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Radiation Hardening in BWR Core Shrouds: Relative Roles of Neutron and Gamma Irradiation

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Abstract: We present a calculation of the displacement rates and freely migrating defect production caused by neutron and gamma irradiation and their roles on causing irradiation hardening in a BWR core shroud. We find that the neutron displacement rate is much higher than the gamma displacement rate, but that the freely-migrating defects produced by gamma irradiation are significant compared to those produced by neutrons. We evaluate the influence of gamma and neutron irradiation on hardening using a point defect clustering model. We find that the influence of gamma irradiation on radiation hardening in the core shroud is small compared to that for neutron irradiation.

Keywords: Gamma displacement damage, freely-migrating defects, BWR core shroud, irradiation hardening.

Introduction

In recent years, extensive cracking has been observed in boiling water reactor (BWR) core shroud welds [1]. The azimuthal cracking pattern suggests a possible role of radiation damage in assisting cracking, and stress-corrosion cracking models in the literature have included a link between irradiation hardening and crack velocity [2]. To help assess the possible influence of radiation damage on core shroud cracking, we performed detailed displacement damage calculations considering both the effects of fast neutrons and gammas. These calculations discriminated the fraction of defects available for long-range migration (freely-migrating defect fraction or FMD), and the clustering fraction. The defect generation rates calculated were used in a hardening model based on the development of point defect clusters under irradiation, to assess the relative influences of gamma and neutron irradiation on the hardening process.

Displacement Damage Calculations

It has long been recognized that the dominant contribution to the generation of atomic displacements in reactor components comes from fast neutrons. In the regions near the reactor core, the number of displacements per atom (dpa) generated by a typical fast neutron flux exceeds the number of displacements generated by the gamma flux by approximately two orders of magnitude [3].

Two recent developments have prompted a reassessment of this picture. First, the accelerated embrittlement of the HFIR reactor pressure vessel steel discovered in the early 1990s [4-6] caused a re-evaluation of the role of other possible irradiation

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embrittlement mechanisms. Analysis showed that the special characteristics of the HFIR reactor (especially the large water gap) caused the gamma contribution to the overall dpa damage to be significant. When the total calculated number of dpa was corrected for the gamma contribution, the observed embrittlement was well predicted by existing embrittlement models [4]. Thus, in certain circumstances, the number of γ dpa can be comparable to or higher than the neutron dpa. Second, a series of experiments indicated that only a small fraction of defects survive the recombination and clustering during the first picoseconds after the displacement cascade forms [7, 8]. Only this small fraction of defects that survives the cascade as individual defects (called the freely-migrating defect fraction – FMD) can contribute to irradiation-induced processes that depend on long-range defect migration through the solid, such as irradiation creep, void swelling and radiation induced segregation [9, 10].

We calculated the neutron displacement damage rate G^{n}_{dpa} (dpa.s^{-1}) using the SPECTER code [11]. The code takes a given neutron flux and calculates the number of dpa, taking into account both the inelastic and elastic scattering due to fast neutrons, and thermal neutron reactions. SPECTER also calculates the recoil spectrum, which we use to calculate the FMD. The rate of gamma displacement damage in dpa/s is given by

$$G^{\gamma}_{dpa} = \sum_{j=1}^{20} \phi_j(E^j_\gamma)\cdot\sigma^{\gamma}_{d}(E^j_\gamma) \qquad (1)$$

where $\phi_j(E^j_\gamma)$ is the gamma flux for energy group j and where $\sigma_d(E^j_\gamma)$ is the total displacement cross section for gammas in the material. The displacement cross section $\sigma_d(E^j_\gamma)$ was evaluated for several materials in a previous reference [12]. Following Baumman [13], we used analytical formulas for the gamma-electron interaction by the three different mechanisms and used Oen's electron displacement cross sections [14] to evaluate the displacement rate. The calculated values of the displacement cross sections are in very good agreement with those calculated by Baumann [13] and in reasonable agreement with the values calculated by Alexander [15].

Using the approach described here we calculated the gamma induced displacement rates at the core shroud beltline weld. For the present calculations, we used a displacement energy of 24 eV, which is near the minimum displacement energy for Fe. This choice of displacement energy overestimates both the neutron and gamma displacement rates by about 60% relative to the value calculated using E_d=40 eV, (the recommended value averaging over all crystallographic directions) [ASTM Practice E521-96 for Neutron Radiation Damage Simulation by Charged-Particle Irradiation].

Freely-Migrating Defect Fraction (FMD) from Neutron Irradiation

Different types of irradiation produce different recoil spectra, which in turn change the cascade size, density, subcascade formation, all of which affect the final fraction of freely migrating defects or FMD. Thus, different recoil spectra result in different FMD. Researchers have determined the relative FMD from radiation-induced segregation (RIS) [16-18] or radiation enhanced diffusion (RED) experiments [19]. Other researchers have performed molecular dynamics (MD) calculations to assess the surviving defect fraction as a function of recoil energy [20-22]. The surviving defect fractions normally found in MD simulations are one order of magnitude higher than the FMD derived from the experiments above, possibly due to the short time scale of the MD simulations, and to complicated sink structure in the experiments. In this work we use the experimentally derived values of FMD. We note that by using the FMD values from experiments rather than from MD calculations, the total number of freely migrating defect generation rate from neutron irradiation G^n_{fmd} decreases, and the relative importance of the defects produced by gamma is magnified. Because of this, the present calculation should be considered an upper limit for the possible effects of gamma induced freely-migrating defects on the core shroud.

We use a function of the form $\mathrm{FMD} = 0.01 + 0.38 \cdot (\mathrm{E_r[keV]})^{-0.89}$, fit the experimental results where E_r is the recoil energy in keV, and where the constants were determined by fitting the experimental data [16-19]. The experimental results are given in terms of FMD versus median recoil energy, which we approximate in this work to the actual recoil energy, i.e., we take the FMD measured from experiments in which there was a recoil distribution, and approximate that distribution by its median value. This is a reasonable approximation for a smoothly varying function. The average neutron and gamma fluxes at the inner surface of core shroud are given in reference [23].

We obtained the neutron recoil spectra by running the SPECTER code for the neutron flux calculated at the inner surface of the core shroud. A typical spectrum, calculated for the average neutron flux in the shroud is shown in figure 1. The bumps in the distribution are an artifact of the energy group collapsing method.

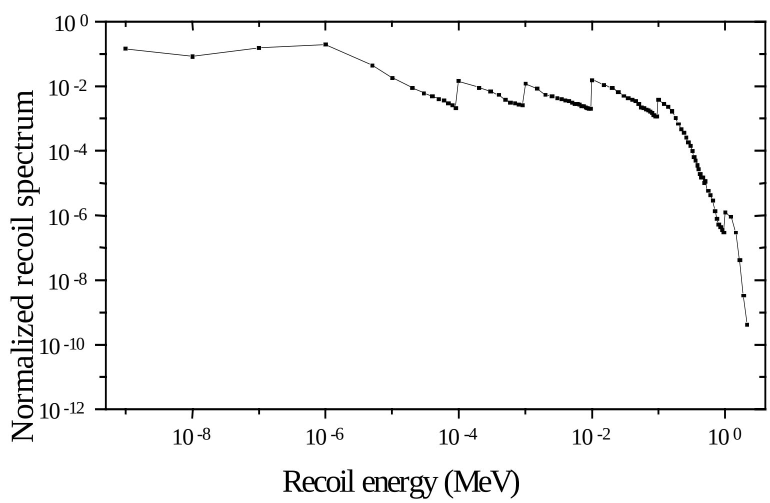

Figure 1: Recoil spectrum from SPECTER for the average shroud neutron flux.

We combined the recoil spectrum with the FMD equation to obtain the neutron FMD. Since the gamma induced median recoil energy is very low, we took the gamma FMD = 1, i.e. all gamma produced defects are freely-migrating. The results for the calculated dpa rate (G_{dpa}) and freely migrating defect production rate (G_{fmd}) from neutron and gamma at the inner surface of the core shroud are shown in table 1, for the highest and lowest azimuthal values of the neutron flux.

The number of displacements per atom produced by neutron irradiation and calculated by SPECTER are 60 to 250 times higher than the total displacements caused by gamma irradiation calculated using the cross sections described above. Thus, the ratio $G^{\gamma}_{dpa} \big/ G^{\gamma+n}_{dpa}$ is about 0.4 to 1.7 %; however, the ratio $G^{\gamma}_{fmd} \big/ G^{\gamma+n}_{fmd}$ ranges from 9.4 to 37.7 %. These results indicate that gamma rays contribute significantly to the total number of freely migrating defects but not to the total number of displacements [24].

Table 1: *Calculated neutron and gamma-induced displacement damage rates at the inner surface of core shroud weld H4 (beltline weld).*

	Highest Azimuthal Flux		Lowest Azimuthal Flux	
	G_{dpa} (s^{-1})	G_{fmd} (s^{-1})	G_{dpa} (s^{-1})	G_{fmd} (s^{-1})
neutron, *n*	3.33×10^{-9}	1.18×10^{-10}	1.91×10^{-10}	5.61×10^{-12}
gamma, γ	1.23×10^{-11}	1.23×10^{-11}	3.40×10^{-12}	3.40×10^{-12}
$G^{\gamma}_{fmd} / G^{\gamma+n}_{fmd}$ (%)	0.4	9.4	1.7	37.7

Radiation Hardening: Point Defect Clustering Model

We now evaluate the relative roles of the gamma and neutron fluxes on irradiation hardening, using a dislocation barrier model. When stainless steels are exposed to fast neutron irradiation below 10^{20} n.cm^{-2} at relatively low temperature (< 300^{o} C), small dislocation loops and defect clusters are created [25]. These clusters contribute to radiation hardening, by adding barriers to dislocation motion, and increasing the yield strength. To describe this process, we used the model developed by Stoller for microstructural evolution and hardening of ferritic steels under irradiation [26]. In this model, interstitial and vacancy loops evolve from defect clusters that nucleate from displacement cascades, or aggregate from individual defects. The defect clusters can then grow or shrink by absorbing one or the other type of point defect. The balances for interstitial and vacancy concentrations (C_i and C_v) can be expressed as:

$$\frac{dC_i}{dt} = P_i - R_{iv} C_i C_v - D_i \sum_j S_{ji} C_i \qquad (2)$$

$$\frac{dC_v}{dt} = P_v - R_{iv} C_i C_v - D_v \sum_j S_{jv} C_v \qquad (3)$$

where P_i and P_v are the interstitial and vacancy production rates, R_{iv} is the recombination constant, the D_i and D_v are the diffusion coefficients for each point defect, and the S_{jx} are the sink strengths for the absorption of point defect x by sink j (j= network dislocations, grain boundaries, and irradiation-induced point defect clusters). We modified Stoller's model such that the interstitial production rate, P_i in Eq.2, is given by:

$$P_i = G^n_{fmd} + G^{\gamma}_{fmd} + \sum_{m=2}^{4} E^m_i C_m \qquad (4)$$

where the E^j_i are the rate constants for interstitial emission from a m-interstitial cluster and C_m is the concentration of a m-interstitial cluster. Following Stoller, we write balances of higher order defect clusters as:

$$\frac{dC_m}{dt} = \eta G^n_{dpa} \frac{f^m_{icl}}{m} + \beta^{m-1}_i \frac{C_m}{m} + \left(\beta^{m+1}_v + E^{m+1}_i\right) C_{m+1} - \left(\beta^m_v + \beta^m_i + E^m_i\right) C_m$$

for m=2,3,4 and where η is the cascade efficiency (fraction of displacements that avoid recombination within the cascade.)

$$\frac{dC_m}{dt} = \beta_i^{m-1} C_{m-1} + \beta_v^{m+1} C_{m+1} - \left(\beta_v^m + \beta_i^m\right) C_m \qquad \text{for } 5 \le m \le 500 \qquad (5)$$

where the β_j^m are the probabilities for an m-size cluster to absorb a defect j. The model includes point defect cluster balances for interstitial clusters up to 500 atoms, and only interstitial clusters contribute to hardening. Vacancy clusters are described by a simple creation and decay model. At low temperature (< 300 ^oC) and irradiation dose, it is unlikely that vacancy clusters grow and become voids. The vacancy clusters are assumed to form in the displacement cascade as microvoids with a constant radius and to change in size depending on the balance of the relevant point defect fluxes.

Table 2. Parameters used in the calculation

Irradiation temperature (T)	560 K	Lattice constant (a_L)	3.68x10^{-10}m
Vacancy migration energy (E_v^m)	1.4 eV	Vacancy clustering fraction f_{vcl})	0.3
Interstitial cluster binding energy ($E_2^B : E_3^B : E_4^B$)	0.75 : 1.0 : 1.25 eV	Interstitial clustering fraction ($f_{icl}^2 : f_{icl}^3 : f_{icl}^4$)	0.15 : 0.1 : 0.05
Effective grain diameter (d_g)	10^{-6}m	Vacancy formation energy (E_v^f)	1.5 eV
Interstitial migration energy (E_i^m)	0.65 eV	Dislocation recombination radius	1.8 x 10^{-9} m
Interstitial pre-exponential factor	7x10^{-6} $m^2.s^{-1}$	Burgers vector (b)	2.07 x 10^{-10} m
Vacancy pre-exponential factor	7x10^{-5} $m^2.s^{-1}$	Shear modulus (μ)	7.6 x 10^4 MPa
Dislocation interstitial bias (z_i^{dis}) and vacancy bias z_v^{dis})	1.25 and 1	Dislocation density (ρ_{dis})	5 x 10^{14} m^{-2}

The parameters used in the calculation of point defect cluster balances (shown in table 2) were modified from those in reference [26], to values relevant to stainless steel [27-31]. The amount of hardening due to PDC was calculated using a dislocation barrier model, in which, the increase in the yield strength caused by PDC is determined by the average distance (λ) between PDC ($\lambda = 1/(Nd)^{1/2}$), where N is the PDC concentration and d is the diameter of the PDC. The change in the yield strength was calculated by $\Delta\sigma_{YS}$=Mμb/$\beta\lambda$, where M is the Taylor factor used to adjust the shear stress to a change of uniaxial stress, μ is the shear modulus, b is the Burgers vector, and β represents the strength of the barrier [32]. We used an average damage level of $G_{dpa}^n = 1.23$ x 10^{-9} dpa.s^{-1} and $G_{dpa}^\gamma = 7.73$ x 10^{-12} dpa.s^{-1}. For a given fluence the yield stress increased by 15% as for the high displacement rate compared to the average displacement rate in table 1. Using the equation above we calculated the increase in yield strength, and fit the results to experimental data in 304 stainless steels irradiated and tested at 288 ^oC, from Refs. [33-36]. There is considerable variation in the values used in the literature for interstitial migration energy [26-29], so we used this as our only fitting parameter. The best fit was obtained with E_m^i equal to 0.65 eV (fig.2 a). There is considerable scatter in the data, so the fit can only be considered an approximate one, but it should give the correct order of magnitude of variation of the irradiation microstructure.

Using the above value of the interstitial migration energy, we calculated the increase in yield strength as a function of irradiation time for the cases where neutron irradiation alone causes damage, and for the case where both neutron and gamma damage are considered. Fig. 2.b shows that the yield stress changes for neutron displacements only is approximately 2-5% lower than when both gamma and neutron displacements are considered. This result was achieved while using the upper limit of the relative contribution of gammas to the production of freely migrating defects, indicating that gamma produced defects do not play a crucial role in hardening and that it is a good approximation to neglect the gamma contribution to hardening. The reason for this result is likely that the cascade production of defect clusters is the crucial step in achieving the observed hardening under in-reactor irradiation.

Because of this the specific effect of gamma displacements and/or neutron and gamma freely-migrating defects on various irradiation induced phenomena will depend on the operating mechanism and on the irradiation conditions (temperature, dose, dose rate, g/n flux ratio, etc.).We note that the rate of irradiation induced processes that are proportional to the accumulated flux of defects at sinks, will likely increase in direct proportion to the number of freely migrating defects available. This means that freely migrating defects could influence directly processes which affect crack propagation velocity, such as creep relaxation and sensitization by irradiation-induced segregation.

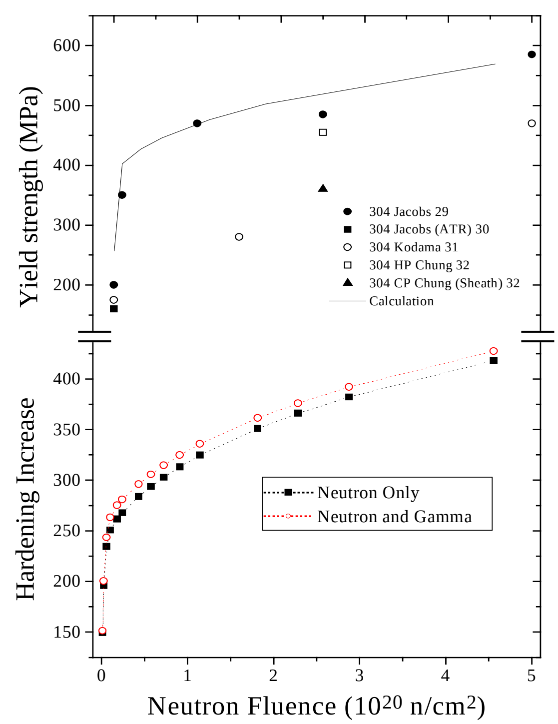

Fig. 2. (a) Yield stress increase fit to experimental data and (b) Calculated change in yield stress as a function of fluence for neutron and neutron + gamma irradiation.

It is interesting to compare the present results to those obtained during irradiation of pressure vessel steels in HFIR [4], and in which the gamma contribution was explained on a straight cumulative dpa basis, as noted above. The operating mechanism (hardening by irradiation induced obstacles) is similar, but the irradiation conditions are very different, as illustrated in table 3 below.

In the hardening mechanism used here two different processes combine to produce hardening: (i) heterogeneous nucleation of defect clusters in cascades and (ii) growth of these clusters by absorption of defects and smaller size mobile clusters. In cases where displacement cascades are few, homogeneous nucleation of clusters by single defect aggregation also becomes a factor. Clearly cascade-producing irradiation, (such as fast neutrons) affects both processes (i) and (ii) while irradiations that have lower average recoil energy such as gamma affect only cluster growth, by affecting the defect fluxes. Both the size of the obstacles and their number density affect the hardening. The HFIR irradiations were conducted at much lower temperature, under a fast neutron flux that was 10^{-5} smaller, and a gamma flux that was 10 times higher, to a much lower total dose. Under these circumstances, it is reasonable that the relative importance of cascade-induced and homogeneous nucleation of defect clusters shifts towards the latter in HFIR. For homogeneous nucleation of clusters, the total number of dpa is the relevant quantity, which agrees with the HFIR observations.

Table 3: Irradiation Conditions for HFIR and the present case

	HFIR PV	Core Shroud	Ratio
Neutron flux (n.cm^{-2}s^{-1})	10^{8}	10^{13}	10^{-5}
Gamma Flux (γ.cm^{-2}s^{-1})	10^{13}	10^{12}	10
Overall dose (n.cm^{-2})	10^{18}	10^{20}	10^{-2}
Temperature (K)	323	573	0.6

Conclusions

We have performed an assessment of the relative roles of neutron and gamma irradiation on hardening. The main conclusions are as follows:

1. Gamma induced displacements in the core shroud are much less numerous when compared on a straight dpa basis, to those produced by fast neutrons.

2. The upper limit of the ratio of freely migrating defect generation from gamma and neutron irradiation, ($G^{\gamma}_{fmd} \big/ G^{n}_{fmd}$) in the core shroud is between 10 and 40%, indicating that in situations where the magnitude of defect fluxes are important, gammas could have a significant effect on microstructure evolution.

3. The radiation hardening caused by neutron damage alone is similar in magnitude to that caused by a combination of neutron and gamma damage, indicating that gamma displacements have a smaller effect on hardening than fast neutrons.

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