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Scientific Issues in Fuel Behaviour

A report by an NEA Nuclear Science Committee Task Force

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PUBLISHER'S NOTE

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- *developing exchanges of scientific and technical information particularly through participation in common services;*
- *setting up international research and development programmes and joint undertakings.*

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Credit of cover picture: Fission gas bubbles at grain boundaries in UO_2 (photo: CEA - Grenoble)

FOREWORD

The OECD/NEA Nuclear Science Committee (NSC) has long recognised the need for improved knowledge and international co-ordination of important scientific issues related to fuel behaviour. Consequently a special Task Force was set up by the NSC in late 1993.

The objectives of the Task Force, as endorsed by the NSC, were to identify areas of high priority to Member countries which would benefit from international co-ordination and co-operation on studies of the basic underlying phenomena of fuel behaviour under normal operating conditions.

The Task Force was asked also to advise the NSC on developments needed regarding data, models and experiments to meet the requirements for better understanding of fuel behaviour and for improved predictive models.

The main findings and conclusions of the Task Force are summarised in this report. It is published on the responsibility of the Secretary-General of the OECD. The opinions it expresses are those of their authors only and do not represent the position of any Member country or international organisation.

Acknowledgments

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SUMMARY

High-priority Issues

The current limits on discharge burnup in today's nuclear power stations have proven the fuel to be very reliable in its performance, with a negligibly small rate of failure. However, for reasons of economy, there are moves to increase the fuel enrichment in order to extend both the cycle time and the discharge burnup. But, longer periods of irradiation cause increased microstructural changes in the fuel and cladding, implying a larger degradation of physical and mechanical properties. This degradation may well limit the plant life, hence the NSC concluded that it is of importance to develop a predictive capability of fuel behaviour at extended burnup. This can only be achieved through an improved understanding of the basic underlying phenomena of fuel behaviour.

The Task Force on Scientific Issues Related to Fuel Behaviour of the NEA Nuclear Science Committee has identified the most important scientific issues on the subject and has assigned priorities. Modelling aspects are listed in Appendix A and discussed in Part II.

In addition, the quality assurance process for performing and evaluating new integral experiments is considered of special importance.

Main activities on fuel behaviour modelling, as carried out in OECD Member countries and international organisations, are listed in Part III. The aim is to identify common interests, to establish current coverage of selected issues, and to avoid any duplication of efforts between international agencies.

Recommendations

Important conclusions and recommendations of the Task Force include:

- Countries and organisations should make special research efforts to reduce further uncertainties in the modelling of thermal behaviour of fuel and to improve modelling at high burnup. The following aspects should be treated with the highest priority:
  - thermal conductivity,
  - fission gas release,
  - fission product swelling and uranium oxide creep,
  - thermomechanical behaviour
  - high burnup fuel in transient conditions.
- A first concrete step in this direction is a review of existing, well characterised experiments covering issues and phenomena of the highest priority. This work is already well under way and is published in a companion report. The Task Force recommends that this be followed by the setting up of an international database with selected experimental data.
- This database should be available for international model validation and computer code benchmarking exercises, providing an international standard for quality assurance of fuel rod models.
- It is strongly recommended that the database be maintained, updated and distributed by the NEA Data Bank.
- International topical meetings covering high-priority issues should be organised by the NEA in co-ordination with the IAEA.

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PART I

INTRODUCTION

1. Background

The OECD/NEA Nuclear Science Committee (NSC) proposed in 1993 “Scientific Issues in Modelling Fuel Behaviour” as a topic for future investigation. Modern reactor fuel in operation today has proven extremely reliable with a very small failure rate. However, extension of plant life is now being considered, together with higher enrichment and higher discharge burnup in the fuel: this aims at improving the economics of the nuclear fuel cycle, but also places stricter demands on the performance of fuel.

The NSC concluded that it is very important to improve predictions of material performance limits in the environment of nuclear reactors; this is especially needed for high burnup fuel and for predicting reactor life-cycles.

From a first overview the NSC inferred that much of the ongoing research was of an empirical nature and that there is a need for a better understanding of the underlying basic phenomena. Consequently it decided to set up a *Task Force on scientific issues in fuel behaviour* which should work in consultation with the Halden Reactor Project, IAEA and other international programmes.

The Task Force has held two meetings, the first one in Paris on 8 December 1993 and the second one in Halden on 7 November 1994.

Task Force Members and co-authors of this report are listed in Appendix A.

2. Scope, objectives and work programme

The scope and objectives as approved by the Task Force and the NSC are the following:

- identify areas of high priority to Member countries which would benefit from international co-ordination and co-operation concerning the basic underlying phenomena of fuel behaviour under normal operating conditions;

- provide advice to NSC on the developments needed (data, models, experiments) to meet the requirements for a better understanding of fuel behaviour and for improved predictive models.

The work programme of the Task Force aimed at producing the present report in which important scientific issues in fuel behaviour are identified, their priority established and concrete actions recommended to the NSC including the type of co-operation and information exchange envisaged.

This report concentrates on phenomena rather than on specific fuels. For each topic, modelling requirements are specified and clarified. Particular attention is paid to modelling assumptions and possible developments in the statistical and mechanical models.

3. Selection of issues

Reliable predictions of fuel behaviour are important in order to achieve improved design and economics of the nuclear fuel cycle. In particular, modelling the long-term fuel rod behaviour during irradiation implies extrapolation of models from current to higher exposure levels, supported by experiments.

Higher discharge burnup may cause major microstructural changes near the edge of the fuel rod and a general degradation of the thermal conductivity; also waterside corrosion and concomitant hydriding impair both the thermal conductivity and mechanical properties of the Zircaloy based cladding. These and other related phenomena can become life limiting for the materials involved. Reliable predictions require a better understanding of the basic phenomena underlying fuel behaviour.

While this report is limited to normal operating conditions, it is pointed out that burnup effects may have strong implications on the fuel behaviour in operational and off-normal transients and accidents. Changes of fuel properties will affect both the initial state and the fuel behaviour during the transient itself. References to high burnup fuel behaviour in transient and accident conditions are made in Appendix B, prepared by S. Kelppe, *VTT*. The conclusion is that there may be an urgent need to elaborate upon several of these issues.

Futuristic approaches were discussed; although of interest, it was agreed that they are not of high priority. They require in addition a big leap forward in basic understanding of fuel behaviour phenomena. It was therefore decided not to include this issue.

The scientific issues in fuel behaviour under normal operating conditions considered of major importance by the task force are given in Part II and listed together with their authors in Appendix A.

PART II

SCIENTIFIC ISSUES AND MODELLING ASPECTS

Chapter 1

Thermal Analysis

Fuel rod behaviour is determined by complex thermal, mechanical, physical and chemical processes depending on design and operational parameters, material selection, loading conditions, burnup etc. All phenomena are highly interdependent and need to be modelled to quantify the complex fuel rod behaviour in all situations. Here we concentrate on the thermal analysis providing local temperatures and temperature gradients that are of relevance for nearly all the processes mentioned above.

The thermal analysis in a fuel rod is governed by the heat conduction equation

$$(1) \qquad c\rho \frac{\partial T}{\partial t} = \nabla \bullet \lambda \nabla T + q''' $$

from which it can easily be seen that the local temperature T depends on local material properties (thermal conductivity λ, specific heat c, density ρ) and the local power density q'''. It also depends on time-dependent boundary conditions and heat transfer coefficients (coolant-to-fuel rod and gap conductance). No single parameter dictates the behaviour of a fuel rod for all real or postulated conditions. Nevertheless, under steady-state normal operation the gap conductance and the thermal conductivity λ dominate. However, this does not imply that the specific heat c and density ρ are of minor relevance. The heat capacity is important in transient conditions and in most cases the thermal conductivity is only indirectly obtained from measurements of diffusivity; these two properties are of equal importance.

1.1 Thermal conductivity of LWR fuel

The thermal conductivity of LWR fuel has been extensively investigated, both theoretically and experimentally. A pioneering work on UO_2 is that of H.E. Schmidt [1] and the theoretical understanding is summarised by Hyland [2]. The many correlations which have been suggested for practical use in fuel rod performance codes need not to be discussed here. Three contributions are identified:

conduction through lattice vibrations (phononic term), conduction through free electrons and a small contribution due to radiation. It is generally accepted that at lower temperatures the phononic term

$$(2) \qquad \lambda_{phonon} = (a + bT)^{-1}$$

dominates. The constant *a* may be interpreted as the thermal resistance due to scattering by residual impurities and to scattering from lattice dislocations and grain boundaries (lattice imperfections). The constant *b* may be interpreted as the thermal resistance due to phonon-phonon interactions [2] & [3]. At higher temperature the electronic contribution is of relevance. For this term different correlations are used; for example,

$$\lambda_{electronic} \propto T^3$$

$$\lambda_{electronic} \propto e^{c_T}$$

are simple correlations for the electronic contribution to λ. Together, both contributions lead to a minimum of the total thermal conductivity λ at temperatures around 2000 K. Experimental data above 2000 K is scarce, difficult to obtain and therefore uncertain. A recommendation for the thermal conductivity of UO_2 has been given by Harding and Martin [4]. The thermal conductivity of molten UO_2, needed in reactor safety analyses, is even more uncertain. A value of $\lambda_{molten} = 1\text{-}2$ *W/mK* was suggested by Tasman [5]. This value is lower than the thermal conductivity of solid UO_2 prior to melting which is ≈ 3.5 W/mK.

Beside the dependence on the temperature, the thermal conductivity of UO_2 depends on the porosity *P*, the oxygen-to-metal ratio *O/M* (stoichiometry) and the burnup *bu*. All dependencies have been known for nearly 20 years [6]. Since point defects (oxygen interstitials or vacancies) and fission products act as phonon scatterers, an extended phononic term of the form

$$(2a) \qquad \lambda_{phonon} = (a + a_x x + a_{bu} bu + bT)^{-1}$$

seems to be justified, where $a_x x$ accounts for the effect of hypo- or hyperstoichiometry and $a_{bu} bu$ for the burnup effect. The variable *x* is the stoichiometric deviation *abs(O/M-2).*

Recently, a new dependence of λ was found which results from a clear structural change of the UO_2 fuel near the surface at high burnup, in the so-called “rim zone”. Thus, the burnup effect consists of two contributions: a general degradation in the thermal conductivity of the fuel and the development of the “rim zone” which may act as a thermal barrier. In the following all dependencies are briefly discussed.

a. The effect of porosity

Porosity in a ceramic material invariably decreases the thermal conductivity. Geometry (morphology, size, shape, orientation) and physical properties (emissivity of the solid, properties of the

gas trapped inside the pores) are of importance. In general the effect of porosity is taken into account by a correction factor f_p

$$\lambda (P) = \lambda_{100} f_p, \quad (3)$$

where λ_{100} denotes the 100% dense material. Correction factors f_p are discussed by Winter and Macinnes [3] and by Hayes and Peddicord [7]. A typical correction factor is $f_p = (1-P)^{2.5}$. The Finite Element Method (FEM) is used by Bakker et al., [8], to determine the effect of porosity on the thermal conductivity of UO_2. This method takes the real shape, orientation and size distribution of the pores of the fuel cross section into account. From these computations a constant value of the porosity correction factor can be determined. This constant depends strongly on the microstructure. The problem, however, is the determination of the details needed. At the beginning of irradiation a given fabrication porosity (pores with a given size distribution and morphology) exists, which decreases during irradiation. Simultaneously, depending on irradiation conditions, new classes of porosity evolve, mainly at grain boundaries but also in the interior of the grains. In addition, macroscopic and microscopic cracking of the fuel takes place, which provides further internal volumes.

Consequently, porosity correction factors will always remain a source of uncertainties and need permanent attention.

b. The effect of stoichiometry

Hypo- and hyperstoichiometric fuel, i.e. fuel with a non-zero value of x has a lower thermal conductivity than stoichiometric fuel since introducing point defects (vacancies or interstitials) increases phonon scattering. A typical value is $a_x = 3.60$ *mK/W*. Philipponneau gives a different form of a correlation for $(U,Pu)O_{2-x}$ [9]. As in the case of the porosity correction, the difficulty arises from how to obtain the necessary details of the local stoichiometry. The redistribution of oxygen in non-stoichiometric fuels is not completely understood. Chemical processes certainly complicate the picture.

Consequently, the effect of stoichiometry needs permanent attention.

c. The degradation effect of burnup

The introduction of solid fission products and the formation of fission gas bubbles should decrease the thermal conductivity λ. This expected decrease of the thermal conductivity of UO_2 with burnup has been studied for a long time on real irradiated fuel and by doping fresh fuel with a wide range of fission products in both metallic and oxide form. Doping experiments were performed (among others) by Schmitz et al. (1970), Kleykamp (1970) and Hartlip et al. (1973). Their work is discussed in the papers of Phillipponeau [9] and Lucuta et al. [10]. Studies on irradiated fuel were first performed in the seventies for Fast Reactor fuel where a high burnup was anticipated. However, the expected degradation of the thermal conductivity with burnup was either not confirmed or only a small effect was found. The reason was that most of these irradiation experiments were inconclusive because of the many interacting phenomena (densification and swelling, formation of a central void, high fission gas release etc.). Today we have clear evidence for the degradation of the thermal conductivity λ with burnup from three sources:

- Fuel centre line temperature measurements performed at Risø on re-fabricated fuel rods with different designs and burnup levels [11];

- Fuel centre line temperature measurements performed at Halden on specifically designed fuel rods with a small pellet-to-clad gap, negligible fission gas release and fuel restructuring proved the degradation of λ with burnup. A reduction of 6-8% per 10 MWd/kgUO_2 was found by Kolstad and Vitanza;

- Measurements performed on SIMFUEL (*SIM*ulated high burnup nuclear *FUEL*) with an equivalent burnup of 3 and 8 at% provided data of the intrinsic conductivity (i.e. without gas bubbles, cracks etc.) and proved a degradation of the thermal conductivity λ with burnup. The reduction was approximately linear with burnup, Lucuta et al. [10].

Direct centre line temperature measurements on irradiated fuel rods give only an integral information over the entire fuel rod, whereas the measurements performed on SIMFUEL give local values.

All findings are in agreement with what is expected from theory. It can be concluded from the general equation (2a) that the degradation of the thermal conductivity λ with burnup is more pronounced at lower temperatures. Since FBR fuel rods operate at higher temperatures, the degradation is lower than for LWR rods, which may explain the findings from FBR irradiations. Kolstad and Vitanza, [12], obtain the factor $a_{bu} \approx 0.014\text{-}0.016$ *mK/W per at%*; Lucuta et al. obtain almost the same factor although their correlation is more complicated. The value deduced by Phillipponeau, $a_{bu} = 0.0044$ *mK/W per at%*, is lower, however the author indicates a high uncertainty.

d. The effect of high burnup at the pellet surface ("rim")

At the pellet surface of LWR fuel the local burnup is enhanced due to the built-up of plutonium. Above a local threshold burnup of ≈ 68 MWd/kgU, microstructural changes (loss of optically definable grain structure, i.e. low angle subgrains on a very fine scale, change of porosity) and both a lower dislocation density and lower density of intragranular fission gas bubbles and precipitates were observed, [13] to [15]. Because of the microstructural changes and the high quantities of fission gas present in this zone, there is a potential for athermal fission gas release. The thermal conductivity of the rim zone is unknown. An increased porosity and a lower concentration of fission gas in the matrix (i.e. a higher concentration of fission gas on the grain boundaries) implies a lower thermal conductivity than would be expected from the burnup effects discussed in section 1.1.c. Since the heat flux density is highest at the pellet surface, such a rim effect would further increase fuel temperatures. A first attempt to quantify this effect was undertaken by C. Bagger, M. Mogensen and C.T. Walker. [16]. The authors used direct centre line temperature measurements, temperature "markers" (grain growth and xenon diffusion data) to construct a radial temperature profile. From known boundary conditions they concluded that the thermal conductivity of the rim zone is considerably lower (factor 5 to 10) than in the rest of the fuel, thus the rim zone acts as thermal barrier.

1.2 Thermal conductivity of the cladding

The thermal conductivity of the cladding is well established. However, the thermal conductivity of zirconium oxide needs further clarification.

1.3 Heat transfer coefficient between coolant and cladding

The heat transfer coefficient between coolant and cladding is a classical problem. The relevance is in connection with corrosion which is a very temperature sensitive process. The problem is being addressed in a number of research programmes and no further actions need to be undertaken.

1.4 Heat transfer coefficient between fuel and cladding

The heat transfer coefficient between fuel and cladding (gap conductance) is one of the important contributors defining the temperature of the fuel. At high burnup where the gap is closed, the influence is not as pronounced as at the beginning of irradiation. Nevertheless, the gap still acts as a thermal barrier. Gap conductance models depend on temperature, emissivity, gas composition, gas and contact pressure on surface morphology (roughness, waviness) which may change significantly at high burnup. Consequently, the surface morphology and composition of LWR fuel and cladding should be investigated. Similar to the discussion of the influence of porosity and stoichiometry on the thermal conductivity of the fuel the problem lies in applying the right data for gap conductance models. At the beginning of the irradiation where the gap is open the gap size is determined by highly uncertain relocation of the pellet fragments. At high burnup the integral effect of all irradiation processes, especially gas release (influence on gas composition) and swelling (influence on contact pressure) contribute to an uncertainty of the gap conductance.

Consequently, the gap conductance needs further investigation, especially at high burnup.

1.5 Conclusions and recommendations

1. The thermal conductivity of LWR fuel has been extensively studied. Clearly*, local measurements* at different burnup levels and temperatures are needed in order to better determine the influence of the accumulation of fission products and changes in stoichiometry. All details of the degradation of the *local thermal conductivity* need better experimental support which must be accomplished by a better theoretical understanding. The thermal conductivity of the rim structure is unknown and needs to be investigated. These recommendations are in complete agreement with the recommendations of Howl made in a specific summary report on fuel rod chemistry and related properties [17]. Local measurements of irradiated fuel are planned at the Institute for Transuranium Elements. All investigations should be extended to gadolinia doped fuel.

2. The surface morphology and composition of LWR fuel and cladding at high burnup should be investigated. The gap conductance at high burnup needs further clarification.

3. The thermal conductivity of zirconium oxide needs further clarification.

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Chapter 2

Fission Gas Release

The natural consequence of the fission process is the splitting of the fissile nucleus into two fragments which eventually become new atoms with appropriate chemical behaviour. The maxima in the doublet distribution of mass numbers occur in the regions 90 - 100 and 140, thus giving high yields to the rare gas species krypton and xenon and the volatile species iodine and caesium. Concerns arise when theses species are released from the fuel matrix because they challenge the clad integrity both through increased internal pressure and also Stress Corrosion Cracking (SCC). Because krypton and xenon have lower thermal conductivities than helium, the release of these gases also causes a worsening in the pellet-clad gap conductance and increased fuel temperatures. The trend towards higher burnup and connected high fission gas release raises the question of operation at internal overpressure that can induce cladding lift-off and gap reopening, events that can occur at very high burnup end of life operation. Although fission gas release has been studied for many years, and the basic mechanisms understood, the subject still presents difficulties in modelling particularly in the construction of a model that handles with the same accuracy, steady-state and transient conditions.

2.1 Low temperature, low burnup [1]

For steady-state operation of fuel at standard power and burnup (20 kW/m and 40 MWd/kgU), the main mechanism for fission gas release (FGR) is considered as “athermal”. It is indeed weakly temperature dependent.

The mechanisms involved are the following:

- *Recoil* [2], where a fission product close enough to a free surface ($d < 6/7\mu m$) will escape from the fuel, due to its high kinetic energy (60 to 100 MeV). These atoms are mainly trapped in the cladding but some will be released in the gap.

- *Sputtering* [3], when the above described fission product travels through the oxide, it looses its energy by “electronic” and “nuclear” interactions with UO_2 at a rate of

$$\sim \delta E / \delta x = 1 \text{ keV/Å},$$

leading to a high local heat pulse along its path. When it leaves or enters a free UO_2 surface, the heated zone will evaporate (*sputter*) [4]. All the volatile fission products available in this volume are released in the gap. Due to the ionic bounding of UO_2, *no quantitative description of this phenomenon exists today*. Since this process is the main process of FGR in a power plant, knowledge and modelling of this physical process is required.

These two processes lead to different release rates for the various radioactive species:

for *recoil* the release rate is independent of decay half-time

$$R_R \propto \lambda^0,$$

while for *sputtering* it is as

$$R_S \propto \lambda^{-1},$$

closer to the experimental value of $R_{exp} \propto \lambda^{-0.7}$.

2.2 Intermediate temperature, higher burnup [5]

In the centre of the pellet some diffusion can occur and fission gases diffuse from the bulk of the grains to the grain boundaries. When the temperature is high enough, bubbles will nucleate, grow and interlink for the gas to escape. At intermediate temperature (700-800 °C), thermal activation is too low to allow bubble nucleation and the limited amount of fission gas, that diffused to grain boundaries, is trapped by the grain boundaries and remains as segregated atoms.

2.3 Power transients

This segregation embrittles the grain boundaries and enhances intergranular fracture induced by any thermal stress. Thus, after a long steady-state operation, the grain boundaries are charged in fission gas, and any change in power leads to cracking of the fuel and release of the segregated fission gas. The higher the burnup, the more efficient the process. *Modelling this process of fission gas release, including fission gas diffusion, grain boundary embrittlement and thermomechanical cracking, would be of high value*. In addition, the modification of this process when several transients have to be considered, is also important due to the more frequent load follow operation. How are the grain boundaries reloaded with fission gas, when will the oxide reopen, etc.? [6]; improved understanding of this mechanism is needed.

2.4 Moderate power

In the range of 900-950°C, diffusion controlled processes are more active and lead to a higher FGR rate. Pure diffusion should lead to a $R_D \propto \lambda^{-1/2}$ law, but due to some contribution of sputtering release, in the outer part of the pellet, to $R_{exp} \propto \lambda^{-0.7}$.

Thus a good modelling of FGR in a fuel rod should consider all types of mechanisms.

2.5 High burnup

In the specific case of very local burnup (above 100 MWd/kgU) found in the rim and in (U, Pu)O_2 aggregates in MOX fuel above 35 MWd/kgU (rod average), the microstructure of the oxide is completely transformed, the grain size drops to ≈ 1μ, a high density of fission gas bubbles develops and a new mechanism of FGR by microstructure change seems to occur. A detailed understanding of the mechanisms involved (fission product super saturation, potential $P_{(O_2)}$ changes inducing a change in UO_2 microstructure or other...) is required for the development of high burnup fuel [7].

2.6 High temperature

Fission gas bubble nucleation at grain boundaries or within the grains (see chapter 3. on Swelling) is physically understood but extremely difficult to correctly forecast. Transmission Electron Microscope (TEM) work is underway to analyse the initial steps of bubble nucleation and to define a critical stable gas nucleus on UO_2.

2.7 Proposed experimental support

Sputtering: • Using UO_2 pellets, an irradiation with heavy ions (as in GANIL, Caen, France) having masses and energies similar to fission products (Xe, ~ 60 MeV) is proposed, the sputtering rate versus heavy ion energy, incident angle, UO_2 stoichiometry should be counted.

Staff at GANIL consider the experiment as an easy one to perform;

• Modelling of the phenomenon could also be carried out using molecular dynamics computation. A lack of valuable interatomic potential in UO_2 could make the computation difficult.

2.8 Proposed analytical work

The new softwares developed for advanced chemistry, by “ab initio” computations, could be used to compute the chemical state of the xenon atom in the UO_2 matrix in various interstitial, substitutional or single- or multivacancy sites. The knowledge of the energy of each configuration will allow a better description of the precipitation kinetics of the fission gas in the irradiated fuel.

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Chapter 3

Fission Product Swelling

Experimental measurement has shown that during irradiation the volume of UO_2 fuel changes continuously with burnup. The two major implications of this are that whilst the fuel to cladding gaps are open, as at the beginning of irradiation, swelling of the pellet tends to decrease the gap dimension providing a benefit by decreasing fuel temperatures. On the other hand, when the gap is small or closed, fuel swelling during an inadvertent overpower transient contributes towards the mechanical stressing of the cladding and under severe conditions can lead to eventual failure.

Initially, at the start of irradiation, there is a contraction in volume as pores remaining from the sintering process continue to shrink. This is most pronounced in low density fuel, especially if the porosity is small, typically less than one micron diameter. This process quickly saturates and is followed by a monotonic increase in volume as more and more fission products take the place of the fissionable uranium. This swelling has several identifiable components:

1. solid fission products,

2. fission gas as individual atoms,

3. fission gas precipitated into intragranular bubbles,

4. fission gas in intergranular bubbles (situated on grain boundaries).

The first two are termed “inexorable swelling” since they cause a volume change which is dependent only on burnup. Swelling caused by the formation of fission gas bubbles (points 3 and 4) only occurs at temperatures sufficiently high to permit atomic migration. The largest contribution to fuel swelling comes from intergranular bubbles, and it is these that are formed during high power transients and contribute along with thermal expansion strains to distend the cladding which in severe cases can ultimately result in rod failure.

The following sections give a brief description of the swelling contributions, data providing quantitative values and indicating dependence on material and irradiation parameters, a selection of modelling approaches and finally the requirements for better quantitative treatment of the phenomenon.

3.1 Solid fission product swelling

An early analysis of this topic was conducted by Anselin [1] and reviewed by Olander in his book [2]. The fission products were divided into groups as follows:

- Soluble fission products e.g., Nb, Y, Zr,

- Metallic inclusions e.g., Mo, Ru, Te, Rh, Pd,

- Other fission products e.g., Cs, Rb, I, Te, Ba, Sr.

These contributed to a change in volume estimated to be around 0.32% per 1% FIMA, (1% FIMA = 10 MWd/kgU). To this should be added the contribution from the rare gases krypton and xenon when in isolation in the lattice. For a fission yield of 25 atoms per 100 fissions and an atomic volume of 85.10^{-24} m^3, these contribute to give a total inexorable swelling from all isolated fission product atoms of around 0.84% per 10 MWd/kgU. This is in excellent agreement with experimental data reviewed by Franklin [3] and obtained from length measurements in Halden reactor experiments (see for example [4]) of 0.8 to 1% per 10 MWd/kgU.

Thus for this contribution to volume change it is necessary only to assume independent of material and irradiation conditions a volume change of between 0.8 to 1% per 10 MWd/kgU. As a possible refinement for regions of fuel experiencing high temperatures, in transient conditions for example, a reduction in this swelling component could be made equivalent to the fraction of fission gases released. Thus for every gas atom released from the fuel, a volume of 85.10^{-24} m^3 should be subtracted.

3.2 Fission gas swelling

By far the largest contribution to volume change arises from the formation of fission gas filled bubbles on the grain boundaries. Even from the earliest days optical microscopy of cross sections of fuel rods operated at high temperatures revealed the presence of grain boundaries “decorated” with cigar shaped pores. However, it was not until the advent of Scanning Electron Microscopy (SEM) that the true nature of this porosity was established.

Examination of fractured surfaces of irradiated fuel by Reynolds and Bannister [5] showed large bubbles a few microns in diameter on grain faces and also along grain edges. At high exposures and temperatures, the grain edge bubbles interlinked to form tunnels, often penetrating the entire grain edge length between four point corners.

The morphology of this tunnel network was studied extensively by Beere and Reynolds [6] and Tucker and Turnbull [7] and [8]. It was concluded that bubbles were first formed on the grain faces by gas atoms diffusing from the grain interior and that the transfer of gas and porosity from these to grain edges was responsible for the formation of the tunnel network. In this way, the acceleration of fission gas release after a given burnup and temperature could be explained as the interlinkage of grain edge tunnels to the free surface. The theory also established that a minimum grain edge swelling of around 5% was necessary to permit a stable network of open tunnels to exist and that this was the criterion necessary for the increase in surface to volume ratio observed in in-pile experiments measuring the release of radioactive fission gases, see for example [9].

a. Experimental data

Although there have been many observations of fission gas swelling there are few with sufficient information on irradiation history and accuracy of measurement that can be of use in formulating a model or validating a code for swelling predictions. Principal sources of early data on steady-state high temperature swelling as used by Hollowell [10] to derive his swelling model are given in Table 3.1.

Zimmermann [11] measured the change in volume after isothermal irradiation of both restrained and unrestrained UO_2 samples. He found that the observed swelling and swelling rate in unrestrained samples was strongly dependent on temperature. The initial rate of swelling at around 1000°C was 1.2% per 1% FIMA whilst at 1700°C it was 20% per 1% FIMA. The swelling increased linearly with

burnup, saturating at just over 10% at 12% FIMA for samples irradiated at 1000°C and 20% at 2% FIMA for samples irradiated at 1700°C. Restrained samples showed a much reduced dependence on both temperature and burnup, saturating at just over 5% and were virtually unaffected by the level of restraint at least up to the maximum imposed value of 50 MPa.

Turnbull [12] irradiated specimens of two different grain sizes at 1750°C up to 0.4% FIMA. He found that swelling was less in large grain size material and that above 7-10% swelling a significant fraction of the porosity was in the form of interlinked tunnels in accord with the theoretical study.

More recently well qualified data on swelling during power ramps have become available through international projects, in particular, the three Fission Gas Release Projects carried out in Risø, Denmark. Here well characterised fuel irradiated in Halden and American BWRs and a PWR were subjected to power ramping to powers around 45 kW/m and held for several hours before unloading and extensively examined by a variety of destructive and non-destructive techniques. Of particular value were the data generated by the third and final set of experiments where the in-pile instrumentation during the power ramp included fuel centreline thermocouples. Unlike the experiments given in Table 3.1, where, apart from a set of Zimmermann's samples, the fuel was unrestrained during the irradiation, the fuel rod geometry and closed gaps at the start of the power ramping in Risø means that the swelling values are those for conditions of restraint where the stresses are imposed by the clad and system pressure. The range of parameters covered in the third project is given in Table 3.2.

Local swelling measurements were made at various locations across pellet diameters and could be correlated with the integrated pellet swelling determined from increase in rod diameter. Typically the diametral changes were in the region of 0.2 to 1% for each fuel type with maximum local fuel swelling values of 5-10% depending on ramp terminal power and hold time.

Table 3.1 **Data base used by Hollowell** [10] **to derive an unrestrained swelling model**

Experiment	**UO_2 grain size** *μm*	**Temperature range** *°C*	**Burnup** *MWd/kgUO_2*	**UO_2 density**	**Fuel Form**
Zimmermann [10]	10	900-1725	3.0-42.0	95	Pellet rings 5.1 mm/2.2 mm OD/ID 1 mm high, also restrained sample 3.0 mm OD 5.7 mm high
Hilbert, Storhok et al. [11]	10-20	1375-2140	2.2-6.6	97	Annular pellets 5.3 mm dia.
				90	Solid pellets 5.3 mm dia.
Turnbull	7 & 40	1750	1.1-2.2	98	Cylinders 3 mm dia. 10 mm long

Table 3.2
Range of parameters covered in the test matrix of the Third Risø Fission Gas Release Project

Test identifier	**Burnup** *MWd/kgU*	**Fill gas composition**	**Rod average power in test** *kW/m*	**Hold time in** *hours*	**Centre Temperature** *T/C °C*
PWR fuel ANF manufacture	43-44	He: 6 rods Xe: 1 rod	16.9-40.7 29.8	4-62 42	1225-1425 1500
BWR fuel GE manufacture	24-46	He: 4 rods	35.5-43.3	4-140	1520-1640
BWR design fuel irradiated in HBWR	16-53	He: 4 rods Xe: 1 rod	40.1-44.7	2-36 24	1500-1575 1800

b. Modelling of intergranular swelling

It is clear that the processes involved in fission gas release and concomitant fuel swelling are many and varied. Providing an adequate mechanistic description of these processes has occupied many man-days of research over the past 30-40 years. The approaches adopted for modelling fuel swelling fall into two broad categories, the first is empirical, using linear regression against an identified data base whilst the second attempt to provide a mechanistic formulation benchmarked against data where parametric values were available.

A good example of an empirical approach is that adopted by Hollowell [10]. Hollowell assumed that fission gas swelling started at 900°C and inspection of the database presented in Table 3.1 lead him to propose an equation for unrestrained swelling based upon temperature, burnup and grain size as follows:

$$\Delta \mathbf{V/V\%} = 2.413.10^{-3}\, T_1 + 1.0634.10^{-5}\, T_1^{\,2} + 0.11509\, B + 2.86.10^{-3}\, T_1\, \mathrm{Ln}(B) - 2.0184.10^{-3}\, T_1\, \mathrm{Ln}(G)$$

where: T_1 = (Local Temperature - 900)°C
B = Burnup in MWd/kg UO_2
G = UO_2 grain size in microns.

This correlation was specifically for unrestrained swelling during near constant irradiation conditions and was applied successfully to the calculation of pellet-clad gap closure in order to calculate gap conductivity and hence fuel temperatures. Thus the correlation had only limited application and was not intended for use where restrained swelling was needed, for example, in calculating clad strain and hence failure propensity in overpower transients.

An early example of a mechanistic treatment of fission gas swelling was in the UKAEA code MINIPAT, [14]. Here it was assumed that gas released from grains accumulated into a fixed areal density of lenticular bubbles situated on grain boundaries. The capacity of each bubble was dictated by the geometry of the bubble, the UO_2 surface energy and the applied external hydrostatic pressure. Thus if the bubble density was specified, the hydrostatic pressure known, then the bubble size and

hence the swelling per unit volume of fuel could be calculated from the amount of gas released to the grain boundaries.

Although this model was successful in predicting the level of swelling in WAGR hollow pellet fuel, it was clearly inadequate when compared with data from the Risø Projects. There were four major developments needed to this approach before a satisfactory mechanistic swelling model was devised and included in the Nuclear Electric version of the ENIGMA code. The first was an improvement in the gas release model such that both steady-state and transient conditions could be treated without recourse to a “flag” or switch of models. A brief description of this model is given by White [15].

The second was an independent treatment of the formation of tunnels, and this was based on simplifications to the geometry investigated by Tucker and Turnbull [7] & [8] and discussed by White and Tucker [16]. In this way both grain face and grain edge porosity are treated separately with different swelling kinetics.

Based on observations made in the second and third Risø Fission Gas Release Projects, the capacity of grain face bubbles was increased such that they behaved as quasi crystalline with the bubble capacity now independent of surface energy or hydrostatic restraint. This was necessary since a comparison of xenon retention profiles measured by Electron Micro Probe Analysis (EPMA) and X-Ray Fluorescence (XRF) could only be rationalised if the grain boundary capacity for retaining fission gas was substantially greater than hitherto assumed.

The swelling rate dependence on restraint was reserved for the grain edge porosity only. The final development therefore was to embed the swelling model in a code structure that calculated the stress and strain state of the pellet and cladding and hence evaluated local hydrostatic pressures. The elements of this model devised by White are given in [15].

The swelling model in ENIGMA outlined above is very much more complicated than the single equation derived by Hollowell. Also, the model cannot stand alone as it requires input from other routines within a code to calculate the fission gas atom flux to the grain boundary and the local hydrostatic stress. This development was necessary in order for the model to be generally applicable directly to: a variety of irradiation conditions, both steady-state and transient, open and closed pellet-clad gap, solid and hollow pellet geometry.

3.3 Future requirements

A fuel swelling model is a necessary component in any fuel performance code. Its purpose is to reflect the dimensional changes of the pellet during the course of irradiation and in doing so, in concert with clad creep down, to calculate the rate of closure of the initial fuel-clad gap. This is an important step in order to provide a good estimate of the evolution of temperature within the pellet as irradiation proceeds and to calculate the time for fuel-clad contact. Under transient conditions, where high power may be sustained for periods to promote fission gas release and the growth of porosity, the swelling model evaluates the strain additional to thermal expansion which impose tensile stresses in the clad which under extreme circumstances could ultimately lead to failure and the release of fission products into the coolant circuit.

For a comprehensive treatment of these effects the goal must be the development of a mechanistic model. Although data are available covering many aspects of swelling, enlargement of the database is desirable to improve the current swelling models and necessary if other than standard pellet geometry and fuel composition and microstructure are to be addressed.

Although the present database covers the effects of grain size and restraint, further data are needed as a function of burnup particularly during power ramp conditions. The effect of restraint is of relevance to different pellet geometries since the hydrostatic pressure is very dependent on the volume available to accommodate the change in pellets. In a solid flat ended pellet all the swelling is directed outwards both axially and radially thus cladding must accommodate the total integrated strain. With hollow pellets the swelling is directed inwards to fill the bore as well as outwards to strain the cladding. Whether or not the swelling is accommodated by outward clad strain or inward bore closure depends on the creep rate of the fuel and its temperature dependence. For very “soft” fuel, all the swelling can be accommodated by bore closure with no appreciable clad strain. A similar effect can occur for solid but dished pellets, where the volume of the dish can accommodate the swelling provided the creep rate is fast enough. Thus the competition between swelling and fuel creep is necessarily linked for a full quantitative description of pellet dimensional changes. To provide information on this, it is necessary to irradiate and power ramp fuel rods of different pellet geometry and cladding thickness in order to provide different levels of restraint and to examine by destructive post irradiation examination (PIE) the amount of local swelling and the direction in which this swelling has been accommodated.

As new fuels are developed, so their swelling properties are expected to differ from those of standard products. Killeen [17] has shown that at low burnup, the unrestrained swelling of niobia and lanthanum oxide doped UO_2 of various grain sizes showed no dependence on dopant. Also, further experiments using large grain size chromia doped UO_2 irradiated to higher burnup behaved remarkably similar to standard UO_2 despite a sevenfold increase in grain size and a markedly different porosity structure (Killeen [18]).

In a recent paper, Howl, Palmer and Topliss of BNFL [19] have shown that rods manufactured with niobia-doped fuel pellets are possible pellet-clad-interactions (PCI) remedy fuel. The paper reports experimental data which show that niobia-doped fuel can withstand considerably larger power ramps without failure than is the case for standard undoped fuel. Enhanced creep of the doped fuel into large voids associated with controlled fabrication porosity as well as into cracks and pellet dimples (dishes) was confirmed to be the mechanism responsible for the improvement. These references show that the presence of additives to standard fuel can affect the swelling and creep properties, thus to proceed with modelling of these and other variants, e.g. gadolinia-doped fuel and MOX fuel requires further dedicated experiments and modifications to the established principles of fuel property models.

In conclusion, it is considered that although no further data are required for solid fission product swelling and that a simple correlation is all that is necessary, further data are required on specific aspects of fission gas swelling. Discussion above has concentrated on intergranular swelling as this is the major contributor to dimensional changes, and although a useful database is available to develop and validate models, additional data are desirable especially for application to new fuel products. There is however, a second order contribution to fuel swelling from intragranular porosity, which can under certain circumstances be significant. Until now little attention has focused on this contribution, but as intergranular swelling becomes better understood and quantified, so it will become necessary to direct attention on the smaller diameter but more numerous intragranular bubble population and its contribution towards pellet dimensional changes.

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Chapter 4

Stress Corrosion Cracking

This type of fuel sheath failure mechanism was first reported in June 1963 during the start-up of the GETR at Vallecitos [1]. The failure occurred on a high rated fuel pin and, at that time, it was regarded as a form of chemical attack by fission products.

After this event, many other similar cases were detected in LWR, PWR, PHWR and CANDU fuel types triggering an intense research effort to better understand this failure mechanism. Initial research identified iodine as the most probable active species for stress corrosion cracking (SCC). Subsequent observations indicated that this type of failure was generic to zirconium alloy clad fuel that was subjected to significant power increases. Today, it is grouped with other types of pellet-clad-interaction (PCI) failure mechanisms observed to occur in zirconium alloy fuel cladding [2].

Power changes are inevitable during the operation of nuclear power plants; hence, the possibility of a large number of fuel failures ensured a large allocation of resources to better understand and solve this problem. The number of internationally sponsored research programs increased dramatically as a growing number of failures was attributed to this mechanism.

Cures were attempted based on the characteristics learned about the SCC process in zirconium alloys. Three type of solutions were widely applied: internal coatings (graphite or siloxane), barrier cladding (pure zirconium) and, equally important, a restriction on the reactor power changes and refuelling schedules. The incorporation of these solutions greatly reduced the frequency of SCC failures. Today, the number of SCC failures is well below the number of failures due to coolant debris.

The success of these remedies lead to a decline in the interest on this subject despite the fact that an understanding of the process at the mechanistic level is still unknown.

4.1 Stress corrosion cracking phenomenology

When the fuel failures were characterised as a manifestation of a SCC phenomenon, it was evident that the concurrence of four factors was necessary for the failure to take place: sufficient stress, sufficient time, a susceptible material and the presence of the right chemical environment.

a. Sufficient stress

The UO_2 pellets used in power plants have a cylindrical shape with length to diameter ratios often greater than unity. With increasing power, the pellet thermal expansion leads to an increase in the pellet diameter, i.e. a departure from the initial cylindrical shape due to hour glassing, and cracking of the pellet periphery. Also, during normal fuel handling, small UO_2 chips can relocate to the space between the pellet surface and the clad. All these processes, independently or in combination, can increase the stress in the cladding. In addition, the geometry of the cladding (wall thickness-to-diameter ratio), its creep rate under reactor operating conditions, and the presence of flaws on the internal clad surface all contribute to produce the resultant stress field. The stress intensities required

to crack irradiated cladding as observed in laboratory experiments, can in fact be reached during certain power ramps experienced during normal reactor operation.

The stresses imposed on the cladding arise from a combination of maximum power seen by the fuel due to expansion and the geometrical state of the pellets, and the change in power during the ramp (that establishes the increment in the clad strain over the period during which the power is raised). The relative importance and the critical levels of these two factors are strongly dependent on the fuel design itself.

Results of power-ramp test projects [3] [4] & [5] have shown the importance of the ramp rate (fast ramps are more severe than slower ones) and the hold time at high power.

All of these factors make the calculation of stress (or stress intensity) sustained by the cladding, and its time dependence, extremely complicated. Consequently, these calculations will always remain as a source of uncertainty, requiring further study.

b. Sufficient time

Based on the results of early laboratory experiments, the time to failure was divided into two periods: an incubation time and a propagation time. The incubation time was the period required for the formation of incipient cracks. The propagation time was the time required for these cracks to grow through the clad wall thickness. The majority of the time to failure is the incubation time, during which no apparent damage to the cladding is observed to occur.

To study these and other aspects of the SCC phenomena, several international in-reactor power-ramp projects have been conducted at Studsvik, Sweden, i.e. various results have been published for the Inter Ramp [3] & [6], Over Ramp [7], Super Ramp [8], Demo Ramp [9] and Trans Ramp [4] & [5] Projects. In the last of these ramp tests, it was observed that partial through-wall cracks can be produced after ramps of about 20 seconds whereas through-wall cracks were observed for ramps of less than a minute. However, these short incubation and propagation times could only be obtained if the tested fuel was irradiated at a relatively high power for at least six hours prior to the ramp. These results strongly indicate that preconditioning of the internal fuel environment is essential. Inferred values for the crack propagation velocities for these power ramp experiments agree with values measured in laboratory tests, giving velocities of the order of 10^{-6} m/s.

A review [10] of laboratory and in-rector experiments support the hypothesis that the incubation time is a manifestation of the chemical conditioning processes that may take place. If the right chemical environment is achieved prior to the stressing of the cladding, the incubation time is close to zero and the time to failure is predominantly due to the propagation time.

c. Susceptible material

A number of metallurgical variables have been studied for their effect on the susceptibility of the zirconium alloys to SCC. The search for a solution to this problem was initially directed towards finding a single metallurgical property, or a combination of properties that would produce a material that was more resistant to SCC attack. Among these properties, the most studied were: yield strength, texture, residual stresses, stress ratio, stress state and alloy composition. All of these properties have some effect on alloy susceptibility; however, all the improvements are drastically reduced with material irradiation.

After a critical irradiation dose, the positive effects of the clad metallurgical properties disappear and all materials become about equally susceptible to cracking [11]. The observation that Zircaloy tubing that will not fail in the laboratory but became susceptible when irradiated, led to the development of protective layers on the inner surface of the cladding.

Two types of protective layers have been extensively studied and implemented in fuel fabrication; a lubricant and chemical barrier (graphite) that is applied as an internal coating after tubing fabrication for CANDU fuel, and a metallic barrier (zirconium) that is applied in the form of an internal thin layer as part of the tube fabrication process for BWR fuel. Comparison between graphite coatings and zirconium barriers in typical BWR fuels [12] have found the two to be almost equally effective, although neither method offers a complete protection against severe power ramps after large irradiation doses.

d. Chemical environment

Due to its high yield, volatility and known properties of the halogens in causing SCC, iodine was first targeted as the chemically-active species in this process. Surveys [13] & [14] of other fission products as potential cracking agents showed that Cs [14] [15] [16] and/or Cd [17] were both capable of cracking zirconium alloys, with a mixture of these species being more potent than either one by itself. With iodine there are two potential crack propagation modes; transgranular pseudo-cleavage and the slower grain boundary attack by removal of volatile iodides. The latter propagation mode for iodine is typically the slowest one. SCC fractures in iodine-containing environments, although predominantly transgranular, always contain at least some intergranular features [10]. When zirconium alloys are cracked in Cs/Cd mixtures, the fractures appear to be entirely transgranular. Examination of PCI fractures in CANDU fuel that is refuelled on-power, always show some intergranular features; hence, it is concluded in these studies that iodine-induced SCC is the primary cause [18].

As the internal fuel chemistry plays a leading role, the presence of other species can act as either inhibitors or promoters. As mentioned earlier, graphite can act as a strong inhibitor. The replacement of graphite by siloxane as a chemical coating has proved to be even more effective. Oxygen has also been proved to be an effective inhibitor that may restore the protective oxide layers on most of the regions that are sensitive to the chemical attack. The presence of iron and organic iodides significantly increase the frequencies of crack initiation and shorten times to failure, thereby classifying them as promoters.

Although iodine has been extensively used in laboratory experiments to simulate in-reactor environments, it is now clear that the time to failure is critically dependent on whether or not a chemical precondition has been reached, the chemical form (speciation) of iodine is also critical to the cracking process. The work of Cubicciotti et al., [19] to [23], on binary zirconium iodides, and other SCC studies using metal iodides and zirconium iodides, show that the time-to-failures in ZrI_4 is much less than that in I_2. Consequently, the formation of the correct active species, the chemistry at the crack tip and the access of this species to the crack tip will ultimately determine whether or not a through-wall crack will develop.

In addition to all these characteristics, both laboratory and in-reactor power-ramp experiments have also indicated that the chemical environment in the fuel rod may change to an inert condition where crack initiation is not possible.

The chemical aspects of the SCC process are poorly understood and because of its dominant role in this process, it is the area where further theoretical and experimental studies are clearly needed.

4.2 Crack initiation and propagation.

Two plausible mechanisms have been identified for crack initiation. Firstly, a slow grain-boundary attack (intergranular cracking) may result in the local removal of material as a volatile compound, together with its subsequent deposition and regeneration of the aggressive iodine species. This deposition might occur within the crack or at its mouth, and could be a mechanism for the accumulation of impurities and alloying additions at these sites. Investigators have also seen extensive intergranular attack at the start of the cracking. Secondly, an initiation process, very similar to the transgranular crack propagation step, may occur at a group of grains whose basal poles are oriented parallel to the principal stress.

The mechanisms responsible for the transgranular crack propagation are less understood. The proposed mechanisms can only explain in part the observed fractographic features. Adsorption-induced cracking, where adsorption at the crack tip reduces the bond strength and permits pseudo-cleavage, is a possible mechanism, i.e. the similarities between transgranular cracking in iodine and Cs/Cd vapour, offers some support for this hypothesis. Restricted slip processes at the crack tip resulting from adsorption of the active species is another possible mechanism. This leads to localised microvoid growth and coalescence in a very restricted band at the crack tip. While evidence at present suggests this mechanisms as a transgranular propagation step, there is no evidence about the form of the iodine compound at the crack tip that can cause this effect.

More research on the identification of the transgranular crack propagation mechanisms is therefore needed.

4.3 Conclusions and recommendations

1. The estimation of the stress (or stress intensity) sustained by the cladding and its time dependence are extremely complex calculations that need further development;

2. The chemical aspects of the SCC process are poorly understood and the concentration of efforts on theoretical and experimental studies would be most beneficial;

3. More experimental work on the identification of the transgranular crack propagation mechanisms is also needed.

The incorporation of solutions to the SCC problem have greatly reduced the frequency of SCC failures. Today, the number of SCC failures is low and no economic incentives are clearly apparent for continued research in this field. However, the development of physico-chemical models for the prediction of operational thresholds are not possible within the current understanding. The assessment of future limits to performance requires an improved understanding of the SCC mechanism.

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Chapter 5

Constitutive Equations

5.1 Constitutive equations of fuel

The structural analysis of a fuel rod is governed by three basic mechanical principles: equilibrium, compatibility and constitutive equations. The constitutive equations, also called stress-strain relations or material equations, are as well known in their simplest form as Hook's law. A further specialisation of the generalised Hook's law is the case in which the material is assumed to be linear, isotropic and elastic. Although the theory of plasticity of a solid is already a very complex and broad subject, the fuel behaviour is much more complex than the behaviour considered by classical thermoelastic-plastic behaviour due to local swelling, densification and cracking. The fuel must be considered as anisotropic, with different behaviour under tension and compression. Plastic deformations may be caused within short time scales by high stresses and by lower stresses within large time scales. These stresses cause cracking of fuel pellets (macroscopic cracks) but may also cause cracking on microscopic scale (microcracks). The volume changes by densification and swelling are extremely complex since structural changes such as shrinkage of pores, formation of new bubble populations, grain growth and grain decoration and specific irradiation induced damage are involved. How to consider this complex material behaviour by a constitutive equation?

The presently adopted approach follows the classical approach by adding strain increments $d\varepsilon_i$ from different mechanisms to obtain the total strain increment $d\varepsilon_{tot}$, for instance:

$$d\varepsilon_{tot} = d\varepsilon_{elastic} + d\varepsilon_{thermal} + d\varepsilon_{plastic} + d\varepsilon_{creep} + d\varepsilon_{s\text{-}d} + \ldots$$

where the index *s-d* refers to swelling and densification. Traditionally, plastic and creep strains are distinguished and it is assumed that the volume is kept constant during a short time plastic or a long time creep deformation.

It is obvious that this concept of material behaviour has some limitations since all contributions are considered as independent. In fact, one of the general difficulties in measuring the behaviour of the fuel during irradiation is to single out one of the specific mechanisms.

5.2 Constitutive equations of cladding materials

The availability of mathematical models able to describe how materials behave under complex loading histories and in aggressive environments, is highly desirable. However, because of the physical complexity of non elastic deformation in metals and alloys, it is practically impossible to develop a complete mechanistic description of the observed macroscopic behaviour of materials (based on fundamental physical processes such as dislocation motion, immobilization and remobilization). The aim has been to study and to understand the physical mechanisms active at the microscopic level and their relation to the property changes enabling the development of models describing the material behaviour in terms of microscopical evolution.

Equations to describe the transient plastic strain rate ε of a metal or an alloy as a function of applied stress σ_a and temperature T are complicated, but in general can be written as:

$$\varepsilon = f(\sigma_a, T, S_1, S_2, ...) \qquad (1)$$

where S_1, S_2 ..... represent the state of the material microstructure affecting plastic strain. Microstructural parameters are: grain size, dislocation density, phase distribution, crystallographic texture, etc.

Already in the 1970's, significant results were achieved on constitutive equations for Zircaloys at the Stanford University (U.S.A.) under an EPRI co-ordinated project and by AECL in the development of CANDU fuel models.

The Stanford model, called MATMOD, [1], is a broad, general purpose, phenomenological model for the non elastic deformation of Zircaloy. A single strain mechanism, or a single unified strain equation (1) is assumed to describe the non elastic strain rate of the material, with three structure variable S_1 (friction stress due to deformation), S_2 (backstress), S_3 (friction stress due to solutes) which accounts for three specific categories of strengthening mechanisms.

The approach of the NIRVANA model, [2], from AECL, is different. This model is a microstructural model for transient deformation of Zircaloy cladding at high temperatures, which takes different strain contributions into account. A different form of equation (1) has been developed for each strain mechanism. At high temperature three deformation mechanisms were identified: athermal glide, grain boundary sliding and dislocation creep. A fourth component, transformation strain, has been found to occur in the alpha+beta phase field when material deforms under stress. The microstructural changes which alter deformation rates (grain structure, recrystallization, phase transformation) are accounted for. All individual components of the model represent known metallurgical phenomena.

Since then, the continuous improvement of techniques available for microstructural characterisation of irradiated materials generated a considerable progress in understanding physical mechanisms associated with the material properties changes, [3]. New insight and advanced modelling techniques suggest that renewed efforts should be devoted to the construction of microstructure based models. Although it is not yet feasible to construct *comprehensive* models explaining the behaviour of materials, once accomplished, they should be extremely powerful and would have the potential of requiring fewer fitting variables, and to easing the incorporation of new data and new understanding.

It is therefore suggested to reconsider this basic concept.

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Chapter 6

Water Chemistry *

The issue of water chemistry is of particular importance. Through the right choice, monitoring and control, not only the fuel cladding but also the primary cooling circuit components can be better protected from corrosion and thus their lifetime extended.

6.1 Status of water chemistry

a. Actual practice of water chemistry in PWR reactors

From several experiences with “low” and “high” lithium water chemistry, it is concluded that “high” lithium chemistry would increase the Zircaloy-4 corrosion. Modelling performed in France shows an increase of corrosion up to 45% when the lithium (Li) effect is more pronounced in two-phase flow and when the void fraction increases.

Therefore, the recommendation is to use the modified Li-B chemistry with Li $\leq$ 2.2 ppm (in Germany the limit is set at 2 ppm).

Two other factors have also to be taken into consideration when deciding on water chemistry. On the one hand, a constant pH around 7.2 throughout the cycle would probably decrease the activity transport in the primary circuit and therefore the dose rate in the plant. On the other hand, high Li chemistry would increase the risk of stress corrosion cracking of primary circuit components, especially those made of alloy 600.

Several experiments have been conducted on alternative pH controlling agents. It was reported that potassium hydroxide (KOH) has shown good results in both autoclave and loop tests in single or two-phase flow even with high void fraction, the difference of ionic radius between Li and K may be the reason; but there is still a long way to go before western PWRs can be operated with KOH; extensive tests on corrosion of in-core components working under irradiation would be needed beforehand. It is also reported from Russia that some laboratory tests are showing a higher sensitivity of Zircaloy-4 to hydrogen pick-up when used in WWER chemistry, therefore, behaviour of cladding materials regarding hydrogen pick-up would have to be checked also.

Addition of zinc can be beneficial for decreasing dose rates to personnel.

b. Actual practice of water chemistry in WWER reactors

In WWER reactors potassium water chemistry associated with zirconium and 1% Nb cladding shows good performance: low corrosion rate and hydrogen pick-up is observed. In WWER reactors a co-ordinated water chemistry at $pH_T \approx 7.0\text{-}7.1$ instead of “drifting” pH in the range 6.6-6.9 was implemented. The result of the application of this new water chemistry was a decrease in personnel

* This section summarizes the results of the IAEA Technical Committee Meeting “Influence of Water Chemistry on Fuel Cladding Behaviour” held in Prague in October 1993

radiation exposures; however, a controversial matter remains over the possible use of ammonia or hydrazine an oxygen scavenger. On one hand it is claimed that the use of hydrazine decreases cladding corrosion and activity transport, on the other hand, some Russian results show that the reactor contamination is higher when hydrazine is used. A comparison between ammonia and hydrazine has been carried out at the PAKS power plants in Hungary but does not show a significant difference between the two types of water chemistry from the point of view of dose rates. In this matter there are very few comprehensive results available and further work is needed to clarify the situation (especially as far as fuel examination is concerned).

c. Effect of low tin content and heat treatment on Zircaloy-4 behaviour

It is recognised that any comparison between “high” and “low” lithium, “high” and “low” tin content or “high” and “low” temperature is somehow impaired by other parameters which seem very sensitive. A wide dispersion of results was obtained, sometimes in the same batch of cladding. Two main parameters are identified as possible culprits:

- Heat treatment during tube fabrication. An improvement of 40% was attributed to optimisation of composition and heat treatment by the French utility;
- Temperature differences in the core.

Further international studies of the heat treatment will be difficult because the values used in practice are very often commercially classified. The study of temperature differences in a core would require a more sophisticated approach to linear power and thermal-hydraulic modelling.

It was also recognised that low tin alloys were showing improved corrosion behaviour. Although the improvement is sometimes difficult to assess, a figure of 15% benefit in the oxide layer thickness at 50 MWd/kgU is announced by EDF.

d. Clad corrosion modelling

Models used up to now are empirical or semi-empirical, their capability to predict the behaviour of a new alloy or the consequence of a moderate increase in temperature (10-15°C) is very limited. The need to go to a more mechanistic understanding of the phenomena involved is therefore apparent.

Two areas have been identified for additional research:

- The effect of radiation and energy deposition related to the dissolution and deposition of oxides and hydroxides;
- The consideration of real corrosion curves, the study of which would lead to a more mechanistic approach to zirconium alloy corrosion. In this study, the influence of annealing parameters and alloying elements would have to be studied in more details.

e. On-line monitoring of water chemistry

Many companies are now working on “on-line monitoring of water chemistry”. It is generally agreed that “on-line” monitoring is the way to go in the future but is extremely difficult to implement. The potential cost benefit is such that it is very surprising that not much effort is directed to this area. Several promising methods of measurement are still under development, they are for instance: conductivity, pH, hydrogen, soluble and insoluble impurities, corrosion potential.

The benefit of this monitoring for safety, minimization of corrosion or man-sievert reduction is widely recognised. As for any new development, many questions are still open. Among these questions the following ones are most important:

- What measurement to perform?
- What kind of sensor to use?
- Where to perform the measurement?
- How to extrapolate the measurement results to other locations in the reactor? (modelling).

6.2 Recommendations

The most likely developments are identified as follows:

a. Water chemistry practice

- In order to minimise activity transport, clad and out of core corrosion, it is recommended:
  - To use a modified Li-B water chemistry with a pH as close as possible to 7.2 in PWR reactors,
  - To use a co-ordinated K-B water chemistry with a pH_T 7.0-7.1 in WWER reactors;
- Evaluation of alternative pH controlling agents, especially potassium hydroxyde for PWR application;
- A further study about the influence of zinc on the activity transport is needed;
- Evaluation of the behaviour of Zircaloy-4 in WWER water chemistry in normal operation and accident conditions, including the influence of the oxygen scavenger used on corrosion and hydriding (influence of nitrogen and ammonia);
- It is recommended to carry on the comparative study ammonia/hydrazine as oxygen scavenger.

b. Cladding corrosion

- More observations on cladding corrosion from power reactors and loops in MTR reactors are needed and an effort has to be made to identify the specific influence on corrosion of each parameter (temperature, fabrication route, annealing temperature, tin content, alkali concentration);
- Special attention has to be given to the in-core assessment of irradiation parameters (linear rate, temperature).

c. Corrosion modelling

- The effect of radiation and energy deposition on the dissolution and deposition of oxide and hydroxide have to be further studied;
- More mechanistic models of corrosion have to be developed starting from the observed kinetics of corrosion on well characterised cladding.

d. On-line monitoring of water chemistry

- Further development of the on-line monitoring of water chemistry is needed with appropriate sensors and the correlated modelling.

Chapter 7

Hydrogen Measurement Techniques (in Cladding)

Zircaloy is widely used as a cladding material for Light Water Reactor (LWR) fuel pins, both Boiling Water Reactors (BWR) and Pressurized Water Reactors (PWR). In the core of the reactor the outer side of the cladding is exposed to cooling water at temperatures between 300°C and 400°C. During its life time the cladding should remain intact to prevent the release of fuel and fission products to the cooling water. Therefore, the Zircaloy cladding should be strong enough to withstand the mechanical and thermal stresses originating from the coolant pressure, the filling gas and the released fission gas pressure, the mechanical interaction with the fuel pellets due to thermal expansion and swelling of the fuel, and from temperature gradients.

During operation the cladding of a fuel pin oxidizes and some hydrogen is trapped by the Zircaloy. High hydrogen concentrations could lead to hydride precipitation. This phase is brittle at low temperatures and could induce fuel clad failure during outage handlings like fuel rod removal and reconstruction of the assembly. In addition, the very high strain rates during reactivity insertion accidents (RIA) could lead to a brittle behaviour of the cladding with high hydrogen contents even at high temperature. Therefore, good knowledge is needed of the hydrogen content and the hydride formation in the cladding.

During normal operation there is a slow reaction between the Zircaloy cladding and the cooling water in the core:

$$(1) \qquad Zr + 2H_2O \rightarrow ZrO_2 + 2H_2$$

The reaction rate is determined by the oxygen diffusion rate through the zirconium oxide (ZrO_2) layer into the Zircaloy. The growth rate of the oxide layer depends on the chemical condition of the cooling water. In BWRs the zirconium oxide growth rate is larger than that of PWRs due to a higher oxygen content in the cooling water of BWRs. The thermal conductivity of ZrO_2 is much lower than that of Zircaloy. Therefore, a thick oxide layer limits the heat transfer and leads to higher cladding and fuel temperatures.

Together with the oxygen also some hydrogen atoms generated according to (1) diffuse to the Zircaloy and are absorbed. The diffusion of H-atoms is enhanced by nickel containing precipitates in the zirconium oxide layer. The solubility limits for hydrogen in Zircaloy at a few temperatures are [1]:

T/°C	25	100	200	250	300	350	400
(H)/ppm	0.05	1.0	20	29	61	114	192

This means that at rather low hydrogen concentrations and at temperatures below about 150°C precipitation of zirconium hydride occurs as needles or platelets. These hydrides are very brittle, do not strongly bond to the surrounding zirconium and have almost no strength. Hence, zirconium hydrides can reduce the toughness of zirconium alloys and induce fracture. The hydrogen solubility limits may be slightly different for other zirconium alloys. Under certain conditions the hydrogen content may exceed the solubility limits without precipitation of hydrides. The phase, size and orientation of the zirconium hydrides depend on the hydrogen concentration, the cooling rate, the present stresses and the microstructure of the Zircaloy [2] to[5]. Large hydride platelets are formed by cooling down slowly from 350 to 200°C. These hydrides are most detrimental for the mechanical properties when tangential stresses are present which force the hydrides to precipitate in radial direction [6]. At cooling rates larger than about 50°C/h the zirconium hydrides precipitate mainly as very small platelets which have much less effect on the mechanical properties. During normal operation and at low enough hydrogen concentrations the zirconium hydrides formed at low temperatures dissolve again. However, cracks formed during low temperature periods remain in the cladding and thus affect the integrity of the fuel pin.

7.1 Hydrogen distribution in the cladding

The hydrogen generated in the cooling water diffuses through the zirconium oxide layer into the Zircaloy. The build-up of a hydrogen concentration at the outer surface causes a hydrogen flow to the inner side of the cladding. An opposite flow arises from hydrogen dissolved in the cladding which diffuses from high temperature to low temperature regions thus from the inner side to the outer side of the cladding. The result is a gradient in the hydride concentration across the clad wall. The hydride structure depends on the temperature history, especially during cooling down (shutdown of the reactor).

At room temperature hydrogen in Zircaloy cladding is mainly present as radial and/or circumferential zirconium hydrides throughout the cladding thickness. At operational temperatures between 300°C and 400°C and at hydrogen concentrations higher than the solubility limit, zirconium hydrides will be formed predominantly at the outer interface Zircaloy-ZrO_2. The exact mechanism of hydride precipitation under temperature gradients is not well understood.

The effect of the amount of hydrogen on the mechanical properties of the cladding depends largely on the size and orientation of the zirconium hydrides platelets formed and on the presence of stresses in the cladding [6].

7.2 Hydrogen measurement

At high burnup the build-up of the hydrogen concentration in the cladding may exceed the solubility limit of the operation temperature. This results in the formation of a zirconium hydride layer at the outer interface Zircaloy-ZrO_2 which also deteriorates the mechanical properties of the cladding. Because of the formation of zirconium hydrides in the Zircaloy cladding, it is important especially for high burnup fuel pins and for long term fuel storage to measure the hydrogen concentration in the cladding, by non-destructive techniques.

The best method to determine the amount of hydrogen and especially the size and orientation of the zirconium hydrides in the Zircaloy cladding is by mass spectrometry and metallography during post-irradiation destructive analysis. However, this method can not be used when the irradiation of the fuel pin has to be continued. For these fuel pins it is important to determine the hydrogen content and distribution in a Zircaloy cladding without affecting the integrity.

Well-known non-destructive examination techniques are: Eddy Current Testing, Ultrasonic Testing, X-ray Photography and Neutron Radiography. However, it is difficult to discriminate between the various possible defects in a cladding like: flaws, holes, inclusions, corrosion products, fabrication defects and hydrogen or hydrides. Another non-destructive technique to measure the H_2 content is to analyse the thermal behaviour during heating and cooling. The concentration of H_2 is connected to the thermal spike occurring at the temperature of the solubility limit (differential thermal analysis), [8] & [9]. However this method is only valid for hydrogen contents below about 300 ppm. A method using the Young's modulus shift induced by hydrogen doping was developed by the Whiteshell laboratories, [10].

Results on hydrogen measurements from these non-destructive examination methods are very scarce. The sensitivity of hydrogen detection in metal was demonstrated by microdensitometer evaluation of neutron radiographs taken from Zircaloy tubes with different hydrogen loadings of the inner wall [7]. Subthermal neutrons from the High Flux Reactor (HFR), Petten, were used due to their higher imaging potential originating from the higher linear attenuation coefficients of hydrogen for subthermal neutrons compared with thermal neutrons. For the measurements four Zircaloy tubes were used with hydrogen concentrations of 0, 700, 1100 and 1800 ppm. A linear relation was established between the hydrogen loading and the differential film density (background-tube wall). This method is also capable of providing information about the depth of the hydrogen ingress into the tube wall.

7.3 Conclusion

Extension of the life time of a fuel pin is not only of economic interest; it is beneficial for the utilisation of materials and it also reduces the amount of radioactive waste.

The irradiation of fuel pins up to a high burnup is only possible if the soundness of the cladding can be monitored. In particular, the presence of high hydrogen concentrations can limit the cladding integrity. Therefore, measurement techniques are needed to detect hydrogen in fuel pin cladding. However, information on non-destructive methods to measure hydrogen concentrations in metals is very scarce. Neutron radiography appears to be promising. Research on hydrogen measurement in metals by non-destructive techniques should be encouraged.

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Chapter 8

Failed Fuel Behaviour

8.1 Background

Improved fuel assembly design and manufacturing as well as operational constraints during service have considerably reduced the occurrence of fuel failures. In BWRs the failure rate has decreased by one order of magnitude from 1985 to 1991. The currently reported BWR fuel failure rate varies from 0.002 to 0.005%. In the same period, the failure rate in American PWRs had decreased from 0.02% to 0.005%. Partly due to the fact that fuel rod failure is a very rare event today, the failure cause remains often unknown. Debris failures and possible manufacturing defects are considered to be the major cause of failure. The industry aim is to achieve a “zero” defect rate by further improvements of the assembly design (e.g., by introduced debris filters) and by tighter quality control routines.

The progress made on fuel failure control has considerably reduced the activity release of radioactive species to the coolant and consequently improved the plant dose control. Sporadically, however, single rod failures can progressively degrade with consequent large activity release to the coolant. A single degraded fuel rod may cause an activity release increase which is orders of magnitude higher than a stable, non-degraded failure. The consequence of fuel failure degradation can be that a plant must be shut down and the degraded rod removed. In recent years, fuel rod degradation has been experienced both in European and American reactors, causing unscheduled shutdowns in some cases. This has considerable economical consequences, in addition to aggravating the plant dose burden.

Operators of nuclear power plants need procedures in order to avoid the propagation of a primary failure of whatever origin into a large secondary failure. The general rule is to localise the core region of the primary failure and then reduce the power locally. This method, however, has shown not to be suitable to control propagation, and degraded secondary failures have been reported also in cases where the local power had been reduced.

In order to identify possible design or operational remedies, a better understanding of the degradation mechanisms is necessary. Efforts are being made by the industry aiming at a model description of the phenomena involved. This requires a comprehensive review of the data already available as well as new ad-hoc experiments. Post-irradiation examinations of failed and degraded rods will also give valuable information, although observations are often masked by processes occurring after the secondary failure.

The modelling effort is to cover two different aspects of the failure behaviour. The first addresses the failure characterisation (e.g., number of failures, average size and fuel burnup) based on coolant radiochemistry data. These correlations help in localising the failed fuel rods and in preparing a suitable unloading plan. The second aspect is the predictive behaviour of the failure in terms of degradation rate. As said earlier, these models are to identify remedy actions to prevent failure degradation.

8.2 Correlation between radiochemistry data and failure characteristics

Utilities monitor leakage of fission products to the coolant very carefully. Monitoring covers gaseous fission products (xenon and krypton isotopes), caesium and iodine isotopes.

The *number of failures* can be roughly inferred from the release of gaseous isotopes with intermediate half life. This is because noble gaseous fission products are readily available for release to the coolant, that is there are no trapping mechanisms in their release path from the fuel. The most suitable isotope is ^{133}Xe which has a half life of approximately 5 days, thus making its measurement rather independent on the transport time within the rod. However, the source term depends on the operating conditions of the fuel and on the possible consequences induced by the water ingress on the fission gas release.

The *degree of defect degradation* can be inferred from the release of gaseous fission products having short half life, of the order of minutes. The detection of short lived isotopes, however, may be considerably affected by the delay time between source and detection point.

The *plutonium content* of the failed fuel can be determined by the ratio ^{134}Cs/^{137}Cs. Algorithms providing the expected burnup range of the leaker can be derived from physics calculations. The picture may become more complicated if more than one leaker (with different burnup) is present in the core.

Mechanistic models and empirical correlations for failure characterisations based on activity monitoring have been developed by different organisations. However, their applicability has been demonstrated on limited data sets only. An effort aimed at validating existing models or correlations against actual plant data appears justified by the potential economical benefit for the industry.

This effort may consist of the following steps:

- Systematic review of existing detection methods (detection systems, isotopes to be monitored, data presentation);
- Literature review of existing models/correlations, including comparison with experimental or plant data;
- Collection of relevant data on activity release and failure characteristics from power plants. The aim should be to construct to a data bank on existing failed fuel data in a standard format;
- Verifications of selected models/correlations against plant data.

The output of such an effort can be as follows:

- Recommendations on monitoring methods for power plants (equipment type, location, isotopes to be measured, data presentation);
- Recommendations on the approach for fuel characteristics predictions. Alternatives can be basic mechanistic models, correlations or expert systems based on existing experience in a given plant type;
- Recommendations on ad-hoc simulations of particular processes or phenomena where data are lacking.

8.3 Failed fuel behaviour modelling

Experience with LWR fuel shows that a primary failure may remain in a stable condition for a long time or it can produce extensive secondary failure. The latter may take place at quite some distance from the primary defect and it generally causes a sudden pronounced increase in the off-gas activity. When it occurs, the secondary failure develops in a time period of a few weeks or months after the appearance of a primary failure. The general mechanism associated with failure degradation is hydriding of the cladding. Steam ingress in the primary failure site promotes oxidation of both fuel and cladding with consequent generation of hydrogen. This is then picked up by the cladding which becomes prone to failure due to embrittlement. The secondary failure is thus usually caused by extensive hydriding and presence of cladding stress.

Although the gross mechanisms are understood, predictive descriptions of the degradation process are lacking. This is partly due to the complexity of the phenomena involved as well as to the lack of critical data. A comprehensive model of the fuel rod behaviour in the presence of water ingress is essential for remedy actions to be taken by the plant operators and/or by the fuel designer.

Early work by Locke points out that the time to secondary failure is strongly affected by the operating power. This is currently acknowledged in operational routines which contemplate power reductions by control rod insertion near the failed assembly. As said earlier, however, this precaution has not always been successful in preventing degradation.

Experimental work has shown that a critical amount of hydrogen must be present in the hydrogen/steam mixture in order to promote cladding hydriding. This is because a sufficient amount of oxidant can maintain an oxide protective layer at the cladding inner surface. It follows therefore that small defects are potentially more dangerous than larger defects. The latter ones, in fact, may provide enough steam (oxidant) to prevent massive hydriding.

The defect size is not the only geometric parameter controlling the hydrogen/steam ratio. The residual pellet-cladding gap size is in fact another critical parameter: a very small or “closed” gap may produce steam starvation conditions along the fuel column and thus more favourable conditions for hydrogen pick-up.

Fuel and cladding oxidation constitute important hydrogen sources. Data on UO_2 or Zircaloy oxidation rate in the presence of steam and radiation are very scarce. The radiation field is important because it may considerably enhance the corrosion rate due to radiolytic formation of hydrogen peroxide. Experiments are being designed at the IFE-OECD Halden Reactor Project with the purpose of characterising the fuel and Zircaloy oxidation rate in steam and in the presence of radiation and fission fragments.

An additional source of hydrogen at the rod interior is the radiolytic decomposition of steam, giving modular hydrogen and hydrogen peroxide as products. The highly reactive hydrogen peroxide enhances the UO_2 oxidation. In this picture, the steam entering the primary defect diffuses along the pellet-to-cladding gap and is gradually enriched in hydrogen, whereas the water molecules are consumed in the oxidation processes. Hydrogen can enter the cladding at sites where the protective oxide layer is damaged or directly as the pick-up fraction of the cladding oxidation process.

Heavily hydrided cladding may undergo extensive failure in the presence of cladding stress. Stress may arise from pellet-cladding interaction since fuel swells due to oxidation. Additional sources of cladding stress are possible changes in gap chemistry (e.g., changes from H_2 to steam in the gap causing high fuel temperatures) or anisotropic expansion of the cladding due to local hydriding.

Experimental and modelling activities should aim at the following results:

- Identification of relevant processes and related available data. When specific data are needed, quality tests should be performed;
- Review of computer models which already incorporate algorithms for failed fuel performance;
- Predictability of selected models against actual data;
- Connection between modelling of failed fuel behaviour and fuel failure characterisation based on water radiochemical fingerprints;
- Recommendations on models upgrading, i.e. processes to be included and relevant data sets.

Chapter 9

Spent Fuel – Long-term Behaviour

Understanding and modelling of ageing effects of long-term burnt fuel storage need to be addressed in order to predict with confidence the lifetime of present and future facilities.

The IAEA has been promoting the international research programme on “Behaviour of Spent Fuel Assemblies during Extended Storage (*BEFAST*)” since 1981. The documents of IAEA-TECDOC series collect many results related to spent fuel storage.

The policy and status of spent fuel storage in various countries is listed in Table 9.1. There are two categories as far as the storage method is concerned, i.e. wet storage and dry storage. The wet method is the main mode, at present.

From the viewpoint of fuel behaviour during long term storage, there are some technical considerations for each method. These are described in the references [1], [2], [3] & [4].

The IAEA has also several programmes on materials problems in spent fuel storage facilities.

9.1 Current status

a. Wet storage

Technical issues during wet storage are as follows:

- cladding behaviour;
- behaviour of fuel with cladding breaches;
- behaviour of fuel assembly structural components.

A schematic picture of mechanisms affecting spent fuel cladding performance during *wet storage* is shown in Figure 9.1, [5].

Waterside corrosion including crud effects from external mechanical loads and chemical attack by fission products from the inside are the main issues. Hydrogen pick-up and redistribution are more or less secondary effects caused by severe corrosion or clad failure. In addition water radiolysis effects on corrosion enhancement and oxide etching have to be considered for long term wet storage.

There is much experience on wet storage of the fuel. The integrity of fuel during the duration of storage has been confirmed so far [3] & [4]. Only defective fuel rods require special consideration.

b. Dry storage

Technical issues that need addressing during dry storage are as follows:

- maximum allowable cladding temperature in inert and oxidising cover gases,
- reactions which affect the cladding on the internal and external surfaces,
- behaviour of cladding defects in inert and oxidising cover gases,
- behaviour of fuel assembly components,
- degradation of fuel matrix.

A schematic picture of the mechanisms affecting spent fuel cladding performance during *dry storage* is illustrated in Figure 9.2, [5].

The mechanisms are principally the same as in the case of wet storage, but the degree at which they contribute to degradation is different. In particular, stress-induced mechanisms like creep dominate among the restrictive parameters in the case of dry storage in inert atmosphere.

Oxidation of UO_2 becomes the most important fuel degradation mechanism of defective rods if oxygen is available in the storage environment. Consequently, many studies have been carried out on this topic, as listed in Table 9.1 For most of the engineering issues, practical solutions were found through the studies. Issues concerning basic mechanisms related to the phenomena caused by dry storage and with new types of fuel rod pellets are the ones that may need further attention.

9.2 Future issues

As mentioned above, for almost all present issues practical solutions have been found. However, for the storage of high burnup fuel and plutonium/uranium mixed fuel under wet or dry storage conditions, it is necessary to obtain mechanical data such as tensile properties, fracture toughness, creep strength, etc., of the corresponding spent fuel, because of the property change in the cladding due to irradiation, oxidation and hydriding.

During dry storage of spent fuel in inert atmosphere, creep deformation is considered to be the critical degradation mechanism of spent fuel.

On the other hand, for the dry storage method in an air-containing atmosphere, oxidation of UO_2 and the following deformation of fuel rods due to volume expansion becomes a critical factor for fuel integrity of defective rods [6], [7] & [8].

As already pointed out [7], the oxidation of irradiated UO_2 is different from that of unirradiated UO_2, and oxidation data of irradiated UO_2 below 200°C are needed to evaluate the allowable storage temperature of the defected fuel rod during dry storage.

Figure 9.1 **Mechanisms affecting spent fuel cladding performance during wet storage**

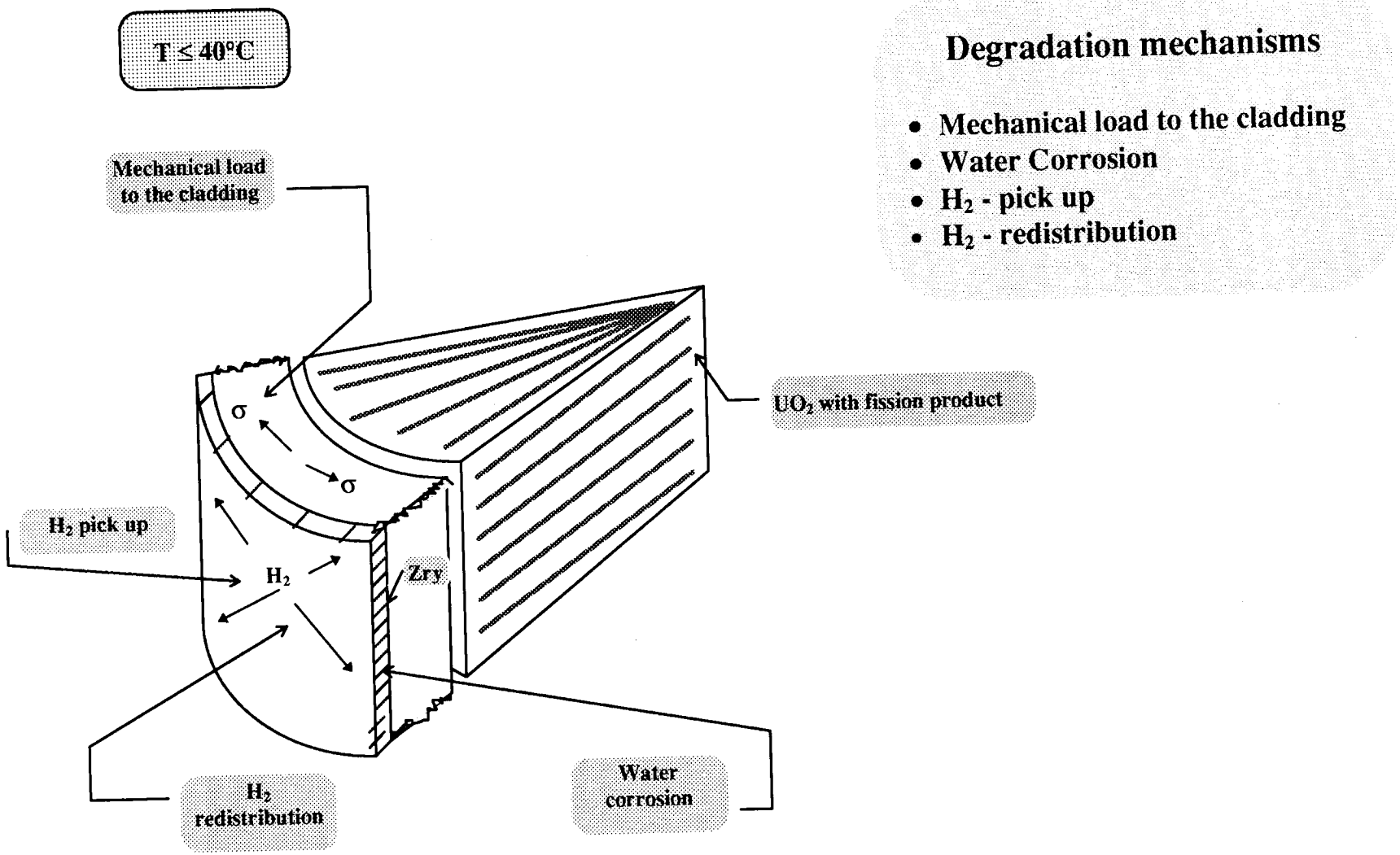

Figure 9.2 **Mechanisms affecting spent fuel cladding performance during dry storage**

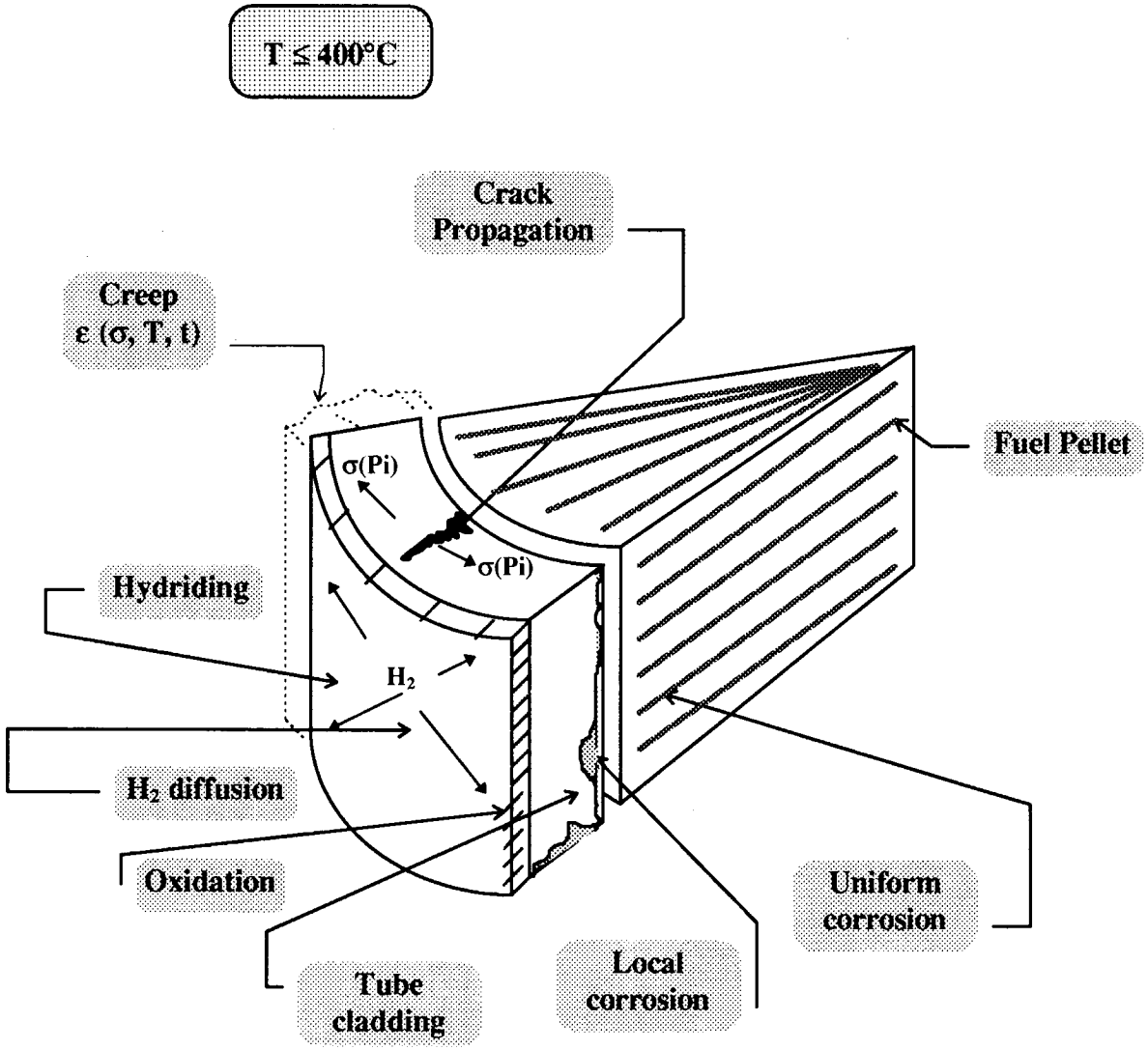

Table 9.1 **Status of Spent Fuel Storage in Each Country**

Country	**Reactor** *Number*	**Policy on Spent Fuel**	**Generation of Spent Fuel** *ton/year*	**Storage method for Spent Fuel** *other than reactor pool*	**Research Activity**
U.S.A.	109	Direct disposal	1, 700-2, 100 Total 84, 000 t until 2020	AR: Cask/Sailo/Vault AFR: Cask/Sailo/Vault/Pool	Dry Storage below 250°C in helium / air including defected rod
Canada	22	Direct disposal	Total 14, 000 t until 1990	Sailo Concrete cask	Dry Storage at 150°C in air, CANDU Fuel
France	55	Reprocessing *pluthermal, FBR*	1, 000-1, 100 t	Vault Pool at reprocessing factory	Dry Storage Cask/Vault
U.K.	35	Reprocessing *Magnox/AGR* undecided on PWR fuel	750 t Magnox until 2002	Vault	Wet Storage: monitoring Dry Storage: oxidation of UO_2
Japan	46	Reprocessing *pluthermal, FBR*	1, 000-1, 100	AR: Cask (R&D) AFR: Cask/Sailo/Vault (R&D)	Dry Storage: cladding creep, cladding oxidation, irradiated rod oxidation
Germany	21	Reprocessing & Direct disposal *optional*	600	AFR Cask (3, 000 t) AFR Pool (560 t)	Dry Storage: heat transfer analysis FA behaviour ≤ 55MWd/kgU
Sweden	12	Direct disposal	Total 7, 800 t	AFR pool (3, 000 t)	Wet Storage: defective fuel rod
Finland	4	Direct disposal Reprocessing in Russia	Total 540 tU in 1992	Wet Storage (1, 200 tU)	
Switzerland	5	Reprocessing & Direct disposal		AFR Cask	
Former USSR	43	Reprocessing & Direct disposal	13, 000	3, 000 t in the pool at reprocessing factory AFR Cask	Wet and Dry Storage: UO_2 - Zr + 1% Nb cladding
Czech Rep	4 *2 under construct.*	Undecided *Reprocessing considered*	56	AFR Cask/Vault	
India	9	Reprocessing	25 from BWR	AFR Pool AR Cask	
Korea	9	Undecided	2, 810 t in 1995	AR Pool	Wet storage: defected rod
Italy	4	Decommission *Reprocessing abroad*	243 t in 1986	AFR Cask	

Table 9.1 **Status of Spent Fuel Storage in Each Country -** (cont.)

Country	**Reactor** ***Number***	**Policy on Spent Fuel**	**Generation of Spent Fuel** *ton/year*	**Storage method for Spent Fuel** *other than reactor pool*	**Research Activity**
Hungary	4	Reprocessing in Russia		AR Pool	Pool monitoring
Argentina	2	Re-evaluation *Reprocessing considered*	160	AR Sailo	Wet Storage Experience
Belgium	7	Reprocessing undecided		AR: Pool/Cask	
Brazil	1	Reprocessing undecided			
Bulgaria	6	Reprocessing undecided			
China		Reprocessing			

AR: at reactor
AFR: away from reactor
FA: fuel assembly

References

[1] IAEA Technical Working Group Behaviour of Spent Fuel Assemblies during Extended Storage, IAEA-TECDOC-414 (1987).

[2] IAEA Technical Working Group Extended Storage of Spent Fuel, IAEA-TECDOC-673 (1992).

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[4] Guidebook on Spent Fuel Storage, second edition, IAEA Technical Report series No 290, IAEA, 1991.

[5] Peehs M., Fleisch J., J. Nucl. Mater., 137 (1985), 190-202.

[6] Peehs M., Einfeld K., Effects of Long-term Dry Storage of Spent Fuel, paper presented at the Intl. Conference on HLWR, Las Vegas, U.S.A., April 1992.

[7] Einziger R.E. et al, J. Nucl. Mater, 190 (1992), 53-60.

[8] Wasywith K., Taylor P., The Canadian Long-term Experimental used Fuel Storage, paper presented at the Intl. Conference on Nuclear Waste Management and Environmental Remediation, Prague, Sep. 5-11, 1993.

Chapter 10

Quality Assurance Process for New Integral Experiments and Their Utilisation in Computer Codes

10.1 Introduction [1]

Because of the need to provide fuel modellers with information on high burnup fuel behaviour in steady-state, transient and accident conditions, specific integral experiments have been carried out during the last few years in MTR reactors using water reactor fuel.

In some cases, in order to shorten the irradiation time, fuel already irradiated in power reactors was used. Here, an operation called “re-fabrication” is needed to adapt these fuel rods for continued irradiation in the MTR reactor.

The actual challenge is to measure fuel parameters (fission gas release, densification, swelling, etc.) during irradiation to complement the observations made once the irradiation is complete (post-irradiation examination). The correlation between the fuel characteristics and the irradiation parameters needed for improved fuel modelling can be obtained with this method. For this purpose, fuel rods have to be instrumented with sensors for measuring fission gas pressure, fuel temperature, fuel column elongation or clad elongation, etc.

In recent years many techniques were developed to assess these parameters on-line in relation to fuel power history. More recently on-line measurements of water chemistry parameters (conductivity, pH, electro-chemical potential) have also been developed.

Instrumentation, re-fabrication and re-instrumentation techniques are now widely used and they have advanced progress in the mechanistic understanding of fuel and cladding behaviour. Although comparable results have been obtained from experiments performed with segmented and re-fabricated rods, irradiated under the same conditions, results from different material testing reactors (MTR) on similar devices are not always compatible. This appeared especially in countries embarking only recently on fuel development and modelling. Several steps in the process can be suspected: different re-fabrication techniques, linear power measurement or the choice of sensors and their calibration. Therefore common specifications are needed in order to ensure that both the re-fabrication methods and instrumentation and re-instrumentation do not perturb the experimental behaviour or the measurement performed.

The quality assurance process applied to data production needs also to be carried through when utilising these data for computer code development and to the application of the resulting models to power reactor fuel.

10.2 Quality assurance for reactor fuel experiments in test reactors

Quality is a global connotation concerning an organisation as a whole and each individual in the organisation. It also relates to subcontractors and to suppliers of parts and components. Quality requirements apply to the entire spectrum of activities and project phases, from the initiation stage to final reporting.

For in-pile fuel experiments, the key elements of a quality control plans are as follows:

- A clear sequence of actions/tasks necessary for the correct identification of the test objective and for the correct and safe implementation and execution of the experiment;
- An organisational structure with a well defined job assignment;
- A suitable document flow covering the entire spectrum of activities ensuring an effective communication exchange and data traceability.

Tentatively, a “check list” of actions/tasks is given in Appendix C.

Internal quality control routines have been in place in all research centres dedicated to in-pile experiments in OECD Member countries. In recent years, however, the need of a standard quality control system has arisen, especially in response to customers’ demands. Future efforts, thus should aim at adapting internal control routines to an acknowledged standard. At the IFE-OECD Halden Reactor Project, for instance, the Quality Control plan conforms to the ISO 9001 [2].

It is recommended that future work in this field addresses the following items:

- Assessment of quality control routines at various research centres, dedicated to in-pile tests exchange of experience and outline of objectives/plans;
- Recommendations for establishing a co-ordinated quality control policy;
- Headlines of a quality control plan for common use in reactor experiments.

10.3 Quality assurance process for data analysis and application

Having introduced Quality Assurance methods and procedures to the production of data, there is a need to continue the assurance process through analysis and application. The steps between data production and application can be broken down as follows:

- Assembly of database,
- Analysis and formulation of models,
- Introduction of model into code or calculational route,
- Application to reactor operation.

The three basic Quality Assurance functions which need to be applied in parallel to these operations are: *verification*, *validation* and *evaluation*.

The process of verification ensures that the final model, calculational route or code performs the calculation as defined in the functional specification. At the highest level of Quality Assurance this entails a line by line check that the physics, mathematics and coding are free from error. In most fuel performance codes used for safety analysis this requires a check by persons independent of the original developers.

Once verification has been performed, it is necessary to demonstrate that the model, calculational route or code provide adequate predictions of experimental data. In principle this requires comparison with a database other than that used in development. In practice this is often not possible because of the few data available. Comparison of predictions with data contain uncertainties due to measuring errors. It is necessary therefore to devise criteria for this comparison to distinguish whether or not the predictions are acceptable within the experimental uncertainties and to ensure that the model or models themselves do not contain fundamental flaws.

It is unlikely that the database used for development and validation encompasses all the situations met in the final application. For this reason it is necessary to perform the final process, that of evaluation. Here predictions are performed for key situations to ensure that the predictions are well behaved, i.e. are not subject to calculational instability and do not provide results significantly different from those in the database. For example, if the calculation is for fission gas release, and a model has been assembled from a database encompassing release fractions up to say 20% yet evaluation produces estimates which are systematically all very high, approaching 80 to 100%, then the predictions must be viewed with caution. It is possible that for the applications for which the model is to be applied, the model is inappropriate and an alternative formulation should be investigated. Alternatively the database may be inadequate and further tests must be commissioned if the predictions indicate possible unacceptable consequences.

Although the Quality Assurance steps outlined above are all common sense it is advisable that formalised procedures are drawn up covering these activities. Nevertheless, the most important aspect is the experience of the staff involved. Even the most rigorous and comprehensive procedures are worthless if not applied constructively by well qualified professional staff.

References

[1] IAEA Technical Committee Meeting In-core Instrumentation and In-situ Measurement in Connection with Fuel Behaviour, Petten, October 1992.

[2] Norsk Standard, NS-ISO 9001, 1st March 1988: Quality Systems; Model for Quality assurance in design/development, production, installation and servicing.

PART III

ONGOING ACTIVITIES

Activities in nuclear fuel behaviour modelling are carried out in most OECD Member countries by fuel vendors, utilities and research organisations.

At the international level the IFE-OECD Halden Reactor Project, the International Atomic Energy Agency and the CEC Institute for Transuranium Elements have a long standing involvement in this field.

Activities carried out in each country and international organisations are listed in this chapter. The list is not intended to be comprehensive, but aims at identifying common interests and the potential for establishing co-operation and avoiding duplication.

1. National activities

Belgium

- rod thermo-mechanical behaviour up to high burnup,
- fission gas release,
- fuel and clad structure analyses,
- power ramping effects,
- extended post-irradiation examinations and modelling MOX versus UOX behaviour at high burnup,
- pellet clad interaction at different power levels,
- Zircaloy high dose irradiation creep,
- Gadolinia fuel,
- conductivity degradation versus burnup and porosity.

Canada

- post-defected fuel behaviour (fission product release and UO_2 and sheath degradation),
- effect of coolant radiolysis on UO_2 and sheath behaviour,
- gap oxygen potential during irradiation for intact and defected fuel elements,
- defected fuel behaviour during power ramps,
- effect of extended burnup on release, UO_2 microstructure, UO_2 chemistry and properties, etc.

Finland

- cladding (Zircaloy, Zr 1%Nb) and structural material studies (irradiated),
- water chemistry monitoring techniques,
- bilateral test reactor irradiations and examinations (WWER),
- poolside examination campaigns,
- fuel performance code validation for high burnups,
- issues of transient and accident conditions and modelling.

France

- fission gas release and clad coolant interaction with burnup increase,
- transient behaviour of pellet clad interaction (thermo-mechanical, fission gas release),
- fuel with recycled plutonium matching the performance of uranium-oxide fuel,
- fission product retention for different classes of incidents and accidents,
- research and development involving research centres, vendors and utilities to improve modelling software,
- basic understanding of UO_2 microstructure (fission product doped UO_2) and of zirconium alloy corrosion under irradiation.

Germany

- fuel design and modelling for high burnup (including experimental support),
  - fuel thermal conductivity,
  - fission gas release,
  - rim effect,
  - clad performance,
- extension of code systems to high burnup application,
- hydriding in cladding and structural materials,
- performance of MOX fuel,
- rod performance in power ramps (PCI),
- probabilistic fuel rod analysis and LOCA analysis,
- dry storage of spent fuel.

Italy

- co-operation with Russia on RMBK fuel,
- studying fuel options for actinides transmutation,
- code system capability extension to cover high burnup.

Japan

- in LWRs:
  - fuel design and modelling for high burnup,
  - rim effect and defect formation studies through high energy ion bombardment,
  - studies on MOX fuel,
- experimental and modelling work in advanced thermal reactor (ATR) fuel,
- MOX and nitride fuel for fast reactors (FR),
- high burnup high temperature gas reactor (HTGR) fuel,
- fuel irradiation and fuel plate stability for research reactors.

Netherlands

- modelling and experimental work on the thermal conductivity effect at high burnup,
- thermal conductivity measurements on irradiated fuel are being envisaged by using a laser flash method.

Spain

- fuel performance at high burnup of standard and advanced fuel or cladding components,
  - corrosion / hydriding,
  - fission gas release,
  - pellet clad interaction,
  - material properties,
- gadolinia performance,
- defective fuel rod cause research,
- fuel rod modelling improvement.

Sweden

- stress corrosion cracking and waterside corrosion studies,
- ramp tests including ultra-fast over-power ramps simulating reactivity initiated transients in high burnup fuel.

Switzerland

- radial distribution of isotopes in UO_2 and MOX fuel pins (study of rim effects),
- fuel thermal conductivity (correlations based on experimental data),
- fission gas release.

United Kingdom

- development of fuel performance code for LWRs and AGRs,
- modelling of fuel temperatures, degradation of fuel conductivity with burnup,
- modelling of the rim effect and its influence on thermal performance and fission gas release,
- fission gas swelling in solid and hollow pellets,
- improved mechanistic models for pellet clad interaction,
- need for Zircaloy mechanical properties data,
- waterside corrosion and hydriding,
- noise analysis applied to time dependent phenomena,
- studies on MOX fuel.

United States

- thermal and mechanical properties of UO_2 and gadolinia fuel up to high burnup,
- mechanical properties of irradiated Zircaloy cladding up to high neutron dose,
- fuel cladding and hydrogen pick-up for different materials and water chemistry conditions,
- thermal and mechanical properties of the pellet rim region,
- fission gas release at high burnup,
- research on reactivity initiated transients in high burnup fuel,
- post-failure severe cladding degradation: mechanism and remedies.

2. International activities

Commission of the European Union

- development and maintenance of fuel rod analysis code,
- local effects in thermal conductivity,
- properties of the rim zone,
- gap conductance at high burnup,
- probabilistic fuel rod analyses,
- co-operation on WWER fuel modelling.

International Atomic Energy Agency

Co-ordinated Research Programmes (CRP) on :

- fuel modelling at extended burnup (FUMEX, a blind code comparison exercise focusing on high burnup fuel modelling issues of temperature, fission gas release, swelling and pellet clad interaction using data from the Halden project),
- modelling of burnable absorber fuel (BAF),
- water chemistry,
- fuel failure,
- Zircaloy corrosion.

Technical Committee Meetings cover:

- high burnup fuel modelling,
- fuel performance utilisation and management,
- performance of MOX fuel, control rods,
- improved pellet material and design,
- fuel reliability,
- influence of water chemistry on cladding behaviour,
- poolside inspection, repair and reconstruction of fuel elements,
- core material behaviour in severe accidents.

IFE-OECD Halden Reactor Project

- high burnup fuel properties,
- hydriding of cladding,
- cladding creep,
- cladding oxide conductivity,
- small dryouts,
- crack growth rate versus stress intensity,
- crack initiation in different alloys,
- stress corrosion cracking,
- in-core water chemistry,
- crud deposition and sensor developments.

OECD/Nuclear Energy Agency

- NSC Task Force on scientific issues in fuel behaviour,
- CSNI-PWG2 on coolant system behaviour: transient behaviour of high burnup fuel,
- NEA Data Bank acquisition, verification and distribution of fuel behaviour modelling codes.

Other bilateral/multilateral activities

- NFIR research projects at EPRI (U.S.A.),
- co-ordination of fuel issues for European Pressurized Water Reactors (EPR),
- Studsvik's international fuel R&D projects.

PART IV

PRIORITIES FOR FUTURE WORK

1. Coverage of the selected topics by international projects

The subjects selected by the Task Force, as described in Part II, are all considered as being important. Some of the issues identified are already covered by activities carried out by other agencies, in particular the IAEA.

It was agreed that the work carried out within the NSC will be co-ordinated with the IAEA and that duplication will be strictly avoided.

In particular, the International Atomic Energy Agency has set up an International Working Group on Fuel Performance and Technology (IWGFPT). Among the important topics identified by the task force, the IWGFPT programme covers the following:

a. Thermal conductivity - Zircaloy oxide

A review entitled *Corrosion of Zirconium Alloys in Nuclear Power Plants* (TECDOC-684, 1993) was published and is currently being updated. A considerable part of the review deals with the formation, structure, physical and mechanical properties of the various zirconium oxides in water-cooled nuclear reactors. While this work does not focus on thermal conductivity in particular, oxide growth rates and thicknesses under various circumstances will be of considerable importance in any assessment of thermal conductivity of cladding.

b. Stress corrosion cracking - Zircaloy

A Co-ordinated Research Programme (CRP) in this specific area has recently been initiated. It is co-sponsored by BNFL and the host laboratory is AEA Technology, Risley. Four participating laboratories from four other countries share the workload of a large experimental test matrix. The aim is to obtain more detailed information on cracking rates for various concentrations of iodine and at various temperatures. The results will yield useful data to include in modelling calculations and may give further insight into the fundamental mechanisms involved.

c. Water chemistry

Water chemistry and zirconium alloys corrosion have been issues of constant preoccupation to the IAEA since the beginning of the 1980's. Two CRPs were carried out: the first one on Coolant Cladding Interactions (CCI) and the second, WACOLIN, on water chemistry, activity transport mechanism and corrosion in the primary coolant system. In 1989 a Technical Committee Meeting on "Fundamental aspects of corrosion on zirconium base alloys in water reactors environments" was held in Portland, U.S.A., and from 1988 to 1991 the IAEA worked on "Corrosion of zirconium alloys in

nuclear power plants" and issued a widely used publication. In 1993 a Technical Committee Meeting on "Influence of Water Chemistry on Fuel Cladding Behaviour" was held.

On-going activities include:

- Updating of the publication: *Corrosion of Zirconium alloys in nuclear power plants*;
- Co-ordinated research programme on "High Temperature On-line Monitoring of Water Chemistry and Corrosion".

d. Hydrogen measurement techniques (in cladding)

Hydrogen pick-up by cladding is inextricably linked to the corrosion processes which take place in water-cooled nuclear reactors. Updated information on this topic will be included in the revised version of TECDOC-684.

In addition, part of IAEA's CRP on the Degradation of Materials in Spent Fuel Storage deals with the fragility of hydrided cladding. The programme in question attempts to model the distribution of hydrides in cladding when it is removed from the reactor to the spent fuel bay, taking into account the important phenomenon of thermal diffusion and known data on the terminal solid solubility of hydrogen in the Zircaloys. This modelling is backed up by actual experimental measurements of hydride distributions in the cladding.

e. Fuel failures propagation (secondary failures)

The subject of fuel failures in water reactors has been on the work programme of the IWGFPT since its beginning. It has been touched upon briefly in most specialists' and topical meetings held, related to fuel behaviour (PCI, high burnup, etc.). On the basis of the conclusions and recommendations of these meetings the IWGFPT organised a specific specialists' meeting on the subject in Dimitrovgrad (1992).

On the same subject a CRP by the IAEA is under preparation. Its main objectives are:

- To review and report on the occurrence, main causes, mechanisms, consequences and management of fuel failures (including detection and location);
- To encourage information exchange;
- To provide information for use of fuel designers, operators and developers of water reactors.

A special manual in preparation will include the following subjects:

- Fuel Failures, Occurrence and Relevance to Reactor Operation,
- Monitoring and Detection of Fuel Failures,
- Characterisation of Causes and Mechanisms of Primary Failure and their Mitigation (fabrication, PCI, corrosion, etc.),
- Fuel Failure Analyses (techniques of evaluation, data needed for evaluation, supporting test and modelling),

- Fuel Failure Evolution
  (secondary effects and mechanisms, release of fission products and loss of fuel, experience with continued irradiation of failed fuel),

- Fuel Failure Management
  (existing types of operating criteria, use of repaired and reconstituted fuel, back end aspects including handling, transport, storage).

This proposal was approved at the IWGFPT Regular Meeting in Vienna, 9-11 May 1994.

f. Spent fuel - long term behaviour

The CRP on BEFAST involves the extended interim storage of spent fuel and compiles the on-going experiences of the participating countries in both wet and dry fuel storage. The IAEA also has several programmes specifically related to material problems in spent fuel storage facilities. These include a CRP on the Irradiation Enhanced Degradation of Materials in Spent Fuel Storage Facilities and the preparation of a document on the experience with various fuel cladding alloys during long-term storage in pools.

All of these programmes address the mounting concerns about ageing irradiated fuel in ageing spent fuel storage facilities. Many such facilities are now operating beyond the service life envisaged at the design stage. Although the experience with fuel storage to date has been generally excellent, (for example, spent fuel from some research reactors has been safely and reliably stored for periods approaching 40 years), there is the knowledge that corrosion is proceeding at some non-negligible rate. Consequently, there is a need to collect, analyse and summarise the existing information both of a quantitative and qualitative nature to help facility operators assess the viability of extending the storage time for several more decades.

2. Identification of areas and means of co-operation and co-ordination

Of particular importance for modelling fuel behaviour at high burnup are the topics listed below. Work on these subjects does not overlap with activities of other international agencies, in particular the IAEA.

a. Issues of high priority

Among the several identified phenomena (see Part II), the understanding of the following is considered as being of highest importance for improved fuel behaviour modelling:

- *Thermal conductivity*

  Although this has been extensively studied, local measurements at different burnup levels and temperatures are needed in order to better determine the influence of the accumulation of fission products and changes in stoichiometry. All details of the degradation of the local thermal conductivity need better experimental support which must be accomplished by a better theoretical understanding. The thermal conductivity of the rim structure is unknown and needs to be investigated. The surface morphology and composition of LWR fuel and cladding at high burnup should be investigated. Of lesser importance to integral behaviour is thermal conductivity of zirconium oxide; it needs further clarification, in particular for corrosion, creep and deformation of clad.

- *Fission gas release*

  No quantitative description of fission product damage to the lattice exists today. Radiation enhanced diffusion through the lattice and sputtering (evaporation of fission products along the heated path created by fission products) are two examples. It is recommended to study the phenomenon by irradiating fuel pellets with heavy ions having masses and energies typical of fission products. The phenomenon could also be modelled using molecular dynamics methods. More research on the very high local burnup and on athermal release from the rim is necessary.

- *Fission product swelling and uranium oxide creep*

  These two topics are related as it has already been established that gaseous swelling is dependent on the hydrostatic stress. Hence the amount of swelling is governed by the space available and the ability of the fuel to fill the space by creep. There is therefore a requirement for two forms of experiments: the first to determine creep properties and the second to provide a matrix of data demonstrating the interplay of swelling and creep.

  The combined effect of swelling and creep are best demonstrated through the power ramping of fuel rods under different conditions. In this context it would be useful to vary: the magnitude of the power ramp and hence the distribution of fuel temperature, the burnup of the fuel rods to investigate the effect of the driving force on swelling, different clad thicknesses, pellet geometries (hollow, dimpled, solid, etc.) and system pressure to investigate the effect of restraint. Post test analysis would include diametral strain and other dimensional changes, e.g. dimple filling and bore closure of hollow pellets, and porosity distribution. Such a series of tests would enable the construction of a mechanistic model for pellet dimensional changes under ramping conditions.

- *Modelling of thermo-mechanical behaviour; constitutive equations*

  The issue of constitutive equations should be addressed; this would be an important step forward in fuel behaviour modelling. Increased availability of well characterised experimental data would improve the knowledge of parameters required by these equations.

- *High burnup fuel in transient conditions*

  The transient and accident issues referred to in Appendix B require clarification and due attention.

b. Means of co-operation and co-ordination

- *Data base for fuel rod modelling:*

  After 30 years of testing and development of fuel rods a large body of data and experience already exists. The relevant, major results of irradiation experiments are usually published, however, due to the proprietary nature in many cases, details of design and irradiation history are restricted or even missing. In other words very few experimental results available in the public domain can be directly used for the validation of fuel rod computer codes.

  Fuel rod behaviour has been investigated systematically by different organisations over the last decades and many basic questions were solved. Most of the results must be considered as standard tests for any fuel rod code. However, the information is spread over many reports that may not be readily available. Clearly, the use of results from diverse projects can be facilitated by establishing a central comprehensive database.

  Such a data base is certainly needed as it could improve the reliability of fuel rod codes and set an international standard for quality assurance for fuel rod models.

- *Computer code benchmarking and validation*

  Computer code validation and benchmark studies are very valuable and are necessary steps in code development. This activity would derive considerable benefit from the computer programme services of the NEA Data Bank and would ensure that the distributed codes have been validated.

- *International topical meetings* covering high priority issues are an effective method for establishing co-operation and co-ordination.

PART V

RECOMMENDATIONS AND CONCLUSIONS

1. Issues of high priority

In order to further reduce the uncertainties in the modelling of thermal behaviour and to improve modelling at high burnup, the following phenomena should be treated with high priority:

- thermal conductivity,
- fission gas release,
- fission product swelling,
- thermo-mechanical behaviour; constitutive equations.

It is recommended that countries and organisations provide effort and resources to address these topics. It is proposed that topical meetings covering these issues are organised by NEA/NSC in co-ordination with the IAEA.

The issue of high burnup fuel behaviour in transient conditions should receive high priority. The CSNI will address this issue in a specialists' meeting to be held from 12-14 September 1995, in Cadarache, France.

2. Means of co-operation and co-ordination

a. Experimental data sets

1. It was suggested that a consultant be engaged to review and evaluate existing well characterised experiments, covering issues and phenomena of the highest priority. The objective is to identify data sets for which quality criteria are met, and assess consistency, completeness and limitation in range of the data parameters. The best existing format for storage and retrieval of the data should be identified.

   The review should concentrate on data sets which can be shared at international level under defined conditions. However, also proprietary data sets should be identified, as it is anticipated that data sets will be more widely released if resources are made available for structuring them into a standard form.

   This proposal was already discussed and approved by the NSC who also agreed that an expert should be engaged to review the well characterised experiments and to propose which ones should be included in a data base that would be generally available. The review is published as a separate report [1].

2. It is recommended that, based on the review of the consultant and discussion within the Task Force on the data sets to be selected, a data base is set up. As a start, data from the IFE-OECD Halden Reactor Project should be included. Experts and consultants should be identified to carry out the work.

   It is estimated that a period of two years is required for carrying out the bulk of the work. Additional data sets of particular importance will be added as they become generally available.

3. It is strongly recommended that the established data base is updated, maintained and distributed by the NEA Data Bank.

b. Computer code validation and benchmarking

International code benchmarking is of great value, for computer code and model validation, for an exchange of ideas and experience among modellers and as a learning process for model users. The FUMEX exercise organised by the IAEA is an excellent example. Further comparison exercises are encouraged.

References

[1] J. A. Turnbull, A Review of Fuel Behaviour Data Available from the IFE-OECD Halden Reactor Project for Development and Validation of Computer Codes, OECD/NEA Report, January 1995.

APPENDIX A

List of Authors and Issues of Major Importance

Topics of PART II		***Co-ordinators/Authors***
Chapter 1	**Thermal analysis**	K. LASSMANN, CEC/ITU
Chapter 2	**Fission gas release**	J. F. MARIN, CEA C. M. LEMAIGNAN, CEA
Chapter 3	**Fission product swelling**	J. A. TURNBULL, U.K.
Chapter 4	**Stress corrosion cracking**	F. IGLESIAS, Ontario Hydro B. COX, University of Toronto B. J. LEWIS, Royal Military College
Chapter 5	**Constitutive equations**	K. LASSMANN, CEC/ITU G. VALLI, ENEA
Chapter 6	**Water chemistry**	P. CHANTOIN, IAEA
Chapter 7	**Hydrogen measurement techniques (in cladding)**	H. KWAST, ECN
Chapter 8	**Failed fuel behaviour**	C. VITANZA, IFE-OECD HRP
Chapter 9	**Spent fuel - long-term behaviour**	T. FURUTA, JAERI
Chapter 10	**Quality assurance process for new integral experiments and their utilisation in computer codes**	C. VITANZA, IFE-OECD HRP P. CHANTOIN, IAEA J. A. TURNBULL, U.K.

APPENDIX B

High Burnup Fuel Behaviour in Transient and Accident Conditions

As regards changes in the fuel condition with increasing burnup, the properties of interest that develop in normal steady-state operation would have implications on the behaviour during transients and accidents as well. It appears that several transient and accident conditions certainly require further attention due to uncertainties in the fuel behaviour at the current high end of burnups and beyond. The great majority of the experimental research on behaviour in accident conditions has been carried out with fresh fuel, and only in few others 30 MWd/kgU is exceeded.

One of the issues calling for particular attention is the reactivity initiated accident (RIA) behaviour. The changes due to exposure can effectuate a rod failure mode that is different from the one of fresh or low burnup fuel; failure limits drastically lower than expected or lower than current acceptance criteria are reported from single tests. When quantifying and modelling of these will be attempted, close account has to be taken of the burnup effects, particularly distributions of fissile nuclides, power densities and burnup, fission gas distribution and release, fission gas swelling during the transient, thermal and mechanical effects of the "rim" zone. Also changes in the cladding mechanical and chemical state, have to be taken into account. Many of the contributing factors are insufficiently known at the relevant burnup levels. Testing and verification of failure limits and margins to fuel dispersion remain to be performed; this is especially demanding due to the difficulties in the preparation of pre-irradiated test rods and the high requirements for representative test conditions and instrumentation. Another complication is the fact that the behaviour may prove to depend much on the reactor type and fuel design. The extent of the effort required to elaborate these aspects of LWR fuel behaviour strongly suggests seeking international co-operation. Japan (JAERI/NSRR) [1] and France (IPSN/Cadarache) [2], at least, have ongoing experimental programmes on this issue.

Concern has also been expressed as to the fuel strong cracking and fission gas release in a later phase of a large break LOCA transient with high burnup fuel in certain conditions (FLASH 5 test at CEN Grenoble [3] and earlier Karlsruhe tests [4]).

Still another area of high burnup fuel transient behaviour deals with BWR flow transients leading to degraded cooling, short term dryout and operation at elevated clad temperatures. There is a need to consider that the consequences of operation at dryout after prolonged irradiation may be dependent on the rod conditions. Again, the details of heat transfer in the path from inside the fuel to the coolant – fuel, rim, gap, surface – is of importance. The issue of possible differences of heat transfer characteristics of irradiated (oxidised) cladding in comparison with fresh surfaces should be addressed. Also here, the importance of careful precharacterisation and post-test examinations are emphasised. A programme to address these phenomena is being outlined in the Halden Project with solicited international participation.

References

[1] Ishijima, K. et al., Behaviour of preirradiated LWR fuel rods under reactivity initiated accident conditions. IAEA TC on Behaviour of core materials and fission gas release in accident conditions in LWRs. Aix-en-Provence 16.-19.3.1992.

[2] Papin, J. et al., Irradiated fuel behaviour during reactivity insertion accidents of light water reactors: research and development studies at the CEA-IPSN, France. Ibid.

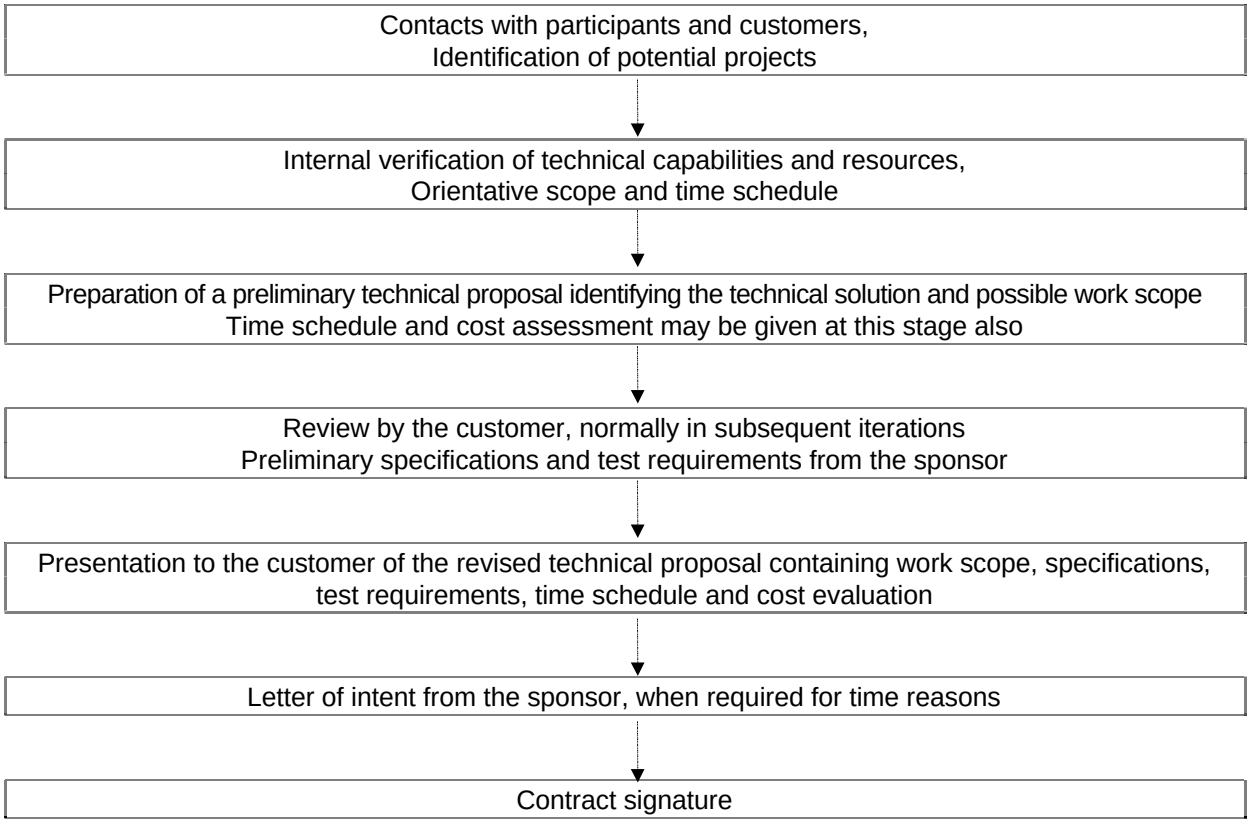
[3] Bruet, M. et al., High burnup fuel behaviour during a LOCA type accident: the FLASH 5 experiment. Ibid.

[4] Karb, E.H., In-pile testing at Karlsruhe of LWR fuel-rod behaviour during the heat-up phase of a LOCA. Nuclear Safety, Vol 21, No. 1, Jan-Feb 1980.

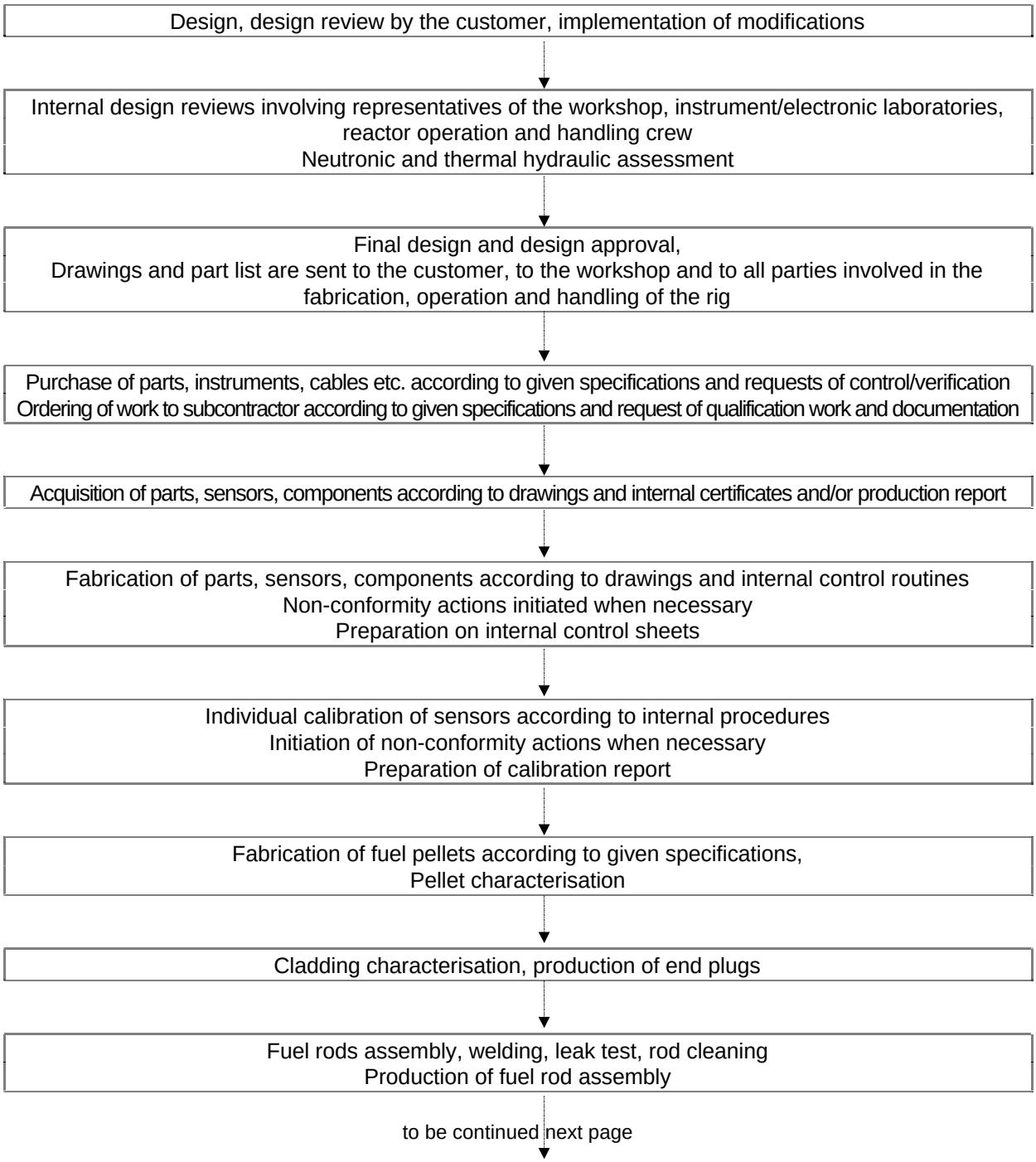
APPENDIX C

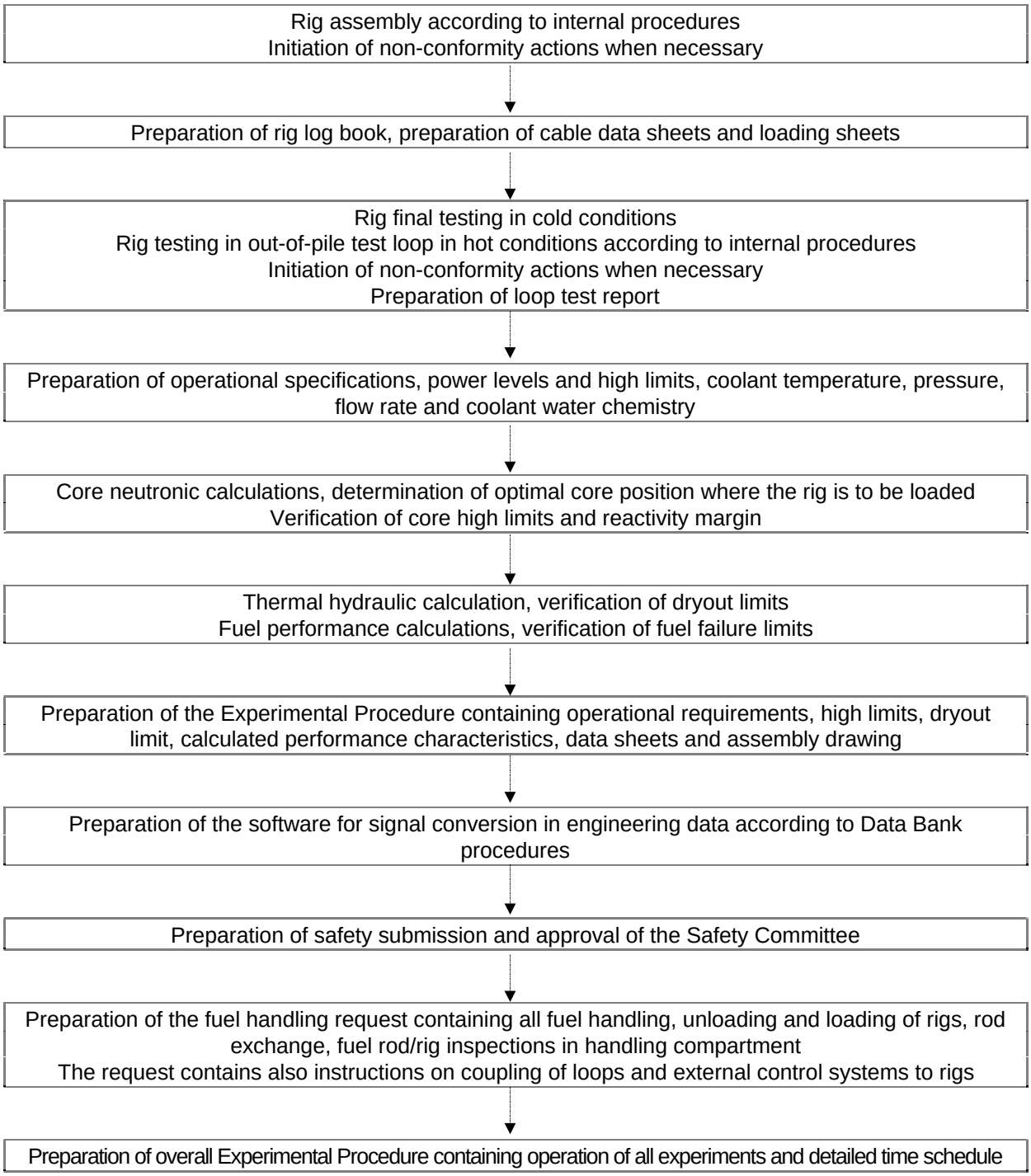
"Check List": Sequence of Actions/Tasks related to In-pile Experiments

1. Job Initiation

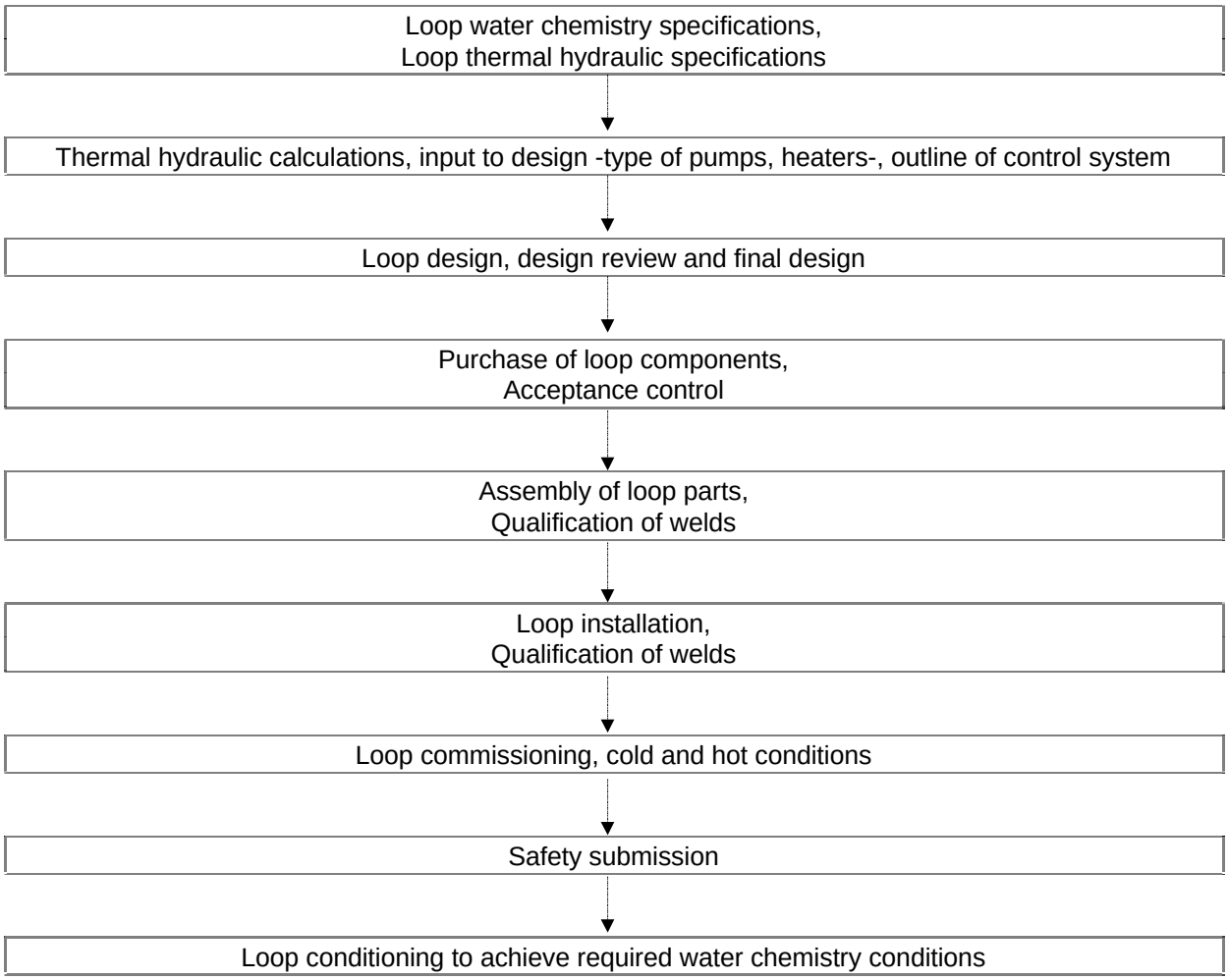


2. Design, Fabrication, Control, Installation

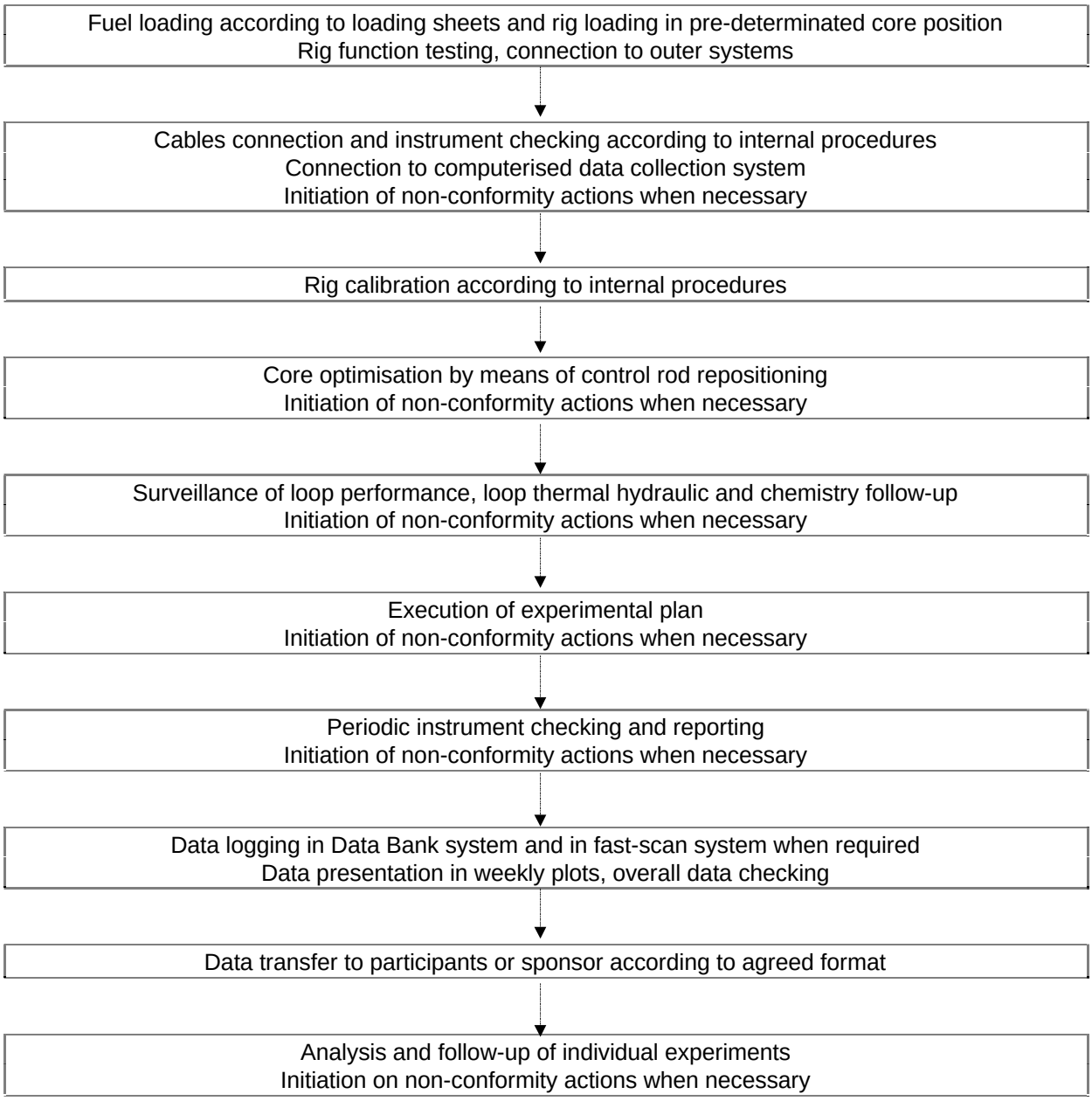



Rig assembly according to internal procedures
Initiation of non-conformity actions when necessary
Preparation of rig log book, preparation of cable data sheets and loading sheets
Rig final testing in cold conditions
Rig testing in out-of-pile test loop in hot conditions according to internal procedures
Initiation of non-conformity actions when necessary
Preparation of loop test report
Preparation of operational specifications, power levels and high limits, coolant temperature, pressure, flow rate and coolant water chemistry
Core neutronic calculations, determination of optimal core position where the rig is to be loaded
Verification of core high limits and reactivity margin
Thermal hydraulic calculation, verification of dryout limits
Fuel performance calculations, verification of fuel failure limits
Preparation of the Experimental Procedure containing operational requirements, high limits, dryout limit, calculated performance characteristics, data sheets and assembly drawing
Preparation of the software for signal conversion in engineering data according to Data Bank procedures
Preparation of safety submission and approval of the Safety Committee
Preparation of the fuel handling request containing all fuel handling, unloading and loading of rigs, rod exchange, fuel rod/rig inspections in handling compartment
The request contains also instructions on coupling of loops and external control systems to rigs
Preparation of overall Experimental Procedure containing operation of all experiments and detailed time schedule

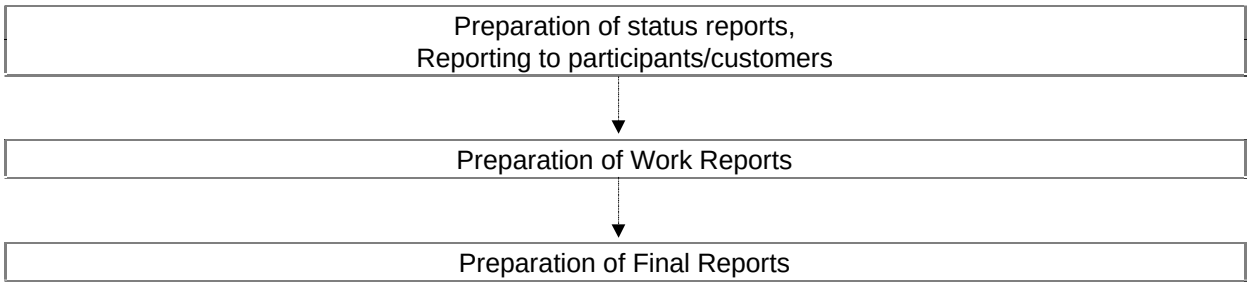
3. Loop Design and Fabrication *(simplified)*



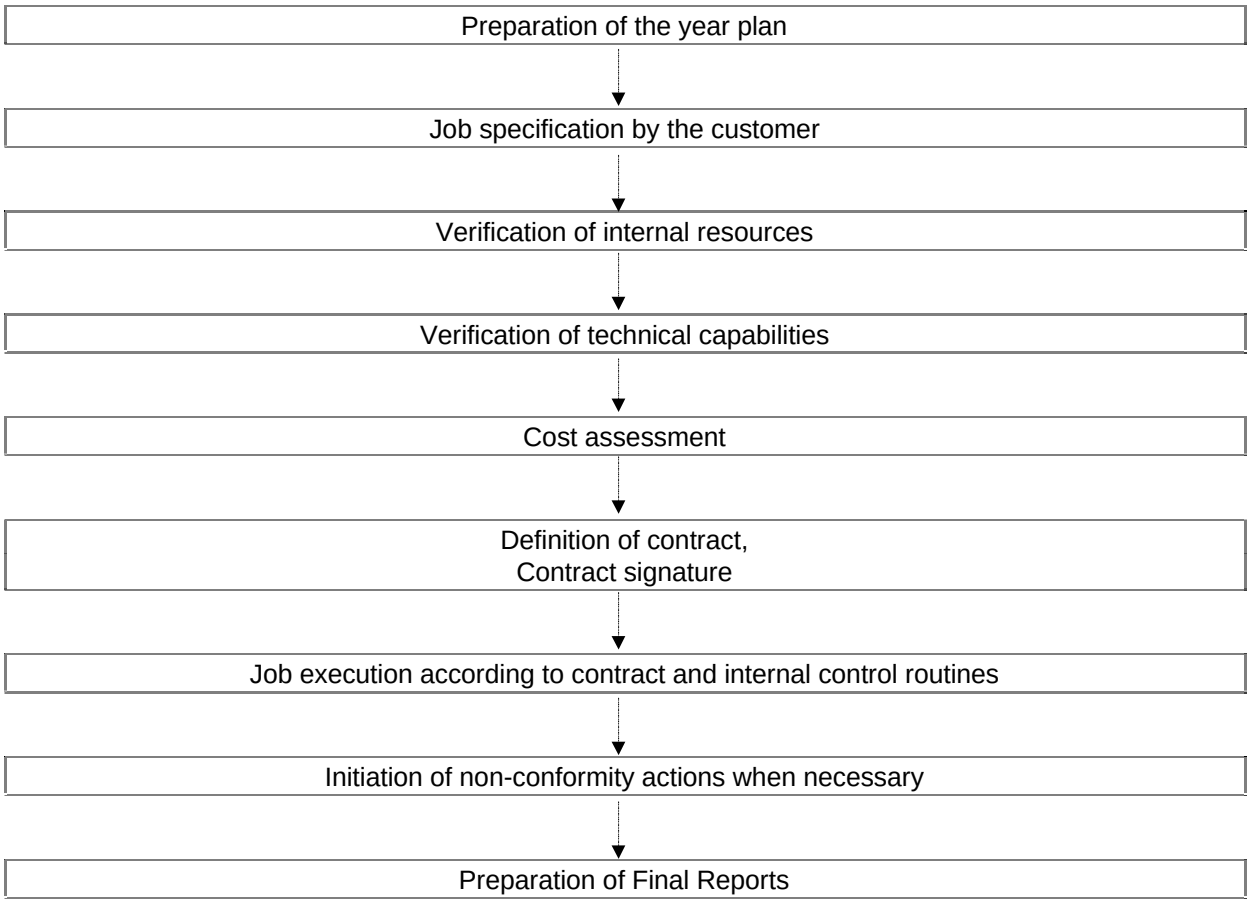
4. In-pile Operation



5. Reporting of In-Pile Test



6. Post-Irradiation Examinations *(simplified)*



APPENDIX D

Glossary of Abbreviations Used

AEA	Atomic Energy Authority (U.K.)
AECL	Atomic Energy of Canada Limited
AFR	away from reactor storage
AGR	advanced gas reactor
AR	at reactor storage
ATR	advanced thermal reactor
Å	Ångström (10^{-10} m)
at%	atomic per cent
BEFAST	*BE*haviour of spent *F*uel *A*ssemblies during extended *ST*orage
BNFL	British Nuclear Fuels Ltd.
BWR	boiling-water reactor
°C	degree Celsius
CANDU	*CAN*ada *D*euterium *U*ranium
CRP	co-ordinated research programme
CSNI	Committee on Safety of Nuclear Installations (OECD/NEA)
EDF	Electricité de France (French electricity utility)
EPR	European pressurised reactor
EPRI	Electric Power Research Institute (U.S.A.)
FA	fuel assembly
FBR	fast-breeder reactor
FGR	fission gas release
FR	fast reactor
GANIL	Grand Accélérateur national d'ions lourds (France)
HFR	high-flux reactor (Netherlands)
HRP	Halden Reactor Project (IFE-OECD)
HTGR	high-temperature gas reactor

IFE	Institute for Energy Technology (Norway)
IPSN	Institut de protection et de sûreté nucléaire (France)
IWGFPT	International Working Group on Fuel Performance and Technology (IAEA)
JAERI	Japan Atomic Energy Research Institute
K	degree Kelvin
keV	kiloelectronvolt (10^3)
LOCA	loss-of-coolant accident
LWR	light-water reactor
MAGNOX	fuel *MAGN*esium-*OX*ide cladded
MeV	megaelectronvolt (10^6)
MOX	mixed oxide (uranium/plutonium) fuel
MTR	material testing reactor
MWd/kgU **MWd/kgUO_2**	megawattdays (10^6) per kilogram of uranium metal megawattdays (10^6) per kilogram of uranium oxide MWd/kg is equivalent to the unit GWd/t, gigawattdays (10^9) per tonne. In Canada MWh/kg is used, which differs by a factor 24 from both MWd/t and GWd/t
µm	micrometre (10^{-6} m)
NEA	Nuclear Energy Agency (OECD)
NFIR	Nuclear Fuel Industries Research (EPRI)
NSC	Nuclear Science Committee (OECD/NEA)
NSRR	Nuclear Safety Research Reactor (JAERI)
O/M	oxygen-to-metal ratio (stoichiometry)
OECD	Organisation for Economic Co-operation and Development
PCI	pellet-clad interaction
PHWR	pressurised heavy-water reactor
ppm	part per million
PWR	pressurised-water reactor
RIA	reactivity-initiated accident
RMBK	Reactor Bolchoe Molchnastie Kipiachie, Russian high-power boiling-water reactor
SCC	stress corrosion cracking

t	tonne (10^3 kg)
T	temperature
UOX	uranium oxide (UO_2)
VTT	Technical Research Centre of Finland
W	watt
WWER	Russian water-water power reactor