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**Task Force on Computing Radiation Dose and Modelling of
Nuclear Radiation-Induced Degradation of Reactor Components**

**ISSUES OF IRRADIATION-INDUCED AGEING OF REACTOR COMPONENTS
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ISSUES OF IRRADIATION-INDUCED AGEING OF REACTOR COMPONENTS

Introduction

1. Currently, the issues of reactor components ageing due to irradiation present some of the most important challenges of nuclear engineering. As nuclear reactors are ageing across the world, the ability to accurately predict the status of various reactor components is becoming critical from the point of view of nuclear safety and extension of reactor operating life.
2. If reliable and accurate computational techniques can be developed to predict accurately doses incurred by various reactor components throughout their lifetime, then utilities can possibly justify extending the life of their reactors. Of course, this could potentially imply considerable financial savings.
3. The issues of reactor component ageing can be divided into two major subfields:
 - materials research on the mechanisms of material degradation due to irradiation and development of its theoretical models,
 - the experimental techniques of measuring and the computational techniques (i.e., neutron transport methods) of calculating doses incurred by various reactor components.
4. In the following sections we give some insight into the problems faced when dealing with irradiation-induced ageing of reactor components.

Phenomena of material degradation with time relevant to reactor components

5. Reactor components sustain damage due to irradiation. The main damaging process[1] is a result of neutron interaction with atoms of the material lattice causing displacement and displacement cascades blocking the movement of dislocations and thus leading to reduction in material toughness. Additionally, precipitates can be formed (e.g. Cu and P) leading to precipitation hardening[1] of the material. The net effect of these processes is the irradiation embrittlement[1] of the reactor components materials (including welds). There are several clearly defined parameters[1] contributing to neutron embrittlement. These are:

- neutron flux;
- material temperature (low temperature neutron exposure increases strongly the neutron damage);
- steel composition (e.g. P, Ni segregate at the grain boundaries);
- steel microstructure.

Additional factors in irradiation embrittlement are:

- thermal ageing;
- strain ageing;
- hydrogen embrittlement.

6. The key to understanding the mechanism of irradiation embrittlement is a model describing the microstructure of materials exposed to neutrons at the atomic level. Unfortunately, such a complete model does not yet exist[1].

In Table 1, we present an overview[2] of various factors and their effects contributing to material ageing. In Table 2, we present the effects that various processes[2] defined in Table 1 have on specific reactor components (a distinction is made in Table 2 between the processes known as damaging and the processes considered potentially damaging to materials). It can be seen in Tables 1 and 2 that the neutron flux is not the only cause of material degradation.

7. In the remainder of this document we shall concentrate on the neutron flux as the leading factor of the irradiation embrittlement.

Neutron Flux And Irradiation Embrittlement

8. The neutron flux is composed of neutrons of varying energy levels. The neutron fluence (a time integrated quantity) is defined as a count of neutrons above a certain threshold energy. The question of what this threshold energy should be so that the "right" part of the spectrum (i.e., neutrons at energies causing material damage) is chosen is still causing a controversy[1]. In general, only neutrons with energies above 1 MeV are considered to contribute to material damage; the value of $E_0=1$ MeV in fluence calculations is accepted in France being the easiest part of the flux spectrum to measure experimentally. In Russia, the threshold is set at $E_0=0.5$ MeV (then the spectrum contains about 30 percent more neutrons) and in the USA at $E_0=0.1$ MeV.

9. The assumption that neutrons below certain threshold energy do not cause any material damage is clearly a crude approximation of the reality as the threshold energy[3] (set somewhat arbitrarily) does not reflect the actual physics of the material lattice. Moreover, the threshold energy concept implies that all neutrons of energies greater than this particular value produce the same damage - yet another crude[3] approximation. Hence, a different fluence parameter called DPA (Displacement Per Atom) was introduced[3] to represent the material damaging effects of neutron at all energy levels. DPA is thus a much more conservative[3] parameter than fluence calculated using a threshold energy value and yields a higher embrittlement value[3] (i.e., a lower fracture toughness) than the threshold fluence calculations. Unfortunately, DPA as much as it accounts for the full neutron energy spectrum is not a precise[3,4,5] parameter either. This is because at operating temperatures a large number of DPA are restored immediately and only an unknown number of DPA actually remain to cause the residual damage. Moreover, the DPA cross-section approach is valid only[4,5] for monoatomic crystalline materials (and NOT for composite materials such as e.g. alloys). In recent studies[5], it was determined that a knowledge of the PKA (i.e., Primary Knock-on Atom) spectra in a crystalline medium is a fundamental information required for the in-depth investigation of the neutron radiation damage effects. In particular, the dimensions of the cascades and stable defects, after the temperature induced recombination of vacancies and interstitials, are strongly influenced by the PKA spectra. These spectra can be further used[5] to define damage parameters (e.g., inter-displacement model) for polyatomic materials which cannot be treated correctly with the DPA approach. A code is being developed[5] at ENEA Bologna

(PRISMA - Primary Spectrum in Materials) to produce PKA spectra from evaluated data libraries. It would be useful[5] to extend this method to nuclides of stainless steels.

Additional confusion[4] arises from the fact that different units of DPA according to different models have been used by different laboratories making the comparison of irradiation effects on materials in different reactors and locations very difficult. Hence, there is a need[4] for an international standard for a displacement dose unit.

10. Furthermore, the evidence of the higher-than-expected irradiation damage[3] of the RPV (Reactor Pressure Vessel) and reactor components of the HFIR reactor[3] (High Flux Isotope Research Reactor - a light-water-cooled, low temperature high performance research reactor at ORNL in the USA) indicates that the importance of the thermal neutron flux has long been underestimated[3].

11. The HFIR neutron spectrum consists in 95 percent of thermal neutrons and its vessel operating conditions are similar to the LWR RPV operating conditions (i.e., low temperature of 70-130 C, low neutron flux, and possibly high thermal-to-fast neutron flux ratio); hence, the importance of the HFIR findings for the PWR reactor components surveillance programs[3].

12. Studies[3] have shown that the mechanism of the thermal neutron induced material damage is that of (n,γ) reactions characterised by recoil energies of about 500 eV and (n,α) reactions (e.g. with boron) characterised by average recoils of approximately 1 keV. In contrast, the mechanism of the fission neutrons ($E > 1$ MeV) induced material damage[3] is that of elastic and inelastic (n,n') reactions or transmutation reactions (n,p) or (n,α) characterised by recoil energies in the range of 10 keV to 100 keV. In particular, the fast neutrons induced recoils produce a large number of material defects of which only a small fraction survive to cause permanent material damage whereas the thermal neutron induced reactions produce a small number of point defects of which most survive to contribute to permanent material damage. In fact, the point defects survival rate[3] for low energy recoils is about 0.3 in comparison to 0.05 survival rate for the recoils originating from fast neutrons of energies above 1 MeV.

13. The evidence[3] presented indicates that:

- more effort should be directed towards better experimental and computational dosimetry of thermal neutrons for reactor components,
- the effects of gamma rays on material ageing should be considered.

Computational problems of reactor components dosimetry

14. The computational schemes for evaluating fast neutron fluence at the RPV and reactor cavity (i.e., a volume outside of the RPV locations filled with air) are quite well established yielding results within 15 percent accuracy in comparison to measurements (this number varies depending on the measurement position and deteriorates for the positions outside of the reactor vessel). The flow of calculations can be divided into three fundamental steps[6]:

- calculation of the multigroup fast neutron flux (including those neutrons which become thermalized);

- calculation of the neutron fluence at desired locations;
- calculation of DPA caused by high energy neutrons.

15. Calculation of the multigroup neutron flux is carried out using a transport code. The transport methods of choice are: state-of-the art S_N or Monte Carlo codes with advanced geometrical modelling capabilities. This calculated multigroup neutron flux is then adjusted by least square fitting[6] within its uncertainty interval by using the responses of dosimeters (sensitive to the desired energy ranges) irradiated at locations of interest in the reactor and reactor cavity. Typical[6] detectors (and reactions) used are:

- $\text{Co}^{59}(n,\gamma)\text{Co}^{60}$, $\text{Fe}^{58}(n,\gamma)\text{Fe}^{59}$, $\text{Cu}^{63}(n,\gamma)\text{Cu}^{64}$ - in the thermal energy range;
- $\text{Np}^{237}(n,f)\text{FP}$, $\text{U}^{238}(n,f)\text{FP}$, $\text{Th}^{232}(n,f)\text{FP}$ - in the energy range 0.67 to 1.52 MeV to estimate the fission reaction rates;
- $\text{Nb}^{93}(n,n')\text{Nb}^{93m}$, $\text{In}^{115}(n,n')\text{In}^{115m}$, $\text{Ti}^{47}(n,p)\text{Sc}^{47}$, $\text{Ni}^{58}(n,p)\text{Co}^{58}$, $\text{Fe}^{54}(n,p)\text{Mn}^{54}$, $\text{Ti}^{46}(n,p)\text{Sc}^{46}$, $\text{Ti}^{48}(n,p)\text{Sc}^{48}$, $\text{Cu}^{63}(n,\alpha)\text{Co}^{60}$, $\text{Al}^{27}(n,\alpha)\text{Na}^{24}$ - for estimating threshold reactions in the energy range 0.5 MeV to 6.5 MeV.

16. In view of the importance of the thermal neutron induced material damage via (n,γ) and (n,α) reactions[6] induced by thermal neutrons (e.g. an (n,α) reaction in thermal energy range with boron) the absence of an (n,α) detector (in the above list and in thermal energy range) may result in the underestimation of the DPA induced by thermal neutrons. In a recent paper[9], the thermal neutron activation in Fe is reported to be underestimated by 25 percent in computations. In general, the accuracy of computations in the thermal energy range is far lower than in the fast energy range. This is due to the energy group structure and approximations used in the thermal energy range (e.g. no upscattering is considered) which was normally neglected in the dosimetry calculations.

17. The measured activation of irradiated detectors is used to calculate the fluence of neutrons which combined with precalculated multigroup neutron flux and displacement cross-sections allows to calculate the number of DPA[6].

Uncertainties implicit in transport calculations

18. There is a number of uncertainties[7] associated with the computations using transport codes. These are:

- Numerical approximations (quadrature used in S_N calculations, scattering cross-section expansion, mesh spacing, energy groups, statistical convergence criteria, etc.);
- Modelling approximations (capsule placement, PV thickness variations, cavity streaming, 3-D flux synthesis, peripheral subassembly source distribution, dimension and material uncertainties);
- Nuclear data uncertainties (cross-sections, dosimeter cross-sections, U^{235} fission spectrum for $E > 6$ MeV).

Most recent advances in dealing with computational uncertainties

19. In recently reported publications[8-15], all of the above defined computational uncertainties have been addressed and many have been reduced to acceptable levels. Thus, based on most recent publications[8-15], we have

- S_g quadrature set is reported to be sufficient[13];
- P_3 (for neutrons) and P_5 (for gamma rays) scattering cross-section expansion is commonly used[10] although a more accurate quadrature may be needed[13];
- as fine as possible group structure should be used and attention should be paid to the actual group boundaries; currently, cross-section libraries from 47 energy groups up to "continuous" Monte Carlo data sets are used[8-15];
- an uncertainty of about 20 percent in the flux in the capsule is reported[8] if the radial position of the capsule is known within 0.5 in;
- cavity streaming is reported[13] not to be a significant issue (S_N codes);
- the 3-D flux synthesis based on 2-D and/or 1-D calculations is reported to be an acceptable procedure[9,10,12,13,14] (S_N vs. Monte Carlo comparisons) but the evidence to the contrary is given[9] for positions in the cavity;
- peripheral subassembly source distribution may differ about 3-5 percent from the average[8] (reported using Monte Carlo method);
- maximum effect of varying dimensions within the manufacturer tolerances is estimated[8] at about 2.5 percent (reported for Monte Carlo results);
- material uncertainties in steel compositions have no significant effect but coolant density variations may cause about 6 percent errors in flux predictions at measurements positions[8] but smaller values are also reported[15];
- cross-sections are reported as the major source of uncertainties[8-15] with the inelastic scattering cross-section for Fe causing as much as 9 to 13 percent errors in high energy neutrons reactions predictions in the RPV and/or cavity (the problem does not exist for the low energy reactions); conflicting conclusions are given for using ENDF/B-V versus ENDF/B-VI data for Fe reactions depending on the computational method used[13] and on the Fe inelastic cross-section correction[15]; the uncertainty in the total hydrogen and oxygen cross-sections result in a 1 to 3 percent and approximately 1 percent uncertainty in the flux prediction respectively;
- dosimeter cross-sections are a source of major errors in the computations - the cross-sections for measured reactions have covariance matrices (for cross-sections) in reduced energy schemes resulting in the overall uncertainty in calculated responses of about 1 to 10 percent; the $Np^{237}(n,f)$ reaction is often underpredicted due to neglect of the photo-fission effects resulting in errors as high as 20 to 30 percent[8]; the photo-fission effect is also reported important for U^{238} [15];

- various ways of dealing with the U^{235} fission spectrum are reported ranging from using a slightly harder spectrum[9] through adjustment procedures[15] to analytical fits to experimental data[12].

Computational uncertainties - discussion

20. In the previous subsection the state-of-the-art minimal values of various uncertainties associated with the computations of flux/fluence in the dosimetry calculations were reported. It has to be stressed that the reported values of uncertainties were picked from publications in the list of references. Often, the computations presented utilise procedures, data sets, and modelling assumptions which are not optimal (arising from availability of particular data sets and tools at particular organisations). Consequently, the computations that follow contain extra errors which adversely affect the results. The problem is dealt with by devising "corrective factors" to force a better agreement of the results with measurements. This approach often leads to ambiguities and difficulties in interpreting the results. A more sensible approach would be to start with the best data available and modelling assumptions as accurate as possible and then proceed with calculations using various computational techniques.

21. Based on the state-of-the-art literature it is very difficult to infer the relative (with respect to each other) merits and the magnitude of associated uncertainties of various computational techniques (and nuclear data sets) used as these are very strongly problem dependent.

Proposal

22. Based on the presented evidence *two* conclusions can be made:

- I. that there is a need for further work on the development of the basic physics models (such as e.g. the PKA[5]) relating particle flux/fluence to material damage, and
- II. that a benchmarking study involving one reference problem and utilising different computational methods could be very useful in comparing relative merits of various reported in literature approaches. Such a study would clearly define the merits and shortcomings of various computational techniques as well as it would define the ranges of their applicabilities for a give problem. The study should utilise the S_N codes and Monte Carlo codes.

23. Recently, progress has been made in the area of simplified P_N [16,17,18] (SP_N) methods which resulted in robust 3-D algorithms performing very well in situations involving highly absorbing media (shielding calculations) where standard diffusion theory fails[16,17,18]. It is recommend that codes based on this new (and already available) technology be tested in the benchmark studies against the S_N and Monte Carlo codes (no such studies have yet been carried out). The extremely attractive feature of the SP_N methods is that they are very fast in comparison to S_N and especially Monte Carlo methods. Thus, if the SP_N technique proved to be sufficiently accurate it could become a valuable and cheap tool in dosimetry computations.

24. The proposed benchmark studies should address the additional research issues not discussed yet in this report such as:

- flux shape variation during the fuel cycle length (two different approaches have been reported in literature);

- impact of the core management on the flux/fluence calculations (new core management schemes alter irradiation conditions from cycle to cycle);
- further research on variance-covariance matrix approach to uncertainties in computations, measurements, and nuclear data;
- studies of flux/fluence at positions other than considered in all reported studies (i.e., inner surface of the RPV, within the RPV wall, and in the cavity) to estimate doses received by reactor structural materials (e.g. screws of the reactor header, control rod mechanisms);
- estimation of the "parasitic" fraction of the dosimeter activation due to neutron reactions with the material a dosimeter is made of;
- investigation of the accuracy of accelerated ageing of capsule material samples in higher flux environment in relation to inferring the material status at positions of lower flux values (i.e., are the effects of accumulating a high radiation dose in the short time equivalent to accumulating the same dose but over a longer period of time from the point of view of material ageing?)

Table 1. Potential effects on materials and components

	Temperature									
	Thermal ageing	Neutron flux irradiation	Load: static, cyclic, dynamic			Relative motion				
			creep	Water environment					wear	
				fatigue	corrosion					
toughness reduction	x	x	x							
grain des-integration							x			
cracking			x	x	x	x	x			
pitting							x	x	x	
swelling		x								
thinning							x	x	x	
denting							x			

Table 2. In-servicing ageing mechanism of LWR components

	Temperature					
	Thermal ageing	Neutron flux irradiation	Load: static, cyclic, dynamic	Relative motion		
				Water environment		
		creep	fatigue	corrosion	wear	
RPV vessel shell	o	x		o	o	
Fuel element clad	o	o	o	o	x	
SG vessel shell				o	o	o
SG tubes				o	x	
Main coolant piping				o	o	
Main coolant pump				x	o	
Feedwater/Wet steam piping				o	x	x
Turbine				x	o	x
Condenser tubes				o	x	

x = Leading ageing mechanism
o = Potential ageing mechanism

REFERENCES

- [1] H. Hänninen, "Phenomena of Material Degradation with Time Relevant to Reactor Pressure Vessels", Proceedings, NEA/CSNI Workshop on Safety Assessment of Reactor Pressure Vessels, Espoo, Finland, 8-9 October 1990, vol. 1, p.13
- [2] K. Kussmaul, J. Föhl, "Review of the State of the Art in the Assessment for Material Degradation with Time", Proceedings, NEA/CSNI WORKSHOP on Safety Assessment of Reactor Pressure Vessels, ESPOO, FINLAND, 8-9 October 1990, vol.2, p.255
- [3] R.J. McElroy *et al.*, "Low Temperature Embrittlement of LWR RPV Support Structures", Proceedings, NEA/CSNI Workshop on Safety Assessment of Reactor Pressure Vessels, Espoo, Finland, 8-9 October 1990, vol. 2, p. 311
- [4] Y. Orechwa, M. Salvatores, "NEACRP Recommendations for Standard Displacement Damage Dose Unit", NEACRP-L-343, September 1989, OECD Nuclear Energy Agency
- [5] M. Pescarini, "NJOY User Group Meeting, NJOY/NEADB-11, 8 June 1994
- [6] C.A. English *et al.*, "Approach and Methodology for Condition Assessment of Thermal Reactor Pressure Vessels", Proceedings, NEA/CSNI Workshop on Safety Assessment of Reactor Pressure Vessels, Espoo, Finland, 8-9 October 1990, vol. 1, p.61
- [7] F.B.K. Kam *et al.*, "Neutron Fluence Calculations and Uncertainty Analysis", Proceedings, NEA/CSNI Workshop on Safety Assessment of Reactor Pressure Vessels", Espoo, Finland, 8-9 October 1990, vol. 1, p.179
- [8] A. Avery *et al.*, "Calculation of Pressure Vessel Fluence in PWR's Using F/B-VI Data, Proceedings, 8th International Conference on Radiation Shielding", April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.677
- [9] B. Baers *et al.*, "Validation and Application of the Fast Kernel Based Program Preview for Out-of-Core Neutron Fluence Estimations in WWER-440 Reactors", Proceedings, 8th International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.686
- [10] E.B. Brodtkin, A.L. Egorov, "Calculated Neutron Spectra and Responses in the Pressure Vessel of a WWER-440 Using ENDF/B-4 and ENDF/B-6 Evaluations and Comparison with the Measured Spectra in WWER-440 Mock-Up", Proceedings, 8th International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.694
- [11] J.C. Nimal *et al.*, "French PWR 900 MWe Pressure Vessel Surveillance Neutron Field Characteristics TRIPOLI-3 Calculations and Experimental Determination", Proceedings, 8th

- International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.699
- [12] D.K. Parsons *et al.*, "Application of the TWODANT Code System to Pressure Vessel Dosimetry Calculations", Proceedings, 8th International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.706
- [13] J.C. Wagner *et al.*, "Comparison of Monte Carlo and Synthesized 3-D Deterministic Models for Reactor Cavity Dosimetry Calculations", Proceedings, 8th International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.714
- [14] B.G. Petrovic *et al.*, "Validation of SN Transport Calculations for pressure Vessel Fluence Determination at Penn State", Proceedings, 8th International Conference on Radiation Shielding, April 24-28, 1994, Arlington, Texas, U.S.A., vol. 2, p.721
- [15] D. Beretz *et al.*, "La Dosimétrie du Programme de Surveillance des Cuves REP", CEA/DRN, Rapport Scientifique 1994, p.89
- [16] D.I. Tomašević, E.W. Larsen, "Variational Derivation of Simplified P₂ Equations with Boundary Conditions", *Trans. Am. Nucl. Soc.*, **70**, 159 (1994)
- [17] D.I. Tomašević, E.W. Larsen, "The Simplified P₂ Approximation", *Nucl. Sci. Eng.*, to appear.
- [18] D.I. Tomašević, E.W. Larsen, "Variational Derivation of the Simplified P₂ Equations with Boundary and Interface Conditions", Proc. ANS Topical Meeting, International Conference on Mathematics and Computations, Reactor Physics, and Environmental Analyses, April 30 - May 4, 1995, Portland, Oregon, vol. 1, p.319 (1995)

