into the less harmful incoherent precipitates, similar to those produced in PWHT or found in overaged model alloys. It is the stability of these large precipitates under irradiation or reirradiation that is of interest to this study as, if they subsequently dissolve under irradiation, they will act as a source of Cu for subsequent fine scale precipitation resulting in increased hardness.
Under irradiation the precipitates formed are similar in size to those generated at peak hardness in thermal ageing experiments. The microstructural investigations conducted on this and other irradiated material suggest that the irradiation induced precipitates remain small, with a coherent bcc structure, and that no over ageing takes place in the irradiated material.

With this as background information, a series of experiments were set up to examine the effect of irradiation on the size and number density of precipitates. Samples were heat treated to produce two types of precipitate, namely small coherent precipitates found at peak hardness and those found in the overaged condition. In addition to this material, as quenched samples with all the copper in solution were included as controls. Samples were examined by both electron microscopy techniques and small angle neutron scattering (SANS) to assess the distribution of copper (matrix or precipitate) and to determine the size and number density of precipitates formed under thermal or irradiation conditions.

2 Material and Irradiation conditions

Binary Fe - 1.3wt% Cu alloy, HE, used in previous studies (1) has been employed here. Samples were given a solution heat treatment of 825°C for 6hrs followed by a water quench (HEQ). Further ageing at 550°C produced a peak in hardness after two hours (HE2), with subsequent overaging after ten hours to produce sample HE10.

All samples were irradiated in the PLUTO MTR at Harwell as part of a major programme of irradiations. The 1154 instrumented rig employed gas mixture and gas gap techniques to maintain the temperature of the sample carrier. Temperatures were monitored throughout by thermocouples with an average recorded temperature of 290±5°C being attained by all the samples. Six sample carriers were loaded to attain a range of doses between 0.1 and 3×10^20 n/m^2 (E>1MeV). However, as a result of the rescheduling of reactor operations and the ultimate closure of the facility, only three out of the six capsules were irradiated. The irradiated samples examined in this study received a dose of 0.53×10^20 n/m^2 (E>1MeV) at a dose rate of 4.6×10^14 n/m^2/sec. The samples are prefixed with T to denote irradiated in future text.

3 Results

3.1 Hardness results

Hardness values were obtained by conventional Vickers hardness testing using 5kg loads, with three indents being performed on each sample. These tests were performed on samples used in the SANS investigations. Samples were 1mm thick and of diameter between 8 and 9 mm. The results of this testing are shown in Table 1 and graphically in figure 1. Examination of the figure, clearly shows the significant rise in hardness from both thermally aged material peaking at 2 hrs. The increase in hardness for all the irradiated material above the starting condition is clearly visible, the effect being strongest in the as quenched condition (HEQ).

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Hardness (1v)</th>
<th>Change due to thermal ageing</th>
<th>Change due to Irradiation</th>
</tr>
</thead>
<tbody>
<tr>
<td>HEQ</td>
<td>95 ± 2</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>IHEQ</td>
<td>238 ± 5</td>
<td>143 ± 5</td>
<td></td>
</tr>
<tr>
<td>HE2</td>
<td>189 ± 2</td>
<td>94 ± 3</td>
<td></td>
</tr>
<tr>
<td>IHE2</td>
<td>221 ± 7</td>
<td>32 ± 7</td>
<td></td>
</tr>
<tr>
<td>HE10</td>
<td>170 ± 3</td>
<td>75 ± 4</td>
<td></td>
</tr>
<tr>
<td>IHE10</td>
<td>204 ± 3</td>
<td>34 ± 4</td>
<td></td>
</tr>
</tbody>
</table>

Figure 1  Hardness measurements of the thermally aged and irradiated samples.

3.2 Microstructural examination

Three types of microstructural examination were performed. The first using TEM, was employed to examine for any large scale changes in microstructure and to determine size distribution data, in particular for the larger precipitates to act as an independent check on the SANS data, this comparison when complete will be presented elsewhere. FESTEM was used to determine the volume fraction of copper in the form of small precipitates. Again this data can be used to independently confirm the data produced by SANS.
3.2.1 STEM Technique and Results

Since our earlier work on these alloys [1,2], the first to use dedicated FEGSTEM examinations on this class of material, considerable advances have been made [3-5]. These concern both advances in equipment such as the use of Parallel Electron Energy Loss Spectroscopy, and in the methodology of the analysis, eg the use of spot and area scan modes. The use of PEELS in the study of RPV steel microstructures has two benefits, namely the ability to analyse for Mn in the precipitates [3-6] and using the annular detector to image small precipitates by changes in atomic weight (Z contrast). These samples have been examined throughout this technique evolution and so are useful indicators of the techniques capabilities.

By conducting analysis in the FEGSTEM, an instrument capable of producing high intensity electron beams some 2nm diameter, chemical analysis with nm spatial accuracy can be obtained by Energy Dispersive X-ray analysis (EDX). The technique employed for matrix copper determination was to position the beam in precipitate free regions of the foil, as assessed by imaging using Z contrast, and collecting the signal until a set number of counts or acquisition time had been obtained. These spectra were then analyzed using commercial 'Link' software to produce composition values. The process was repeated several times depending upon the accuracy required and the amount of spatial variation in the sample.

The sample is held in a specially modified holder which is of copper-free construction thus eliminating any copper signal arising from beam/holder interactions. Calibration checks with an Fe sample have shown that the background copper level after a 300 second acquisition time spectrum is unsolvable and is therefore considered to be <0.05%. The quoted values have therefore not be adjusted for any background copper effects.

The errors on the analysis can be split into two components, those from the analysis technique, considered for Ca to be ±0.02, and those arising from spatial inhomogeneity of the sample at the nm level due to ultrafine precipitates etc. The latter can produce differences in spectra to a maximum of ±0.2 wt%. It is these errors that can be minimised by conducting more analysis under well defined conditions, eg variation in foil thickness, knowledge of beam drift etc., and application of statistical methods to deconvolute the measured distribution of spectra to obtain appropriate matrix values.

As an indicative guide, for 5 matrix spot mode values we would not expect an accuracy of better than ±0.1, while 15 to 20 values now typically produces an error of ±0.03. However, this latter technique requires foil thickness information readily obtainable from PEELS, not fully developed at the time of making the STEM measurements reported here and is reflected in the accuracy of the data. Even so the data have improved over previous reported work [1] on this alloy and are comparable with accuracy inferred from other techniques such as SANS, making comparisons possible between the two techniques. Data from small area scans and large area scans provide valuable information on the distribution of copper in the form of small and large precipitates. The large area scan can be compared to bulk values directly, while the difference between large and small can be related to the volume fraction of large copper precipitates.

Differences between the spot and small area scan modes can therefore be used to assess the volume fraction of copper associated with small precipitates. The positioning of the spot and small area scan is shown by the crosses and boxes in figure 2. The large area scan would typically cover an area the size of the micrograph. This figure was obtained on the STEM from sample IHE10 using Z contrast, here the precipitates appear as white spots. Examination of the figure shows a bimodal distribution in precipitate size, both small ~2-5 nm diameter and larger ~10nm precipitates can be seen.

In developing the technique, one deficiency of the above is manifest by comparison of the data produced by large area scans compared with a bulk value of 1.3wt%. In all cases the large area scans under predict the bulk value, the effect being strongest in the thermally aged material. We have now attributed this to the preferential leaching out of the foil, in the electropolishing process, those large copper precipitates intersecting the surface, thereby reducing the copper level measured in the scan. This loss of large copper precipitates does not affect the small area scan and spot mode measurements as these are positioned to avoid any large precipitates.

Figure 2. Polaroid image obtained from the FEGSTEM on sample IHE10, the image was produced using Z contrast. The precipitates appear as white spots. The spot, small area and large area scans are represented by crosses, dotted box and the whole micrograph respectively.
The results of the STEM analysis are shown graphically in figure 3. The figure show the histograms for spot, small and large area scan. From these values average data were calculated and are shown in Table 2.

### Table 2

<table>
<thead>
<tr>
<th></th>
<th>Spot</th>
<th>Small Area</th>
<th>Large Area</th>
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<tbody>
<tr>
<td>HEQ</td>
<td>1.24±0.08</td>
<td>1.23±0.06</td>
<td>1.28±0.03</td>
</tr>
<tr>
<td>IHEQ</td>
<td>0.15±0.09</td>
<td>*</td>
<td>1.21±0.02</td>
</tr>
<tr>
<td>HE2</td>
<td>0.40±0.04</td>
<td>0.92±0.13</td>
<td>1.07±0.07</td>
</tr>
<tr>
<td>IHE2</td>
<td>0.19±0.05</td>
<td>0.86±0.03</td>
<td>1.20±0.04</td>
</tr>
<tr>
<td>HE10</td>
<td>0.32±0.04</td>
<td>0.49±0.06</td>
<td>1.05±0.09</td>
</tr>
<tr>
<td>IHE10</td>
<td>0.17±0.08</td>
<td>0.30±0.01</td>
<td>1.17±0.05</td>
</tr>
</tbody>
</table>

* Not measured - assume same as large area value

#### 3.2.2 SANS Results

SANS experiments were made with the SANS instrument in the neutron guide house of the DR3 reactor, Risø, Denmark. The measurements were made using a wavelength of 0.6nm and sample to detector distances of 1.25 and 2.5m to cover the required scattering vector (Q) range. In addition to the samples, a calibration sample of water (1mm thick) and backgrounds were measured at both detector positions. The PLUTO-SANS electromagnet was installed in the sample vacuum enclosure of the instrument to provide the required 1T field for magnetic measurements. These measurements are necessary to produce A ratio values and hence compositional information on the precipitates in addition to the size and number density values. Figure 4 shows intensity versus Q² plots in zero field. This data together with that obtained in magnetic field has been processed using the maximum entropy methods[6-7] employed by Buswell et al[11] to produce the precipitate size distribution plots presented in Figure 5. The figures, for clarity, show line diagrams taken at the mid point of the histogram bin. The line structure observed in the distribution is an artefact of the maximum entropy technique and the bin width chosen for display. The volume fraction information is derived independently and not subject to such choice of bin width.

The SANS data are summarized for all the conditions examined in Table 3. The table presents both diameter (d), volume fraction (Vf) and number density (Nd) values for the total population, for the small coherent bcc precipitates (d<6nm) and the large incoherent precipitates (d>6nm). The choice of 6nm is in keeping with previous work on this alloy and permits comparisons to be made. The precise transition from bcc to 'fcc' is not well

---

Figure 3 Copper determination from FEGSTEM analysis. The data show the values obtained in spot, small area and large area scan modes.
Figure 4  Differential scattering cross sections as a function of Q² for unirradiated and irradiated HEQ, HE2 and HE10.

Table 3  Summary of SANS data

<table>
<thead>
<tr>
<th></th>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IHEQ</td>
<td>11.1</td>
<td>100%Cu 90%Vac 10%Cu+voids</td>
<td>3.7</td>
<td>0.82</td>
<td>3.10</td>
<td>3.7</td>
<td>0.82</td>
<td>3.09</td>
<td>0.76</td>
<td>2.94</td>
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<td>0.62</td>
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<td>0.30</td>
</tr>
<tr>
<td>HE2</td>
<td>5.5</td>
<td>90%Cu 90%Vac 10%Cu+voids</td>
<td>2.2</td>
<td>0.93</td>
<td>8.99</td>
<td>2.25</td>
<td>0.93</td>
<td>8.90</td>
<td>0.75</td>
<td>5.90</td>
<td>0.57</td>
<td>3.22</td>
<td>0.57</td>
<td>0.32</td>
<td>0.30</td>
</tr>
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<td>IHE2</td>
<td>7.5</td>
<td>100%Cu 90%Vac 10%Cu+voids</td>
<td>1.1</td>
<td>0.93</td>
<td>8.99</td>
<td>2.25</td>
<td>0.93</td>
<td>8.90</td>
<td>0.75</td>
<td>5.90</td>
<td>0.57</td>
<td>3.22</td>
<td>0.57</td>
<td>0.32</td>
<td>0.30</td>
</tr>
<tr>
<td>HE10</td>
<td>12.3</td>
<td>100%Cu 90%Vac 10%Cu+voids</td>
<td>1.3</td>
<td>0.11</td>
<td>0.11</td>
<td>1.5</td>
<td>0.14</td>
<td>0.04</td>
<td>0.55</td>
<td>13.10</td>
<td>0.85</td>
<td>0.36</td>
<td>0.06</td>
<td>0.35</td>
<td>0.30</td>
</tr>
<tr>
<td>IHE10</td>
<td>8.7</td>
<td>100%Cu 90%Vac 10%Cu+voids</td>
<td>1.0</td>
<td>0.20</td>
<td>0.20</td>
<td>1.5</td>
<td>0.14</td>
<td>0.04</td>
<td>0.55</td>
<td>13.10</td>
<td>0.85</td>
<td>0.36</td>
<td>0.06</td>
<td>0.35</td>
<td>0.30</td>
</tr>
</tbody>
</table>
defined and is the subject of ongoing research activities\[8-10\]. The absolute volume fraction is dependant upon the assumed precipitate composition, the table gives some alternatives, e.g. 100%Cu, a copper 4% vacancy alloy and a mixture of pure copper precipitates and microvoids having identical size distributions. In addition, from a knowledge of the volume fraction of precipitate and starting copper available for precipitation, it is possible to calculate the copper remaining in the matrix, these values are shown together with those obtained by direct measurement with FEGSTEM.

4 Discussion

4.1 Copper precipitation

The data obtained in this study are in excellent agreement with earlier studies on thermal ageing (550°C) experiments using this alloy. Both studies confirm a rise in hardness to a peak at approximately 2hrs, with over ageing and associated reduction in hardness being observed in the 10hr condition. Microstructural analysis confirms that for conditions of peak hardness, not all the copper has been removed from solution. Analysis by SANS and by the differencing techniques adopted in the FEGSTEM, confirm a large volume fraction of small precipitates are present. Further analysis also reveals a population of larger d>6nm are also present, but with a considerably lower number density.

Under irradiation, the population of small precipitates is enhanced, the effect being the most dramatic in the HEQ sample where all the copper was available for precipitation at the start of the irradiation. In other samples where copper matrix values were lower at the start of the irradiation, the incremental hardness associated with irradiation is smaller. In all the irradiated alloys, the final copper matrix value measured by FEGSTEM is similar, with an average of 0.17 wt%, indicating that full precipitation has not yet been achieved under these irradiation conditions, i.e. not yet achieved full depletion of the matrix copper. This is in keeping with our earlier work and will be discussed in detail elsewhere.

Comparison of the two techniques employed to determine volume fraction of copper precipitate is shown in Table 4. In general the agreement is very good, given the uncertainties of the measuring methods and the assumptions employed, e.g. that the precipitate composition is 100% copper. Further work is required on sample HE2 to determine the source of the difference between the two techniques, but the initial assessment points to the arbitrary split of small and large precipitates at 6nm in the SANS data. A repeat STEM examination is planned, using a sample cut from the SANS specimen examined as part of this study.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Volume Fraction (at%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>FEGSTEM</td>
</tr>
<tr>
<td>S/A - Spot</td>
<td>L/A - S/A (Bulk S/A)</td>
</tr>
<tr>
<td>HE2</td>
<td>0.46</td>
</tr>
<tr>
<td>HE10</td>
<td>0.15</td>
</tr>
<tr>
<td>IHEQ</td>
<td>0.93</td>
</tr>
<tr>
<td>IHE2</td>
<td>0.59</td>
</tr>
<tr>
<td>IHE10</td>
<td>0.11</td>
</tr>
</tbody>
</table>

4.2 Effect of irradiation on precipitates

The size of the thermally aged and irradiation induced precipitates is in agreement with previous studies on these alloys\[1\]. However, the effect of irradiation on existing precipitates has not been previously examined, although the good agreement obtained does give confidence to the accuracy of this new data. Under irradiation the SANS data suggests that the size of the small precipitates induced at peak hardness remain relatively unchanged, but increase in number. The larger precipitates in the averaged sample HE10 apparently undergo a reduction in mean size from 14 to 12 nm, however, the volume fraction of precipitate (d>6nm) remains constant as a result of increase in number density of the smaller precipitates in this size distribution.

In welds, microstructural examination shows that the precipitates produced in PWHT are typically 10 to 20nm diameter, and under the irradiation conditions experienced by the vessel are essentially unchanged, as measured qualitatively by TEM in unirradiated and irradiated material. Quantitative information on the large precipitates in commercial material is not available. SANS studies generally are concerned with the small precipitates, and in typical commercial material with copper values above ~0.3, little information exists in the open literature. As the bulk copper levels are not as high as the model alloys employed here, even if SANS data were available, the volume fraction of large precipitates is relatively small, making any comments on the change in this part of the SANS signal statistically invalid.

A better opportunity for comparison comes from work on irradiated, annealed and reirradiated material, where a high percentage of the copper during the annealing stage is in the form of large precipitates. Again the only SANS analysis of such material in the open literature is the data of Kampsman et al\[11\] that unfortunately does not permit comparisons of the large precipitate distribution to be made due to the use of total volume fraction and size information, i.e. the presence of a bimodal distribution is acknowledged but not analysed in terms of the two components that make up the overall distribution.
It is therefore extremely difficult to rationalise these observations of precipitate refinement with observed effects in commercial-type material. The applicability of the model alloy with extremely high, by RPV standards, copper content and the extremely high dose rates of the irradiation need to be considered before over interpretation of the data to suggest that large copper precipitates in RPV welds are unstable under irradiation. It is thought that the data are more relevant to plant life extension studies employing annealing, as not only will the time and temperature of the anneal affect precipitate size and hence stability under subsequent irradiation, but the surrounding microstructure will be considerably cleaner than in the as fabricated condition. This 'clean' microstructure local to the precipitate will therefore be more like that observed in the model alloys examined in this study. It may therefore explain the rapid re-embrittlement observed in high copper material[12] after annealing. Here the rate of embrittlement is close to that observed at start of life condition, even though matrix copper values should be considerably lower in the annealed material. Microstructural examination of this material to provide matrix copper values would be required to understand further the role of annealing and the subsequent response of the material to irradiation.

4.3 Mechanical properties from microstructural data

The volume fraction and size information can be used in combination with the Russell-Brown[13] modulus hardening theory to predict yield strength and subsequent hardness change. The data have been calculated in accordance with Physihan et al[2] using the modulus values for bcc copper. This has been done for all the conditions examined using the size information from SANS, and the volume fraction information that generated for the small precipitates <56nm by SANS and the small area - spot mode differenting technique developed for FEGSTEM. The information on the large precipitates has not been used as these incoherent precipitates harden by a different mechanism where use of the modulus hardening theory is not appropriate. The hardness data and volume fraction information used in the analysis are presented in Table 5.

The data predict hardness changes associated with bcc copper precipitates making a direct comparison with experimental data difficult especially so in the irradiated material where in addition to copper precipitation, matrix damage (loops and/or microvoids) are formed. The copper contribution is in keeping with the values predicted for this irradiation condition by models of embrittlement.[14-16]. To determine the partitioning of the observed hardness into these two components, a series of post irradiation annealing experiments are now underway and will be reported elsewhere. When available these should act as further independent checks on the validity of the Russell-Brown model for this type of work. For the unirradiated samples, the only appropriate comparison with experiment is the change in hardness between HEQ and HE2, here the experimental value of 94 is in good agreement with that predicted from microstructural data. Values of 87 and 65 are predicted when using the FEGSTEM and SANS volume fractions respectively, giving further support to the use of the differencing technique employed with the FEGSTEM data.

The observed increase in hardness between the unirradiated and irradiated HE10 can be explained by the increase in both the number density and mean size of the small coherent precipitates observed in this alloy. Application of the R-B model to this data predicts an increase in hardness between 22 to 34Hv depending upon the size of precipitate used (1.5 or 2.2nm), this compares well with the measured increase of 34Hv that also includes a contribution from matrix defects. The change in the size distribution for the larger precipitates is of importance to understanding the effects of PWHT and re-embrittlement after annealing. The data for HE10 suggest that under irradiation a new equilibrium position is set up where the mean precipitate size is smaller than that observed under thermal conditions alone.

Table 5
Comparison of experimental hardness data with predicted values using microstructural volume fraction data and modulus hardening theory.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Change in Hardness</th>
<th>Volume fraction of precipitate (at%)</th>
<th>Mean precipitate Diameter (nm)</th>
<th>Calculated hardness from precipitate Russell-Brown</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>STEM</td>
<td>SANS</td>
<td>SANS</td>
<td>STEM</td>
</tr>
<tr>
<td>HEQ</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>HE2</td>
<td>94</td>
<td>0.46</td>
<td>0.26</td>
<td>2</td>
</tr>
<tr>
<td>HE10</td>
<td>75</td>
<td>0.15</td>
<td>0.14</td>
<td>1.5</td>
</tr>
<tr>
<td>IHEQ</td>
<td>143</td>
<td>0.93</td>
<td>0.82</td>
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<td>IHE2</td>
<td>126</td>
<td>0.59</td>
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<tr>
<td>IHE10</td>
<td>109</td>
<td>0.11</td>
<td>0.14</td>
<td>2.2</td>
</tr>
</tbody>
</table>
Conclusions

Techniques have been developed to assess directly and indirectly the copper remaining in the matrix as a result of thermal or irradiation induced precipitation.

In general good agreement is achieved between the two techniques for precipitate volume fraction assessment.

Modelling the hardness change can be successfully accomplished using the modulus hardening theory and appropriate bcc modulus data.

Thermal ageing produces a range of precipitate sizes, the peak occurring in this model alloy after 2hrs at 550°C. In this condition, the matrix is not depleted of copper, peak hardness being achieved by the optimum number and size of bcc precipitates. Overaging results in an increase in precipitate size and relatively modest increase in precipitate volume fraction.

Irradiation of the material independent of the starting copper matrix results in a similar matrix depletion of copper under irradiation. The irradiation condition examined here does not appear to have reached peak hardness, with ~0.17wt% remaining in solution.

The size and number density of larger (d=6nm) is refined by irradiation, bringing average diameters down from 14 to 12nm, with associated increase in the number density of precipitates at the smaller end of this distribution. The net result is a relatively constant volume fraction of precipitate.

Acknowledgements

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REFERENCES

SESSION E

MITIGATION OF

IRRADIATION EFFECTS
IRRADIATION EMBRITTLEMENT MITIGATION

by
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ABSTRACT. Neutron irradiation affects the material properties and hence the structural integrity of reactor pressure vessels in nuclear power plants. Mitigation of irradiation damage is one of the major issues within the nuclear plant life management. An overview is given on the proposed and utilized mitigation methods.

Low leakage loading schemes are commonly used in PWRs to mitigate reactor pressure vessel embrittlement. Dummy assemblies have been applied in VVER 440-type and in some old western power plants, when exceptional fast embrittlement has been encountered. Shielding of the pressure vessel has been developed, but the method is not in common use. Prestressing of the pressure vessel has been proposed to be a potential method for preventing PTS failures, but the applicability of the method for nuclear pressure vessels has not yet been demonstrated. The large number of successful annealing treatments performed in VVER 440 type reactors as well as the intensive research and development work done in the methods and effect of annealing treatments suggest that more applications will be seen in the near future also in western PWRs. The emergency core cooling systems have been modified in VVER 440-type reactors in connection with other mitigation measures. Efforts to extend the service life of reactor pressure vessels further increase the weight of plant specific surveillance programs.

Keywords: reactor pressure vessel (RPV), light-water reactors, pressurized-water reactors, neutron irradiation, irradiation embrittlement, neutron fluence reduction, low leakage fuel management, shielding, emergency core cooling, prestressing, annealing, recovery, pressurized thermal shock, surveillance programs
Introduction

Reactor pressure vessel (RPV) integrity is assured by several arrangements including the setting of operational limits for normal and different emergency states, surveillance of irradiation embrittlement of beltoine materials as well as in-service inspections and other maintenance procedures.

The pressurized thermal shock (PTS) is generally regarded as the most severe, although very improbable loading situation for the RPVs. During a postulated PTS the pressure vessel is subjected both to thermal stresses and those caused by the repressurization.

The major reason for RPV embrittlement is the fast neutron flux from the core. Besides neutron fluence embrittlement is a function of the impurity contents of the RPV material. Different guides have been developed to evaluate the fluence dependence of materials with certain impurity contents. The USNRC Regulatory Guide 1.99 provides correlations for evaluating the nil-ductility transition temperature \( RT_{\text{NDT}} \) shift and the upper shelf toughness change for different steels and fluences [1]. However, most frequently RPV embrittlement is followed mainly by the plant specific surveillance programs.

Some old PWR vessels (built in 1960-70) are particularly susceptible to embrittlement [2]. Typical reasons are as follows:

- The design end-of-life (EoL) fluence of the RPV is high.
- The steel composition, especially the contents of Cu, Ni and P, is unfavourable.
- There are welds (circumferential and/or longitudinal) in high flux areas, and welds are usually most susceptible to embrittlement.

The effect of some of these factors on the 41 J transition temperature shift is demonstrated in Fig. 1.

Measures to extend the RPV service life include those reducing the fast neutron flux to the RPV, recovery annealing of the material, modification of the emergency core cooling system (ECC) and some other predictive procedures. A literature review on the use and applicability of these measures is presented in this paper.

Flux reduction techniques

There are two principles available to reduce the fast neutron flux to the RPV. The core, i.e. the irradiation source, can be modified or reduced to give lower flux. The second way is to place irradiation shields between the core and the RPV.
Fig. 1. Effect of fluence reduction and material improvement on the transition temperature shift [2].

**Core modification and/or reduction**

Neutron flux to the RPV can be reduced most efficiently by reducing power in the critical peripheral fuel assemblies, i.e. in those which lie nearest the RPV wall. About 85% of the fluence to the RPV is estimated to come from the core peripheral assemblies [3]. Following procedures are applicable in reducing the flux [2,4-6]:

1. Low leakage fuel management. Some or all of the peripheral fresh fuel assemblies are replaced by low reactivity fuel assemblies, i.e. those spent one to three cycles in the reactor.

2. Some of the peripheral fuel assemblies are replaced by dummy assemblies, which contain stainless steel or zirconium rods instead of UO₂ pellets. Either partially or fully replaced assemblies can be used. Typically 5-10% of the fuel assemblies need to be replaced to maintain circumferential symmetry.

3. Installation of neutron absorbing materials on the core periphery. For example peripheral control rods or burnable absorber placed at critical locations can be used to reduce the flux.

When power is reduced at the core periphery, power derating can be avoided only, if power of the remaining assemblies is increased. Generally this means an
increase in power peaking and, if power is not reduced, a decrease in thermal margin.

If there is no concern about RPV embrittlement, the normal loading scheme is the out-in-pattern, where the fresh (most highly enriched) fuel is placed for its first cycle on the core periphery and the exposed fuel in the interior. This was previously the standard loading scheme also for PWRs. The disadvantage of this scheme is a high neutron leakage from the core and a high neutron flux to the RPV and hence, a loss of reactivity and fast embrittlement of the RPV.

However, the standard loading scheme leads to an even core power distribution, i.e., minimum power peaking, and hence to maximum core power. Usually there is also no must to use burnable absorber fuel to make the power distribution more even in the beginning of the cycle [4,5].

The size and configuration of the core, as well as the location of the welds in the RPV affect the applicability and benefits possible to be achieved by different low leakage schemes. The increase in power peaking due to a certain low leakage scheme is larger for small cores. In a large core there are also more low leakage loading schemes available than in a small one. The critical location of the RPV is often on the inside surface of a longitudinal weld, if existing. Flux reduction at a fixed circumferential location (e.g., longitudinal weld) is generally more difficult to be achieved than the same reduction in a circumferential flux peak [7].

The simplest way to reduce the flux locally would be to replace only the adjacent fuel assemblies by assemblies with high burn-up (e.g., two cycles exposed). This, so-called low fluence scheme, can be performed even without marked increase in power peaking, if the number of replaced assemblies is small [5]. However, the reduction in the overall neutron leakage remains small. The effect in RPV lifetime may also remain smaller than expected, when another location becomes critical.

A flux reduction factor (FRF) of up to 2 with little or no increase in power peaking (and without reducing power) seems to be achievable locally for most PWRs, when fresh fuel is replaced by two cycles exposed fuel adjacent to the critical locations. The overall neutron leakage reduction is slight [5].

Previously the main objective of the low leakage schemes was typically to minimize the overall neutron leakage from the core, i.e., the motivation was to improve fuel economy. Present low leakage loading schemes are often modified to minimize the flux particularly at the critical location(s) of the RPV for mitigating irradiation embrittlement, although this may increase somewhat the overall neutron leakage from the core and thus increase fuel costs. About 30-40% local reduction in the flux to the RPV and 1.2% reduction in the overall neutron leakage (compared to the out-in-pattern) can be achieved with only slight increase in power peaking (less than 3%) by following a modified low leakage fuel management scheme, where one and two cycles exposed assemblies are loaded in selected peripheral locations, while power of certain other assemblies is increased to avoid reduction in the core power (CE 217 assembly core) [5]. As a consequence changes in assembly enrichments and the use of burnable absorber
fuel are required. Examples of different low leakage loading schemes (and the use of them) are given in Table 1.

Table 1. Fuel vendor low-leakage management schemes [4].

<table>
<thead>
<tr>
<th>VENDOR</th>
<th>NAME</th>
<th>PATTERN TYPE</th>
<th>TYPICAL FLUX REDUCTIONS(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Babcock &amp; Wilcox</td>
<td>LBP(1)</td>
<td>IN-OUT-IN</td>
<td>30-40%</td>
</tr>
<tr>
<td>Combustion</td>
<td>SAV-FUEL</td>
<td>IN-OUT-IN(4)</td>
<td>20%</td>
</tr>
<tr>
<td>Engineering</td>
<td>IN-IN-OUT</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Exxon</td>
<td>LRL(2)</td>
<td>Mixed</td>
<td>50% Locally</td>
</tr>
<tr>
<td>Westinghouse</td>
<td>L²P or LLLP(3)</td>
<td>IN-OUT-IN</td>
<td>10% to 50%</td>
</tr>
</tbody>
</table>

(1) LBP: Lumped Burnable Poison
(2) LRL: Low Radial Leakage
(3) LLLP: Low-Leakage Loading Pattern
(4) SAV-FUEL was initially IN-OUT-IN but as IN-IN-OUT has become attractive, it has been used as a general name for CE low-leakage plans.
(5) These schemes are intended to improve fuel cycle economics. CE estimates that a scheme designed to improve economics and reduce flux at vessel welds would reduce neutron flux at the vessel by 20 - 50%

A flux reduction factor up to ca. 3-5 can generally be achieved (without need to reduce power) by applying low leakage fuel management, if also part of the remaining peripheral assemblies at selected locations are replaced with dummy stainless steel assemblies (with stainless steel rods instead of UO₂), which not only reduce the neutron production, but also to a some extent reflect neutrons back to the core interior [5]. Need to use burnable absorber fuel and an increase in power peaking is obvious, when dummy assemblies are used [5,8].

The calculated effect of flux reduction in a case where the implementation occurs after 7 full power years (FPPY) is shown in Fig. 2. After that operation time most of the expected transition temperature shift has already occurred. The BoL RTNDR shift for the 10:1 flux reduction scheme is about 45 °C. The horizontal lines show the USNRC screening criteria for longitudinal welds (132 °C) and circumferential welds (149 °C).

In some plants it has been possible to reduce the flux to the RPV even by a factor of 10, when both low leakage fuel management and dummy assemblies have been applied [2,4]. Generally flux reduction exceeding a factor of 5 is not possible without power reducing [5]. In general, the maximum achievable and realistic flux reduction depends on thermal margins.
Fig. 2. The RT<sub>NDT</sub> shift over time for a range of flux reduction schemes [4].

The selection of fuel designs for a low leakage scheme is based on the evaluation of various parameters, which include cost, safety and impact on plant operations [9]. The number of flux reduction assemblies depend on the number and location of critical welds. The number of these assemblies will be larger, if core symmetry has to be maintained. Assembly types used in fluence reduction programs supported or analyzed by Advanced Nuclear Fuel Co. are listed below [9]:

1. Assemblies with high burn-up (usually three cycles exposed).
2. Low enriched assemblies in which the bottom third of all the fuel rods contain stainless steel.
3. Reconstituted assemblies with high burn-up and with multiple rows of stainless steel rods.
4. Low enriched assemblies with multiple rows of stainless steel rods.
5. Assemblies with high burn-up and in which Hf inserts are placed in the guide tubes.
6. Low enriched assemblies in which Hf inserts are placed in the guide tubes.

Shielding of the pressure vessel

Neutron flux to the RPV can be reduced by fitting new materials between the outer fuel elements and the RPV, because neutron diffusion depends on the
detailed neutron absorption and scattering cross-sections of the materials. In fact, the use of dummy assemblies can also be regarded as shielding. One possibility is to modify the core support barrel or the core shroud so that for instance stainless steel shields (patches) can be attached to selected locations [5]. Also the use of materials like tungsten or some metal hydrides have been considered [10]. For example, a 50 mm thick stainless steel patch is estimated to reduce the flux to the RPV by a factor of ca. 1.5.

The modification costs of the core (including the loss in power production) may be significant. A comprehensive coolant flow analysis is also necessary. Technical solutions to make shielding possible have been developed [2,11]. There are expected to be no applications in commercial PWRs.

Factors affecting flux reduction

In general, the aim of flux reducing procedures is to reduce the flux at critical locations of the RPV without limiting too much the operational flexibility of the reactor and, if possible, without reducing power. The minimum flux reduction factor required for a certain design service life depends on

- the design EoL fluence of the RPV,
- the circumferential flux distribution and the initial value of flux,
- composition of the RPV base metal and welds, i.e. the irradiation embrittlement sensitivity,
- location and number of welds in the RPV,
- years of operation before the intended flux reduction measures.

The restricting boundary conditions in applying different flux reduction schemes are for example

- the availability of thermal margins and the resulting possibility to increase power peaking without operational restrictions and without reducing power,
- the operational margins (pressure-temperature windows),
- reactivity margin, for example long cycle length (18 moths),
- core configuration and size.

It is evident that the applicability of different flux reduction methods is very plant specific.

Modification of the emergency core cooling and other systems

The primary measures to mitigate RPV embrittlement in operating power plants are those reducing the neutron flux to the RPV. As the most severe expectable loading situation for RPVs is considered to be a PTS, extra safety margin can be achieved also by changing the emergency cooling system so that the maximum loading in the RPV during such events is reduced.
RPV failure risk can be reduced

- by minimizing the probability of abnormal events such as PTS,
- by minimizing the maximum expectable stress concentration in the RPV during possible abnormal events.

Emergency core cooling systems can be modified in order to reduce stresses during PTS

- by increasing coolant temperature and/or mixing (for example the location of coolant inlet(s) can be changed) in order to reduce thermal stresses,
- by limiting the maximum pressure increase.

Pressure-temperature limiters together with programmable controllers are used to protect RPVs against overpressure transients during heat-up and cooldown stages [12].

Some applications of modified emergency cooling systems are presented afterwards.

Other mitigation methods

**Prestressing**

Prestressing of the RPV has been suggested to be a potential method for preventing PTS failures [13]. The stresses during postulated PTS events are lowered in this method by prestressing the RPV with a memory alloy band wound on the outside surface.

The predictions made for postulated (and somewhat simplified) PTS events and RPVs have shown that the increase in the RPV temperature margin due to prestressing could allow a significant service life extension without exceeding the embrittlement criterion [13]. One prediction has been made for a RPV with following dimensions:

<table>
<thead>
<tr>
<th>Dimension</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>inside radius</td>
<td>218 cm</td>
</tr>
<tr>
<td>wall thickness</td>
<td>216 mm</td>
</tr>
<tr>
<td>cladding</td>
<td>6.4 mm</td>
</tr>
<tr>
<td>active core height</td>
<td>483 cm</td>
</tr>
<tr>
<td>total RPV height</td>
<td>1245 cm</td>
</tr>
</tbody>
</table>

The RPV material was SA533 B Class 1 plate with RT_{PTS} = 143 °C (exceeding thus the USNRC screening criterion 132 °C for axial welds). Only an axial weld (and crack) was considered. Prestressing was assumed to affect the stresses only in the hoop direction. The prestress was induced by 100 mm thick rings put on the belttine region of the RPV so that they covered most of the core region. The heat transfer and elastic properties of the band were taken to be identical with those of the RPV. The yield strength of the band was 483 MPa at 20 °C and 420 MPa at 316 °C. The resulting hoop prestress of 221 MPa was computed to induce the prestress of 104 MPa in the RPV. The pressure in the postulated PTS was
assumed to drop quickly from a normal operation level to 6.9 MPa and remain constant.

The stress intensity factor ($K_I$) for a 25 mm crack (in base material) with and without the prestress and the $K_e$-curve for the RPV material are shown in Fig. 3. The results demonstrate that at least 17 °C (30 °F) increase in the temperature margin can be achieved by using prestressing. For a 50 mm crack this margin was 28 °C, which suggest that larger margins could be obtained, if the analyses were based on crack arrest.

![Graph showing $K_I$ vs. temperature for a PTS transient (25 mm crack) with and without the prestress band [13].](image)

It is evident that the proposed prestressing alters significantly the stress and temperature distributions in the RPV not only during PTS events but also under cooldown and heat-up stages. These give subject for a comprehensive plant specific evaluation, including at least analyses of different transients for each RPV type, tensile stresses on the inside surface of the RPV during the cooldown stage and the tensile stress peak in the RPV near the border of the band and RPV contact region.

Other things to be examined are the long term thermal properties of the band material and possible changes in properties (especially the memory effect) due to irradiation, as well as the behaviour of the bound during different transients. Also the problem how to determine the prestress should be solved.

**Applications at some nuclear power plants**

At the Loviisa NPP in Finland (VVER 440, 349 assembly positions) the first surveillance test results revealed unexpected fast embrittlement on the circumferential weld. Neutron flux was retarded by replacing 36 peripheral fuel assemblies (ca. 10% of the core) by dummy stainless steel assemblies in 1980 after ca. three
years (Lo1) and one year (Lo2) of operation. Due to exceptional large margins this could be performed without reducing power, even if the number of replaced assemblies was large. In addition, the emergency injection water temperature was raised. These measures were assumed to be sufficient for achieving the design service life of the RPV [4,14].

Both low leakage fuel management schemes and dummy assemblies have been applied to reduce flux in the old KWU plants [2]. At a 360 MWe power plant (probably the Obregenheim plant) with the 121 fuel assembly core the water gap between the core and the RPV was originally small and hence the neutron flux to the RPV high (EoL fluence 7x10^19 cm^-2) [2,4]. A local flux reduction factor of 10 was achieved (compared to the original out-in scheme), when 12 fuel assemblies were replaced after the ninth operating cycle by stainless steel dummy assemblies (in addition, a change from out-in to in-out scheme had been performed after five operating cycles). The EoL fluence after these operations was reduced to 3x10^19 cm^-2. Flux reduction was possible without power reduction.

A KWU 670 MWe power plant (probably the Stade plant) with the 157 fuel assembly core also suffered from the high local neutron flux to the RPV [2,4]. The original EoL fluence of 4x10^19 cm^-2 could be reduced to 2x10^19 cm^-2 by replacing 12 peripheral fuel assemblies with partially burnt-up fuel assemblies after five operating cycles and later (after the seventh operating cycle) by inserting absorber rods into the replaced assembly positions. The total flux reduction factor after these measures was 4. Also in this case power reduction was avoided.

At the Public Service Electric & Gas 1100 MWe power plant (RPV manufactured by CE) in USA, 18 years extension in the service life of the RPV could be achieved (corresponding EoL fluence 2x10^19 cm^-2) by replacing eight corner fuel assemblies with exposed ones [15]. The local flux reduction factor of 2 (50%) was achieved by using this low leakage scheme.

At the Maine Yankee 855 MWe power plant (217 fuel assembly core, RPV manufactured by CE) in USA, the out-in scheme was replaced by a low leakage scheme, where one and two cycles exposed fuel assemblies were loaded at the core periphery after six operating cycles in 1980 [16]. As a result the fluence at expiration-of-license was reduced by 33%.

In VVER 440/230 type reactors the excessive embrittlement is mainly due to the small water gap between the fuel and the RPV, as well as the impurity contents of the weld material. Several remedial measures were proposed, when the first surveillance tests revealed the higher than expected transition temperatures [17], i.e.

- modification of operation pressure-temperature limits,
- low leakage fuel management and core reduction using dummy assemblies,
- temperature increase in the emergency cooling tanks,
- replacement of the injection pipes from the cold to the hot leg of main circulation loop and installation of fast-closing valves in the main steam system,
- recovery annealing.
At the Jaslovske Bohunice power plants in Czech Republic (four VVER 440/230 reactors in two units) the measures which have been or are to be implemented consist of [18]

- emergency coolant temperature increase from 20 °C to ca. 55 °C,
- placement of 36 dummy assemblies on the core periphery,
- installation of quick operating valves and a pressure-temperature limiter,
- recovery annealing of the RPV.

The effect of the flux reduction measures and annealing treatments on the calculated critical RPV brittleness temperature ($T_b$) for the base material (BM) and weld (WM) of two RPVs (V1/1 and V1/2) are shown in Fig. 4. The value of $T_b$ is determined by the impurity contents (only weld material) and neutron fluence for both weld and base materials. Due to the core reduction performed in 1985 and the planned annealing treatment in 1992 the allowable critical temperature of brittleness ($T_{b,a}$) for the defect size 16 mm (181 °C) is not expected to be exceeded in the RPV of V1/2 until the year 2002. In V1/1 dummy elements are to be inserted in 1992.

![Graph showing critical temperature changes](image)

**Fig. 4.** Flux reduction and recovery annealing scheme for two RPVs (V1/1 and V1/2) of the Jaslovske Bohunice power plant. Curves "WM" are for welds and curves "BM" for base materials at two circumferential locations [18].

**Methods**

The first RPV annealings were realized using primary coolant and nuclear heat (US Army SM-1A) or pump heat (Belgian BR-3). The annealing temperatures were about 80°C above the service temperature. The degree of recovery in these cases was about 60%. The planned annealing of the Yankee Rowe vessel was estimated to give 45-55% recovery when using the temperature of 343°C, which is 83°C higher than the service temperature [19].
In the "wet"-annealing method the maximum temperature will be limited to about 350°C. Hence it can be used only in reactors with a low service temperature. Due to a rather limited recovery and a high re-embrittlement rate the wet annealing method cannot be a solution for power reactors.

In dry annealing the RPVs have been heated by electric resistance heaters. Proposals for using e.g. induction heating and superheated steam have also been suggested.

**Accomplished annealings**

Up to autumn -93 some 14 thermal annealings in VVER 440 reactors (plus a prototype annealing for decommissioned Novovoronezh 1- RPV) have been carried out.

**Table 2. Annealings of VVER 440-type RPVs.**

<table>
<thead>
<tr>
<th>Reactor</th>
<th>Year</th>
<th>Temperature/time (°C/h)</th>
<th>SS clad</th>
</tr>
</thead>
<tbody>
<tr>
<td>Novovoronezh 3</td>
<td>1987</td>
<td>430 ±20°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Armenia 1</td>
<td>1988</td>
<td>450 +50°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Greifswald 1</td>
<td>1988</td>
<td>475 -10°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Kola 1</td>
<td>1989</td>
<td>420-60°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Kola 2</td>
<td>1989</td>
<td>420-60°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Kozloduy 1</td>
<td>1989</td>
<td></td>
<td>yes</td>
</tr>
<tr>
<td>Kozloduy 3</td>
<td>1989</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Greifswald 2</td>
<td>1990</td>
<td>475 -10°C / 150h</td>
<td>no</td>
</tr>
<tr>
<td>Greifswald 3</td>
<td>1990</td>
<td></td>
<td>yes</td>
</tr>
<tr>
<td>Novovoronezh 3 (re-ann.)</td>
<td>1991</td>
<td>475 ±15°C / 100h</td>
<td>no</td>
</tr>
<tr>
<td>Novovoronezh 4</td>
<td>1992</td>
<td></td>
<td>no</td>
</tr>
<tr>
<td>Kozloduy 2</td>
<td>1992</td>
<td></td>
<td>no</td>
</tr>
<tr>
<td>J. Bohunice 2</td>
<td>1993</td>
<td>472-503°C / 160h</td>
<td>yes</td>
</tr>
<tr>
<td>J. Bohunice 1</td>
<td>1993</td>
<td></td>
<td>yes</td>
</tr>
</tbody>
</table>

The heat treatment in all the RPVs above had been focused for only one circumferential seam weld in the core zone as seen in Fig 5. This makes it possible to use a rather narrow heating zone. The width of the temperature zone has been approximately 1.5 m or less. One of the most critical points in annealing is to keep thermal stresses within acceptable values. In the realized dry annealings this has been calculated to be possible when the heating-up and cooling-down rates do not exceed 20°C/h and 30°C/h, respectively. The limits for temperature in surrounding structures seemed not to be a critical issue.
Fig 5. The annealing arrangements in Novovoronezh 3 [20].

In many reactors there have not been available actual archive material from the RPV or surveillance test programs. Then the exact degree of embrittlement has been unclear. For this reason it has been very important to make material testing, e.g. chemical analysis and toughness tests with samples cut out from the pressure vessel itself. For old VVER 440 reactors this has been possible also from inner surface because they have not a stainless steel cladding. The boat samples have been typically about 5 mm deep and the other dimensions have been a few centimetres. The samples have been used for subsize impact specimens for evaluating the toughness before and after the anneal. Additionally milling chips for chemical analysis has been removed. In cladded RPVs a material removing has also been carried out. The gap between the outer surface of the vessel and the biological shield tank in the reactor cavity is sometimes only a few centimetres which makes the removing of boat samples or chips as well as the hardness measurement rather troublesome. In some reactors the surface beads have been welded with a lower alloyed filler material which prevents the sampling of the actual weld metal.
In many old western RPVs the situation is much more complicated. They usually have also axial weld seams which means that the entire core zone requires a thermal treatment. This brings difficulties in avoiding unacceptable high stresses and residual strains [21,22]. The axial temperature gradient in the vessel may produce a "cork bottle" shape and bends the primary piping at the vessel nozzles. These problems seem not yet to be resolved for all types of vessel constructions.

In addition to present guides and rules concerning thermal annealings (e.g. ASTM E 509, 10 CFR Part 50 Appendix G & H) new regulatory documents are being developed. A Regulatory Guide on "Form and content for thermal annealing RPVs" (Draft Guide 1-027) and a rule on thermal annealing (anticipated to be 10 CFR 50.66) are now under preparation and are planned to be completed in 1994.

**Recovery and re-embrittlement of VVER 440 RPVs**

High phosphorous and fairly high copper contents in weldments have caused a serious radiation embrittlement problem in many VVER 440-type RPVs resulting in 14 annealings until now. Problems with the base material have not been reported. The first in-service annealing was made at 430°C (about 165°C above irradiation temperature), which was later found to be too low for an adequate recovery and nowadays 475°C has been typically applied. Results from several investigations done with base and weld materials; irradiated in research or commercial power reactors are seen in Figs 6 and 7.

![Graph showing recovery (%)](image)

**Symbols:**
- △ ● base metal
- □ ○ weld metal
- + Novovoronezh 1 weld metal
- △ □ irradiated in research reactor
- ○● irradiated in VVER 440

**Fig 6.** The effect of the difference between annealing and irradiation temperatures on the recovery of transition temperature in VVER 440 RPV steels [23].
Fig 7. The effect of the rate of fluence and the difference between annealing and irradiation temperatures on the recovery of ductile-brittle transition temperature in VVER 440 RPV steels [24].

a) irradiated in research reactor VVR-M
b) irradiated in VVER 440
φ: Θ, ◊, +, x =3x10¹⁶ n/m²s
O, ◊ =3x10¹⁵ n/m²s
■ =5x10¹⁴ n/m²s

The results reveal that the lower bound of the recovery percentage after annealing at 475°C is about 80%. It can also be found that after a slow irradiation rate (e.g. RPV wall) the recovery is retarded. The content of phosphorus has a great effect on the residual embrittlement. A high annealing temperature is needed especially with high P contents (Fig 8). A clear evidence of thermal ageing (Tₐₐₙ ≤ 475°C) during recovery annealings cannot be found in the literature but investigations of the coarse grained HAZ have not been seen, too.
Fig 8. The effect of phosphorus content and the annealing temperature on the residual embrittlement of VVER 440 steels [25].

The investigations show that the re-embrittlement rate usually does not exceed that in the first irradiation. In Figs. 9-12 results of VVER 440 RPV steels after various irradiations and heat treatments are shown.

Fig 9. The shifts in transition temperatures of base metal (P=0.020%; Cu=0.11%) of VVER 440 RPV after irradiations in VVER 440 reactor and thermal annealings [25].
Fig. 10. The shifts in transition temperatures of base metal of VVER 440 RPV after irradiations in VVER 440 reactor and thermal annealings [17].

Fig. 11. The shifts in transition temperatures of weld metal (P=0.028%; Cu=0.18%) of VVER 440 RPV after irradiations in VVER 440 reactor and thermal annealings [25].
Fig. 12. The shifts in transition temperatures of weld metal (P=0.028%; Cu=0.18%) of VVER 440 RPV after irradiations in VVER 440 reactor and thermal annealings [17].

Recovery and re-embrittlement of western RPV steels

The irradiation embrittlement in western reactors has been mainly caused by a high copper content in weldings. Because the older RPVs are usually constructed of hot-rolled plates, they have also axial joints and then weld metal in all the reactor core area. For limiting the risks originating from thermal stresses the studied annealing temperatures have been chosen to be as low as possible. Then the majority of test results are from 400°C annealings, which seems not to be adequate for high copper welds. In Fig. 13 it can be seen the recoveries in the transition temperature by 400°C and 455°C post irradiation heat treatments. The fluence has been 1.4x10^{19} n/cm². The degree of recovery depends strongly on the content of copper. Nickel and phosphorus have a smaller reducing effect on recovery. The influence of impurities is similar in both base and weld metals [26-29].
Fig. 13. Notch ductility changes for various welds after irradiation (288°C) and postirradiation annealing. The content of P=0.010-0.014 %; F= 1.4x10^19n/cm^2 [26].

The recovery of Charpy V-notch upper shelf energies has been easier than the most other properties (Fig.14). The degree of recovery measured by fracture toughness is much less than done by Charpy transition temperature [28,29]. As seen in Fig. 14 the degree of the recovery of J_{IC} may be less than a half of that. No explanations for this subject which is important for the evaluation of RPV integrity has been reported. More light in this respect can be expected from Heavy-Section Steel Irradiation (HSSI) Program where fracture toughness specimens up to 4 inch size will be tested after irradiation, annealing and reirradiation [30,31].
Contents of Cu (%):  
EP-19: 0.40  
EP-23: 0.23  
EP-24: 0.35

Fig. 14. Comparison of annealing results of different welds (Ni=0.59%) [29].

The re-embrittlement rate after recovery anneal is usually equal or smaller than in the first irradiation. The high annealing temperature is more favourable in this respect. In Figs. 15-16 results of welds having various Cu and Ni contents are seen.
Fig. 15. Transition temperature changes observed after reirradiation to a fluence of $2.7 \times 10^{19}$ n/cm$^2$ (IAR$_2$) or $1.8 \times 10^{19}$ n/cm$^2$ (IAR$_1$) vs. first exposure cycle. The left-hand and centre bars indicate the total transition temperature elevation with the IAR treatment [26].
Fig. 16. Effect of re-irradiation after various anneals on the transition temperature shift of weld EP-19 (Cu=0.40%; Ni=0.59%) [32].

The annealing process may also have detrimental effects on fracture toughness. In the case of fine grained base metal the influence of thermal ageing will not be large [33,34] unless the content of phosphorus is high, but in coarse grained HAZ the situation will be different. Already a fairly low content of phosphorus may then due to the thermal ageing increase the ductile-brittle transition temperature e.g. by 35°C when aged at 450°C for 100 hours [33] or 210°C when annealed at 450°C for 2000 hours [35]. Fortunately the prior ductility in the HAZ is usually clearly higher than in the normal base metal which gives more margin for the embrittlement (Fig. 17).
Fig. 17. Comparison of Charpy transition curves for unirradiated, irradiated, post-irradiation annealed (475°C/168h) and unirradiated thermally aged (450°C/2000h) material in the simulated (1200°C/0.5h + stress relieving heat treatment) coarse grained HAZ condition [35].

Evaluation of mitigation methods in PWRs

LL fuel management schemes can be regarded as basic procedures to reduce the flux to the RPV. Any low leakage fuel management scheme would probably be an advantageous option for the utilizer, if it could be done without significantly increasing (or even with decreasing) the fuel cycle cost and without need to reduce power. It is expected that already in 1993 most PWRs follow some kind of low leakage fuel management. The new fuel designs have evidently enhanced this trend.

The evaluation becomes much more complicated, when further flux reduction procedures, for example combinations of low leakage loading and some other method(s), are to be implemented in order to extend the RPV service life, because there will usually be a marked increase in the fuel cycle and/or implementation costs. Up to now the installation of dummy assemblies has been implemented only in few power plants. It should also be noted that flux reduction measures are most effective when applied at an early stage of the plant history.
In any case the measures should be implemented as early as possible. The significance of the timing is demonstrated in Fig. 18. The decision on the method(s) should be made after the first or second fuel cycle [37]. It takes usually two to three years to complete a change into a low leakage loading scheme.

Fig. 18. Curves for evaluating the effect of different options (Zion Unit 1, limiting circumferential weld) [36].

For U.S. nuclear plants the extension of the operating licenses from 40 years by 20 years is considered to be technically feasible. The 60 years service life is already being pursued by the nuclear utility industry [15].

A lot of research have been done on the methods and parameters of different annealing heat treatments. It seems that annealing is becoming the most effective means for extending the RPV service life in cases, where different low leakage loading schemes are not sufficient. This is true especially for those RPVs which do not have longitudinal welds. The modification of the emergency cooling system and related measures are likely regarded as complementary rather than primary methods in extending RPV service life.

A detailed plant specific analysis of different postulated PTS (and possibly some other emergency) events is necessary before a final evaluation can be made on the effect of the proposed prestressing procedure.

The management of RPV failure probability should include also other actions, for example the reduction of uncertainties in the RPV embrittlement material data and a coherent analysis of PTS events. Especially plant specific surveillance programs are becoming more important both in defining the required mitigation measures and in verifying the influence of implemented measures.
Conclusions

1. Low leakage and low fluence fuel management schemes are commonly used for PWRs to mitigate RPV embrittlement. Special fuel assembly designs have been provided in order to reduce the neutron flux at critical RPV locations with minimum loss in power. When flux reduction measure(s) is adopted, the implementation should occur at a very early stage of the plant history.

2. Fuel assemblies have been replaced with dummies, when a large reduction in the flux to the RPV has been required in order to achieve the design service life of the RPV. A core reduction together with modifications in emergency cooling systems has been a normal procedure to mitigate the exceptional fast RPV embrittlement and to extend the service life in VVER 440-type reactors. The number of applications in western PWRs is expected to be small.

3. Flux reduction by using shielding patches between the core and the RPV has been designed but there are expected to be no applications in PWRs.

4. Numerous successful thermal annealings have been carried out in former Soviet Union, East Germany, Bulgaria and Czechoslovakia. According to reports the thermal stress limits have not been exceeded. All those reactor pressure vessels have had only one circumferential weld to be annealed, which greatly simplifies the procedures. In many cases the most problematic question has been resolving reliably the actual chemical analysis and the state of embrittlement. These points are essential for evaluating the degree of recovery and the rate of re-embrittlement.

The annealing treatment of the old western RPVs is more difficult because they usually have axial welds to be treated, too. Hence, the high temperature zone extends close to the thick nozzle course resulting in high thermal stresses. A detailed plant specific thermal stress analysis must be done for the verification that the heat treatment procedure is safe for the pressure vessel and piping.

Other open questions are the observed lesser recovery of J-R curves compared with Charpy results and the possible embrittlement due to thermal ageing of coarse grained HAZ during recovery anneal.

5. The significance of plant specific surveillance tests is emphasized. The scope of surveillance test programs for evaluating the effect of implemented annealing treatments should correspond to those required for the original PWRs.

References


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UNITED STATES DEPARTMENT OF ENERGY PROJECTS RELATED TO REACTOR PRESSURE VESSEL ANNEALING OPTIMIZATION

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ABSTRACT. Light water reactor pressure vessel (RPV) material properties reduced by long-term exposure to neutron irradiation can be recovered through a thermal annealing treatment. This technique to extend RPV life provides a complementary approach to analytical methodologies to evaluate RPV integrity. RPV annealing has been successfully demonstrated in the former Soviet Union and on a limited basis by the U.S. (military applications only). The process of demonstrating the technical feasibility of annealing commercial U.S. RPVs is being pursued through a cooperative effort between the nuclear industry and the U.S. Department of Energy (USDOE) Plant Lifetime Improvement (PLIM) Program. Presently, two projects are under way through the USDOE PLIM Program to demonstrate the technical feasibility of annealing commercial U.S. RPVs, (1) annealing re-embrittlement data base development and (2) heat transfer boundary condition experiments.

Presently, limited information is available regarding the re-embrittlement behavior of typical RPV plate and weld materials following a 454°C (850°F) anneal. This project involves a series of test reactor irradiation-anneal-reirradiation experiments performed on RPV plate (ASTM-type A302 Grade B and A533 Grade B) and weld (Linde 80) materials. The re-embrittlement behavior of these materials is being studied as a function of fluence level, annealing temperature, and time. Results from this study will help determine the proper combination of annealing temperature and time to minimize the re-embrittlement rate and maximize material property recovery. A summary description of the re-embrittlement data base effort is presented.

The DOE PLIM Program is providing benchmark data for the evaluation of analytical models used to characterize and predict RPV response (thermally induced stresses and strains) during annealing. This project involves an experiment at Sandia National Laboratories' Radiant Heat Facility to provide heat transfer boundary condition inputs to analytical models and a future experiment to obtain strain measurements on RPV components during a simulated anneal. The experiments include extensive characterization of material response during annealing simulations performed on an unirradiated section of an RPV. The test apparatus also includes a mock-up of the insulation surrounding the RPV and the reactor cavity wall. The experimental setup is described.

Keywords: radiation embrittlement, nuclear reactor pressure vessel steels, thermal annealing, re-embrittlement

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INTRODUCTION

The continued viability of the nuclear power option (i.e., life extension, or the next generation of plants) is very dependent on the continued safe and economic operation of existing plants without premature shutdown. The resolution of RPV neutron radiation embrittlement issues, in a cost-effective manner without excessive conservatism due to a lack of clear scientific information, is critical given the RPV's safety significance and high replacement cost.

Several options exist to manage the embrittlement of RPV materials. These options can be grouped into four categories [1]:

1. demonstrating embrittlement susceptibility to be less than predicted,
2. reducing the embrittlement rate,
3. removing the embrittlement, and
4. demonstrating that plant-specific variables permit greater levels of acceptable embrittlement.

Category 1 includes, for example, an enhanced surveillance program to obtain more knowledge of the actual critical material irradiation exposure. This information could be used to reduce uncertainty in subsequent analyses to predict embrittlement trends. Category 2 involves flux reduction techniques including fuel management, shielding and derating. Category 3 includes thermal annealing, vessel weld replacement and vessel replacement. Category 4 includes techniques to demonstrate the benefit of particular plant conditions, i.e. vessel weld sampling, analytical methods to demonstrate continued RPV integrity under specified loading conditions.

Thermal annealing, as described in Category 3 above, is one possible means of RPV embrittlement management that will result in the removal of neutron radiation damage. In fact, it is the only mitigative measure that restores the mechanical properties of the RPV materials. Thermal annealing, as applied to a commercial RPV would not be a traditional "full" anneal. RPV annealing temperatures are expected to range from approximately 315-480°C (600-900°F). The effectiveness of a thermal annealing treatment in recovering material properties will depend upon the original RPV irradiation temperature, annealing temperature, annealing time at temperature ("hold" time), original material chemistry and the degree of embrittlement prior to annealing [2,3]. A limited number of thermal anneals have been performed, both in Europe [4,5] and by the U.S. for military reactors only [6].

The technical feasibility of annealing commercial U.S. vessels has been studied [3,7]. Based on these preliminary studies, annealing of U.S. RPVs is technically viable. More recently, thermal annealing has also been shown to be economically desirable under certain embrittlement management scenarios [1]. However, only limited detailed material performance data and information characterizing the general response of the RPV and surrounding components to the annealing treatment has been established to support these preliminary studies. This suggests the need to perform additional confirmatory metallurgical and material behavior research, develop an appropriate annealing process, and ultimately demonstrate thermal annealing technology on a commercial U.S. RPV. The U.S. Department of Energy (USDOE) Plant Lifetime Improvement (PLIM) Program is pursuing the technical
demonstration of annealing commercial U.S. RPVs through a cooperative effort with the nuclear industry. Activities are presently under way through the USDOE PLIM Program to perform confirmatory metallurgical and general material behavior research in support of U.S. nuclear industry efforts to ultimately demonstrate thermal annealing technology on a commercial U.S. RPV. Specifically, these efforts involve preliminary development of a re-embrittlement data base for RPV materials following an anneal and providing benchmark data for the evaluation of analytical models used to characterize and predict RPV response (thermally induced stresses and strains) during an annealing treatment.

RE-EMBRITTLEMENT DATA BASE DEVELOPMENT

One important aspect of a successful annealing demonstration program is the proper characterization of RPV material properties before and after an anneal. A limited amount of metallurgical research has been performed regarding the amount of material property recovery (Charpy impact and tensile properties) anticipated following an annealing treatment \(\text{(3)}\). These studies focused on determining the optimum annealing time and temperature, and the amount of anticipated property recovery. Little effort has been put forth to investigate the rate at which RPV materials may re-embrittle following an anneal, i.e., the embrittlement rate of RPV materials following annealing compared to the rate of embrittlement prior to anneal. The determination of RPV material re-embrittlement rates is critical if the economic viability of annealing is to be evaluated for U.S. commercial pressure vessels.

Test Plan

The USDOE PLIM Program is pursuing initial development of a re-embrittlement data base through an irradiation-anneal-reirradiation (IAR) project involving typical U.S. RPV materials (base plate and weld). The plate materials under study include two types, American Society for Testing and Materials (ASTM) type A 533 Grade B and type A 302 Grade B. These materials are representative of those used in the fabrication of commercial U.S. RPVs. The weld being studied is a low Charpy upper-shelf impact energy material fabricated with Linde 80 flux. This particular flux was chosen for weld fabrication of several early vintage U.S. RPVs because it resulted in very small, finely dispersed nonmetallic inclusions that resulted in fewer required weld repairs. Unfortunately, the fineness of the inclusions, as a result of the Linde 80 flux, led to a relatively low Charpy upper-shelf impact energy. Therefore, this weld material would be more adversely affected by exposure to neutron radiation. Due to its sensitivity to neutron irradiation, Linde 80 weld material was included in the project and is expected to provide bounding information for typical RPV weld materials. The chemistries of the materials being studied under this project are given in Table 1.

The complete test matrix for the materials included in this study is shown in Table 2. The IAR project consists of two separate sample capsules containing standard Charpy V-notch and tensile test specimens. Charpy V-notch samples will be used to determine the radiation-induced changes in the ductile-to-brittle transition temperature and upper-shelf energy. Tensile samples are included to characterize changes in yield and tensile strengths due to neutron radiation. Results obtained through the irradiation, annealing, and reirradiation activities of this study will be compared with previously published testing results regarding unirradiated material properties of the identical materials \(\text{(8)}\) to determine the amount of
<table>
<thead>
<tr>
<th></th>
<th>Cu</th>
<th>Ni</th>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>Mo</th>
<th>S</th>
<th>P</th>
<th>Fe</th>
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<td>0.25</td>
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<td>0.011</td>
<td>0.008</td>
<td>Rem</td>
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<tr>
<td>(A 533B)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Plate Material</td>
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<td>0.19</td>
<td>0.17</td>
<td>1.28</td>
<td>0.22</td>
<td>0.50</td>
<td>0.022</td>
<td>0.026</td>
<td>Rem</td>
</tr>
<tr>
<td>(A 302B)</td>
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<td></td>
<td></td>
<td></td>
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<td></td>
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<td></td>
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<td>1.00</td>
<td>0.08</td>
<td>1.39</td>
<td>0.89</td>
<td>0.50</td>
<td>0.015</td>
<td>0.015</td>
<td>Rem</td>
</tr>
<tr>
<td>(Linde 80)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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material property recovery due to annealing and the re-embrittlement rate during subsequent reirradiation.

As shown in Table 2, the test plan involves the simultaneous irradiation of the two sample capsules to a fluence of approximately $3 \times 10^{19}$ n/cm$^2$, $E > 1$ MeV at a target irradiation temperature of 288°C (550°F). Following initial irradiation, one-half the contents of capsule A will be removed and tested to obtain as-irradiated data. The remaining samples in Capsule A will be annealed at 454°C (850°F) for 168 hours (one week) and tested. Results will be compared with previous mechanical property tests (performed on the same testing equipment) on unirradiated samples from the same material piece [8] to obtain annealing recovery data. The second capsule (Capsule B) will be annealed intact with the samples from Capsule A and reinserted into the test reactor to obtain re-embrittlement data at a single target fluence level of $1 \times 10^{19}$ n/cm$^2$, $E > 1$ MeV. Initial irradiations are under way on the two capsules and should be completed in November 1993. The anneal of Capsule B and half the Capsule A samples will be performed by the end of 1993. Reirradiation of the annealed samples, mechanical testing and project completion is anticipated by mid-1994.
**Table 2. IAR Project Test Matrix**

Capsule A - Part I, Irradiation Only (As-irradiated condition)
Target Fluence - $3 \times 10^{19}$ n/cm$^2$, E > 1 MeV
Irradiation Temperature - 288°C (550°F)

<table>
<thead>
<tr>
<th>Material</th>
<th>Type</th>
<th>Samples</th>
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</thead>
<tbody>
<tr>
<td>Weld (Linde 80)</td>
<td>Charpy</td>
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</tr>
<tr>
<td>Plate (A 533B)</td>
<td>Charpy</td>
<td>8</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Charpy</td>
<td>8</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Tensile</td>
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</tr>
<tr>
<td><strong>Total - Capsule A, Part I</strong></td>
<td></td>
<td><strong>27</strong></td>
</tr>
</tbody>
</table>

Capsule A - Part II, Irradiation-Anneal (As-annealed condition)
Annealing Temperature - 454°C (850°F), 168 hrs.

<table>
<thead>
<tr>
<th>Material</th>
<th>Type</th>
<th>Samples</th>
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</thead>
<tbody>
<tr>
<td>Weld (Linde 80)</td>
<td>Charpy</td>
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</tr>
<tr>
<td>Plate (A 533B)</td>
<td>Charpy</td>
<td>8</td>
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<tr>
<td>Plate (A 302B)</td>
<td>Charpy</td>
<td>8</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Tensile</td>
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<tr>
<td><strong>Total - Capsule A, Part II</strong></td>
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<td><strong>27</strong></td>
</tr>
</tbody>
</table>

Capsule B - IAR (Re-embrittled condition)
Irradiated, annealed with Capsule A, Part II
Reirradiated target fluence - $1 \times 10^{19}$ n/cm$^2$, E > 1 MeV

<table>
<thead>
<tr>
<th>Material</th>
<th>Type</th>
<th>Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weld (Linde 80)</td>
<td>Charpy</td>
<td>14</td>
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<tr>
<td>Plate (A 533B)</td>
<td>Charpy</td>
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<tr>
<td>Plate (A 533B)</td>
<td>Tensile</td>
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</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Charpy</td>
<td>8</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Tensile</td>
<td>2</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Charpy</td>
<td>8</td>
</tr>
<tr>
<td>Plate (A 302B)</td>
<td>Tensile</td>
<td>2</td>
</tr>
<tr>
<td><strong>Total - Capsule B</strong></td>
<td></td>
<td><strong>48</strong></td>
</tr>
<tr>
<td><strong>Total Samples Tested</strong></td>
<td></td>
<td><strong>102</strong></td>
</tr>
</tbody>
</table>

*Microstructure characteristic of RPV inside surface, i.e., finer grain size.*
HEAT TRANSFER BOUNDARY CONDITION EXPERIMENTS

Accurate prediction of material behavior for the RPV, including the nozzle and flange regions, via thermal/stress computer models is also an important element of successful demonstration of thermal annealing technology on U.S. commercial RPVs. Validation and verification of annealing computer model(s) is an important step in that accurate prediction.

The following sections describe a set of 1-dimensional (1-D) experiments designed to provide heat transfer boundary condition and RPV material temperature response data to help benchmark thermal/stress finite-element annealing computer model(s). Heat transfer data generated can be used to verify the thermal boundary conditions used as model inputs.

The experiments subjected an RPV test specimen to a linear temperature rise from ambient to 454°C (850°F) at two rise rates, namely 14°C/hr (25°F) and 28°C/hr (50°F/hr). After thermal equilibrium was reached, the temperature was reduced to ambient at approximately the same rate as the rise. For redundancy, two tests were performed at each rise rate.

Test Setup

A 1.2 m x 1.2 m x 17.1 cm thick (4 ft x 4 ft x 6.75 in) piece of the Phipps Bend RPV (Boiling Water Reactor) was obtained from the Electric Power Research Institute Non-Destructive Evaluation Center. The base material is ASTM A 533 Grade B with a 3.2-4.8 mm (0.13-0.19 in) thick stainless steel (SS) cladding on the concave (inside) surface.

Figure 1 shows a side view of the test setup (not to scale). The RPV test specimen and heater assembly were mounted on individual supporting frames. The heater assembly

![Diagram of heat transfer boundary condition experiments]

Figure 1. Side View of 1-Dimensional Heat Transfer Boundary Condition Experiments
consisted of 9 individual heaters each 0.6 m x 0.6 m (2 ft x 2 ft) square in a 3 x 3 array configuration. Each heater is capable of 3.1 W/cm² (2.7 Btu/ft²·sec) heat flux output, and they are collectively referred to as "infrared panel heaters". This type of heater was chosen because of its similarity to that used by the former Soviet Union to anneal their RPVs. See Reference [5] for a description of the Russian heater hardware.

Figure 2 shows a top view of the setup. The curvature is exaggerated but shows that the heaters were contoured to match the curvature of the RPV wall section. The radius of curvature of the RPV is 2.8 m (110 in).

![Diagram of infrared panel heater and RPV wall section]

**Figure 2.** Top View of 1-Dimensional Annealing Thermal Model Benchmarking Experiments

The total heater assembly is 1.8 m (6 ft) square, whereas the test specimen is 1.2 m (4 ft) square. The overlap ensures heating uniformity on the RPV test specimen and more nearly simulates 1-D conditions. The 9 total heater panels are controlled by 3 power channels, i.e., 3 heaters controlled per channel. Each set of 3 heater panels oriented horizontally are connected to a single channel to avoid convective effects on heater control. Automatic, precise temperature profile control will be achieved with programmable power channels.

All edges of the RPV wall section were insulated with a ceramic-fiber type of insulation. This type of high temperature insulation has very good insulating properties (i.e., very low thermal conductivity). The top, bottom and sides between the heater assembly and the RPV test specimen were enclosed with insulation to simulate anticipated in-situ RPV annealing conditions.

The space between the unheated side of the RPV test specimen and the concrete wall simulating the reactor cavity was also insulated. Typical U.S. commercial RPVs are insulated with 7.6 cm (3 in) of "mirror" insulation made of SS sheets with an aluminum foil filler. The
thermal conductivity of this type of insulation is about twice that of the ceramic-fiber type insulation described above and is much more expensive. Therefore, approximately 3.8 cm (1.5 in) of the ceramic-fiber insulation was substituted for the mirror type insulation and placed between two aluminum sheets that simulate the thermal heat transfer properties of stainless steel. The insulation was placed 1.9 cm (0.75 in) away from the unheated side of the RPV wall section.

A concrete wall was placed 5.1 cm (2 in) behind the insulated, unheated side of the RPV test specimen to simulate the reactor cavity. This wall is solid concrete, 2.1 m x 2.1 m x 25.4 cm (7 ft x 7 ft x 10 in) thick. For ease of fabrication purposes, the concrete wall did not possess a curvature matching that of the RPV test specimen, i.e., the concrete wall was flat. However, based on the size of the RPV test specimen and the overall objectives of this experiment, a flat wall was assumed a reasonable approximation.

Typical specifications call for maximum air flow of about 736,000 l/min (26,000 cubic feet per minute (CFM)) to a minimum of 130,000 l/min (4600 CFM) at 38°C (100°F). Assuming a 5.1 cm (2 in) gap surrounding a 4.1 m (160 in) diameter RPV, the velocity through the gap would range from about 1,135 m/min to 204 m/min (3724 ft/min to 670 ft/min). Fans and ductwork will be used to simulate upward-directed flow at typical flow rates through the 5.1 cm (2 in) space between the insulation and the concrete wall at the test setup. Air flow will be measured once per test at the top of the RPV section using a velometer (air flow velocity meter) located between the insulation and the RPV test specimen.

Instrumentation/Measurements

Figure 3 shows the instrumentation layout on the heated side of the RPV wall. The bulk of the instrumentation are type K (chromel-alumel) thermocouples (TCs), 1.6 mm (0.13 in)
diameter and inconel sheathed. The TCs used in this study are accurate to ± 0.75% of the reading, which is sufficiently accurate for the purposes of these experiments. Therefore, calibration was performed only at a single temperature, not throughout the entire temperature range. All thermocouples were within the ± 0.75% limits. They were attached to the RPV test specimen via nichrome strips tack welded to the surface. The TCs were bonded to the concrete wall with cement.

Three "pyrheliometers" (heat flux gages) will be used to measure heat flux incident on the heated RPV wall surface. This type of gage is typically used for solar energy applications to measure radiative heat flux, but were configured to measure total heat flux (radiative + convective) in these experiments.

As Figure 3 shows, 32 TCs were mounted on the heated surface of the RPV test specimen. The bulk of the TCs were concentrated in the center where the heat transfer was closest to 1-dimensional (to minimize "edge" effects). The heat flux gages were mounted to obtain (2-point) vertical and horizontal profiles of the heat flux. A TC was mounted in near proximity to each heat flux gage and will be used to estimate the total (convective + radiative) absorbed flux. Each heat flux gage will be used to measure the total incident flux. Power to each of the heater elements is recorded to provide a further estimation of the heat flux.

The TC layout on the unheated side was identical to that on the heated side. Heat flux gages were not mounted on the unheated side. Nine TCs were mounted on the concrete wall to determine concrete face temperature. The air temperature in the gap between the insulation and the concrete wall will not be measured.

Temperature Profile Imposed on RPV Wall Section Heated Surface

A maximum temperature rise rate of approximately 14°C/hr (25°F/hr) will be utilized [9]. Assuming an initial ambient temperature of 21°C (70°F) and a maximum experiment temperature of 454°C (850°F), the ΔT is 416°C (780°F). At 14°C/hr (25°F/hr), the rise time to maximum temperature is about 31.2 hrs. Preliminary modelling showed that, at this very slow rise rate, the temperature gradient through the thickness of the RPV wall section would be small, less than 5°C (10°F). Therefore, once the maximum temperature of 454°C (850°F) is reached, the test specimen should be very close to equilibrium, and it will not be necessary to hold the maximum temperature for a long time (greater than 1 hr). The temperature will then reduced to ambient at approximately the same rate as the rise, 14°C/hr (25°F/hr). The test time for a typical test will be about 63-64 hours using a 14°C/hr (25°F/hr) rise rate. This profile will be programmed into the power controllers. The power system typically controls to within ± 3°C (± 5°F) or better.

A total of four tests will be performed. The principal test conditions are as follows:

- Two tests @ 14°C/hr (25°F/hr) rise rate on the heated surface (two for redundancy).
- Two tests @ 28°C/hr (50°F/hr) rise rate on the heated surface (two for redundancy).

The 28°C/hr rise rate was proposed in order to examine a larger through-wall temperature difference.
Data Analysis

Table 3 gives an overview of the a) measurements to be made, b) data reduction plan and c) final output. Details are given below.

The TC measurements will provide a large amount of data to analyze the heat transfer to the RPV wall section and determine the impact of annealing on the concrete wall. The following data will be provided:

1. Heated surface temperature vs. time at a maximum of 32 locations (Multiple traces on a single plot).
2. Unheated surface temperature vs. time at a maximum of 32 locations (Multiple traces on a single plot).
3. Temperature difference (from heated and unheated side data) through the RPV wall thickness vs. time at a maximum of 32 locations (Multiple traces on a single plot).
4. Temperature "maps" on the heated face to check for "hot" and "cold" spots at a maximum of 32 locations and specified times.
5. Temperature at weld location vs. time, to compare with temperature at non-weld locations vs. time. (3 locations).
6. Concrete surface temperature vs. time at a maximum of 9 locations (Multiple traces on a single plot).
7. Temperature "maps" on the concrete surface (specific form of output to be determined).

The TC measurements, in conjunction with a Sandia National Laboratories developed computer code, "SODDIT" (Sandia One-Dimensional Direct and Inverse Thermal) [10], will be used to predict the heat flux actually absorbed into the heated surface vs. time (sum of radiative and convective parts). As a result, wherever a TC is located, absorbed heat flux will be determined. The following heat flux data will be available:

1. Absorbed heat flux vs. time on the heated surface from heated surface TCs (maximum of 32 locations).
2. Absorbed heat flux vs. time on the heated surface from unheated surface TCs (maximum of 32 locations).
3. Absorbed heat flux map on the heated surface at specified times from heated surface TCs (maximum of 32 locations).\(^1\)
4. Absorbed heat flux map on the heated surface at specified times from unheated surface TCs (32 locations).
5. Absorbed heat flux on the concrete surface (specific form of output to be determined).

Considerable redundancy will be maintained during measurements. This is intentional, as it allows comparison of data from the heated and unheated sides and provides backup data to check for anomalies. Under ideal conditions, only the heated side TCs would be used to

\(^1\) Data from selected locations depending on the results.
Table 3. Data Analysis/Data Reduction Plan

<table>
<thead>
<tr>
<th>Transducer/Measurement</th>
<th>Data Reduction</th>
<th>Final Output</th>
</tr>
</thead>
<tbody>
<tr>
<td>TC/heated face temperature (total - 32 TCs)</td>
<td>Use TC data from heated face as input to SODDIT(^1) which calculates heat flux absorbed into heated face of RPV test specimen</td>
<td>Plots of temperature vs. time at 32 locations on heated face. Plots of absorbed heat flux vs. time for 32 locations on heated surface.</td>
</tr>
<tr>
<td>TC/unheated face temperature (total - 32 TCs)</td>
<td>Use TC data from unheated face as input to SODDIT to estimate absorbed heat flux on heated face.</td>
<td>Plots of unheated face temperature vs. time for 32 locations on unheated face. Plots of absorbed heat flux on heated face vs. time for 32 locations, used as backup for heat flux data from heated face TCs.</td>
</tr>
<tr>
<td>Pyrheliometer(^2)/heat flux (total - 3 heat flux gages)</td>
<td>Direct measure of incident heat flux on heated surface. Compare with absorbed flux at same locations.</td>
<td>Plots of incident heat flux on heated surface for 3 locations. Will be used to compare with absorbed heat flux at same locations to check efficiency of overall heat transfer.</td>
</tr>
<tr>
<td>TCs on heated/unheated faces</td>
<td>Use unheated face TC measurements as inputs to SODDIT to estimate the temperature profile through the thickness. Compare with heated and unheated face TC measurements.</td>
<td>Plots of temperature profile through the wall thickness as estimated by SODDIT and those measured from the heated and unheated face TCs.</td>
</tr>
<tr>
<td>TC/concrete wall temperature (total - 9 TCs)</td>
<td>Direct measure of concrete wall temperature.</td>
<td>Plots of temperature vs. time for 9 locations on concrete wall, including air flow.</td>
</tr>
<tr>
<td>Power to heaters</td>
<td>With known heater area, estimate maximum heat output.</td>
<td>Power used by heaters vs. time. Compare with heat flux incident on test specimen.</td>
</tr>
<tr>
<td>Velometer/air flow (1 velocimeter - total)</td>
<td>One-time, direct measurement of air flow in gap between insulation and concrete.</td>
<td>Comparison of concrete temperature/air flow with anticipate plant conditions.</td>
</tr>
</tbody>
</table>

\(^1\) SODDIT = Sandia One Dimensional Direct and Inverse Thermal computer code.

\(^2\) Pyrheliometer = heat flux gage that can measure low values of heat flux typical of these experiments (0-1 Btu/ft\(^2\)-sec).
estimate absorbed heat flux on the inside surface of the RPV test specimen. However, because the heated side TCs are mounted on top of the surface (rather than flush with the surface) they will indicate a higher temperature than the actual surface temperature of the RPV test specimen. This will result in estimated heat flux values higher than actual. Comparisons of the absorbed flux estimated from both the heated and unheated side TCs will be made to determine the "best" values.

Pyrheliometers will be used to measure total incident heat flux to the heated surface. This allows comparison with the total absorbed heat flux to determine the amount of heater energy actually absorbed by the RPV wall section. The expected incident flux level for the 1.4°C (25°F) rise rate is about 0.24 w/cm² (0.21 Btu/ft²·sec). In a completely enclosed setup, such as this study, virtually all of the heater energy should be absorbed by the RPV wall.

Output Data for Thermal/Stress Modelling Efforts

To achieve the goals of providing boundary condition data for subsequent modeling efforts, the following information will be provided:

1. Temperature vs. time on the heated surface of the RPV wall section.
2. Temperature map of heated surface at selected times during heat-up and cool-down.
3. Heat flux absorbed into the surface vs. time, including the sum of the radiative and convective parts (maximum of 32 locations).
4. Incident heat flux on the heated surface vs. time, including the sum of the radiative and convective parts (3 locations).
5. Other data as required.

The following additional information to be provided will assist in further code benchmarking efforts:

1. Temperature profiles through the thickness vs. time from measured data (front and back sides).
2. Temperature profiles through the thickness vs. time from predicted data (SODDIT).
3. Temperature on beltline weld material, compared with that on the base metal.
4. Temperature on the concrete wall behind the RPV wall section.
5. Temperature vs. time on the heated surface of the RPV wall section.
6. Temperature map of heated surface at selected times during the heat up and cool down.
7. Other data as required.

At time of preparation, experiments at the Sandia National Laboratories Radiant Heat Facility were under way. Data reduction and analysis will be completed at the conclusion of the experiments.

CONCLUSIONS

The USDOE PLIM Program, through the individual projects described above, is pursuing its objective of demonstrating the technical feasibility of annealing commercial U.S. RPVs.
Efforts are under way to (1) begin development of an annealing re-embrittlement data base and (2) provide benchmark data for the evaluation of analytical models used to characterize and predict RPV response (thermally induced stresses and strains) during an annealing treatment. Future efforts may include additional experiments to measure strains induced in RPV components during an annealing treatment, establishment of the technical basis for RPV requalification following an anneal, and additional re-embrittlement data base activities to optimize the amount of material property recovery and re-embrittlement rate of RPV materials. By working cooperatively with the U.S. nuclear industry, the USDOE is contributing to the technology and information needed to establish the technical groundwork for a successful demonstration of annealing technology in the U.S.

ACKNOWLEDGEMENTS

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REFERENCES


Irradiation Embrittlement of Reactor Pressure Vessel Steels: Considerations for Thermal Annealing

IAEA Specialist Meeting on Irradiation Embrittlement and Optimization of Annealing

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1.0 Introduction/Background

The material properties of reactor pressure vessel (RPV) steels are known to be degraded by exposure to high energy neutron bombardment during the operation of pressurized water reactors. This degradation of the vessel material restricts the allowable operating temperature and pressure conditions for the reactor and also leads to concern regarding the response of the pressure vessel to possible pressurized thermal shock transients. The Code of Federal Regulations (CFR) establishes formal fracture toughness criteria for the acceptability of RPV materials. 10CFR Part 50 Appendix G, "Fracture Toughness Requirements," [1] establishes a limit of 50 ft-lbs as the minimum Charpy upper shelf energy (USE) that must be maintained throughout the life of the vessel while 10CFR Part 50.61 [2] addresses fracture toughness requirements for pressurized thermal shock (PTS) conditions.

One of the ways to manage or alleviate irradiation embrittlement is to thermally anneal the reactor pressure vessel. The restoration of material properties of embrittled vessels has been demonstrated to be feasible, both in the laboratory and in the field. Although the RPV of a commercial U.S. nuclear power plant has yet to be thermally annealed, this process is being performed on reactor vessels in the former Soviet Union. A program sponsored by the Electric Power Research Institute (EPRI) in the U.S. was carried out by Westinghouse to determine the feasibility of and methodology for thermally annealing an embrittled U.S. vessel.[3] It should be noted that this EPRI study did not address the numerous licensing, code and standard issues involved with such an anneal. Now, however, renewed interest in thermal annealing has prompted further consideration of a proposed annealing demonstration on a full-scale unirradiated vessel.

It has been shown that a thermal treatment of 450°C (842°F) for 168 hours recovers most of the mechanical properties that are lost due to irradiation embrittlement and results in RPV material with reduced radiation sensitivity. These studies have not however, addressed questions concerning variations in chemistry and/or fluxes and the effect of these variations on the material's recovery and re-embrittlement rates. Because of the differences in chemistry from heat to heat and weldment to weldment, and variations in fluxes from reactor to reactor, all vessel materials do not recover to the same degree or at the same temperature. By identifying the mechanism(s) by which the embrittlement phenomenon occurs, it will be possible to develop the most appropriate monitoring techniques and recovery procedures. A better understanding of the mechanism(s) of irradiation embrittlement should also lead to improved embrittlement behavior prediction capabilities.

In this paper, an overview of the irradiation embrittlement phenomenon is presented from a structure-properties viewpoint. In addition, the application of thermal annealing to effectively restore the original properties of an embrittled reactor pressure vessel is discussed.

2.0 Effect of Irradiation Conditions on Embrittlement

Embrittlement of the reactor pressure vessel is a major concern for the nuclear industry because it severely restricts safe plant operating margins, i.e., pressure-temperature limits for heat-up and cool-down. This embrittlement is most severe in the belt-line region of the vessel which is exposed to a large dose of high-energy neutrons.
The effect of this neutron bombardment is generally measured by a conventional Charpy V-notch impact test and is manifested by an upward shift in the ductile-to-brittle transition temperature (DBTT) and by a decrease in the upper shelf energy. A large amount of data has been generated which examines the effects of various irradiation conditions such as irradiation temperature, neutron fluence, flux and material composition on embrittlement behavior. However, these data are often difficult to interpret because of the complex interactions between the material and the high energy neutrons. This section discusses the effects of these parameters on embrittlement.

2.1 Irradiation Temperature

The temperature at which a material is irradiated significantly impacts the degree to which a material is embrittled. Specifically, irradiation embrittlement is enhanced by irradiation at lower temperatures with the maximum embrittlement occurring at temperatures ~<230°C (450°F).[4,5] Embrittlement at higher temperatures is less pronounced because of simultaneous partial annealing of the irradiation defects. Most of the irradiation embrittlement data generated to date has been obtained for irradiation temperatures of ~290°C which is the typical operating temperature for pressurized water reactors.

The effects of irradiation temperature (150°C, 300°C, 400°C) as a function of fluence, flux and material composition where systematically studied by Pachur [6,7]. For a given fluence range (10^{18} to 10^{20} n/cm^2 (E>1MeV)), he showed that a lower irradiation temperature of 150°C produced the greatest increase in yield strength, transition temperature, and Vickers hardness. Lower increases in these parameters were observed for irradiations at 300°C with even smaller increases for irradiations at 400°C, consistent with simultaneous partial annealing of defects at higher irradiation temperatures.

2.2 Fluence

The effect of fast neutron fluence (or accumulated dose) on irradiation hardening and embrittlement has been reported to be significant, particularly in the range 10^{18} to 10^{20}n/cm^2 (E> 1MeV). However, because many studies of fluence effects only varied the dose within an order of magnitude, subtle and quantitative influences of the effect of neutron dose are difficult to evaluate.

In general, data from surveillance and test reactor irradiation studies show that increases in DBTT, yield strength, and hardness are related to the neutron fluence [8-49]. These increases in embrittlement are reported to be proportional to the square root [9] or cube root [14,50] of the fluence. Furthermore, there exists a threshold fluence below which the steel is insensitive to irradiation hardening. Additionally, a saturation of embrittlement (transition temperature shift) within certain fluence ranges for both low and high flux irradiations at a variety of irradiation temperatures was observed by Pachur [6] for several RPV steels. Lucas et al. [33] observed a similar saturation at a fluence of ~10^{19}n/cm^2.
2.3 Flux

The effect of flux on the embrittlement behavior of pressure vessel steels is not well-defined. Data from numerous investigators on a variety of alloy compositions indicate that the flux effects are related to the material, the fluence, and the temperature of irradiation. In particular, the copper and nickel contents of the steel appear to have a pronounced influence on dose rate effects.

A great portion of the irradiated data generated to date has been obtained from samples irradiated in high flux test reactors to fluences simulating end-of-life doses. The application of this accelerated test data to power reactor materials assumes that the embrittlement of the materials is not a function of flux. Mager and Lott, [51] using weld and plate materials irradiated in both power and test reactors to fluences of $2.5 \times 10^{18}$ to $8.8 \times 10^{19}$ n/cm$^2$ ($E>1$ MeV), have shown no apparent dependence on flux. Trend curves were developed for the irradiated material; despite the mixture of irradiation environments, the data appeared to indicate a smooth trend curve. Recent investigations however, show that the embrittlement may indeed be dependent on flux. Oak Ridge National Laboratory (ORNL) has observed significant acceleration of embrittlement in routine PV surveillance specimens from the High Flux Isotope Reactor (HFIR) as compared to prior test reactor data.

Mansur and Farrell [52], and Stoller and Mansur [53] have described the effect of flux (displacement rate) and neutron spectrum by the number of point defects which avoid recombination. Recombination of these point defects should be strongly influenced by flux and spectrum. A lower level of recombination may be experienced by RPV materials than by test reactor materials because of the lower displacement rates and softer spectra experienced by RPV materials. Less recombination would provide more point defects per displaced atom to cause greater embrittlement.

2.4 Chemistry

The alloy composition of RPV steels has a significant effect on the embrittlement response for materials irradiated at ~290°C. This sensitivity to alloy chemistry appears to be less significant at lower irradiation temperatures (~230°C). [4]

Both commercial steels and model alloys have been studied to verify the role of specific elements in the embrittlement phenomenon. There is considerable data in the literature pertaining to the effect of alloy composition on irradiation embrittlement, in particular the detrimental effects of Cu and P. Significant levels of Cu promote the formation of irradiation-induced solute-rich features in the material, which are associated with the degradation in impact properties. Increased levels of P have been associated with increased shifts in DBTT for irradiated materials. Recently, there has been renewed interest in the non-hardening aspect of irradiation embrittlement, i.e. the role of segregation and intergranular fracture. Microanalytical data indicate that P is an important contributor to non-hardening embrittlement. Other elements such as Mn and Ni have been the focus of various embrittlement studies. Manganese has been identified as a notable variable which differentiated the embrittlement behavior of A302B and A212B steels. [18] The effect of Ni in irradiation embrittlement is unclear, with contradictory reports in the literature: for welds and plates, the effect of Ni is dependent upon the presence of Cu.
3.0 Identification/Characterization of Irradiation-Induced Microstructural Features

The phenomenon of irradiation embrittlement is well-documented through the extensive mechanical properties studies which have been performed over the past 40 years. However, the changes in the microstructure caused by neutron irradiation of these materials are not as readily evaluated. The identification of these chemical and physical changes is important in that it is the microstructure of the material which controls the observed mechanical properties of the steel. The major analytical techniques which have been applied to the evaluation of irradiated steels include transmission and scanning transmission electron microscopy (TEM, STEM), small angle neutron scattering (SANS), atom probe field-ion microscopy (APFIM), and positron annihilation lifetime spectroscopy (PALS). It is important to note that each technique has its advantages as well as disadvantages in the characterization of irradiation-induced microstructural changes.

3.1 TEM/STEM

Transmission electron microscopy (TEM) has been employed in numerous studies of irradiation embrittlement, with marginal success. Although TEM provides important microstructural data concerning the material (i.e., identification and distribution of major phases and precipitates, dislocations, loops and voids, if present), it is limited by its inability to resolve ultrafine features (<2 nm).

Irradiation to fluences which produce significant hardening in ferritic alloys can not be correlated with TEM-observed microstructural changes within the material. However, defect structures formed by higher fluence irradiation have been observed and studied extensively by TEM. Of particular importance in the investigation of irradiation effects is the identification of dislocation loops and microvoids at high fluence (~10²⁰n/cm²). To date, microvoids have not been reported in irradiated RPV materials to end-of-life fluences although nanovoids have been observed in irradiated austenitic steels.

Recently, the dedicated field emission gun scanning transmission electron microscope (FEG-STEM) has been employed in the evaluation of irradiated pressure vessel steel from the Gundremmingen reactor.[54] With very specialized sample preparation techniques, it was possible to prepare thin-foil specimens which were amenable to STEM-EDS microanalysis. The fine probe size (~1 nm) attainable with a FEG-STEM permitted the detection of localized enrichments of Cu, Ni and Mn which were interpreted by Buswell and co-workers as Cu-rich precipitates. The dedicated FEG-STEM provides a complementary analytical tool to APFIM and SANS for the characterization of irradiated materials.

Lattice images obtained from very thin irradiated specimens have been presented by researchers at EDF [55] and Harwell [56]. These bcc regions appear to be associated with the "precipitates" containing Cu, Mn, and Ni as reported from FEG-STEM studies. Fundamental microstructural analyses utilizing high resolution electron microscopy have been performed on aged model alloys at Oxford [57,58]. Othen and co-workers [58] have proposed a complex transition from coherent bcc Cu precipitates to an orthorhombic 9R structure, which subsequently transforms to an fcc structure.
High resolution TEM may be able to provide further information concerning the nature of irradiation-induced features in suitably thin specimens prepared from real RPV steels.

3.2 Small Angle Neutron Scattering

The technique of small angle neutron scattering (SANS) can provide information on the size and number density of structural features within a material. SANS is a diffraction process which is sensitive to very fine-scale structural detail, and arises from fluctuations in scattering density within the sample. With this technique, it is possible to resolve features approximately 1 nm in size. However, data interpretation is extremely complex, due, in part, to the need to model the "unknown" scattering centers and to predict their effects on the SANS data, and also to the wide variety of inhomogeneously distributed microstructural features present in commercial steels (carbides, nitrides, etc.). In consequence, the use of SANS to detect irradiation-induced microstructural changes requires alternative supplementary microstructural analysis from TEM and APFIM.

Several ambitious investigations, including those performed by Buswell and colleagues, Odette and co-workers, and Kampmann et al., have successfully used SANS to assess the hardening features present in irradiated RPV steels. Specifically, SANS techniques were employed in order to determine the nature of the scattering centers in the irradiated steels, i.e., voids vs. precipitates. The volume fraction and size of voids and precipitates was reported to increase with increasing fluence. The authors note, however, that their data interpretation requires additional microstructural data from independent analytical techniques other than TEM.[9]

3.3 Atom Probe Field-Ion Microscopy

The application of atom probe field-ion microscopy (APFIM) to the characterization of neutron irradiated reactor pressure vessel steels has permitted the identification and characterization of ultrafine irradiation-induced microstructural features. APFIM permits the detailed microstructural and microchemical evaluation of materials on the atomic scale. The instrument consists of a field-ion microscope coupled with a time-of-flight mass spectrometer. There is no mass limitation for microchemical analysis; all elements from H upward can be detected. For microstructural studies, both field-ion imaging and atom probe analysis techniques are employed. These techniques rely on the processes of field ionization (for image formation) and field evaporation (for controlled removal of surface atom layers). APFIM is a suitable analytical technique for the identification and quantification of ultrafine microstructural features which are present in a number density \( \approx 10^{17} \) per cm\(^3\). This value is the effective "detectability limit" for the analysis of discrete features by APFIM. The disadvantages of this technique include the limited sampling capability (only a small amount of material is analyzed) and the time-consuming nature of the technique.

Field-ion microscopy (FIM) had been used to study defects produced during irradiation in a variety of metals [59,60] but it should be noted that this technique is not suitable for the analysis of dislocation loops which form in the material during irradiation. APFIM studies of surveillance welds have shown the existence of irradiation-induced Carich clusters with Ni and Mn enrichments.[61,62] Figure 1 shows a field-ion micrograph
of irradiated A533B weld metal containing a Cu-rich cluster.[61] Some APFIM results indicated that the clusters consisted of a Cu-rich core surrounded with a cloud of Ni and Mn atoms. In addition, APFIM analyses have provided information on the Cu content of the matrix; specifically, approximately 60% of the available Cu was associated with clusters in the irradiated surveillance material, as compared with 55% in the test reactor samples. An example of an atom probe composition profile through a Cu-rich cluster is shown in Figure 2.[63]

Importantly, the APFIM identification of Mn in the irradiation-induced clusters, and the semi-quantification of cluster chemistries has had a significant impact on SANS research in irradiation embrittlement. Previously, SANS data from irradiated steels and model alloys had been interpreted in terms of microvoids or solute-vacancy clusters, and Cu precipitates. (In SANS data analysis, a vacancy has the same signal as a Mn atom.) Therefore, SANS information on irradiated RPV steels has required re-interpretation in light of more recent APFIM results.

3.4 Positron Annihilation Lifetime Spectroscopy

Positron Annihilation Lifetime Spectroscopy (PALS) is a technique which has proven to be an effective method for investigating vacancy-type atomic defects and is particularly useful for analyzing RPV steel surveillance program specimens where only the most limited amount of material is available. Much of the recent interest in PALS is based on the technique's sensitivity to the measurement of vacancy-type defects which act as trapping centers for the positrons. PALS data can be used to accurately estimate not only the concentration of vacancy-type defects present in the material, but also the size of the defects. The key to the technique is the precise measurement of the positron lifetime within the material of interest since this lifetime is directly related to the local electron density within the material, and hence, to the presence of vacancies. Therefore, the relationship between electron density, free volume (i.e. vacancies), and positron lifetime is unique and quantifiable. An example of recent PALS data for unirradiated and irradiated modified A302B base metal is shown in Figure 3.[64] A third lifetime resulting from irradiation-induced defects can be resolved.

Ghazi-Wakili et al. [65] used positron annihilation to measure neutron irradiation effects on a high Cu weld (0.3%) irradiated in a power reactor to ~5 x 10^{17} (E>1MeV) and a medium Cu RPV forging irradiated in test reactor to 1.9 x 10^{19} n/cm². The positron data was interpreted as measuring irradiation-induced Cu precipitates which coarsened at 650°C and later dissolved at ~750°C. Lopez et al. [66] later used positrons to measure distinct recovery processes in A533B steel during isochronal annealing. In this work, positron data was interpreted as measuring irradiation-induced carbon coated microcavities (containing ~10 vacancies) and smaller vacancy-type defects for steels irradiated at 150°C to a dose of 3.5 x 10^{18} n/cm². The temperatures at which these defects annealed out corresponded well with stages of recovery in the mechanical properties. It was concluded that the recovery stages correspond directly with the disappearance of the two types of defects. For irradiations at 290°C to a dose of 2 x 10^{19} n/cm², only microvacancies with no small vacancy clusters were observed. The nonexistence of the small vacancy clusters was attributed to the increased point defect mobility at the higher irradiation temperature.
In another study by Brauer et al. [67], a number of different Soviet steels, both in the unirradiated and neutron irradiated condition, were evaluated using PALS. Brauer describes the positron annihilation results in terms of positron trapping at irradiation-induced precipitates (carbides). In addition, for irradiations at intermediate temperatures, i.e., 60 to 160°C, positron data indicated the presence of irradiation-induced vacancy clusters (voids) containing 15 or more vacancies. The size of these voids was correlated with the Cu content of the two materials studied. For higher temperature irradiations, i.e., 265 to 270°C, Brauer found that vacancy clusters were no longer present in the materials and all the positron annihilation data could be completely interpreted by the production of irradiation-induced precipitates (carbides).

4.0 Mechanism(s) of Embrittlement

The identification of mechanism(s) controlling irradiation embrittlement enables the formulation of quantitative predictive models which are based upon actual physical phenomena as opposed to models which are solely based upon statistical correlations of mechanical properties data. A variety of mechanisms have been proposed to explain embrittlement phenomena.

Most of the embrittlement mechanisms are based upon the formation of irradiation-induced defects in the material (i.e., vacancy-rich regions, micro or nanovoids, interstitial dislocation loops, point defect complexes) and on the formation of features such as precipitates or solute-rich zones which affect dislocation motion.[8,9,16,19,68-74] The complex nature of these defects is reflected in recent speculation of the existence of unstable and stable matrix defects which are formed during irradiation; the former defects, however, can "anneal out" at the irradiation temperature making detailed analyses extremely difficult.[7,75,76] It is widely accepted that Cu plays an important role in the embrittlement and hardening of RPV steels due to the formation of irradiation-induced Cu-rich features throughout the material.

The interaction of complex carbonitrides and P in the formation of intergranular films has been suggested to be a significant factor in the non-hardening embrittlement of irradiated RPV steels. The extent of this non-hardening embrittlement due to neutron irradiation is reflected in the proportion of intergranular fracture in the charpy V-notch specimens, thereby highlighting the importance of basic scanning electron microscopy (SEM) evaluations of fracture surfaces. The application of microanalytical techniques such as TEM, SANS, APFIM and PALS has provided critical data necessary for the elucidation and identification of the subtle structural changes associated with neutron irradiation of these materials.

The complex nature of the irradiation-induced defects which control embrittlement has been examined through a series of post-irradiation annealing experiments. Pachur [7] has proposed the existence of four specific "mechanisms" or defect types, each occurring over specific temperature ranges, which caused reductions in Vickers hardness. Each "mechanism" or defect type was identified by its particular activation energy (as determined via annealing experiments).

The temperature regimes over which the various "mechanisms" or "defect types" were operative were analyzed to identify the nature of the defect. This analysis suggested that "No. 3 defects" are associated with Cu, and "No. 4 defects" involve Ni and Cr. "No. 3
defects' are assumed to be responsible for the upper shelf energy decrease, and part of the increase in both yield and tensile strengths during irradiation. It has been suggested that the "No. 4 defects' become important as the fluence increases. However, there has been no direct observation of these "defect types' to date. Recent positron annihilation studies by Brauer and co-workers support the idea of an unstable defect which anneals out at the irradiation temperature. [77] Additional positron and APFIM studies would assist in the characterization of the various defect-types proposed by Pachur.

The influence of Ni and Mn concentrations was addressed by Grant et al. [62] during their structure-modelling study of surveillance weld specimens. The APFIM observations of Ni and Mn in association with the Cu-enriched clusters supports the premise that Ni and Mn increase the irradiation sensitivity of RPV steels. [61,62] The extension of APFIM studies to low fluence BWR surveillance materials has provided information on the precursory stage of clustering or "atmosphere" formation stage which has been associated with irradiation hardening.

A possible physical model of irradiation hardening/embrittlement based upon the microstructural observations involves the irradiation-enhanced nucleation of solute-rich clusters/precipitates and the subsequent growth of these features during continued irradiation. The "atmospheres" detected after low fluence irradiation and the solute-rich clusters observed in both surveillance and MTR specimens can be interpreted in terms of "precipitate" nucleation: the presence of "atmospheres" and clusters is merely a "freeze-frame" picture of the nucleation process, as illustrated in Figure 4.

Such observations cannot be duplicated by conventional thermal aging studies because the nucleation event occurs too rapidly. Recently, additional support for this mechanism has been obtained using SANS, FEG-STEM, and high resolution TEM [56]. Lattice images of bcc "features" enriched in Cu, Ni and Mn have been acquired in irradiated RPV steel. Whether these features are the APFIM "clusters or zones" [61-63,70] or true "precipitates" [10,54,56,78] is still the subject of considerable discussion. Interestingly, recent APFIM research at the University of Rouen on RPV steel irradiated to a fluence of $\sim 1 \times 10^{20}$ n/cm$^2$ (E>1 MeV) has provided additional confirmation of the existence of solute-rich clusters (with increased concentrations of Si as well as Cu, Ni and Mn) within the matrix. [79]

Several mathematical models have been developed to predict the effect of irradiation on the mechanical response (DBTT) of RPV steels. These models may be based either upon the physical changes occurring in the material due to irradiation or upon a statistical analysis of the mechanical properties and chemistry data for the irradiated RPV steel. Two such models are the Fisher et al. [73,74] and NRC Guide 1.99 [80], respectively.

The predictive mechanistic model by Fisher and colleagues [73,74] is based upon the assumption that irradiation hardening is due to the summation of displacement damage hardening and the strengthening produced by Cu precipitates. In addition, the model has been modified to include the effect of non-hardening embrittlement. This model incorporates the effect of irradiation temperature, fluence, flux, time, and Cu and P contents.

On the other hand, the NRC Guide 1.99 (rev. 2) provides a procedure for
predicting the change in the nil-ductility reference temperature, $RT_{NDT}$, based upon the Cu and Ni contents of the steel and the fluence. This model was developed from the statistical analysis of surveillance capsule data, and is applicable for irradiations at 288°C.

The general consensus is that the dominant mechanisms for hardening and embrittlement consist of a "precipitation-type" and a "damage-type" component, with a growing awareness that "non-hardening" embrittlement can play a notable role in the degradation of RPV materials.

5.0 Annealing

5.1 Overview of Annealing

Many options are available to utilities for management of vessel embrittlement including shielding of the vessel beltline region, fuel management, changes in operating procedures, improved evaluation techniques, and thermal annealing. Of these options, only thermal annealing can restore most of the original pressure vessel material toughness. Of specific concern are older light water reactors which have pressure vessels fabricated from steels with relatively low Charpy V-notch upper shelf energies. In addition, these older steels contain relatively high copper and phosphorous levels which are now known to enhance irradiation embrittlement. Not only are continually restricting operating margins a problem for these plants, it is also possible that some of these plants may subsequently fail to meet Nuclear Regulatory Commission 10CFR Part 50 Appendix G requirements for continued operations. It is these concerns, along with the possibility of plant life extension, that are driving the current interest in thermal annealing.

5.1.1 Temperature/Time Results

Studies on the effects of annealing temperature have shown that the anneal must occur at a temperature well above the irradiation temperature for any significant recovery to occur. Furthermore, annealing generally produces more marked levels of recovery in the upper-shelf energy than in the transition-temperature.

A great deal of data exists which suggests that an anneal time of 168 hours (1 week) would be sufficient for significant material toughness recovery following irradiation at 288°C. A joint EPRI/Westinghouse study [3] showed that the transition temperature recovery ("shift") for three submerged arc weldments with high Cu concentrations was between 80 and 100% and the upper shelf recovery was 100% after annealing at 454°C (850°F) for 168 hours.

Mader et al. [81] studied the effects of anneal time and temperature on intermediate Ni level weldments and a number of A533B plate type alloys. They discovered that there is an overall trend towards increased recovery with higher annealing temperatures and longer annealing times. From this work, the authors suggest that annealing involves the recovery of multiple irradiation-induced features. Each of these features requires various annealing times for recovery to occur. Specifically, during postirradiation treatment, relatively short annealing times would be required to dissolve small cascade clusters with longer times needed for the recovery of larger microvoids. Even longer times still would be required for the recovery of precipitates by both dissolution and coarsening. In addition, they suggest that annealing recovery involves at least two mechanisms.
Work by Mancuso et al. [82] on three A533B welds with different weld fluxes and levels of Cu showed that most recovery of material properties occurs within the first few hours of the anneal. Furthermore, annealing experiments at the Royal Naval College in Greenwich [83] on irradiated SA533B plate and weld showed that the majority of material property recovery occurred during the first 6 to 8 hours of the anneal at temperatures between 320°C and 370°C, with no further recovery observed for anneal times up to 336 hours.

5.1.2 Material Chemistry Effects

Pachur's investigation [6,7] of the effect of material chemistry on annealing behavior showed that four different defect types are generated during irradiation with each type annealing out at a different temperature and with a characteristic rate. The recovery of USE during annealing may be associated with a single defect type that is sensitive to Cu levels under conditions of low to intermediate fluence. At higher fluences, an additional mechanism that is sensitive to Ni may become important.

Hawthorne [84] showed that a 343°C (650°F) 168 hour anneal provided only small transition temperature recoveries in a variety of materials regardless of the level of Cu, P, or S. However, the same anneal produced full USE recovery for most of the impurity compositions looked at. In addition, a Cu content of 0.30% but not 0.16% was found to be detrimental to USE recovery for S contents ~ 0.017%. In another study, Hawthorne [85] showed that the influence of P on post irradiation heat treatment recovery is not pronounced when the Cu content is low, at least for fluences ~ 2.5 x 10^19 n/cm². In addition, high Ni content (0.68%) can be detrimental to the recovery of high Cu steels in terms of residual embrittlement and percentage recovery.

5.2 Proposed Mechanism(s)

The dramatic improvement in DBTT and recovery in USE which occurs as a result of post-irradiation annealing must be related to the fine-scale changes in microstructure. Based upon the results of an extensive annealing program for EPRI, it was proposed that the annealing process promotes the coarsening of the irradiation-induced Cu-rich features present in the microstructure. This coarsening process is diffusion-controlled and is accompanied by the dissolution of other Cu-rich features in the matrix. Lott and co-workers [86] have proposed a model based on copper precipitate coarsening that predicts the observed recovery in yield stress and DBTT for annealing temperatures between 350°C and 450°C. Their model also accounts for the low re-embrittlement rates in annealed materials. The formation of discrete Cu-rich precipitates has been previously observed in model alloys aged at 550°C [78,87,88] but these features have not yet been documented in post-irradiated annealed RPV materials. Thus, it is likely that the Cu-rich clusters and zones which have been identified via APFIM analysis will develop into discrete stable Cu-type precipitates which should be amenable to detailed microstructural and microchemical characterization. Annealing at elevated temperatures (~454°C) should also result in the elimination of most matrix damage due to irradiation. The effect of postirradiation annealing on the extent of intergranular segregation may be unclear in that temper embrittlement-type segregation may be promoted.
5.3 Applicability/Feasibility of Annealing

The restoration of material toughness through postirradiation thermal annealing treatments of reactor vessels is not without precedent. Because of a radiation sensitive material (A350-LF1, Modified) and a low operating temperature (220°C), the U.S. Army SM-1A reactor became embrittled and was annealed in 1967 using nuclear heat.[89] It was ascertained from SM-1A surveillance capsules that recovery was 61%. Approximately twenty years later, the BR-3 reactor in Mol, Belgium, with an operating temperature of ~260°C, was annealed at 330°C using nuclear heat.

Currently, there are no commercial reactors operating at temperatures below ~280°C. Therefore, with the delta between the reactor system design temperature and the reactor operating temperature becoming much smaller, the use of nuclear heat to anneal the vessel may not be economically attractive.

In addition to the two above named reactors, it has been reported that approximately fifteen full size commercial power reactor vessels of the VVER-440 type have been successfully "dry annealed" in Russia and other former COMECON countries. The use of electrical resistance heaters in Russia to supply the required heat for a thermal anneal was the first application of this dry anneal methodology. The use of electrical resistance heaters was also recommended by Westinghouse in the EPRI supported program "Feasibility of and Methodology for Thermal Annealing an Embrittled Reactor Vessel."[3] The Westinghouse study concluded that "dry" in-situ thermal annealing of an embrittled vessel using an annealing temperature of 454°C for a period of 168 hours is feasible and can be performed on most of the existing U.S. vessels without jeopardizing plant integrity.

A major design difference between the VVER and U.S. vessels is that the VVER type reactors only require annealing of the mid-core plane circumferential weldment whereas the U.S. vessels require annealing of both the circumferential and longitudinal weldments. Perhaps a more significant difference is that the design of the VVER reactor pressure vessels is such that the vessel nozzles are relatively distanced from the vessel beltline region; thus, thermal stresses at nozzle and piping locations are of little or no concern. In contrast, RPV designs outside of Russia and the COMECON countries is such that the vessel nozzles are relatively close to the mid-core plane. This design creates a concern as to nozzle and piping dimensional stability during thermal annealing at 454°C. An in-place thermal annealing study performed by EG&G for the U.S. Nuclear Regulatory Commission evaluated this potential stability problem including both vessel distortion during 454°C annealing and the extent of residual stresses imposed on the vessel after the anneal. From this work, EG&G recommended that further analytical and/or experimental studies should be performed to resolve these questions.

Unanswered questions concerning the dimensional stability/structural integrity of the vessel during the anneal has been a significant hindrance to the culmination of RPV thermal annealing in the U.S. Until it is clearly demonstrated experimentally that distortion will not occur during the anneal, no U.S. utility will choose annealing as an option for vessel embrittlement management. To answer these important questions, Westinghouse has proposed to use the nuclear steam supply system of a cancelled nuclear power plant to perform a "dry" anneal at 454°C to demonstrate the structural integrity of the vessel during the anneal. Westinghouse has also recommended a 3-D stress analysis of
stresses developed during the thermal anneal cycle complimented with actual strain gage measurements of the vessel during the anneal. The results from the actual in-vessel strain gage measurements can then be compared with the results from the 3-D stress analysis. Westinghouse is currently soliciting $4 million in funding for this effort.

5.4 Economic Impact

The use of a thermal heat treatment to recover mechanical properties degraded by neutron radiation exposure has been shown by many investigators to be a technically feasible method for assuring RPV compliance with regulatory and license renewal requirements. The utilities decision to thermal anneal any vessel will be based on economic advantages for long-term plant operability; economic performance improvement is currently a major industry initiative in the U.S. Thus, the value of annealing will be examined on a plant specific basis rather than on a general industry-wide basis.

While those who advocate the use of thermal annealing push for a full-scale demonstration of the methodology, others propose mitigative options for controlling vessel embrittlement. These options include flux reduction either by fuel management or by shielding of the reactor vessel wall. However, it should be made clear that flux reduction options also have an economic impact on the continued operation of a commercial nuclear power plant. For example, fuel management schemes require removing fuel rods from the perimeter of the core and replacing the fuel rods with "dummy" rods. These fuel management schemes are expensive because of reduced plant megawatt thermal capacity and/or costs associated with increased fuel shuffling. Perhaps more importantly, flux reduction options are only of value early on in plant life prior to embrittlement of the vessel. Once the vessel has become embrittled, thermal annealing is the only method capable of recovering material properties.

EPRI has developed software to perform economic scoping studies for evaluating embrittlement mitigation options. This software package, called VTester, permits a utility to perform initial cost and benefit analyses for a given strategy. EPRI, as well as small individual utilities, have utilized VTester for plant specific evaluation. The results show that the value of thermal annealing can vary depending on the level of vessel embrittlement and the remaining years of plant operation. As one would expect, thermal annealing is of high value for plants approaching or expecting to approach the PTS criteria limit. However, as mentioned previously, thermal annealing is not always the most economic option compared with other alternatives for plants operating earlier in plant life. In cases where plants are considering life extension, the economic benefits of thermal annealing becomes even more attractive.

In 1982, EPRI estimated that a thermal anneal of a reactor pressure vessel would cost between $30 to $60 million. In 1992, Westinghouse estimated the cost of thermal annealing a vessel to be within a range of $12 to $15 million. Today, based on modern technology, Westinghouse estimates the basic cost of the anneal as approximately $5 to $7 million. However, associated operational and qualification aspects may result in a total process cost of $8 to $10 million per vessel. Even though the annealing cost estimates have dropped drastically since the early 1980's, it is most likely that utilities will not commit to an anneal of their vessels in the absence of a prior full-scale annealing

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demonstration to show that the structural integrity of the vessel will not be impaired. The cost of performing such a full-scale annealing demonstration on an actual unirradiated vessel will be $4 million.

6.0 Summary/Recommendations

Irradiation embrittlement of the reactor pressure vessel is a major concern for the nuclear industry because it can severely restrict plant operating margins in addition to causing the plant to fail to meet regulatory compliance. Each reactor pressure vessel is, however, somewhat unique because of effects of various irradiation conditions such as irradiation temperature, neutron fluence, flux, and material composition on embrittlement behavior.

The irradiation sensitivity and thermal annealing behavior of reactor pressure vessel materials have been assessed by many investigators. Research programs conducted in the USA, Russia, and Germany have demonstrated the ability of thermal annealing to recovery the material properties of a variety of irradiated steels and weldments. Perhaps just as importantly, thermal annealing is a technically feasible method which will assure vessel compliance with regulatory licensing rules and therefore permit license renewal.

The Soviets and other countries in eastern Europe have performed "dry anneals" at higher temperatures (454°C) than design temperatures (343°C) for a number of commercial vessels. Under the sponsorship of EPRI, Westinghouse clearly demonstrated the benefits of annealing in terms of mechanical property recovery and developed a methodology for thermal annealing an embrittled vessel.

Westinghouse has concluded that it is technically feasible to thermally anneal a vessel designed to USA standards however, each reactor pressure vessel is somewhat unique. Therefore, thermal annealing should be "tailored" (plant-specific) rather than generic. This tailoring requires a better understanding of the mechanism(s) of irradiation embrittlement as well as the mechanism(s) of mechanical property recovery.

The identification of chemical and physical changes occurring in the material is vital in that it is the microstructure of the material which controls the observed mechanical properties. The major analytical techniques such as transmission and scanning electron microscopy (TEM, STEM), small angle neutron scattering (SANS), atom probe field-ion microscopy (APFIM), and positron annihilation lifetime spectroscopy (PALS) are available to develop a better understanding of the mechanism(s) of irradiation embrittlement and recovery phenomena.

EPRI has developed software to perform economic scoping analyses. The software package called VTester was used by Griesbach and Server to show that thermal annealing of an embrittled vessel is of economic benefit.[90] The results show that the value of thermal annealing can vary depending on the level of vessel embrittlement.

Plans are underway for Westinghouse to perform a thermal annealing demonstration on a unirradiated reactor pressure vessel to establish the dimensional stability of the vessel during the anneal. The cost of such a demonstration is estimated as $4 million while the cost of thermally annealing an irradiated vessel is estimated at $10 million.

It is recommended that 1. Systematic studies be continued and/or initiated to develop a better understanding of irradiation embrittlement and recovery phenomenon, and 2. Support be given to the full-scale demonstration planned by Westinghouse in the USA.
Figure 1. Field-ion micrograph of irradiated A533B weld metal from HB Robinson II containing a Cu-rich cluster (arrowed). [61]
Figure 2. Atom Probe composition profile through a Cu-rich cluster in an irradiated A533B weld [63]
Figure 3. Schematic representation of PALS results for modified A302B base metal.[64]
MICROSTRUCTURAL DEVELOPMENT DURING NEUTRON IRRADIATION OF RPV STEELS

Increasing Exposure

Solute-Rich Atmospheres  Clusters  Precipitates

Figure 4. Schematic diagram of the transition from "atmosphere" to precipitate during neutron radiation based upon APFIM data.[71]
References


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Preprint

ANALYSIS OF MECHANICAL TENSILE PROPERTIES OF IRRADIATED AND ANNEALED RPV WELD OVERLAY CLADDING

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ANALYSIS OF MECHANICAL TENSILE PROPERTIES OF IRRADIATED AND ANNEALED RPV WELD OVERLAY CLADDING

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ABSTRACT. Mechanical tensile properties of irradiated and annealed outer layer of RPV weld overlay cladding, constituted by Cr19Ni10Nb alloy (Sv08Ch19N10G2B), were experimentally determined by usual method using laboratory specimens and by nondestructive technique, indentation testing, on RPV's. Experimental results were analyzed within the constitutive properties homogenization framework.

Keywords: pressure vessel, cladding, mechanical properties, analysis

1. Introduction

Including the effect of weld overlay cladding properties into the analysis of both the RPV residual life and RPV defects acceptability requires, among others, determination of weld overlay cladding mechanical tensile properties which take into account both technology of cladding deposition (and heat treatment) and service conditions at certain location in RPV under consideration.

In the first part of this paper, the results of experimental determination of mechanical tensile properties of RPV weld overlay cladding obtained by two different methods are described and mutually compared:

Fundamental information concerning "typical" mechanical tensile properties of weld overlay cladding of VVER-pressure vessel (Cr19Ni10Nb) was obtained by conventional tensile testing of specimens in initial state as well as specimens irradiated in VVER-440, using standard technology of surveillance specimens irradiation. Special attention was given to the influence of subsequent annealing under conditions identical with the accepted regime of RPV-annealing. Further, special method for indentation testing of the RPV weld overlay cladding and evaluation of indentation diagrams was used. This method permits to obtain "specific" mechanical tensile properties of weld overlay cladding, which respect influence of both technology of cladding deposition and service conditions at certain location.

Experience obtained by measurements at the first and second unit of NPP V-1 at Jaslovské Bohunice, Slovakia, has shown that mechanical tensile properties are affected by both technological factors (i.e. initial properties of weld overlay cladding) and service conditions (neutron fluence, annealing of the RPV). The influence of the cladding deposition technology on mechanical tensile properties of cladding may be caused by differences in chemical composition of welding filler metal (in range given by material specification) or by differences in deposition
regime. An important resulting structural parameter, ferrite
volume ratio, varies in certain limits.
In the second part of this paper, analysis of mechanical
tensile properties of RPV weld overlay cladding is carried
out:
For the purpose of the analysis of ferrite volume ratio
influence on mechanical tensile (constitutive) properties of
weld overlay cladding two theoretical models of constitutive
properties homogenization for elastic-plastic
matrix-inclusion composites were chosen, evaluated and
applied. Properties of these models were tested on a more
wide set of materials with similar properties - the
appropriate analysis was performed also for cast
austenitic-ferritic materials with coarser two-phase
structure.

2. Experimental determination of mechanical tensile
properties of RPV weld overlay cladding

The outer layer of the weld overlay cladding of the
VVER-440 RPV is constituted by the Cr19Ni10Nb alloy (for the
filler metal the Russian designation Sv08Ch19Ni10G2B - in
latin transcription - is used). Information about chemical
composition, ferrite volume ratio and fineness of ferrite
distribution is given in table 1.

<table>
<thead>
<tr>
<th>material</th>
<th>chemical composition [wt. %]</th>
<th>a)</th>
<th>b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cr19Ni10Nb</td>
<td>C 0.08, Mn 1.8+, Si 0.2+, Cr 18.5+, Ni 9.5+, Mo -</td>
<td>0.9+</td>
<td>6</td>
</tr>
<tr>
<td></td>
<td>Nb 2.2, Fe 0.45, Nb 20.5, Ni 10.5, Fe 1.3</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a) ferrite volume fraction [%], measured
b) ferrite particle size [μm]

Mechanical tensile properties of weld overlay cladding were
determined by conventional tensile testing of specimens
prepared from thick cladding obtained in laboratory by
repeated weld layer deposition. These specimens were tested
1) in initial state (i.e. after stress-relief annealing
665°C/43h), 2) after subsequent annealing 475°C/168h, 3)
after irradiation, 4) after irradiation and subsequent
annealing 475°C/168h. Irradiation was performed in VVER-440,
using standard technology of surveillance specimens
irradiation. Conventional mechanical characteristics Rm,0.2,
Rm and A m, together with information about irradiation
conditions in case 3) and 4), are summarized in table 2.
These data are designated as "typical" mechanical tensile
properties of cladding corresponding to the selected
"typical" histories.
### Tab. 2.
Data obtained from the experimental study of the weld overlay cladding [1]

<table>
<thead>
<tr>
<th>History</th>
<th>(R_s) [MPa]</th>
<th>(R_{0.2}) [MPa]</th>
<th>(A_r) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(initial state)</td>
<td>604±20</td>
<td>415±25</td>
<td>29.7±4.6</td>
</tr>
<tr>
<td>Regeneration annealing</td>
<td>613±15</td>
<td>434±7</td>
<td>27.0±1.8</td>
</tr>
<tr>
<td>(3.5\times10^{23} \text{n/m}^2 ) (E&gt;0.5 MeV) in 308 days + regeneration annealing</td>
<td>623±14</td>
<td>451±6</td>
<td>29.2±1.8</td>
</tr>
<tr>
<td>(1.1\times10^{23} \text{n/m}^2 ) (E&gt;0.5 MeV) in 308 days, without annealing</td>
<td>604±33</td>
<td>475±6</td>
<td>17.3±6.8</td>
</tr>
</tbody>
</table>

The more representative "specific" mechanical tensile properties of weld overlay cladding, which result from the combination of individual filler metal batch, cladding deposition regime used at certain location and local history (technological annealing after cladding deposition, neutron fluence, annealing of the RPV), were deduced by indentation testing of the RPV weld overlay cladding and evaluation of indentation diagrams.

For indentation testing, a special device was developed and used. This device is equipped by the last type of Remote Hardness Tester, developed by Scientific and Research Institute for NPP’s Operation, Moscow, Russia, with spherical indentor. The Fixation and Fine Positionning Module, permitting measurements inside RPV, was designed and realized by Nuclear Research Institute (NRI), Ústí, Czech Republic. Method of evaluation of indentation diagrams was developed by NRI; this method was not described until now, therefore we inform about its essence in this paper (the method will be described in more details with supporting arguments elsewhere).

Term "indentation diagram" is used for the experimentally obtained curve (or data file) indentation force \(F\) - indentation depth under load \(t\, F=F(t)\) or \(t=t(F)\). With the diameter \(D\) of the spherical indentor, the simple formula \(d_{nom}=2\left[t*(D-t)\right]^{1/2}\) permits to obtain the nominal indentation diameter \(d_{nom}\). For evaluation of indentation diagrams, it is important to obtain the true indentation diameter \(d\); this is based on empirical calibration which gives, in certain important interval of \(d/D\)-values, the proportionality relation \(d_{nom}=K*d\). As a result of this calibration, Meyer hardness \(HM=4*F/(\pi*d^2)\) is obtained as a function of the ratio \(d/D\). Meyer hardness shows interesting properties, which permit simple calculation of certain section of the true stress-strain diagram \(\sigma=\sigma(e)\); these properties are usually described as properties of "hardness - flow stress ratio" \(HM/\sigma\) (called also "constraint factor" or "normalized indentation pressure"). Meyer hardness \(HM=HM(d/D)\) and true stress \(\sigma=\sigma(e)\) are related with the use of "mean strain under the spherical indentor", defined as \(e=0.2*(d/D)\). The key feature of \(HM/\sigma=CF\) is the following one: in certain interval of \(d/D\), usually at least for 0.4≤\(d/D\)≤0.5, the approximate equality \(CF=3.0\) is valid, for both austenitic and ferritic-carbidic steels (for recent information about properties of CF see [2], [3]). Extrapolation of \(\sigma=\sigma(e)\) to lower and higher strain values and the subsequent analytic
determination of yield stress, tensile strength and uniform strain (or uniform elongation) is based on the following observation: For austenitic-ferritic welds and weld overlay claddings, the plastic part of \( \sigma = \sigma(\varepsilon) \) can be approximately described as composed of two parts: 1) linear, 2) power law, with continuous tangent \((d\sigma/d\varepsilon)\) in the transition point. The 3 parameters defining the theoretically possible true stress-strain relations are not independent in the class of materials under consideration, i.e. the confinement condition between them exists; this condition is based on results of tensile tests performed with the class of materials under consideration. With confinement condition, only 2 independent parameters are necessary for characterization of the complete stress-strain curve; for determination of these 2 parameters, 2 values of true stress obtained in the region of constant CF are sufficient.

Indentation tests performed on 1. and 2. unit of NPP V-1 at Jaslovské Bohunice, Slovakia, before and after annealing of RPV's, reveal mechanical tensile properties of weld overlay cladding affected by both continuous range of histories (height profile of neutron fluence corresponding to the geometry of the active zone and another height profile of annealing temperature) and local technological factors. Due to the continuous range of histories, only global comparison with data in table 2 is possible; this comparison is shown on figures 1 and 2 (1 experimental point represents 3+5 indentation tests). The scatter of experimental values represents practically the true scatter of mechanical properties, because the measurements show existence of areas on the RPV wall with extremal properties consistently both before and after annealing of the RPV. We consider the inhomogeneity of ferrite volume ratio to be a possible source of the scatter of mechanical properties; estimation of the ferrite volume ratio with the use of DeLong diagram [4], based on chemical composition limits given in table 1, gives the range \((2+12)\%\). For verification of this hypothesis and for the assessment of the maximal possible effect of thermal ageing of the ferritic phase on the mechanical tensile properties of weld overlay cladding after annealing, the analysis of mechanical tensile properties of weld overlay cladding was performed. We consider the problem as a special case of the homogenization of constitutive properties of elastic-plastic matrix-inclusion composites.

3. Modelling of constitutive properties of weld overlay cladding

Several basic types of theoretical models for elastic-plastic properties homogenization exist, but few if any critical evaluations have been presented. Recently, a study was performed [5] with the aim to contribute to critical evaluation of the basic types of these models and to show results obtained by its use in the important technological area: analysis of thermal ageing of two-phase stainless steel castings and welds or weld overlay claddings. In this paper, we summarize main results of study [5], related to weld overlay cladding:

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Two models [6] and [7] of homogenization of constitutive properties of system with one phase dispersed in other polycrystalline phase were chosen and mutually compared. From both versions of the model [7] only the second version ("finite" formulation) is acceptable; this version characterizes constitutive properties of phases by deformation theory (i.e. the generalization for elastic-plastic properties consists of substitution of elastic moduli by secant moduli).

Homogenization models are classical continuum mechanical ones and, consequently, they do not describe the influence of structural dimension of the modeled system dispersed phase - matrix on its constitutive properties. This approach ceases to correspond to reality, if the characteristic length in two-phase structure is not significantly greater than the characteristic length on the micromechanical level (geometric slip distance or characteristic size of the dislocation source of the Frank-Read type). The micromechanical size effect may be manifested at comparison of properties of castings (with sufficiently coarse structure) and welds or weld overlay claddings (with finer structure). Data [8] used for experimental verification of the model [6] show that the yield stress of both one-phase (austenite or ferrite) and two-phase (austenite and ferrite) alloys Fe-Cr-Ni depend on grain size, or phase particle size: the Petch relation

$$\sigma_y = (\sigma_y)_0 + Kd^{-1/2}$$

is valid with value of $K$ universal for different alloys and for different degree of thermal ageing of ferrite (for definition of $d$ for two-phase alloys see [8]). Considering the relationship between the models of constitutive properties of polycrystalline materials and of two-phase composites, we may suppose that the dependence of the yield stress of the dispersed phase on the particle size complies the relation (1). A difficult problem arises with quantification of effective constitutive properties of the matrix which is forced to flow round less deformable small inclusions, when the composite is undergoing deformation. It seems optimal to determine the effective yield stress of the matrix using experimental measurement of the yield stress of the composite - at small strains the less deformable inclusions deform only elastically and, in the ideal case of sharp yield stress and identical elastic properties of both inclusions and matrix, the matrix yield stress is equal to composite yield stress. Increment of the yield stress of the matrix caused by size effect is not, of course, dependent on the degree of thermal ageing of the dispersed phase.

Application of both models requires calculation by a computer program. For Weng's model, the algorithm is described in detail in [6]; in an analogous manner a computer program was written and run for Hervé and Zaoût's model [7]. The loading curve consists of three parts: in the first part both phases deform only elastically, in the second part the matrix deforms also plastically, and in the third part both phases deform both elastically and plastically. In the second and third part of loading curve the values of matrix plastic strain are
prescribed; in the second part the individual values of homogenized stress and strain are determined without iterations, while in the third part iterations are needed. Constitutive properties of both austenitic and (aged) ferritic phase may be taken - in the zeroth approximation - from [8]; values of parameters correspond to austenite and ferrite, respectively, in system Fe-Cr-Ni with very low content of other components and with grain size (19+20)μm. The constitutive equation for plastic strain has a form

$$\sigma = \sigma_0 + h\cdot (e_p)^n$$  \hspace{1cm} (2)

Values of parameters \( \sigma_0, h, n, E \) and \( v \) are given in tab. 3 (values \( E \) and \( v \) are taken from [6]).

<table>
<thead>
<tr>
<th>phase</th>
<th>( E [\text{GPa}] )</th>
<th>( \nu )</th>
<th>( \sigma_0 [\text{MPa}] )</th>
<th>( h [\text{MPa}] )</th>
<th>( n )</th>
</tr>
</thead>
<tbody>
<tr>
<td>austenite</td>
<td>193</td>
<td>0.308</td>
<td>137</td>
<td>617</td>
<td>0.60</td>
</tr>
<tr>
<td>ferrite</td>
<td>207</td>
<td>0.293</td>
<td>1107</td>
<td>382</td>
<td>0.43</td>
</tr>
</tbody>
</table>

Taking into account the ferrite particle size in weld overlay cladding (see tab. 1), it is evident that it is necessary to respect influence of the size effect on constitutive properties. Reduction of dispersed particle size from 20μm to 1μm gives, with the aid of data [8] and relation (1), the size effect with dispersed phase yield stress \( \Delta \sigma_y = 300 \text{MPa} \).

Comparison of yield stress of weld overlay cladding (values given in tab. 2) with yield stress of austenitic phase according to table 3 shows the influence of solid solution hardening, dispersion hardening by carbide particles and size effect of ferritic inclusions. Probably it may be expected as well that the yield stress of the aged ferrite of "high purity" from table 3 will not represent sufficiently truly the properties of hardened ferritic phase in individual materials, even if we include, for weld overlay cladding, the above mentioned modification of yield stress with respect to the size of dispersed ferrite particles.

What concerns the deformation hardening, we suppose that it is independent of chemical composition of phases or of the degree of ageing; therefore we will represent deformation hardening of both phases by values of \( h \) and \( n \) (in eq. (2)) given in table 3. This corresponds to generalized experience; an analogous procedure was used in work [9] at prediction of homogenized constitutive properties of ferritic-martensitic steels. For specific conditions the matrix yield stress was modified, taking into account the grain size and the carbon content in solid solution, while \( h \) and \( n \) were supposed to be independent of these parameters.

(Of course, \( E \) and \( v \) for austenite and ferrite are also taken as independent of chemical composition or ageing.)

In the first series of calculations, the individual values of matrix yield stress (see table 2) together with universal
values of $E$, $\nu$, $h$ and $n$ of both phases, and appropriate value of dispersed phase yield stress according to table 3 were used as input parameters characterizing properties of both phases. In the case of weld overlay cladding, the modification of dispersed phase yield stress for particle size, $4\sigma_0=300\text{MPa}$, was used. In the second series of calculations, the dispersed phase yield stress was increased by $600\text{MPa}$ (besides the modification of the dispersed phase yield stress in the case of weld overlay cladding taking into account the particle size); the value of the ferrite yield stress represents realistic assessment of possible hardening of ferrite with respect to the really reached values for ferrite with controlled inhomogeneities [10]. Results of both series of calculations and corresponding experimental data are summarized in table 4. Figures 3 and 4 show calculated behaviour of nominal stress in dependence on strain for both alternative hypotheses concerning yield stress of the dispersed phase, together with experimental data. Results for both models [6] and [7] are shown.

Tab. 4. Comparison of predictions based on Weng's model [6] and Hervé-Zaoui's model [7] with experiments for weld overlay cladding

<table>
<thead>
<tr>
<th>material</th>
<th>Cr19Ni10Nb</th>
</tr>
</thead>
<tbody>
<tr>
<td>ferrite vol. ratio [%]</td>
<td>6</td>
</tr>
<tr>
<td>history</td>
<td>c)</td>
</tr>
<tr>
<td>yield stress [MPa]</td>
<td>434</td>
</tr>
<tr>
<td>calculation a) Weng</td>
<td>$R_m$ [MPa] 591.9</td>
</tr>
<tr>
<td></td>
<td>$A_m$ [%] 21.19</td>
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<tr>
<td>calculation a) Hervé-Zaoui</td>
<td>$R_m$ [MPa] 592.0</td>
</tr>
<tr>
<td></td>
<td>$A_m$ [%] 21.14</td>
</tr>
<tr>
<td>calculation b) Weng</td>
<td>$R_m$ [MPa] 601.4</td>
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<tr>
<td></td>
<td>$A_m$ [%] 26.12</td>
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<td>calculation b) Hervé-Zaoui</td>
<td>$R_m$ [MPa] 602.6</td>
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<tr>
<td></td>
<td>$A_m$ [%] 26.15</td>
</tr>
<tr>
<td>experiment</td>
<td>$R_m$ [MPa] 613±15</td>
</tr>
<tr>
<td></td>
<td>$A_m$ [%] 27.0±1.8</td>
</tr>
</tbody>
</table>

a) 1. series of calculations - see text
b) 2. series of calculations - see text
c) specific history without irradiation - see text
d) specific history with irradiation - see text

4. Discussion

Results given in table 4 show satisfactory accordance between experiments and model calculations of both tensile strength and uniform strain for both histories including final annealing $475^\circ/168$h. This accordance is obtained even if it may be questioned if the model of ferrite inclusions in austenitic matrix characterizes sufficiently truly the topology of phases. Both models give practically the same results, due to the low ferrite volume ratio in the case studied. Calculations also demonstrated that modifications of annealing conditions, leading eventually to more
pronounced hardening of ferrite, cannot significantly affect mechanical tensile properties of weld overlay cladding. Sensitivity study shows that, with constant yield stress, ferrite volume ratio increment of 1% causes tensile strength increment of 1%. In view of the sensitivity of ferrite volume ratio on alloy chemical composition, this may explain scatter of the tensile strength observed for (practically) uniform history. Mechanical tensile properties after neutron irradiation, without subsequent annealing, are characterized by lower uniform strain; in terms of homogenization models this fact is caused by lower strain hardening of austenitic matrix (strain hardening parameters given in table 3 cannot be used in this case). Heterogeneous two-phase structure affects not only constitutive (or mechanical tensile) properties, but also ductile fracture. Comparison of "specific" mechanical tensile properties of weld overlay cladding with "typical" ones gives information about "specific" structural properties (ferrite volume ratio) at certain location in the RPV under consideration. After determination of "specific" mechanical tensile properties and "specific" structural properties, prediction of "specific" fracture-mechanical properties may follow.

Conclusions

Both models of constitutive properties homogenization for elastic-plastic matrix-inclusion composites are suitable for simple prediction of stress-strain relation (and consequently of related values of ultimate tensile strength and uniform elongation), corresponding to the state after saturation of thermal ageing of stainless steel weld overlay cladding. In this case, the only individual input parameters are both yield stress of austenitic matrix and volume ratio of the dispersed phase. Other input parameters (elastic properties of both phases, yield stress of dispersed phase, parameters describing strain hardening of both phases) are universal.

In view of both the verification of the models and the specification which parameters of constitutive equations describing properties of individual phases are structure sensitive and which are not, we may suppose that the analysis may be applied also to the weld overlay cladding of different chemical composition. Mechanical tensile properties after neutron irradiation can be modelled in an analogous way, but all constitutive properties of phases as input parameters are not at disposal in this case.

References


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Fig. 2. Experimental values of tensile strength versus uniform strain, determined by tensile tests and deduced from indentation tests, for different histories.

Fig. 3. Dependence of nominal stress $R$ on strain $\varepsilon$. 1,2...1. series of calculations, see text
3,4...2. series of calculations, see text
Mean values of tensile strength and of uniform strain found experimentally for specific history without irradiation.

Fig. 4. Dependence of nominal stress $R$ on strain $\varepsilon$. 1,2...1. series of calculations, see text
3,4...2. series of calculations, see text
Mean values of tensile strength and of uniform strain found experimentally for specific history with irradiation.
yield stress vers. uniform strain
all histories

```
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0.05
0
100 200 300 400 500 600 700

yield stress [MPa]
```

- lab.-hist.1
- lab.-hist.2
- lab.-hist.3
- lab.-hist.4
- V-1 2.unit
- V-1 1.unit

Fig. 1.

tensile strength vers. uniform strain
all histories

```
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
100 200 300 400 500 600 700

tensile strength [MPa]
```

- lab.-hist.1
- lab.-hist.2
- lab.-hist.3
- lab.-hist.4
- V-1 2.unit
- V-1 1.unit

Fig. 2.
R vers. eps
weld overlay cladding

nomin. stress [MPa]

--- b) Weng --- a) Weng * experiment
--- b) Her-Za --- a) Her-Za

Fig. 3.

R vers. eps
weld overlay cladding

nomin. stress [MPa]

--- b) Weng --- a) Weng * experiment
--- b) Her-Za --- a) Her-Za

Fig. 4.
Post-irradiation annealing of coarse-grained model alloys

C.A. English
P.H.N. Ray, C. Wilson and R.J. McElroy

AEA Technology,
Reactor Services,
Harwell, Didcot, Oxon OX11 ORA, U.K.

As part of the UK contribution to the Phase 3 IAEA Reactor Pressure Vessel Embrittlement Co-ordinated Research Programme thermal ageing and irradiation studies have been carried out on three of the model alloys (JPC, JPB, JPG). The composition of these alloys is identical except for differing levels of phosphorus and/or copper. They have been irradiated in three conditions, as-received, heat treated to produce a coarse grained microstructure, and in this condition further aged at 450°C to produce a temper embrittled condition. This coarse grained microstructure is similar to that often encountered in weld heat-affected zones (HAZ's). Further, one alloy JPC with copper of 0.14wt% and 0.017 wt%P has been subject to a post-irradiation anneal (see below). The effect of these treatments on mechanical property changes has been characterised by Charpy testing and Vickers hardness measurements, and in addition the phosphorus segregation has been studied by a combination of STEM and Auger techniques.

All three materials exhibit a severe deterioration in Charpy impact properties after thermal ageing at temperatures around 450°C, the $\Delta T_{40J}$ increased by in the order of 100°C. This has been shown to be a consequence of the familiar temper embrittlement phenomenon and is due primarily to the thermally induced segregation of phosphorus to prior austenite grain boundaries, producing a transition from cleavage to intergranular fracture. A slightly higher increase was found in the alloy containing both copper and high phosphorus, suggesting a possible synergistic effect of copper and phosphorus.

The effect of irradiation was as expected to create an increase in ductile to brittle transition temperature and the hardness. The most noticeable effect was the increased embrittlement in the Copper containing alloy.

A pilot post-irradiation annealing programme was carried out on the copper, high phosphorus containing alloy, JPC, with the coarse-grained microstructure to understand the effect of annealing on simulated HAZ microstructure. For this pilot study it was decided to employ the highest annealing temperature (475°C) and longest time (1 week) used in actual RPV anneals. In addition to Charpy tests on reconstituted specimens, hardness was used to determine the level of irradiation hardening recovery, since this is common practice in RPV annealing.

The hardness tests indicated that, as expected, complete recovery of irradiation hardening was achieved by the anneal. Charpy tests, however, indicated a further
increase in transition temperature, such that the transition temperature shift due to irradiation, 69°C, was more than doubled to 155°C after post-irradiation annealing. The latter embrittlement has been shown to be consistent with a segregation model developed for temper embrittlement of similar materials due to phosphorus segregation and intergranular failure. Fractographic and microstructural studies are proposed to test this conclusion.

In addition the phosphorus segregation was measured by STEM and Auger in all conditions and it was shown that small increases in phosphorus segregation occurred in the coarse-grained microstructures after irradiation. It was possible to obtain good agreement between the measured segregation and the predictions of models which described segregation under either thermal or irradiation conditions.

A full report on this work is contained in AERE Report AEA -RS-2426.
COMISION NACIONAL DE ENERGIA ATOMICA
BUENOS AIRES
ARGENTINA

ATUCHA I NUCLEAR POWER PLANT SURVEILLANCE PROGRAMME

Dario JINCHUK

To be presented at the Specialists Meeting on

IRRADIATION EMBRITTLEMENT
AND
OPTIMIZATION OF ANNEALING

Paris, FRANCE-20-23 September 1993
ATUCHA I NUCLEAR POWER PLANT SURVEILLANCE PROGRAMME
D.JINCHUK
CNEA, BUENOS AIRES (1429) ARGENTINA

ABSTRACT

Mechanical properties of steels employed in reactor pressure vessels (RPV) change with neutron irradiation. The main macroscopic effects are a decrease in the material toughness and an increase in the brittle-ductile transition temperature, with the consequent increase in the probability of a sudden crack propagation in a brittle fracture mode.

To monitor these changes a set of capsules, containing specimens made with the same material of the RPV, are placed in certain positions inside the reactor with an adequate lead factor compared with the fluence received by the RPV inner wall.

With the periodic testing of probes contained in the capsules during reactor lifetime, a complete and advance knowledge of RPV material behaviour is obtained.

In order to monitor Atucha I RPV embrittlement a representative coupon (2400 x 900 mm) was welded with the most sensitive plates employed in the PV construction. From this coupon, 60 tensile specimens, 138 impact and 52 fracture mechanic samples were cut and placed in specially designed capsules, including temperature and radiation monitors.

This paper describes the Atucha I Surveillance Programme and results obtained during testing of the second set of capsules.

1- INTRODUCTION

Atucha I is a 370 Mw Nuclear Power Plant located 100 km. North of Buenos Aires city, connected to the grid in 1974 (Fig. 1).

The reactor is a PHWR type employing natural uranium as fuel, cooled and moderated with heavy water.
The core contains 253 fuel elements each one placed in a coolant channel, the refuelling is performed on line at an average of 1.3 fuel elements per FPD.

The primary circuit is a 2 loops system with one main pump and one U tube vertical steam generator in each loop. (Fig. 2).

The reactor is a Pressure Vessel type with an inner stainless steel tank, containing the heavy water moderator, traversed vertically by the coolant channels, welded through ports to the closure head to enable the on line refuelling. The coolant media circulates from the bottom to the top of the coolant channels in a system isolated from the moderator loop. (Fig. 3).

The main Pressure Vessel Characteristics are:

**ATUCHA 1**

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Height (cm)</td>
<td>1216</td>
</tr>
<tr>
<td>Ext. diam. (cm)</td>
<td>620</td>
</tr>
<tr>
<td>Wall thickness (cm)</td>
<td>22</td>
</tr>
<tr>
<td>Weight (Ton)</td>
<td>470</td>
</tr>
<tr>
<td>Oper. Temp. (oC)</td>
<td>290</td>
</tr>
<tr>
<td>Int. Pressure (Atm)</td>
<td>115</td>
</tr>
</tbody>
</table>

The PV is built from a low alloy ferritic steel 22 NiMoCr37, with improved composition for low sensitivity to neutron irradiation. The inner wall is cladded with 5 mm of austenitic steel 1-4541 (similar to AISI 347) applied with automatic welding technics. Table 1 show typical material composition.
TABLE I

CNA 1 PRESSURE VESSELS MATERIALS COMPOSITION

<table>
<thead>
<tr>
<th>Composition</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Cu</th>
<th>Mo</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>%</td>
<td>0.2</td>
<td>0.2</td>
<td>0.7</td>
<td>0.016</td>
<td>0.012</td>
<td>0.4</td>
<td>0.12</td>
<td>0.63</td>
<td>0.8</td>
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</table>

The special design of the Atucha RPV, with cooling channels welded to the closure head, and the existence of a moderator tank filled with heavy water, with a high tritium content, makes the closure head removal an almost impossible task.

In consequence the plant designer placed surveillance capsules at the bottom end of the coolant channels (Fig. 4).

2.- SURVEILLANCE CAPSULES

General requirements of the Surveillance Programme are in accordance with ASTM E-185. A representative coupon was welded with the most sensitive plates to radiation embrittlement, taking into account the Phosphorus and Copper content (Fig. 5-6).

From this coupon 60 tensile specimens, 138 Charpy impact and 52 WOL-1X fracture mechanics samples were cut from the base material, heat affected zone and welding area, and placed in specially designed capsules (Fig. 7 and 8). Capsules include Nb, 02Th and Fe as dose monitors. [2]
The 30 resulting capsules were located in the coolant channels under the fuel elements core. This position is certainly not the most appropriate but the only one accessible due to the particular characteristics of this plant.

The first set of 15 capsules, representing 50% of full power life, was removed from the reactor in March 1980 and the second set (>100% FPL) in August 1987.

3. IRRADIATION CONDITIONS

Atucha 1 reactor may be considered basically as stationary, this means that the neutron flux in one location per unit of thermal power (n/cm² seg/Mw) can be considered practically constant in time. This occurs mainly because in the online refuelling process only a small amount of fresh fuel element is introduced in the core.

With this approach neutron fluence, at the end of a certain period, may be obtained knowing neutron flux and full power days (fpd) from:

\[ \Phi (n/cm^2) = \Phi (n/cm^2 \ seg)* fpd (days) * 66400 (seg/days) \]  

Unfortunately, due to the special position where the surveillance capsules were located, the neutron spectrum in the samples differs greatly as compared with the RPV inner wall (Fig. 4).

At the surveillance position the contribution of fast neutrons (E>1 MeV) to the total fluence is lower than 0.1% (Fig. 9).

Another disadvantage produced due to the location of capsules below the core is a strong flux axial gradient in each capsule.

To take into account this effect the capsule was divided in three zones: upper, medium and lower, and only samples from each zone were considered as being irradiated by the same flux.
4. - TESTING

Capsule holders were dismantled in the spent fuel elements pool at Atucha, and the closed capsules sent to CNEA Ezeiza Atomic Centre hot cells for opening and testing.

Charpy impact tests were conducted in a 300 J Wolpert pendulum according to DIN 50 115. Samples heating and cooling was performed in a specially designed environmental chamber to cover the range-100 C to 3000C in order to have a full ductil-brittle transition curve. Heating was produced by electric resistance and cooling by blowing liquid nitrogen into the chamber. Temperatures were measured by a thermocouple in contact with the notch of the Charpy specimen.[3]

An automatic feeder was employed to position samples in the pendulum anvil.

Tensile tests were performed, according to DIN 50145, and fractomecanic, according to ASTM E399-81, employing a 10 Tn MTS servohydraulic machine. The desired testing temperature was reached employing an environmental chamber in the range-100 to 3000C.

5. - RESULTS AND DISCUSSION

As mentioned before, capsules were divided in three zones, the displacement per atom (dpa) calculated for the Charpy notch in each zone were:

upper: 11,3 \times 10^{-2} \text{ dpa}
medium: 7,7 \times 10^{-2} \text{ dpa}
lower: 4,6 \times 10^{-2} \text{ dpa}

Table 2 show typical results for Charpy tests while Figures 10, 11 and 12 show the transition curves for welding and base materials 31.3 and 41.1 for different
fluences. Lateral expansion and ductile rupture were also
determined for different temperatures.

Upper shelf energy as well as transition temperatures
values for 41J, 68J, 0.9 mm lateral expansion and 50% du-
tile rupture are shown in Table 3 and Figures 13 and 14.

Figure 15 show the shift in transition temperature for
the 32 fpy end of life expected is approx. 920C for the most
desfavourable base material 41.1

Results obtained for tensile test at 00C for base
metals 41.1 and 31.3 are show in Figures 16 and 17 where
yield stress, ultimate tensile stress, reduction in area and
elongation are plotted for different dpa values.

Yield stress for different temperatures and different
dpa are show in Figure 18 for base metal 41.1. A \( \sqrt{0.2} \) value
of approx. 700 N/mm2 was obtained for the reactor operating
temperature of 2900C for 10.7 x 10^-2 dpa.

Table 4 resumes tensile test results obtained for base
metals 31.3 and 41.1 and welding metal for room temperature.

Fracture mechanics results are resumed in Table 5
where a value of \( K_{IC} = 1648 \) N/mm3/2 is show for base metal
41.1 irradiated at 10.7 x 10^-2 dpa and tested at 200C. In
similar conditions a value \( K_{IC} = 1979 \) N/mm-3/2 was obtained
for base metal 31.3

In Figure 19 a reference KWU curve is plotted for \( K_{IC} 
\) as a function temperature. Experimental values show good
agreement with the reference curve.

6.- NEUTRON CALCULATIONS

Table 6 show theoretical and experimental values for
flux (\( \phi \)) and dpa. Experimental values were obtained from
the measured activities of Nb-94 dosimeters placed in the
Charpy-V samples notch. [4], [5], [6]
It is necessary to point out that the reaction \( \text{Nb-93} \) \( (n, \gamma) \) \( \text{Nb-94} \) takes place basically in the range of thermal energies \((E < 0.4 \text{ ev})\) which contributes to approx 97% of the total flux in the surveillance capsules positions.

The Nb-93m activity was also measured for some monitors to obtained flux values \((\bar{\phi})\) for \( E > 1 \text{ Mev} \). In Table 7 a comparison is made between theoretical and experimental calculated flux values.

The decrease in upper shelf energy for base metal 41.1 is approx. 30 J when compared with the non irradiated condition.

7.- CONCLUSIONS

Tensile tests show a saturation of tensile properties with neutron fluence for values greater than \( 4 \times 10^{-2} \text{ dpa} \). This property is valid for base metals 31.3 and 41.1 as well as for welding material.

A constant increase in transition temperature is show for both base metals with increasing neutron fluence.

Results obtained confirm the trend show with the results from the first set of capsules validating the extrapolation of a shift in transition temperature \( \text{RTNDT = 920C} \) for \( 3.8 \times 10^{-2} \text{ dpa} \) corresponding to 32 fpy. [1]

A decrease in upper shelf energy of approx 20 J was obtained after irradiation at \( 11.3 \times 10^{-2} \text{ dpa} \) for the welding material and 30 J for the base metal 41.1 when compared with the non irradiated condition.

Displacement per atom (dpa) values obtained from theoretical calculations agree within 20% with experimental values, this can be considered as a good result for this type of calculations.
Values obtained confirm calculations that contribution of fast neutrons (E > 1 MeV) to total fluence is less than 0.1% for the position where the surveillance capsules were placed.

8.- REFERENCES

[1].- Leitz et al; "KWU - CNA I, RPV-Surveillance Program, Set 1" KWU/R413/82/e 83.


[6].- Volkis, J.E., "CNA I Pressure Vessel Surveillance Program, Comparison of Measured With Calculated Neutron Fluences."
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Fig. 13/14.- Shift in Charpy Transition Curves.

Fig. 15.- Shift in Transition Temperature for 32 fpy.

Fig. 16/17.- Tensile Test Curves.

Fig. 18.- Yield Stress as a Function of Temperature and dpa.

Fig. 19.- Fracture Mechanics Reference Curve.
**ENSAYOS DE IMPACTO**

<table>
<thead>
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<th>Clase de probeta: ISO-V</th>
<th>Máquina utilizada: WOLPERT-AMSLER</th>
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<tr>
<td>Material: BASE 41.1 (22CrNiMo37)</td>
<td>Diskette y archivo:</td>
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<tr>
<td>Norma de ensayo: DIN 50 115</td>
<td>Flujo neutrónico: 11,3 E-2 dpa</td>
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**PARAMETROS DE ENSAYO**

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<th>Temperatura de ensayo</th>
<th>Energía Charpy</th>
<th>Expansión Lateral</th>
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<td>mm</td>
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<td>Aᵥ J</td>
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<td>%</td>
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<td>77</td>
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<td>77</td>
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<td>94</td>
<td>1,06</td>
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<td>77</td>
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<td>64</td>
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<td>E 152</td>
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<td>77</td>
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<td>E 153</td>
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<td>77</td>
<td>273</td>
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</table>

**TABLE 2**

453
ENSAYOS DE TRACCION

<table>
<thead>
<tr>
<th></th>
<th>Flu. Neut. E-2 (dpa)</th>
<th>$R_{P0.2}$ (N/mm²)</th>
<th>$R_m$ (N/mm²)</th>
<th>A (%)</th>
<th>Z (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mat. Base</strong></td>
<td></td>
<td></td>
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<td>828</td>
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<td>875</td>
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<td><strong>Mat. Base</strong></td>
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<td>670</td>
<td>783</td>
<td>18</td>
<td>58</td>
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<tr>
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<td>4,5</td>
<td>695</td>
<td>810</td>
<td>16</td>
<td>56</td>
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<tr>
<td></td>
<td>5,5</td>
<td>750</td>
<td>850</td>
<td>17</td>
<td>51</td>
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<td>10,7</td>
<td>770</td>
<td>860</td>
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<td>49</td>
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<td><strong>Mat. Soldadura</strong></td>
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<td>545</td>
<td>634</td>
<td>25</td>
<td>68</td>
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<td>4,5</td>
<td>659</td>
<td>733</td>
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<td>67</td>
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<td></td>
<td>10,7</td>
<td>680</td>
<td>750</td>
<td>18</td>
<td>52</td>
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</table>

- Valores característicos (extrapolados) de las propiedades tensiles a temperatura ambiente.

<table>
<thead>
<tr>
<th></th>
<th>Flu. Neut. E-2 (dpa)</th>
<th>$R_{P0.2}$ (N/mm²)</th>
<th>$R_m$ (N/mm²)</th>
<th>A (%)</th>
<th>Z (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mat. Base</strong></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>41.1</td>
<td>2,2</td>
<td>23</td>
<td>17</td>
<td>-22</td>
<td>-9</td>
</tr>
<tr>
<td></td>
<td>4,5</td>
<td>26</td>
<td>24</td>
<td>-22</td>
<td>-12</td>
</tr>
<tr>
<td></td>
<td>10,7</td>
<td>45</td>
<td>31</td>
<td>-30</td>
<td>-2</td>
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<td><strong>Mat. Base</strong></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>31.3</td>
<td>2,2</td>
<td>27</td>
<td>18</td>
<td>-22</td>
<td>-12</td>
</tr>
<tr>
<td></td>
<td>4,5</td>
<td>31</td>
<td>22</td>
<td>-30</td>
<td>-15</td>
</tr>
<tr>
<td></td>
<td>5,5</td>
<td>42</td>
<td>28</td>
<td>-20</td>
<td>-23</td>
</tr>
<tr>
<td></td>
<td>10,5</td>
<td>45</td>
<td>29</td>
<td>-39</td>
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<td><strong>Mat. Soldadura</strong></td>
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<td></td>
<td></td>
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</tr>
<tr>
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<td>4,5</td>
<td>21</td>
<td>16</td>
<td>-20</td>
<td>-9</td>
</tr>
<tr>
<td></td>
<td>10,7</td>
<td>25</td>
<td>18</td>
<td>-28</td>
<td>-23</td>
</tr>
</tbody>
</table>

- Cambio porcentual de valores característicos de las propiedades tensiles a temperatura ambiente.

\textbf{NOTA:} Los valores correspondientes a 0; 2,2 y 4,5 E-2 dpa se obtuvieron de Referencia \[1\] KWU.
# ENSAYOS FRACTOMECANICOS

<table>
<thead>
<tr>
<th>Material</th>
<th>dpa(E-2)</th>
<th>T (°C)</th>
<th>K_Q (N/mm²)</th>
<th>Rₚ₀,₂** (N/mm²)</th>
<th>P_Q (°)</th>
<th>K_IC (N/mm²)</th>
<th>b (mm)</th>
<th>B (mm)</th>
<th>2.5(Kₑ/ρₑ)**²</th>
</tr>
</thead>
<tbody>
<tr>
<td>41.1</td>
<td>10,7</td>
<td>20</td>
<td>1648</td>
<td>765</td>
<td>18780</td>
<td>1648</td>
<td>15,62</td>
<td>25,4</td>
<td>11,6</td>
</tr>
<tr>
<td>31.3</td>
<td>10,7</td>
<td>0</td>
<td>1455</td>
<td>798</td>
<td>1455</td>
<td>15,52</td>
<td>25,4</td>
<td>8,3</td>
<td>16,4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20</td>
<td>1974</td>
<td>770</td>
<td>22182</td>
<td>1974</td>
<td>15,13</td>
<td>25,4</td>
<td>12,3</td>
</tr>
<tr>
<td>4,0</td>
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<td>0</td>
<td>1555</td>
<td>700</td>
<td>17621</td>
<td>1555</td>
<td>15,38</td>
<td>25,4</td>
<td>35,7</td>
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<td>20</td>
<td>2627</td>
<td>695</td>
<td>30153</td>
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<td>15,67</td>
<td>25,4</td>
<td>35</td>
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<tr>
<td>Sold.</td>
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<td>20</td>
<td>2572</td>
<td>687</td>
<td>27290</td>
<td>---</td>
<td>14,28</td>
<td>25,4</td>
<td>35</td>
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</tbody>
</table>

* Valores obtenidos mediante extrapolación

** TABLE 5 **
<table>
<thead>
<tr>
<th>CANAL CAPSULA</th>
<th>Pos.</th>
<th>Detector</th>
<th>Actividad-esp. Nb-94 10^7 Bq/gr (*)</th>
<th>10^13 n/cm² seg</th>
<th>10^21 n/cm²</th>
<th>dpa experim.</th>
<th>dpa calculado</th>
<th>cal.-exp. exp</th>
</tr>
</thead>
<tbody>
<tr>
<td>P-26</td>
<td>sup. 51 / 54</td>
<td>3.375 ± 0.1</td>
<td>1.98</td>
<td>6.34</td>
<td>0.1054</td>
<td>0.1117</td>
<td>+6.0 %</td>
<td></td>
</tr>
<tr>
<td>M21 A/D</td>
<td>media 52 / 54a</td>
<td>2.205 ± 0.07</td>
<td>1.29</td>
<td>4.14</td>
<td>0.0671</td>
<td>0.0762</td>
<td>+13.6 %</td>
<td></td>
</tr>
<tr>
<td></td>
<td>infer. 53 / 55</td>
<td>1.415 ± 0.05</td>
<td>0.83</td>
<td>2.66</td>
<td>0.0428</td>
<td>0.0453</td>
<td>+5.9 %</td>
<td></td>
</tr>
<tr>
<td>O-27</td>
<td>sup. 45 / 48</td>
<td>3.910 ± 0.11</td>
<td>2.29</td>
<td>7.35</td>
<td>0.1221</td>
<td>0.1165</td>
<td>-4.6 %</td>
<td></td>
</tr>
<tr>
<td>M21 A/B</td>
<td>media 46 / 49</td>
<td>2.745 ± 0.06</td>
<td>1.61</td>
<td>5.16</td>
<td>0.0635</td>
<td>0.0795</td>
<td>-4.8 %</td>
<td></td>
</tr>
<tr>
<td></td>
<td>infer. 47 / 50</td>
<td>1.765 ± 0.05</td>
<td>1.05</td>
<td>3.35</td>
<td>0.0539</td>
<td>0.0473</td>
<td>-12.3 %</td>
<td></td>
</tr>
<tr>
<td>C-20</td>
<td>sup. 66 / 67</td>
<td>3.910 ± 0.11</td>
<td>2.29</td>
<td>7.35</td>
<td>0.1221</td>
<td>0.1098</td>
<td>-10.1 %</td>
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</tr>
<tr>
<td>G18 A/D</td>
<td>media 65 / 66</td>
<td>2.480 ± 0.07</td>
<td>1.45</td>
<td>4.66</td>
<td>0.0754</td>
<td>0.0748</td>
<td>-0.9 %</td>
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</tr>
<tr>
<td></td>
<td>infer. 64 / 63</td>
<td>1.545 ± 0.05</td>
<td>0.906</td>
<td>2.9</td>
<td>0.0467</td>
<td>0.0446</td>
<td>-4.5 %</td>
<td></td>
</tr>
<tr>
<td>N-6</td>
<td>sup. 74 / 77</td>
<td>4.610 ± 0.14</td>
<td>2.50</td>
<td>8.66</td>
<td>0.1440</td>
<td>0.1148</td>
<td>-20.3 %</td>
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</tr>
<tr>
<td>L12 A/B</td>
<td>media 75 / 76</td>
<td>2.98 ± 0.06</td>
<td>1.67</td>
<td>5.60</td>
<td>0.0907</td>
<td>0.0783</td>
<td>-13.6 %</td>
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<tr>
<td></td>
<td>infer. 76 / 79</td>
<td>1.955 ± 0.06</td>
<td>1.00</td>
<td>3.67</td>
<td>0.0591</td>
<td>0.0466</td>
<td>-21.1 %</td>
<td></td>
</tr>
<tr>
<td>H-5</td>
<td>sup. 80 / 85</td>
<td>4.26 ± 0.13</td>
<td>2.50</td>
<td>8.00</td>
<td>0.1330</td>
<td>0.1100</td>
<td>-17.3 %</td>
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</tr>
<tr>
<td>L12 C/D</td>
<td>media 81 / 84</td>
<td>2.84 ± 0.08</td>
<td>1.67</td>
<td>5.34</td>
<td>0.0804</td>
<td>0.0751</td>
<td>-13.1 %</td>
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<tr>
<td></td>
<td>infer. 82 / 83</td>
<td>1.85 ± 0.06</td>
<td>1.00</td>
<td>3.48</td>
<td>0.0559</td>
<td>0.0446</td>
<td>-20.2 %</td>
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</tr>
</tbody>
</table>

(*) Actividad promedio de los dos detectores al 10-08-87.
| CANAL CAPSULA | Posic. | Detectores | Act \(_{\text{Nb-94}}\) \(10^6\) Bq/gr | Act \(_{\text{Nb-93m}}\) \(10^6\) Bq/gr | \(\Phi_1\) \(\text{experim.} \times 10^{13}\) n/cm\(^2\)seg | \(\Phi_{1\text{e}}\) \(\text{experim.} \times 10^{10}\) n/cm\(^2\)seg | \(\Phi_1 / \Phi_{1\text{e}}\) \(\text{unidad} = 10^{-4}\) | | | |
|-------------|-------|------------|-----------------|-----------------|-----------------|-----------------|----------------|---|---|
|             |       |            | (*)             | (*)             |                 |                 |                 | | |
| O-27        | sup.  | 45         | 4.19 ± 0.11     | 3.63 ± 0.10     |                 |                 | < 9.9          | 11.316 |
|             |       | 48         |                 |                 |                 |                 |                 | | |
|             | media | 46         | 2.80 ± 0.08     | 2.69 ± 0.06     |                 |                 | < 7.8          | 9.132  |
|             |       | 49         |                 |                 |                 |                 |                 | | |
|             | infer.| 47         | 1.82 ± 0.05     | 4.132 ± 0.24    | 1.0676          | 0.8448          | 7.913          | < 7.2  | 8.621 |
|             |       | 50         | 1.75 ± 0.05     |                 |                 |                 |                 | | |
| C-20        | sup.  | 66         | 3.91 ± 0.11     | 10.829 ± 0.12   | 2.2936          | 2.1685          | 9.455          | < 9.9  | 11.316 |
|             |       | 67         | 3.91 ± 0.11     |                 |                 |                 |                 | | |
|             | media | 65         | 2.47±0.07       | 6.940 ± 0.15    | 1.4489          | 1.4190          | 9.794          | < 7.8  | 9.132 |
|             |       | 68         | 2.49±0.07       | 6.211 ± 0.27    | 1.4606          | 1.2699          | 8.694          | < 7.2  | 8.621 |
|             | infer.| 64         | 1.53±0.05       |                 |                 |                 |                 | | |
|             |       | 69         | 1.56±0.05       |                 |                 |                 |                 | | |

(*) Actv. al 10-08-87
FIG. 2 HEAT TRANSPORT AND MODERATOR SYSTEMS

1 Reactor
2 Steam generator
3 Main coolant pump
4 Moderator cooler (heat exchanger)
5 Pressurizer
6 Moderator pump
7 Feed element
8 Moderator
9 Filter elements
10 Control rods
11 Refueling machine
12 Main coolant loop (D₂O)
13 Moderator loop (D₂O)
14 Feedwater (Secondary loop H₂O)
15 Live steam loop H₂O
CENTRAL NUCLEAR
ATUCHA I

Fig. 3
Key to reactor cutaway (Clave para la sección del reactor)

A. Control rods (Barras de control)
B. Channel pole (Cuerdas de los canales)
C. Moderator downcomer (Tubo de líquido del moderador)
D. Upper filter piece (Cuerpo de retenciones superior)
E. Inner flow control sump (Entrada de la bomba de refrigeración)
F. Outlet to steam gate (Salida hacia el generador de vapor)
G. Injection line upper element (Tubería de inyección del recinto superior)
H. Inlet line downcomer (Tubería de inyección del tubo de base)
I. Moderator core (Zócalo del moderador)
J. Fuel element suction system (Sistema de dirección de los elementos combustibles)
K. Moderator tank (Depósito del moderador)
L. Pressure vessel (Recipientes de presión)
M. Fuel rods and control channels (Barras de combustible y canales de control)
N. Control rod guide tubes (Tubos guía de las barras de control)
O. Moderator distribution tube (Tubo de distribución del moderador)
P. Support grid (Perilla apoyo)
Q. Lower filter piece (Cuerpo de retenciones inferior)

Fig. 4

\[
\begin{array}{ccc}
S_r & S_{spl} & S_{th} \\
\end{array}
\]

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
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</thead>
<tbody>
<tr>
<td>$S_r$</td>
<td>$S_{spl}$</td>
<td>$S_{th}$</td>
</tr>
<tr>
<td>$5 \times 10^{16}$</td>
<td>$1 \times 10^{16}$</td>
<td>$5 \times 10^{17}$</td>
</tr>
<tr>
<td>$2 \times 10^{19}$</td>
<td>$5 \times 10^{20}$</td>
<td>$2 \times 10^{21}$</td>
</tr>
<tr>
<td>$2 \times 10^{18}$</td>
<td>$7 \times 10^{19}$</td>
<td>$6 \times 10^{19}$</td>
</tr>
<tr>
<td>$3 \times 10^{16}$</td>
<td>$3 \times 10^{18}$</td>
<td>$6 \times 10^{17}$</td>
</tr>
<tr>
<td>$2 \times 10^{19}$</td>
<td>$5 \times 10^{20}$</td>
<td>$3 \times 10^{22}$</td>
</tr>
</tbody>
</table>
COMPARACIÓN DE ESPECTROS NEUTRONICOS CNA-1

RANGO DE ENERGÍA
- $E > 1$ MeV
- $1$ MeV $> E > 0.1$ MeV
- $0.1$ MeV $> E > 0.4$ eV
- $E < 0.4$ eV

PAPEL INTERIOR DEL RECIPIENTE DE PRESIÓN.
POSICIÓN DE LAS PROBETAS DE VIGILANCIA.

Fig. 9
ENSAYOS DE IMPACTO

Energía Absorbida [J]

Temperatura [°C]

\[ E = 3.00 \quad R_s = 0.8 \quad e_d = 101 \quad r_d = 0.6 \]
\[ t_0 = 38 \quad b = 48.00 \quad T_{11} = 24 \quad T_{66} = 58 \]

MATERIAL: Material de Soldadura
CAPSULAS: M 21A/B - M 21C/D
POSIICION DE IRRADIACION: O 27 - P 26
FLUENCIA NEUTRONICA TOTAL: 7,7 E-2 dpa

Fig. 10
ENSAYOS DE IMPACTO

Energía Absorbida [J]

Temperatura [°C]

\[ \varepsilon_s = 32.00 \quad \eta_s = 0.0 \quad \varepsilon_d = 118 \quad \eta_d = 0.0 \]
\[ T_0 = 101 \quad b = 28.00 \quad T_{41} = 66 \quad T_{68} = 95 \]

MATERIAL: 22NiMoCr37 (Material Base 31.3)
CAPSULAS: M 21A/B - M 21C/D
POSICION DE IRRADIACION: O 27 - P 26
FLUENCIA NEUTRONICA TOTAL: 4,6 E-2 dpa

Fig. 11
ENSAYOS DE IMPACTO

Energía Absorbida [J]

Temperatura [°C]

es = 14.50   Rs = 0.9   wd = 110   Ad = 0.9
T0 = 164   b = 52.80   T41 = 133   T68 = 172

MATERIAL: 22NiMoCr37 (Material Base 41.1)

CAPSULAS: M 21A/B - M 21C/D - G 18C/D

POSICION DE IRRADIACION: O 27 - P 26 - C 20

FLUENCIA NEUTRONICA TOTAL: 11,3 E-2 dpa

Fig. 12

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ENSAYOS DE TRACCION

Variación de las Propiedades Tensiles con la Fluencia Neutónica (dpa) para el Material Base 41.1 a 0 °C.

Fig. 16
Variación de las Propiedades Tensiles con la Fluencia Neutrónica (dpa) para el Material Base 31.3 a 0 °C.

Fig. 17
ENSAYOS DE TRACCION

Variación de la Tensión de Fluencia con la Temperatura para el Material Base 41.1.

Fig. 18
ENSAYOS FRACTOMECANICOS

\[ \text{Tenacidad a la Fractura, } \text{Klc (E-3) [N/mm}^2] \]

\[ \text{Temperatura (°C)} \]

\[ \times \quad 10.7 \times 10^{-2} \text{ dpa} \quad \circ \quad 4.0 \times 10^{-2} \text{ dpa} \]

Material Base 31.3

\[ ^1 \text{Curva de Referencia definida por los resultados de investigaciones previas de KWU} \[1\]

Resultados de Tenacidad a la Fractura (Klc) del 2o Juego de Probetas para el Material Base 31.3, orientación L-S.

Fig. 19

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SESSION F

MECHANICAL TEST PROCEDURES
CONCLUSIONS REGARDING FRACTURE MECHANICS TESTING AND EVALUATION OF SMALL SPECIMENS - AS EVIDENCED BY THE FINNISH CONTRIBUTION TO THE IAEA CRP3 PROGRAMME

by
KIM WALLIN*, MATTI VALO*, RAUNO RINTAMAA*, KARI TÖRRÖNEN*, RALF AHLSTRAND**
*Technical Research Centre of Finland (VTT), P.O. Box 26, SF-02151, Espoo, Finland
**IVO INTERNATIONAL LTD, 01019 IVO, Finland

ABSTRACT. Within the IAEA coordinated programme on optimizing of reactor pressure vessel surveillance programmes and their analysis, phase 3, a specially tailored "radiation sensitive" correlation monitor material, a Japanese steel plate with code designation JRQ, a French forging material (FFA) and a Japanese forging material (JFL) were selected for the investigations to be carried out in Finland.

An extensive evaluation of the materials in the as-received and irradiated condition composed of material property characterization. The mechanical properties measured at different temperatures include Charpy-V notch and instrumented precracked Charpy data and especially, static and dynamic elastic-plastic fracture toughness based on the J-integral. The specimen size and geometry were varied in the fracture mechanical tests.

Based on the evaluation of the experimental results the following conclusions can be drawn regarding the use of small specimen fracture mechanical tests for investigating irradiation effects. CVN<sub>pe</sub> and RCT type specimens are suitable for determining the materials fracture toughness even in the ductile/brittle transition region provided the elastic-plastic parameter K<sub>pe</sub> is applied together with a statistical size correction. Additionally, CVN<sub>pe</sub> and RCT type specimens yield equivalent results for the fracture toughness transition shift. It was demonstrated that the dynamic fracture toughness transition shift is comparable to the Charpy-V shift, but the static fracture toughness transition shift may be considerably larger than the dynamic shift. Thus, Charpy-V is not suitable for estimating the static fracture toughness transition shift.

These findings have a strong impact upon the design of future surveillance programmes.

Keywords: fracture toughness, surveillance testing, Charpy-V testing, size effects, reliability.

the material by standard methods, but to advance quantitative fracture mechanics methodology and to assure the quality of reactor vessel surveillance testing.

![Diagram of JRQ plate sectioning](image)

Fig. 1 Sectioning of the JRQ plate at VTT.

2.2 The French forging (FFA)

The French forging material is an ASTM A 508 Class 3 steel with code designation FFA. Finland was supplied with 8 blanks suitable for 100 mm CT-specimens. The marked surface of the blanks were located at a distance of 60 mm from the outside surface of the 275 mm thick forging. After testing the 100 mm specimens, the remaining material from one specimen was cut into various surveillance type specimens like 12.5 mm RCT, CVN, CVN_{bc} and tensile. Additionally, 25 mm CT-specimens were manufactures from other broken 100 mm CT-specimens. All specimens tested were oriented in L-T-direction, for the same reason as in the case of JRQ.
4 IRRADIATION CONDITIONS

The specimens were irradiated in the surveillance position of Loviisa NPP. The irradiation took three years giving an end fluence ranging from approximately $1.4 \times 10^{19}$ n/cm$^2$, $E > 1$ MeV. The nominal flux is approximately $2.0 \times 10^{11}$ n/cm$^2$s, $E > 1$ MeV. The irradiation temperature was 265 °C. The temperature was measured in the irradiation position with an instrumented mock-up surveillance capsule having a thermocouple. For each specimen the flux and fluence ($E > 1$ MeV) and dpa was determined. A detailed description of the dosimetry is presented elsewhere [3].

5 ANALYSIS METHODS

The Charpy-V impact test results were analyzed in the form of impact energy ($E$), fracture appearance (SA) and lateral expansion (LE) - temperature curves and a number of transition temperatures were determined from the results. The transition temperatures were obtained from the curve representative of a hyperbolic tangent function fit of the form

$$ E = \frac{1}{2}E_{us} \cdot \left\{ 1 + \tanh \left[ \frac{T - T_{50}}{C} \right] \right\} $$

where $E_{us}$ corresponds to the upper shelf energy, $T_{50}$ corresponds to the 50% upper shelf energy transition temperature and $C$ is a fitting parameter. The fitting procedure minimizes the sum of the absolute differences $\Sigma_i(\Delta R_i)^2$.

The static and dynamic $J_c$ values corresponding to cleavage fracture initiation were translated into elastic plastic $K_{ic}$ values and treated by a special statistical estimation methodology. The scatter of brittle fracture toughness results can be described with the equation [4]

$$ P_f = 1 - \exp(- \left( \frac{K_c - K_{min}}{K_0 - K_{min}} \right)^4) $$

where $P_f$ is the cumulative failure probability at a stress intensity factor level $K_c$ and $K_0$ is a specimen thickness and temperature dependent normalization parameter and $K_{min}$ is the lower bound fracture toughness which for steels is close to 20 MPa$\sqrt{m}$.

The temperature dependence of $K_0$ in MPa$\sqrt{m}$ can successfully be described with [5]

$$ K_0 = \alpha + \beta \cdot \exp(\gamma \cdot (T - T_0)) $$

where $\alpha + \beta = 108$ MPa$\sqrt{m}$ (for 25 mm thick specimens), $T_0$ is the temperature (in °C) at which the mean fracture toughness is 100 MPa$\sqrt{m}$ and $\gamma$ is a material constant.
Fig. 2 Effect of specimen location on the static fracture toughness transition temperature $T_o$ for the JRQ material.

Fig. 3 Effect of specimen location on the dynamic fracture toughness transition temperature $T_o$ for the JRQ material.
Fig. 6 Thickness corrected dynamic fracture toughness for unirradiated JRQ material. All specimens correspond roughly to a material depths of 70 and 85 mm.

In the case of static JRQ results only the 4T-CT specimens yield a different transition temperature than the smaller specimens that all yield essentially the same result. The dynamic test results are essentially identical. Practically no size effects are visible in the dynamic results. The deviating behaviour of the large static specimens is not assumed to be caused by constraint effects.

All 4T-CT specimens of JRQ had the cleavage initiation site located close to the plate center side of the specimen. Thus the lower toughness of the 4T-CT specimens is actually likely to be caused by the toughness gradient present in the material. Actually, if Fig. 2 is used to extrapolate to the center plate toughness behaviour one ends up with a transition temperature in the range of -40...-30 °C. This would be fully in line with the 4T-CT specimen results.

In the case of the FFA material the possible toughness gradient was not investigated. Because all 4T-CT specimens were taken from within the center portion of the plate, the thickness gradient was not assumed relevant. The CVN_{pe} specimens yield a slightly lower transition temperature than the larger specimens. This difference is assumed to be caused by constraint effects. The materials JRQ and FFA differ in two aspects. FFA has a lower tensile strength and higher ductile tearing resistance than JRQ. Both of these differences have a tendency to intensify constraint effects. However, for both materials the measuring capacity of even the small CVN_{pe} specimen reaches well a level of 150 MPa√m and the measuring capacity of the small RCT specimen seems to reach a level of 200 MPa√m.
Fig. 7 Fracture toughness irradiation shift (155 °C) for static CVNₚc specimens of JRQ. Specimens included in the analysis correspond to depths 70 and 85 mm in the plate.

Fig. 8 Fracture toughness irradiation shifts for static and dynamic loading rates in relation to material depth for JRQ.
Fig. 10 Irradiation response for FFA, based on different types of transition temperature definitions and types of tests.

Fig. 11 Irradiation response for JFL, based on different types of transition temperature definitions and types of tests.
8 ACKNOWLEDGEMENTS

This work is carried out as part of the project "Properties of Structural Materials" in the scope of the Nuclear Power Plant Structural Safety Research Programme being carried out at VTT, funded by the Ministry of Trade and Industry in Finland, the Technical Research Centre of Finland (VTT), the Finnish Center for Irradiation and Nuclear Safety (STUK) and Imatran Voima Oy (IVO).

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Anomalous fracture toughness of irradiated CrMoV-Reactor Pressure Vessel steel.

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Abstract: The surveillance programme of a VVER-440 plant type 213 is comprehensive including totally almost 1000 test specimens. The base metal COD (Crack Opening Displacement) specimens of the surveillance programme in Loviisa 1 revealed anomalous behaviour of $K_{ic}$ compared to the Charpy-V results and to expected results according to standards /1/ and /7/. About 20% of the COD specimens showed an exceptionally low fracture toughness. The experimental fracture toughness results were analyzed simultaneously by combining a novel statistical $K_{ic}$ estimation method and a normalization temperature, including temperature shift by irradiation /2/.

Abnormal test specimens were carefully analyzed by performing fractography, metallography and carrying out repeated tests using reconstitution technique. In the microscope examinations no remarkable difference could be seen between normal and anomalous COD specimens. The reconstituted specimens did not reproduce anomalous behaviour. As a result of our detective work we could conclude, that the anomalous behaviour is caused by incorrect fatigue cracking of base metal COD specimens.

Keywords: Neutron irradiation, irradiation embrittlement, transition temperature, surveillance programme, COD testing, intergranular rupture.

1 Introduction

In Loviisa NPP (Nuclear Power Plant) there are two VVER-440 units of type 213. The 1st unit was commissioned in 1977 and the 2nd unit in 1980. The power stations are situated on the south coast on the island Hästholmen, 110 km east of the capital Helsinki. Both units have been successfully operated by the utility IVO. The mean load factor today in Loviisa 1 is 82% and in Loviisa 2 90%.

The water gap between the RPV wall and the core is small due to a small diameter of the reactor. Consequently the neutron flux in the core region of the RPV wall is rather high. The integral neutron fluence after 40 years of operation is about $2 \times 10^{20} \text{n/cm}^2 (E > 1 \text{ MeV})$ when using full core loading. This fluence is much higher than in western PWR reactors. For this reason a very comprehensive surveillance programme has been adopted for most of the 213 type of reactors, including totally almost 1000 test specimens, to record the embrittlement rate of the RPV-steel.

The embrittlement of the RPV steel, especially the core weld, has been one of the most serious problems of older VVER-440 plants. The embrittlement of the core weld has been
Due to the big variation in the neutron flux it was difficult to analyze the test results at first. Later on when more specimens were tested in late 1980's, and all the test results were analyzed, a mathematical dependence of the fracture toughness on the neutron fluence could be found (fig. 2). A new mathematical treatment of the heterogenous test results were carried out by multidimensional regression analyses described in /2/. In fig. 2 $T_e$ is the temperature at which the fracture toughness is 100 MPa$\sqrt{m}$ for unirradiated material, and $T_o$ is the same for irradiated material.

Fig. 2 $K_{Ic}$ of Loviisa 1 base metal fitted to the normal population of test results.

3 Anomalous behaviour

In fig. 2 the test results can be divided into two populations; one which follows normal temperature dependence and another with low $K$-values and no temperature dependancy. About 20 % of the $K_{Ic}$ results showed an exceptionally low toughness behaviour. The $K_{Ic}$ curve in fig. 2 has been fitted to the results showing a normal fracture toughness behaviour. The anomalous results can be seen as a separate group under the curve. The normal population in fig. 2 shows a slightly higher irradiation shift than the Charpy-V transition temperature shift.

If all test results, including the anomalous part, are considered in the curve fitting, the curve will be as shown in fig. 3. According to fig. 3 the shift of $T_e$ would be the double compared to the normal $K_{Ic}$ population and even more compared to the shift in the Charpy-V transition temperature.
The anomalous results were from test specimens with typically very high fluence, even much higher than the fluence at the RPV inner wall at the end of life (reduced core). Another typical feature was that the crack stopped immediately after crack initiation. The load at the moment of crack arrest was systematically much higher than for the normal population with high initiation toughness. Such a typical crack arrest behaviour could almost be considered a "pop in" (fig. 4).

4 Non destructive investigations of the anomaly

A lot of effort was concentrated on trying to explain the anomalous test results. First the quality of test specimens and the testing procedures were inspected systematically as regards geometry, cutting depth, elastic modulus, fatigue crack curvature, time of testing, irradiation position, irradiation history (full and reduced core) and the test temperature. No explanation to the anomalous behaviour could be found from these inspections.

The fracture surfaces were analyzed by SEM (Scanning Electron Microscope) /4/. It was recognized, that the crack initiation as well as crack propagation could partly be at grain boundaries (fig. 5).

![Fracture surface of an anomalous specimen, partly grain boundary fracture.](image)

This mixed mode of crack initiation and propagation was, however, typical for both normal and anomalous specimens. The same feature could also be seen in the reference specimens without irradiation. Consequently some degree of intergranular fracture is typical for this type of CrMoV-steel.

In order to find out segregations at grain boundaries and precipitations in the matrix microscope examinations were carried out by AEA Technology in England (FEGSTEM) /5/. In these investigations specimens with normal (E116) and anomalous behaviour (E109) as
Fig. 7  The new $K_{jc}$ curve fitting according to results from reconstituted specimens.

Fig. 8  Test results from reconstituted specimens tested in exactly the same temperature as previously anomalous test specimens.

As can be seen all repeated tests show quite normal behaviour, not a single result was anomalous. In fig. 7 and in eq 2 a new curve fit for the irradiation shift of $T_o$ has been used. This curve fit yields higher toughness values than the old one shown in fig. 2. It also includes a much broader data base than the original ones only.

$$T_o = -123 + 18.8 (\Phi^{10^{-10}})^{0.35}$$
of the material /7/ only. Accordingly making of fatigue cracks for fracture mechanics specimens, especially on small Charpy sized specimens, was probable a rather new task.

Normally the fatigue procedure is not recorded, one shall only follow the requirements of the standards. We greatly suspect, that there were difficulties in controlling the load and thus the stress intensity within proper limits during the fatigue of the COD specimens. We have inspected the fatigue crack surfaces of the specimens in order to find out possible overloading during the fatigue. No traces of overloading could, however, be seen. On the other hand overloading might have occurred only during the last few cycles and cannot be seen in fractographic inspections. A possible overloading even in only one cycle, may produce a large plastic zone in the ligament ahead of the crack tip.

The fatigue crack of the reconstituted specimens were made in VTT in Finland. During the fatigue the load was restricted so, that the stress intensity at the crack tip was 10-12 MPa√m. The COD specimens of the surveillance programme of Loviisa 2 as well as of research programmes, such as testing of the Novovoronesh 1 trepan in VTT, did not show any anomalous behaviour. The fatigue cracks on the COD specimens of Loviisa 2 were made in 1977 in the Izorsky factories in St. Petersburg. Accordingly our conclusion here after all investigations and speculations is the following; the original test results appear to be irrelevant due to incorrect pre-fatigue of the COD specimens. Therefore only the new test results which were received with the reconstituted test specimens should be considered.

9 Conclusion and discussion

The shift in the Charpy-V transition temperature of the base metal due to neutron irradiation of the Loviisa 1 RPV is much higher than specified in the old standard \((A_f = 7) /1/\) and close to the upper limit \((A_f = 18) /7/\) of the present standard /7/. The shift in \(K_{pe}\) due to neutron irradiation was much higher than the shift in Charpy-V. This is due to anomalous behaviour of about 20 % of the tested COD specimens. Repeated tests on reconstituted test specimens showed, that the anomalous \(K_{pe}\) results most probably are due to incorrect pre-fatigue of the COD specimens. A new curve fit based on the results from the repeated tests gave about the same irradiation shift of \(K_{pe}\) as Charpy-V values. The repeated tests also show, that the recovery of the \(K_{pe}\) is excellent when standard annealing parameters are applied. Accordingly the base metal of Loviisa 1 is no problem when considering the service life of the RPV.

We think, however, that the anomalous results are quite interesting and need to be looked at more carefully. The phenomena seems to be connected with plastic deformation of the crack tip due to overloading. The irradiation embrittlement properties of such deformed material are quite different from virgin material without plastic deformation. One could call the phenomena "strain irradiation ageing". It is important to know the strain at the tip of a hypothetical crack surface in the RPV wall and to compare with the strain in the test specimens. The experience has shown, that when using controlled loading in the pre-fatigue, no anomalous embrittlement can be seen.
INVESTIGATION OF IRRADIATION EMBRITTLEMENT AND ANNEALING BEHAVIOUR OF JRQ PRESSURE VESSEL STEEL BY INSTRUMENTED IMPACT TESTS

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ABSTRACT. Seven series of A533-B type pressure vessel steel specimens irradiated as well as irradiated - annealed (460 °C x 18 h when 50% of the target fluence had been reached) - re-irradiated to different fast neutron fluences (up to $5 \times 10^{19}$/cm$^2$) have been tested with a new type of instrumented impact test machine. The radiation embrittlement and the effect of the intermediate annealing was assessed by using the ductile and cleavage fracture initiation toughness. Although the ductile fracture initiation toughness exhibited scatter, the transition temperature shift corresponding to the dynamic cleavage fracture initiation agreed well with the 41 J Charpy-V shift. The results indicate that annealing is beneficial in restoring mechanical properties in an irradiated nuclear pressure vessel steel.

Keywords: Pressure Vessel Steel, Neutron Embrittlement, Optimising Annealing Parameters, Instrumented Impact Tests, Plant Life Management.

The increasing integral neutron dose on the pressure vessel of a nuclear power plant manifest itself by causing progressive changes in mechanical material properties such as increase in hardness and tensile yield stress and in ductile to brittle transition temperature. For some plants these changes can lead to operational restrictions or even to a need to resort to mitigating measures.

Thermal annealing is a powerful means to restore the damaged structure by thermally activated processes which can remove or restructure some of the hardening or embrittling microstructural elements. The choice of optimum parameters for the heat
Figure 1. Perspective view of an ASTM 533B Class 3 steel block. All dimensions are in mm. Note that the top layer has been removed in this diagram for clarity; the Charpy specimen were taken from the 1/4 and 374 T positions.

Irradiations

Irradiations were performed in the 10 MW (thermal) SAPHIR swimming pool type materials testing reactor. The specimens were loaded into a fully instrumented stainless steel capsule which also contained copper, iron, nickel and niobium neutron fluence monitors in addition to thermocouples distributed throughout its length and volume. The irradiation temperature of 290 °C was achieved by gamma heating and maintained by automatic control of a helium-nitrogen gas mixture composition in a gap between the specimens and the capsule wall. The neutron energy spectrum was tailored to a typical pressurized water reactor spectrum by sandwiching a stainless steel plate (2 cm thick) between two aluminium sheets (each 1 cm thick) and placing the irradiation capsule just
type of impact test more suitable for determining fracture mechanics parameters. A more detailed description of the new instrumented testing facility is given in [1, 2].

The method generally used for the evaluation of dynamic fracture toughness values is based on the measurement of stored energy up to crack initiation. The main problem associated with this method is the detection of the moment in time when true ductile crack initiation occurs. In cleavage initiated fracture, the fracture toughness can be derived fairly reliably (up to a specific limit) by using the energy at the cleavage load-drop point. Ductile crack growth however, starts prior to the maximum load limit without showing a load drop.

The fracture toughness measurement technique employed here uses the Double Displacement Ratio (DDR) method which is the ratio of crack opening displacement change (dCOD) and specimen deflection change (dD) [2, 3, 4]. The maximum change in the slope of COD versus deflection corresponds to the onset of ductile crack propagation.

After determining the crack initiation point on the load-time/load-deflection curve, the stored energy needed for fracture initiation can be derived. This is the integrated area below the load deflection curve from the start of the test to the onset of crack propagation (cleavage or ductile). The fracture initiation toughness value can be calculated using the stored energy value (Eid) in Rice's equation as shown in Figure 2. The graphs in the figure are typical examples of the final test plots.

![Graph showing Crack initiation, COD, Load, Energy, and Load vs. Displacement](image)

**Figure 2.** A typical final test results and method for determining the dynamic fracture toughness through the VTT instrumented impact testing facility.
Figure 4. Standard Charpy V-notch impact in the irradiated-annealed-reirradiated condition at different fluence levels.

Figure 5. The total normalised impact energy ($E_{\text{tot}}/A$) measured with irradiated precracked specimens (I) at different fluence levels.
Figure 8. The ductile fracture initiation toughness \( (J_t) \) measured with irradiated-annealed-reirradiated precracked specimens at different fluence levels.

The cleavage fracture initiation toughness \( J_c \) is calculated from the absorbed energy at cleavage initiation and is expressed in \( K_{IC} \)-units. The transition curve is analysed by a statistical procedure described for example in [5]. The fracture toughness transition curves derived from the analyses are presented in Figures 9 and 10.

Figure 9. Dynamic cleavage fracture initiation toughness corrected to correspond to a specimen thickness of 25 mm. The transition temperature (To) represents the temperature where the median fracture toughness is equal to 100 MPa m. Reference (Ref) and irradiated specimens at different fluence levels.
DISCUSSION

The dynamic ductile initiation fracture toughness values given in Figures 7 and 8 demonstrate rather high scatter. It is difficult to separate instrumental scatter from scatter in the true material parameter. As a physical phenomena, the initiation of ductile tearing is not sharply defined because ductile crack formation is a process which develops from microcracking through coalescence of voids into physical separation. Further the load parameter i.e. the local J-value at the crack tip varies along the crack front and hence the initiation is a local phenomenon which is sensitive to crack front geometry. Due to the nature of ductile crack initiation, high sensitivity is required from the measuring system and the measured average initiation value will inevitably have some scatter.

The measured average $J_i$ for irradiated material conditions is about 150 kJ/m$^2$ and it does not appear to be sensitive to irradiation, i.e. no dependence on the specimen fluence or on the irradiation-annealing history is found out. Irradiation condition I and the irradiation-anneal-reirradiation condition give about the same average value for $J_i$. However, the measured $J_i$ values for the unirradiated specimens are definitely higher than for the irradiated specimens.

When technical transition temperature criteria like $T_{42f}$ in Charpy-V tests and $T_{525kJ/m^2}$ in tests with pre-cracked specimens are applied, the measured transition temperature is a monotonous function of the fluence as shown in Figures 3 - 8. At these rather high absorbed energy levels significant components, both from the ductile fracture initiation and from crack growth before the onset of cleavage fracture, are included in the measured impact energy. The onset of cleavage fracture is strongly temperature and fluence dependent.

The transition temperatures of cleavage fracture initiation toughness are shown in Figures 9 and 10. The transition temperature has a definite fluence dependence.

Comparison of the fluence dependence of the dynamic transition temperatures is given in Figure 11.

Irradiation dependence of the transition temperature and even the absolute temperature values measured with Charpy-V and $K_{IC}$ parameters in a dynamic mode are very nearly the same. This is in agreement with other data presented in this conference [8].

Transition temperatures of irradiated specimens exposed to the intermediate annealing heat treatment are about 25 °C lower than transition temperatures of virgin specimens as shown in Figure 11. This temperature difference is the safety margin at the end of the life of the reactor pressure vessel achieved by annealing applied in the middle of the reactor life. In Figure 8 the total fluence is the sum of the fluences experienced by the specimens before and after the annealing. Specimens were annealed when approximately half of the target fluence was achieved. In the figure the transition temperatures of IAI-specimens are also drawn with the reirradiation fluence i.e. with total fluence / 2 -values. These points fall on the same curve as Charpy-V and brittle fracture initiation fracture toughness values. If it is assumed that the degree of recovery in annealing is nearly
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EVALUATION OF FRACTURE TOUGHNESS OF VESSEL MATERIALS
USING SMALL-SIZE SPECIMENS AND FULL STRESS-STRAIN CURVES

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"Irradiation Embrittlement and Optimization of Annealing"

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1. INTRODUCTION

To determine the reliable parameters on the low and average strength steel fracture one faces the considerable methodical difficulties. Nevertheless, presently, the works of a large number of specialists in the fracture mechanics field are restricted to the matters of solving the specific boundary-value problems in the elastic and elastic-plastic area for the plane and three-dimensional cracked bodies. At the same time no proper attention is given to a number of important scientific and practical matters. To such matters it should be referred a mode of stressed state, effect of material temperature and structure.

The present report deals with a short description of method of determination of fracture toughness performances of the small-size specimens under the conditions of equilibrium deformation developed at the Institute of Strength Problems of Ukraine Academy of Science. Presently, this method is used for evaluation of VVER reactor vessel material crack resistance at OKB "Gidropress".
3. METHODS OF DETERMINATION OF MATERIAL CRACK RESISTANCE

Fracture energy density is assumed as a physical parameter correlating with the material crack resistance

\[ \lambda = \frac{P_k \cdot \Delta \rho}{2F_k} \]  \tag{1}

Where \( F_k \) - cross-section at the moment of macrocrack formation, and sense of parameters \( P_k \)
and \( \Delta \rho \) is shown in Fig. 2.

If one makes use of concept of critical length \( \rho \) (kp) of specimen working part at which the elastic energy accumulated in the specimen at the moment of macrocrack initiation is equal to fracture work, the material resistance to crack propagation may be related to fracture elastic energy density during propagation over section \( F_k \) of the macrocrack having occurred and started. Hereat, the averaged material modulus of elasticity determined by the method of unloads from the section equal to specimen critical length is assumed to be equal to the Young's modulus at the moment of macrocrack initiation. Then by analogy with the well-known relation

\[ G_{lc} = \lambda = \frac{P_k \cdot \Delta \rho}{2F_k} = \frac{(1-v^2) K_a^2}{E} \]  \tag{2}

Reducing parameter \( \Delta \rho \) to one fracture area we obtain

\[ K_a = \sigma \sqrt{\frac{P_k \cdot \Delta \rho \cdot E}{F_k (1-v_k)}} = \sigma \sqrt{\frac{S_k \cdot \Delta \rho \cdot E}{1-v_k}} \]  \tag{3}

where \( E \) - Young's modulus
\( v_k \) - narrowing of specimen at the moment of macrocrack initiation.
4. CORRELATION WITH K1c

Reference data analysis shows that the most systematic investigation of crack resistance characteristics using large scale specimens was performed with steels 15X2MFA, 15X2NMFA, 22K, 40X, St3.

These materials were used to define criterion $K_A$ with force criterion $K1c$.

About stable correlation of the specified criteria one may judge by dissipation of average coefficient values found from the condition $K = K1c$ (6) for different materials.

When solving the equation (6) with taking account of (3) with respect to $\alpha$ we obtain

$$d = \frac{K1c}{\sqrt{\frac{S_k \cdot \Delta \varepsilon \cdot \varepsilon'}{1-\gamma K}}}$$

As the calculations for different grade steels showed, $\alpha$-parameters values were within the range of 0.23-0.25, i.e. they were practically similar.

Thus, as to plastic materials of average and low strength it is found a good correlation between parameter $K_A$ determined by material fracture energy density under the conditions of volumetric stressed state with small specimens (material "brittlement" occurs due to ultimate plastic deformation under equilibrium loading) and critical stress intensity factor $K1c$, determined by fracture energy density of the same initial material with considerably larger specimens (material "brittlement" occurs due to initial crack larger thicknesses and lengths).

Graphical interpretation of this correlation is given in Fig. 3.
6. CONCLUSION

6.1. Physically substantiated dependences between crack resistance characteristics determined by the parameters of descending sections of full stress-strain curves and stressed state rigidity at crack initiation moment have been experimentally obtained. The possibility of crack resistance trustworthy estimation based on full stress-strain obtained using small-size specimens with different concentrators has been experimentally substantiated.

6.2. There is a satisfactory conformity between the results obtained according to the supposed method and the actual temperature dependence of irradiated steel 15X2NMFA crack resistance characteristics.

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Кинетика разрушения металлов на заключительной стадии деформирования.
Fig. 5 Stress-strain curves for 15Kh2NMFA steel specimens test under irradiation at different temperatures:

1 - 25°C
2 - 77°C
3 - 84°C
4 - 90°C
Fig. 7 Temperature dependences of fracture toughness of 15Kh2NMFa vessel steel under various conditions calculated by preposed method

- initial material
△ - irradiated material after annealing

Kic, MPa·m^{1/2}
Fig. 1 Crack structure at different fracture stages of steel 15X2MFA
a-e - fracture stages
f - full steel strain-stress curve

0 4 8 12 16 20

504
SPECIALISTS MEETING ON
IRRADIATION EMBRITTLEMENT AND OPTIMIZATION OF ANNEALING

PROPOSED RULE PACKAGE ON
FRACUTURE TOUGHNESS AND THERMAL ANNEALING
REQUIREMENTS AND GUIDANCE FOR
LIGHT WATER REACTOR VESSELS

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September 22, 1993
CAUTIONARY NOTE

- This information is under review by NRC staff and management, and by the Commission.

- By the end of 1993 it is expected that the NRC will issue for public comment proposed rules and a draft Reg. Guide similar to those described here.

- The versions published for public comment may be different from those described in this presentation.

- This presentation does not represent NRC POLICY but is a review of information presented publicly during the normal NRC review process.
PRESSURIZED THERMAL SHOCK RULE

10 CFR 50.61
PROPOSED REVISIONS TO 10 CFR 50.61

- Make RTPTS analysis identical to Regulatory Guide 1.99, Revision 2 -- embrittlement estimates are consistent
  - Method for determining initial RT_{NDT}
  - Equation for margin term

- Incorporate thermal annealing as acceptable option -- greater flexibility to the rule

- Restructure PTS Rule -- clarity
  - (a) Definitions
  - (b) Requirements
  - (c) Calculation of RTPTS
10 CFR PART 50 APPENDIX G
FRACTURE TOUGHNESS REQUIREMENTS:

BACKGROUND AND CONTENT

- Published 1973, last technical amendment 1983
- Fracture toughness requirements for ferritic materials of pressure boundary components
- References Appendix G Section III of ASME Code
- Requires evaluation of pressure-temperature limits and minimum metal temperature
- Requirements for Charpy upper shelf energy
  - 75 ft-lb pre-service
  - 50 ft-lb throughout the vessel life
• Specifically require that pressure and leak tests required by ASME Code be completed prior to core criticality

  - Implements CRGR recommendation to the EDO
    June 7, 1990

• NUBARG backfitting claim and appeal

• Safety concerns:
  - Hindrance of finding leaks at high temperature

• Delete the "design to permit annealing" requirement

• Change reference from Appendix G of Section III to Appendix G of Section XI of the ASME Code

• Delete existing annealing language and reference the proposed thermal annealing rule
CONTENTS OF
10 CFR PART 50 APPENDIX H
REACTOR VESSEL MATERIAL SURVEILLANCE
PROGRAM REQUIREMENTS

- Material surveillance program required to monitor irradiation embrittlement of the RPV beltline materials
  - References ASTM Standard E 185
  - Integrated surveillance programs
PROPOSED RULE AND DRAFT REGULATORY GUIDE
ON THERMAL ANNEALING
NEED FOR THERMAL ANNEALING REGULATION AND REGULATORY GUIDE

- Existing Appendix G addresses annealing only in context of 50 ft-lb criterion
  - annealing also appropriate to reduce RT

- Appendix G requirements on annealing inadequate
  - evaluation of recovery from materials tests only
  - method not practical in all cases
  - no requirements on engineering aspects

- Proposed annealing of Yankee Nuclear Power Station
  - highlighted need for more complete regulatory framework
OVERVIEW OF PROPOSED ANNEALING RULE

- Application for Thermal Annealing
  - Subject to approval by the Director, NRR
  - Submitted three years prior to anneal

- Certification of Annealing Effectiveness
CERTIFICATION OF ANNEALING EFFECTIVENESS

DRAFT

- Certify that annealing was performed within the proposed annealing conditions
  
  - Provide post-anneal $RT_{NDT}$ and Charpy upper shelf energy values
  - Estimate reembrittlement trends for $RT_{NDT}$ and Charpy upper shelf energy
  - Project $RT_{NDT}$ and Charpy upper shelf energy for end of proposed period of operation

- If cannot certify, submit justification for subsequent operation
  
  - Approval by Director, NRR, required before restart
DESCRIPTION OF DRAFT REGULATORY GUIDE 1-027

Provides guidance on format and content of applications for thermal annealing

- Describes the criteria that the NRC staff will use in evaluating annealing applications
- Elaborates the provisions of the thermal annealing rule

- Thermal Annealing Operating Plan
- Requalification Inspection and Test Program
- Fracture Toughness Recovery and Rebrittlement
- Rate Assurance Program
- Certification
THERMAL ANNEALING OPERATING PLAN (cont.)

- Equipment, components and structures affected
  - Biological shield
    - Loss of strength, neutron absorption
  - Concrete
    - Design temperature
    - Properties
    - Irradiated concrete
  - Piping
    - Description (materials, dimensions, restraints)
    - Design requirements
    - Known indications of potential flaws
  - Other equipment or instrumentation
  - Storage of core internals
    - Overall layout of containment
    - Coffer dam (if needed)
THERMAL ANNEALING OPERATING PLAN (cont.)

- Thermal and stress analysis
  - Establish time and temperature profiles
  - Maximum concrete temperature
  - Should evaluate
    - Residual deformations
    - Residual stresses
    - Elastic-plastic creep effects
    - Distortions and bending
    - Piping displacements
    - Effects of thermal gradients
    - Effects of restraints (nozzles, piping)
  - Specify limiting conditions
    - Highest temperature
    - Highest stress and strain
    - Limiting heatup and cooldown rates
REQUALIFICATION INSPECTION AND TEST PROGRAM

- Monitoring program
  - Assure annealing within proposed annealing conditions
  - What to measure (temperatures, strains, deflections, etc.)
  - Measurement frequency during heatup, steady-state and cooldown
  - Record retention

- Inspection program
  - Pre- and post-anneal visual examination
  - NDE program for the RPV beltline

- Test program
  - Demonstrate effectiveness of annealing
  - Assure no degradation of RPV and other affected structures, components or equipment
GUIDANCE FOR REEMBRITTLEMENT RATE ASSURANCE PROGRAM

- "Lateral shift"
  - Same embrittlement trend as pre-anneal operating period
  - Limited US data indicate reembrittlement bounded by initial embrittlement trend
  - Analysis is "mean curve" analysis plus margin term
  - Explicit equations provided

- Surveillance method
  - Results must meet credible data requirements in PTS rule
  - Use of MTR irradiations for specimen preconditioning
POST-ANNEAL CERTIFICATION

- Description of the overall annealing process
  - Details sufficient to evaluate the annealing

- Results and evaluation of inspections and tests

- Annealing effectiveness:
  - Percent recovery
  - Reembrittlement rate
  - Allowable operating period
Warm Prestressing - An Efficient Means for Enhancing Brittleness Fracture Resistance of Power Water Reactor Shells

Pokrovsky, V.V., SPE "Resource", Kiev, Ukraine

Warm prestressing is an efficient means to enhance the brittle fracture resistance of PWR shells and thus to extend their life under irradiation conditions. The lifetime of PWRs is limited by an increase in the critical transition temperature ($T_c$) of the shell metals and welds caused by neutron exposure of the shell wall in the reactor active section during its operation. The shells metal annealing used nowadays for the recovery of their critical transition temperature has some limitations and cannot be applied for all grades of reactor steels. Thus, in particular, neutron exposure of reactor steels with nickel recudes their $K_{IC}$ values in the temperature range below the critical transition temperature ($T_c$) (at the lower shelf of the steel $K_{IC}$ temperature dependence). Annealing shifts the metal $K_{IC}$ temperature dependence to the left with no increase in the $K_{IC}$ values in the lower shelf of the $K_{IC}$ temperature dependence. Annealing is of low efficiency or inefficient for reactor steels with the content of nickel or manganese exceeding 1 percent and with an essential content of copper, phosphorous etc. owing to their tendency to thermal embrittlement.

A method of warm prestressing of PWR shells has been developed at the Institute for Problems of Strength of the Ukrainian Academy of Science together with the Institutes "Gidropress" and "Prometey". The method allows the brittle fracture resistance of the shell metal to be enhanced 1.5 ..... 3 times and by this to increase appreciably the lifetime of reactor shells.

The method proposed eliminates initiation of cracks with a relative depth $\alpha$ - 0.05, 0.1 and 0.25 when filling the PWR shell in with cold water at various combinations of thermal stresses and residual pressure.

At present for WWER-440 and WWER-1000 shell metals optimal load and temperature conditions of warm prestressing have been substantiated, the stability of warm prestressing positive effect has been studied under real service mechanical and thermal loading conditions for the materials with cracks after warm prestressing, the determining mechanisms of hardening for a metal with cracks under specified WPS conditions have been established and a calculation procedure has proposed for the estimation of the increase in the shell metal brittle fracture resistance depending on the degree of their embrittlement, the loading and temperature conditions of warm prestressing (WPS).

Examples of fracture toughness ($K_{IC}$) temperature dependences for the embrittled by simulated thermal treatment 15x2MFA, 15x2NMFA reactor steels and weld metals IOXMST and 08XGHTMA prior to and after warm prestressing are shown in the Figure.
CONCLUSIONS
SESSION A

NEUTRON IRRADIATION EFFECTS
P. Petrequin, K. Törönen

Mr. Davies presented a wide review of the problems related to irradiation embrittlement. He indicated that the solution of these problems is a challenge in both economy and safety. There are nearly 500 nuclear reactors in operation in the world and he showed that the many of them were built before 1972 when the role of copper and phosphorous in irradiation embrittlement was identified. The description of the behaviour of pressure vessels remains a long term problem, needing predictions and he said that in many cases, the basic hypothesis are not always sufficiently validated and that many of the parameters had not been evaluated.

This situation was illustrated by numerous questions:

- What is the most relevant measure of neutron exposure: neutron 0.1, 0.5 or 1MeV? The usefulness of a reference sensitive material for surveillance programmes was emphasized.

- Are the prediction formulae validated by a sufficiently representative database?

- Is the shift of RT_{NDT} measured properly by the different techniques?

- Is the change in upper shelf energy correctly explained mechanistically?

- Are all the copper atoms in the material acting in the embrittlement?

- Should the correlation of irradiation embrittlement be matrix copper?

- What is the role of phosphorous and how relevant is hardness measurement of annealing when non-hardening embrittlement may exist?

- What is the behaviour at high exposure (beyond the EOL fluence)?

- What should be the content and scope of surveillance programme after annealing of the Reactor Pressure Vessel (RPV).

Some answers to these questions can be provided by mechanistic modelling but it remains very complicated. The mitigation of irradiation effects is evoked essentially on the aspect of annealing. The uncertainties of the re-embrittlement after annealing were indicated. He reviewed the more important international programmes and working groups on the subject showing how international cooperation is active in this field. As a final commentary, he asked whether it is now possible to add effectively to all the knowledge accumulated on the two types of steels used in the world for reactor pressure vessels (Mn, Ni, Mo and Cr, Mo, V steels).

During the discussion, it was indicated that there is material from plate HSST03 available at ORNL.
Mr. Brillaud described the more significant results obtained from the surveillance programme of the 54 reactors in operation in France. Base metal, HAZ (heat affected zone) and weld metal were introduced in the surveillance programme. The results presented are essentially Charpy results as, up to now, it is assumed that $\Delta RT_{NDT} = \Delta T_{CV}$.

The toughness results were not yet available. All the materials had low levels of residual elements but there were some (corresponding to 6 units) where Cu reaches 0.13, P0.019 and Ni 0.51%. The range of fluence is 0.3 - 5.510^{19} \text{n/cm}^2(1\text{MeV}). The maximum embrittlement observed was an increase of $\Delta T_{CV}$ of 72°C for 3.5 × 10^{19} \text{n/cm}^2 (weld with $P = 0.014$). The scatter was relatively large. A difference reaching 60°C was observed on base metal on two very similar materials. A high embrittlement at relatively low fluence was associated to intergranular fracture. The results are compared with the FIS formula used in France. $\Delta T (^\circ C) = 8 + [1537 (P-0.008) + 238 (Cu-0.08) + 191 Ni^2Cu] (\Phi \times 10^{19})^{0.35}$. There were only 10 out of over 139 data points exceeding the prediction and then by less than 10°C (1 of 21°C). The R&D actions of support to the surveillance programme were described. They were related to studies on dosimetry and temperature evaluation (essentially in relation with the use of MOX fuel), on materials heterogeneities and on the CHOOZ A vessel examination.

Mr. Platonov gave an extensive overview of the radiation damage of WWER-type reactor vessel materials. The main conclusions were as follows: no saturation effect is seen in radiation strengthening; main contributors to Charpy V transition temperature shift are impurity contents (mainly Cu and P), neutron flux and fluence. The combined effects are rather complicated including an observation of embrittlement saturation with low flux values; the recovery during annealing is dependent on P-content; full recovery requires annealing temperatures 150-200°C above operating temperatures; successive irradiation and annealing cycles do not seem to increase the radiation embrittlement.

**SESSION B**

**NEUTRON IRRADIATION EFFECTS**
H. Horowitz, F.-G. Gillemot

Session B was a direct continuation of Session A dealing with neutron irradiation effects. Five papers were presented. They summarized the up-to-date results about radiation embrittlement obtained on worldwide used RPV steels.

Mr. Soulant presented the French participation with the Phase 3 of the IAEA Coordinated Research Programme and focused mainly on the study of a low sensitive French Forging (FFA) and a high sensitive "monitor" Japanese plate (JQ).

Shifts of Charpy V curves showed good agreement with the estimated shifts given by prediction formulas and good agreement with Finnish results was found. The effects of irradiation on $J_t$ was also investigated, but the main focus of the study was on the "local approach". For cleavage rupture, Weibull parameters were determined and used to obtain a determination of probabilistic values of toughness. Difficulties due to an
insufficient number of notched specimens were encountered but a satisfactory representation was obtained using the Beremin value of Mn. In the ductile range the critical ratio of void growth at rupture \((R/R_0)\)c was determined and used to estimate \(J_{lc}\).

**Mr. Kryukov** from Russia presented the results of investigations made on the templates cut out of the Kozloduy Unit 2 reactor pressure vessel. The specimens were taken before and after annealing of the pressure vessel from base metal and from the weld. Chemical analysis showed no big differences in residual elements such as phosphorous and copper between weld and base metal (\(-0.036\%\, P\) 0.18\%Cu). Annealing was done at 475°C 150 h. Testing of the specimen showed substantial recovery on weld specimens. As subsized specimens were used, correlations had to be used to evaluate DBTT just before and after annealing, and for the future.

**Mr. Brumovsky** presented "pre-preliminary" results from the Phase III of the IAEA CRP. After a brief presentation of the different steels tested he first focused on the results obtained on JRQ material (reference sensitive steel which was to be tested by all participants). If grouped by type of orientations and depth, the Charpy results showed a small scatter, tensile results were also very similar. A first evaluation of the different shifts of Charpy curves under irradiation was given and different kinds of prediction formulas have been fitted for the acquired data.

**Mr. Lowe** presented the results obtained on Linde 80 weld metals. The measurements are a part of the B & W Owner Group Reactor Vessel Integrity Programme. The data obtained during testing materials irradiated in surveillance capsules and research reactors were compared. The neutron embrittlement results were presented as the function of irradiation temperature and in the function of copper content. For B & W designed reactor plants a new chemistry formula was described, showing a correlation level of 0.9. This new formula includes the effect at various elements, such as C, Cu, Ni, P, Mo. A plan of the improved B & W Owners Group RPV Integrity Programme was presented.

**Mr. Valo** presented the results obtained on toughness properties of 15 Ch MFA pressure vessel steels before and after different neutron irradiation. At Loviisa NPP dummy elements were inserted into the core of both units. The results obtained on specimens irradiated before and after the installation of the shielding were compared. No measurable flux rate effect was found. The neutron embrittlement results obtained are relevant to WWER-40 reactors where mitigating efforts like fluence rate reduction have been implemented.

**SESSION C**

**SURVEILLANCE**
S.T. Rosinski, C. Brillaud

In addition to discussions regarding results of various international surveillance programmes, Session C also included brief discussions on thermal ageing of reactor pressure vessel (RPV) materials, activities of the IAEA International Working Group on
Life Management of Nuclear Power Plants (IWG-LMNPP), and activities of the European Action Group on RPV Materials Irradiation Effects and Studies (AMES) group. Presentation summaries are provided below.

Mr. Falcik presented information regarding on-going surveillance programmes for six WWER-440/213 type reactors (PWRs) in the Czech and Slovak Republics. The surveillance programmes discussed experienced a rather high lead factor (> 10) which allowed for the prediction of NPP remaining life with only a small irradiation time (five years). The materials tested exhibited embrittlement trends well below that proposed by applicable Russian Codes, thus indicating acceptable material behaviour for the proposed plant design life. In addition, analysis of the surveillance programme result indicated a fluence dependence of transition temperature shift near 1/2 as compared to 1/3 provided in Russian Code. Subsequent annealing produced nearly full recovery for the surveillance materials tested ductility and less recovery for tensile properties and transition temperature.

Mr. Kupca discussed information regarding surveillance programmes under way in Units 3 and 4 at Bohunice WWER-440/213. The surveillance capsules at Bohunice also experienced a lead factor > 10. The relatively low Cu and P content of the surveillance materials produced embrittlement within acceptable limits for predicted plant lifetime. Efforts to improve neutron fluence and irradiation temperature monitoring and to decrease total fluence through improved fuel loading schemes were discussed.

Mr. Krompholz discussed the surveillance programme for the Swiss RPV. A rather large shift, $\Delta T_{50}$ and $\Delta T_{30}$, was obtained for the automatic weldment material at a relatively low fluence. This shift was attributed to the inhomogeneity of the differing positions within the weldment from which the samples were taken. No significant changes were observed in tensile properties for the relatively low fluence surveillance materials.

Mr. Gillemot discussed the advantages and disadvantages of the surveillance programme for WWER-440/213 RPVs. The surveillance extension programme under way will more closely monitor capsule dosimetry and temperature than the original surveillance programme at the Paks Power Station. The extended surveillance programme also includes reconstituted Charpy samples and will more closely conform to ASTM E-185 specifications.

Mr. Lowe discussed thermal ageing activities in progress on RPV materials in the USA. Samples tested after ageing at 288° - 293°C (550-560°F) for approximately 100,000 hours indicated relatively minor changes in transition temperature. Subsequent annealing at 343°C (650°F) and 454°C (850°F) recovered properties better than that of the original material. Additional activities are under way to continue the thermal ageing effort up to 200,000 hours.

Mr. Ianko presented a brief summary of the IAEA IWG-LMNPP. Included was information regarding the scope and membership of the IWG and planned specialists meetings. In addition the IWG sponsored effort to develop an international database on RPV surveillance results and the anticipated publication of an IAEA book on irradiation
effects were discussed.

Mr. von Estorff presented a brief summary of the activities of the newly formed European Action Group on RPV Materials Irradiation Effects and Studies (AMES). The objectives of this group include acting as an European review group for this subject area and to advise Regulatory Bodies and provide a base for development of common European Studies.

Mr. Kopea described the efforts to anneal the RPVs at Bohunice Units 1 and 2, WWER-440/230, in early 1993. Both RPVs were annealed at a temperature of 475°C ± 20°C for 168 hours. Based on hardness measurements taken prior to and after the annealing process full recovery of original material properties was obtained. Mössbauer spectroscopy investigations also indicated a high degree of lattice order following the anneals indicating a high level of recovery. Further comparison of the annealed RPV material with that located on lower sections of the RPV also indicated full recovery of material properties.

Mr. Krompholz presented two separate approaches to determine crack initiation for J-integral investigations. The first approach involved an experimental method (potential drop technique) and a mathematical procedure based on ASTM E 399. Agreement between both methods was found to be excellent.

SESSION D

VARIATION IN IRRADIATION CONDITIONS AND MECHANISMS MODELLING
B. Houssin - C.A. English

The session comprised five papers, three were concerned with the microstructural changes that occurred on irradiation the thermal ageing, one with the micromechanics associated with RPV embrittlement, and one with the correlation between chemical composition and radiation hardening. In providing a summary it is sensible to group the first three together and deal with the other two separately. A general comment is that, as in the rest of the meeting, the papers covered both the “Western” and “Eastern” steels.

The microstructural papers covered a variety of techniques and various elements of the microstructure from the copper rich precipitates to the dislocation loops formed on irradiation and annealing.

Mr. Kocik reported on a careful Transmission Electron Microscopy (TEM) study of irradiated and annealed WWER-440 RPV. The visible radiation induced defects consisted of very fine vanadium carbide precipitates, small dislocation loops and black dots. The radiation induced defects in power reactor are concentrated to dislocation substructure which become more complex as the irradiation proceeded. Differences were found between accelerated and surveillance irradiation. It was inferred that a high density of small clusters was also present but they were below the resolution limit.

Mr. Auger and co-workers illustrated the use of the Field Ion Microscopy (FIM)
to examine both thermally aged and irradiated model Fe-Cu alloys, as well as steels from the Chooz A and Dampierre surveillance programmes. The results from model alloys followed convention in showing a density of Cu rich clusters in both the thermally aged and irradiated alloys. A major result was that under irradiation the composition was Cu/Fe. The studies of Chooz A showed that irradiation had induced the formation of a high density of local enrichment of elements such as Ni, Mn, Si and Cu, where the average solute contents were 4 at % or less, the balance being made up of iron. Defects had also been detected by SANS, and these data also confirmed that they were not simple copper precipitates.

Mr. Dumbill and co-workers had studied the irradiation of simple binary copper alloys which had been thermally aged to precipitate copper before irradiation. The STEM technique was employed to detect not only individual particles but to perform sophisticated measurements of the levels of copper in the matrix which were still available for embrittlement. The hardening from the copper precipitates had been estimated from the microstructural data and shown to be consistent with the observed macroscopic increases in Vickers hardness.

A strategy to improve pressure vessel surveillance was proposed by Dr. Fabry and co-workers. The method entails an enhanced test matrix and incorporates statistical fracture mechanics and damage modelling. Basically, the ductile-brittle temperature and initiation fracture toughness (K_C and K_Ic) are governed by the stress-strain properties, the critically microscopic cleavage stress and the size distribution of cracked particulates. These parameters are assessed using the load-time diagrams of instrumented Charpy V impact tests in combination with classical tensile tests and K_c measurements at selected temperatures. The approach is also successfully applied to reconstituted Charpy V specimens.

Mr. Novosad presented results on the influence of Cu and a far less significant effect of P on the sensitivity of radiation embrittlement of Cr-Ni Mo-V steel used for manufacturing WWER-1000 reactors. The shifts in transition curves were found to be the same for CVN and K_C but higher for K_Ic. The recovery rate was found to be higher for higher Cu contents without detectable effect of P content.

The only general comment that can be made on the basis of the papers presented in the session, is that the microstructural characterisation of Western PWR steels is such that the clusters responsible for 2 portions of the hardening and embrittlement have been identified and studied, whereas for the WWER RPV steels this has still to be achieved. A point for discussion is how important is it to undertake this.

SESSION E

MITIGATION OF IRRADIATION EFFECTS
D. Miannay, M. Brumovsky

Mr. Novosad presented the results of the mechanical properties of six heats of Cr, Ni, Mo, V steel doped with copper and phosphorous and irradiated. The transition shift
is clearly correlated to the copper content. The role of phosphorous is less evident. However, the transition shift determined statically is higher than the transition shift determined dynamically. As for recovery after irradiation, copper affects the rate but not the final level.

Mr. Törrönen gave a general presentation of mitigation measures which are well documented in the open literature. The first measures are evident: for the same power, decrease the flux coming from the core by low leakage fuel management, by dummy elements or by absorber rods. These measures are in use currently. A further method, vessel wall shielding, has been proposed, but not yet applied. The second way is to decrease the severity of the PTS by improving mixing and increasing temperatures of injection water, or by pre-stressing the pressure vessel (PV), which is a solution under prospect. The last method is to anneal embrittled parts of the PV without introducing effects such as residual stresses, temper embrittlement or segregations which may introduce stress corrosion cracking. Moreover, with annealing, the implementation of surveillance programme has to be defined. The behaviour of the cladding has also to be detailed. As the annealing procedure is considered in the ASTM recommendations, it is suggested that the Eastern block experience is taken into account.

Mr. Rosinski presented US DOE PLIM Programme activities related to annealing including an Irradiation -Anneal-Re-irradiation (IAR) project studying the behaviour of specific American RPV steels and the investigation of heat transfer boundary conditions for an RPV section subjected to an annealing treatment. Strain measurements are envisaged in future experiments on a section containing a nozzle. The results will be made available as "information to the ASME Code".

Mr. Brumovsky mentioned the experiment on a rig consisting of a whole WWER-440 pressure vessel performed by SKODA. The results will be given at the workshop on annealing planned in the Slovak Republic early next year.

Mr. Mager emphasized the great advantage that can be obtained from the understanding of mechanistic modelling on fine microstructural observations to predict embrittlement and the recovery. Thermal annealing conditions appears to be specific to each plant. He also presented the proposed Westinghouse annealing demonstration to be done in cancelled nuclear power plant to resolve the structural integrity dimensional stability issue. Software such as VTESHER can be used for economic assessment of thermal annealing.

Mr. Novak presented a short survey of the method of "indentation testing" that was used for a characterisation of cladding tensile properties in WWER reactor pressure vessels. He explained a theoretical background of such measurements that result in material (cladding) stress-strain curves. Comparison of several calculation models were shown and mentioned that they are sensitive to some input parameters. Real values from measurements in the RPV of NPP Jaslovske Bohunice before and after annealing were presented and shown that this method can be used for the determination of cladding tensile properties as well as of their changes during operation (irradiation hardening) and of annealing but also of technological features during production. In discussion a problem of cladding integrity (in the presence of possible underclad cracks) was opened.
as well as an influence of possible irradiation damage.

Mr. English concentrated his presentation on results from studies of phosphorous segregation in experimental RPV steel heats. Materials were heat treated to obtain coarse grain structure (similar to weld HAZ) and then either irradiated and/or annealed or thermal aged. It was found that phosphorous (0.017 wt%) can cause extremely high embrittlement after thermal ageing at 450°C for 2000h, if material has a coarse grain structure. This effect can be increased by a simultaneous higher content of copper. It was shown that Charpy shift recovery after annealing is not consistent with hardness recovery. A model of phosphorous segregation developed for Magnox RPVs was applied with good results and agreement with experiment. Phosphorous segregation results also in a change from cleavage into intergranular fracture, in some amount. Segregation of phosphorous (up to 35%) of a monolayer was found on grain boundaries. Recommendations for thermal recovery regime were given in conclusions.

Mr. Jinchuk provided information about the RPV surveillance programme for Atucha 1 NPP. Characterization of the reactor (PHWR) and its RPV was given with their implications for its material surveillance programme, mainly in terms of location of specimen capsules. Results from the first capsules were discussed. Because of very different neutron spectra in surveillance location (high flux of thermal neutrons) and RPV, dpa is used as a damage measure in this case. Irradiations in a test reactor for lifetime evaluation were also carried out.

SESSION F

MECHANICAL TESTS PROCEDURES
P. Soulat, C. Leitz

In this session four papers were presented with the aim of evaluating fracture toughness of irradiated specimens and determining embrittlement with small size specimens.

Mr. Wallin presented the Finnish results of IAEA CRP 3 programme with different static and dynamic fracture testing techniques compared with Charpy V results on three different materials. The following conclusions were drawn.

1. It is possible to obtain the same results with large and small specimens with a statistical size correction.

2. For these materials the dynamic transition shift is comparable to the Charpy shift, but the static shift is larger.

Mr. Ahlstrand presented results of Loviisa surveillance programme with some anomalous low fracture toughness. After different examinations, the conclusion was that the specimens with lower values were probably overloaded during fatigue precracking. The influence of irradiation was discussed. For example, Westinghouse found similar results on unirradiated specimens overloaded in the same manner.
Mr. Tipping & Mr. Valo presented results of irradiation annealing and re-irradiation of JRQ steel for the IAEA CRP-3 and fracture toughness test results based on instrumented precracked Charpy specimens. The conclusions of this presentation were:

1. The same shifts were obtained with pre-cracked and notched Charpy tests.

2. After annealing, at the mid end of life (EOL) fluence, the re-embrittlement trend was not higher than during the first irradiation. So, annealing was demonstrated as a valuable method for mitigation of irradiation embrittlement.

Mr. Getmanchuk gave a method of evaluation of the fracture toughness in the brittle and the ductile range based on tensile tests to determine stress-strain curve. The crack resistance calculated from this stress-strain relationship is correlated with linear elastic $K_{IC}$.

One important problem is to determine fracture toughness properties of irradiated materials to improve the perceived margin to failure of pressure vessel. For that, surveillance programmes are undertaken to Charpy V transition curves to determine the shift of RT$_{NDT}$ under irradiation. Some studies are in progress to measure directly fracture toughness with small specimens having results comparable with those obtained on large specimens.

It was concluded that these tests are validated, with the following parameters taken into account:
- size effects
- strain rate
- statistical determination for a probabilistic approach.

Mr. Hiser presented the US NRC plans to revise the PTS rule and appendices G and H of IOCFR50; and to propose a rule and a draft regulatory guide on thermal annealing. These proposed rules and draft guide may be published for public comment by the end of 1993. He also reviewed the background of Reg. Guide 1.99, Rev. 2, including the fact that the guide is based on U.S. surveillance data only. NRC research efforts to develop improved irradiation embrittlement correlations were described, including efforts to collect non-US surveillance data to extend the range of variables such as composition.

It is recognized that the assumed one-to-one correlation between irradiation-induced embrittlement measured with Charpy specimens and that of the fracture toughness is not always correct.

It is therefore recommended that for the future surveillance programmes incorporate more direct determinations of fracture toughness i.e., beyond the current Charpy-based indexing procedure. Investigations are in hand to investigate such tests.
RECOMMENDATIONS
RECOMMENDATIONS

1. When reporting fluence levels values for >0.1, >0.5, >1.0 Mev and dpa should be reported.

2. Internationally accepted reference materials should be included in all surveillance programmes and test irradiations. The assistance of relevant organizations is requested to enable the provision of such material or to enable interested individuals to gain access to reference materials.

3. An International guide, making use of a more comprehensive database, for predictions of irradiation response of RPV materials, would be valuable.

4. Hardness tests are insufficient to evaluate annealing effectiveness. A broader range of mechanical properties data is required, to assess possible non-hardening embrittlement mechanisms.

5. Due to obvious differences in irradiation response of WWER- and Western type of steels, and for radiation damage and annealing predictions, a careful comparison of the behaviour and damage mechanisms in these steels should be carried out.


7. Post-annealing surveillance programmes should be developed.

8. Subsize Charpy and small size notched tensile bars are now used to evaluate the embrittlement. The use of small specimen testing is a promising approach to obtain data from operating plants, and further work is suggested on the field.

9. The effect of fluence rate on neutron embrittlement of 15h2MFA steels in the fluence rate range used at Loviisa was not found, but was found at a wider fluence rate range used in Russian experiments. Further international efforts to study the fluence rate efforts should be useful.

10. Results from the IAEA CRP -Phase III are very promising even in the early stage of the evaluation of the data supplied by the participants. Due to the importance of the radiation embrittlement in the life management of the PWR’s the continuation of the international cooperation in the frame of an IAEA coordinated programme is suggested. The exact topics for Phase IV could be decided after the evaluation of Phase III is finished.

11. Investigations should continue to estimate the fluence dependence on transition temperature shift on a larger variety of materials to determine appropriate trend prediction equations.

12. Development of the international database on surveillance results should be continued. Composition and properties of materials in the database should be verified.
13. Limited quantity of information exists on irradiation and ageing behaviour of claddings. Further tests are needed to increase the quality of integrity (PTS) analysis.

14. It is important to continue with microstructural characterisation of surveillance and specimens irradiated at higher dose rates to build up a more complete characterization of the mechanisms and microstructural changes created by irradiation. In particular, the detailed link between the observed microstructural changes and the changes in mechanical properties should be pursued.

15. An extensive programme for understanding (determination) of a correlation between microstructural and mechanical properties changes (at least qualitatively, i.e. to elaborate a model of radiation damage mechanism) should be initiated.

16. Direct fracture toughness measurement techniques should be validated taking into account size effects, strain rate and statistical aspects.

17. It is recognized that the assumed one-to-one correlation between irradiation-induced embrittlement measured with Charpy specimens and that of the true fracture toughness is not always correct. It is therefore recommended that surveillance programs should incorporate more direct determinations of fracture toughness indexing beyond the current Charpy-based indexing procedure.

18. Measures such as leakage core, dummy elements etc. to degrade the rate of embrittlement should be applied. This should be done as early as possible and specify after annealing.

19. Operating records for the evaluation of dosimetry data as well as temperature histories should be carefully registered.

20. This particular Specialists Meeting was judged to be both timely and successful in terms of content. It was recommended that another similar meeting be held in 2 to 3 years time.
LIST

of

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<td>Dr. Van de Velde J.</td>
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<td>Dr. Van Walle Eric</td>
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<tr>
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<td>Dr. Brumovsky Milan</td>
<td>Czech Rep., Nuclear Research Institute REZ</td>
<td>02-685351</td>
<td>02-6857567</td>
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</table>
SPECIALISTS MEETING on IRRADIATION EMBRITTLEMENT and OPTIMIZATION of ANNEALING
Paris, France, 20-23 September 1993

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<th>Contact Details</th>
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<td>USA, Oak Ridge National Laboratory</td>
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<th>56. Dr. HISER Allen</th>
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<th>57. Dr. LOWE A.</th>
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<th>58. Dr. MAGER T.R.</th>
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<th>59. Dr. ROSINSKI Stan T.</th>
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<td>USA, Sandia National Laboratories</td>
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<th>60. Dr. WICHMAN Keith</th>
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SPECIALISTS MEETING
on
IRRADIATION EMBRITTLEMENT
and
OPTIMIZATION OF ANNEALING

PARIS, FRANCE
20–23 SEPTEMBER 1993

PROGRAMME
IRRADIATION EMBRITTLEMENT and OPTIMISATION OF ANNEALING
PARIS, FRANCE - 20 - 23 SEPTEMBER 1993

20 September 1993

09:30 Registration of participants
10:00 Opening ceremony
1. Mr. P. Petrequin, Chairman of the Organisation Committee
2. Mr L. Ianko, Scientific Secretary, IWG - LMNPP, IAEA
3. Dr A.G. Miller, PWG3, OECD/NEA
4. Acad. L.M. Davies, Chairman of the Specialist Meeting

COFFEE BREAK

11:00 Session A - Neutron Irradiation Effects
Chairmen: Mr Petrequin, France
           Pr Törrönen, Finland

1. Davies L.M. (UK) - "Irradiation effects in pressure vessel steels and weldments"
2. Brillaud C.-de Keroulas F.-Pichon C.-Teissier A. (France) - "Overview of French activities on neutron radiation embrittlement of pressure vessel steel"
3. Kryukov A.-Platonov P. (Russia) - "Radiation embrittlement of WWER-440 vessel materials"

12:30 LUNCH

14:00 Session B - Neutron Irradiation Effects
Chairmen: Ms Horowitz, France
           Mr Gillemot, Hungary

1. Soulant P.-Miannay D.-Marini B.-Schill R.-Horowitz H (France) - "The irradiation embrittlement of two pressure vessel steels - Contribution of local approach"
2. Kryukov A.-Platonov P.-Shtrombach Ya.-Nikolaev V. (Russia) - Klausnitzer E.-Leitz C. (Germany). Rieg C.Y. (France) "Investigation of the templet cut out of the Kozloduy Unit 2 reactor pressure vessel"
6 Ullrich G.-Krompholz K. (Switzerland) - "Fracture mechanics investigations within the swiss surveillance programme for the pressure vessel of modern nuclear power plants"

12:30

LUNCH

14:00

Session D - Variations in Irradiation Conditions and Mechanisms - Modelling
Chairmen: Mr Houssin, France
Mr English, UK

1 Kocik J.-Keilova E. (Czech Rep.) - "Radiation damage structure in irradiated and annealed WWER type reactor pressure vessel steels"

2 Auger P.-Pareige P.-Akamatsu M.-VanDuyssen JC. (France) - "Microstructural characterization of irradiation induced precipitation in pressure vessel steels and model alloys"

3 Fabry A.-Petrova T.-Van Walle E.-Verstrepen A.-Chaouadi R.-Wanninj J.P.-Van de Velde J.-Van Ransbeek T. (Belgium Bulgaria) - "RPV steel embrittlement : damage modeling and Micromechanics in an Engineering Perspective"

COFFEE BREAK

4 Dumbill S.-Phythian-W.J. Brown P.-Sinclair R. (UK) "Stability of thermaly induced copper precipitates under neutron irradiation"

5 Novosad P. (Czech Rep.) - "Effect of chemical composition on irradiation embrittlement and annealing in Ni-Cr-Mo-V reactor pressure vessel steel"

22 September 1993

09:00 Session E - Mitigation of Irradiation Effects
Chairmen: Mr Miannay, France
Mr Brumovsky, Czech Rep.

1 Törrönen K.-Pelli R.-Planman T.-Rintamaa R.-Vallo M.-Wallin K. (Finland) - "Irradiation embrittlement mitigation"
Hiser A. (USA) "Proposed NCR rule package on reactor vessel integrity and annealing" (USA)

23 September 1993

09:30  Session G - Final Session
Chairman: Acad. L.M Davies, UK

Conclusions and Recommendations

12:30  Closure of the Meeting

LUNCH