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THE ROLE OF GAMMA RAYS AND FREELY-MIGRATING DEFECTS IN REACTOR PRESSURE VESSEL EMBRITTLEMENT*

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The Role of Gamma Rays and Freely-Migrating Defects in Reactor Pressure Vessel Embrittlement*

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ABSTRACT

Gamma ray effects are often neglected when evaluating reactor pressure vessel (RPV) embrittlement. However, recent analyses indicate that in newer style light water reactors, gamma damage can be a substantial fraction of the total displacement damage experienced by the (RPV); ignoring this damage will lead to errors in embrittlement predictions. Furthermore, gamma rays may be more efficient than fast neutrons at producing freely-migrating defects and as such can impact certain embrittlement mechanisms more effectively than fast neutrons. Consideration of these gamma effects are therefore essential for a more complete understanding of radiation embrittlement.

1. Introduction

The desire to more accurately predict nuclear power plant lifetimes has driven the need for a more fundamental understanding of radiation embrittlement of reactor pressure vessel (RPV) steels–a key lifetime-limiting degradation phenomenon in light water reactors (LWRs). Previous work reported in this symposium series[1] has contributed important understanding in this area. This understanding includes the more accurate characterization (modeling and measurement) of radiation flux-energy spectra in reactor environments, as well as an improved correlation with embrittlement of the damage produced by these radiation fluxes.

Traditionally the damage correlation effort has focused on the effects of fast (e.g. E≥1 MeV) neutrons. Empirical correlations have been established which predict embrittlement as a function of an exposure variable, principally fast neutron-induced displacements per atom (n-dpa), i.e.,

$$\text{embrittlement} = f(\sigma_{\text{DAM,n}}\ \Phi_n). \tag{1}$$

Here "embrittlement" is a measurable change in a particular mechanical property of interest such as nil-ductility transition temperature, tensile yield strength, hardness, fracture toughness, etc. Neutron dpa is the product of the damage cross section, $\sigma_{\text{DAM,n}}$, and the

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neutron fluence, Φ_n. This type of correlation is reflected in various regulatory guides of embrittlement.[2]

It has long been recognized that in addition to neutrons, other types of radiation present in a reactor environment, such as gamma rays, produce displacement damage in a RPV.[3] Gamma rays are known to generate atomic displacements in medium-Z metals indirectly, through the production of energetic electrons and positrons primarily via Compton scattering and pair production interactions. The damage cross sections for these processes however are typically 2-3 orders of magnitude smaller than that for fast neutrons over the 1-15 MeV gamma/neutron energy range of interest in reactors. This large disparity in cross-sections led to the presumption that damage from n-dpa ($\sigma_{DAM,n}\ \Phi_n$) overwhelmed any gamma damage contribution (γ-dpa $= \sigma_{DAM,\gamma}\ \Phi_\gamma$), particularly for those reactors in which it was believed that embrittlement was of most immediate concern, i.e. pressurized water reactors (PWRs) with small diameter RPVs where the fast neutron flux and high energy (E>1 MeV) gamma fluxes in the vessel were of similar magnitude. Experimental support for this reasoning was provided by Gold and co-workers[4] whose evaluation of gamma spectra in the Poolside Critical Assembly (PCA) facility determined that the fraction of γ-dpa to the total dpa (neutron- plus gamma-induced) was about one-half a percent.

While this percentage is indeed small, it was recognized by Gold et al. that the PCA experiments were only relevant to a subset of reactors, i.e. PWRs with small water gaps. A later analysis by Baumann[5], for example, indicated that as much as 10% of the total damage in the Savannah River Site, heavy water moderated reactor, is from gamma rays. Indeed there are other LWRs (e.g. newer model boiling water reactors (BWRs)) in which the presence of a considerably larger water gap separating the core from the vessel can give rise to a significantly greater contribution of gamma rays to the total displacement damage in the RPV.

The purpose of this paper is to discuss gamma damage effects and their significance to radiation-induced embrittlement. Inclusion of gamma effects is necessary, not only to account for all the sources of damage in an RPV, but more importantly because the γ-dpa may correlate with embrittlement in a significantly different manner than n-dpa.

2. Gamma ray damage in RPV's

Unlike PWRs, the γ-dpa in LWRs with large water gaps may constitute a substantial fraction of the total dpa experienced by the pressure vessel. Ignoring, this component will lead to errors in estimating damage exposure, and concomitant errors in predicting embrittlement when applying standard empirical correlations. The significance of γ-dpa is demonstrated by recent analyses of computer transport calculation results for RPV neutron and gamma flux spectra in various LWRs.[6,7] Fig. 1 shows the fraction of total dpa rate produced by gamma rays at the 1/4-T location of the RPV as a function of water gap thickness calculated for various reactors. The solid line is a simple model fit assuming exponentially decaying gamma and neutron fluxes emanating from the reactor core. Also shown in the figure are the total damage rates (n-dpa rate plus γ-dpa rate) at the same 1/4-T location as a function of water gap. Clearly, the overall damage rate and, likewise the embrittlement rate, decrease with increasing water gap. Importantly however, the contribution of damage from gamma rays increases substantially with increasing water gap. In the General Electric Advanced Boiling Water Reactor (GE ABWR), for example, over one-third of the total dpa rate (for gamma and neutron energies $\geq$1 MeV) experienced by the vessel will be contributed by gamma rays. This gamma component of dpa damage is presently overlooked in regulatory guides and models of radiation embrittlement.

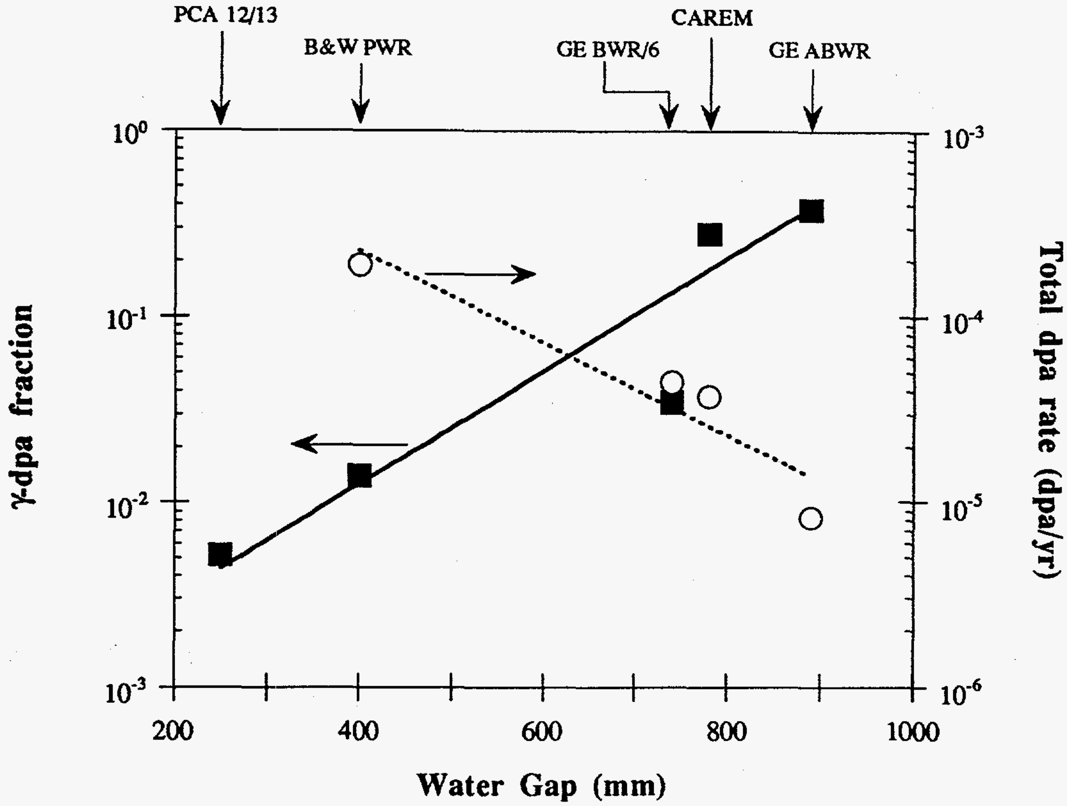

Fig. 1. Variation in the gamma fraction of total dpa rate and the total (gamma plus neutron) dpa rate as a function of the water gap intervening between the core and RPV calculated for various LWR configurations. [6-8] Calculations performed for the 1/4-T location of each RPV.

The reason for the increasing contribution of gamma damage with water gap is physically straight forward: water moderates the energies of fast neutrons, hence attenuates the fast neutron flux, much more readily than it attenuates the high energy gamma flux. A larger water gap will result essentially in an exponentially increasing ratio of gamma/fast neutron flux incident on the RPV, in turn increasing the γ-dpa ($\gamma\text{-dpa} = \sigma_{\mathrm{DAM},\gamma}\,\Phi_\gamma$).

Aside from an accurate accounting of all dpa sources in a RPV, an even more important reason to study gamma ray effects lies in the recognition that not all dpa are necessarily equivalent in terms of their effectiveness for inducing various microstructural changes contributing to embrittlement. There is reason to believe, as outlined below, that gamma rays are more efficient than fast neutrons at producing freely-migrating defects (FMDs), i.e. defects which contribute to long range mass transport during irradiation. Therefore in environments where γ-dpa is significant, embrittling microstructural features which rely on FMDs might form more readily than those which evolve during fast neutron irradiation alone. Depending on the mechanism of interest, gamma rays may be more efficient, on a per dpa basis, at inducing embrittlement. Such a possibility has important implications, even in RPVs where the γ-dpa fraction is small.

3. Freely-migrating defects: their role in the evolution of embrittling microstructures

The simple engineering correlation represented by Eq. (1), while useful, is an empirical one, concealing the underlying mechanisms responsible for embrittlement during irradiation. Embrittlement predictions based on such correlations are subject to uncertainties that can be minimized with a more fundamental, mechanistic understanding of radiation embrittlement. It is in such an approach that recognition of the role of FMDs becomes important and the distinctions between gammas and neutron effects are clarified.

At the elevated temperature of interest to commercial LWR pressure vessels (288°C) it is generally recognized that two types of radiation-induced microstructural features contribute to embrittlement in steels containing Cu impurity: matrix damage and Cu precipitates.[9] The matrix damage component consists of defect clusters formed either directly in displacement cascades or which result from the interaction of migrating defects. The embrittling contribution of Cu results in part from radiation-enhanced diffusion- (RED) induced precipitation and growth of Cu clusters. The evolution of both these microstructural features does not depend exclusively on the total number of displaced atoms generated (i.e. dpa). Rather, an important exposure parameter is the fraction of the total displacements which become FMDs and contribute to long-range mass transport.

Fundamental experiments have demonstrated the inadequacy of dpa for correlating microstructural changes induced by long-range mass transport phenomena such as RED[10] or radiation-induced segregation (RIS).[11-13] An example of this is provided by the recent RIS results[14] shown in Fig. 2. The segregation rate, i.e. the amount of surface segregation induced per dpa (the slope of the lines in Fig. 2), is considerably larger for the light-ion, He irradiation than for the heavier ion, Cu irradiation. In light of such results, a more appropriate exposure parameter for correlating microstructural change in these cases is $\alpha_i \sigma_{DAM,i} \Phi_i$ where α_i represents the FMD production efficiency fraction for an

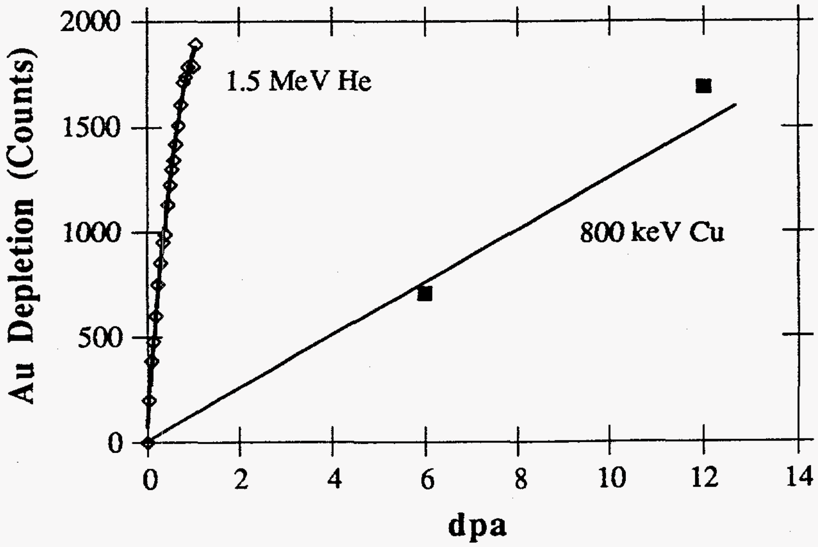

Fig. 2. Radiation-induced segregation, measured by the depletion of Au atoms from the sample surface, caused by irradiation with light and heavy ions in a Cu-1 at.% Au alloy.[14]

irradiating particle, i. It is clear from the results shown in Fig. 2 that light ions are more efficient at producing FMDs than heavier ions, i.e. $\alpha_{He} >> \alpha_{Cu}$.

Experiments[10-13] examining RIS and RED have been used to quantify FMD production efficiencies, α_i, for different irradiation environments. These studies have demonstrated a strong correlation between production efficiency and the median recoil damage energy, $T_{1/2}$, which is a parameter used to characterize the recoil energy dependence of defect production for an irradiation environment. Fig. 3 displays the primary recoil energy dependence of defect production for Cu and He irradiations pertinent to the RIS results of Fig. 2. Also known as integral recoil damage spectra, W(T), these dependencies give the fraction of displacement damage produced by primary recoils with energies less than or equal to T. The median recoil damage energy, $T_{1/2}$, is simply the recoil energy at which W(T)=0.5, i.e., one-half of the displacement damage is produced by recoils with energies above, and below, $T_{1/2}$. Previous RIS experimental results[11-13] shown in Fig. 4 demonstrate a dramatic decrease in the relative efficiency for FMD production with increasing $T_{1/2}$ values. With a small $T_{1/2}$ value for He (2.0×10^3 eV) and a large $T_{1/2}$ value for Cu (6.0×10^4 eV), the RIS rate differences observed in Fig. 2 are consistent with this trend.

The decreasing efficiency with increasing $T_{1/2}$ is rationalized by examining differences in defect production on an atomistic level. For irradiations with large values of $T_{1/2}$, defects are typically produced in dense displacement cascades generated by high energy recoil atoms. The high spatial correlation of defects within the cascade results in substantial recombination and cluster formation, leaving only a small fraction of the total defects produced available for subsequent migration. In contrast, for low $T_{1/2}$ irradiations typically only a few, isolated defects are generated. These defects have a much higher probability to escape annihilation through recombination and clustering and thus become FMDs.

With this background, the potential different embrittling effects of gamma rays and fast neutrons can be understood. Fig. 3 shows the W(T) spectrum recently calculated for an energy spectrum of gamma rays present in a RPV.[15] Also shown is the W(T) curve for fast neutrons at an in-core location of the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory as previously calculated using the SPECTER code.[16] W(T) curves for fast-neutrons in an RPV were not found in the literature so the in-core spectrum was used here for comparison purposes. A somewhat softer spectrum (smaller $T_{1/2}$) is to be expected in the RPV. As seen in Fig. 3, the $T_{1/2}$ value for the gamma ray spectrum (70 eV) is well over 2 orders of magnitude smaller than that observed for the fast neutron spectrum (4.0×10^4 eV). This difference is even larger than that observed between He and Cu RIS irradiations described above. The fast neutrons, with a large $T_{1/2}$, are similar to the Cu ion irradiation and their location on the curve in Fig. 4 indicates a low FMD production efficiency. Gammas, with a very small $T_{1/2}$, will favor the formation of isolated defects and hence have a much higher FMD production efficiency compared to fast neutrons. Hence, when weighted by this production efficiency, the contribution of gammas to embrittlement may be greater than that presumed using dpa as the standard measure of exposure.

Questions remain regarding the validity of extrapolating the results of fundamental experiments performed on high-purity metals and alloys to the real steels of interest to RPVs. Furthermore, mechanisms relying on mass transport (e.g. RED) contribute only partially to radiation embrittlement and hence an exact measure of the effect of enhanced FMD production is at present difficult to assess. In order to address these uncertainties, experiments[17] have been undertaken using high-energy electron irradiations to reproduce, under highly controlled laboratory conditions, the damage generated by the electrons and positrons initiated by gamma interactions. It is anticipated that these studies will provide a better understanding of the correlation of gamma damage to embrittlement.

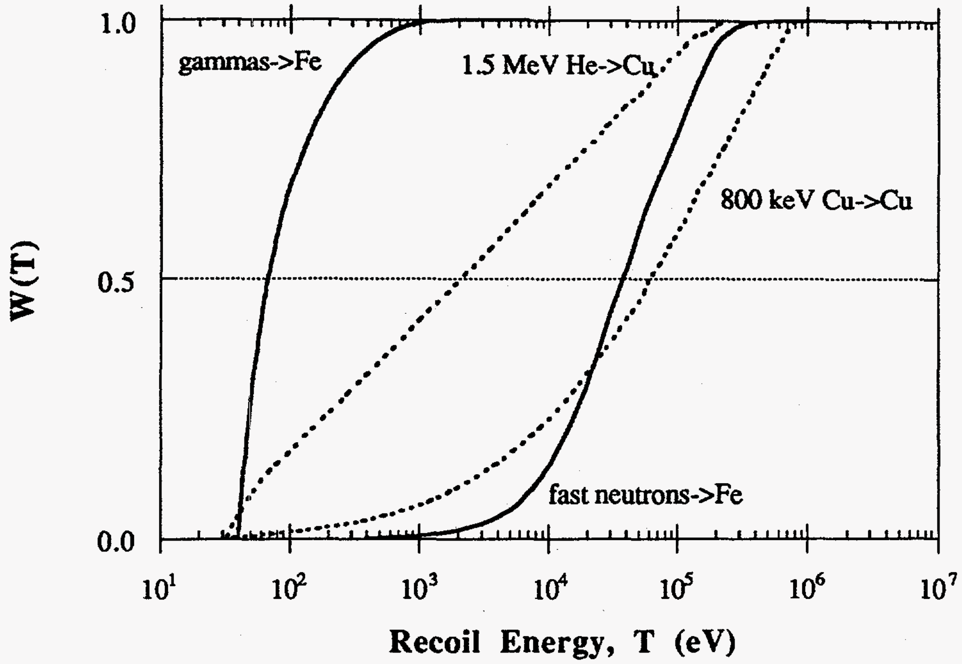

Fig. 3. Integral recoil damage spectra for various irradiation environments discussed in text.

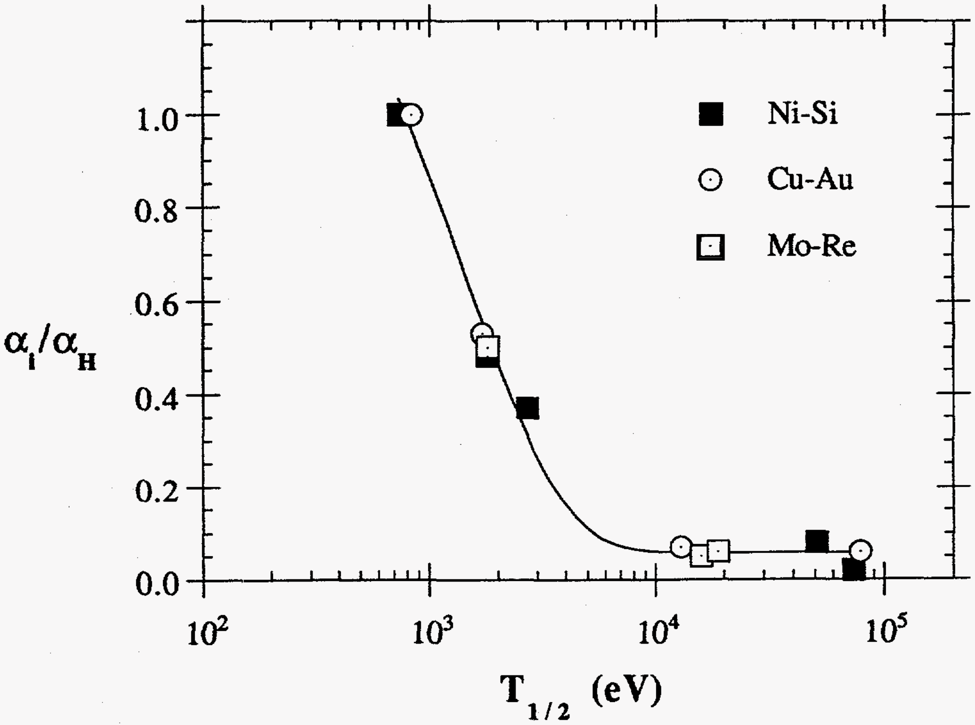

Fig. 4. Relative efficiencies (relative to proton irradiation) for FMD production measured in RIS experiments on alloys as a function of median recoil damage energy.

4. Summary and Conclusions

The importance of considering gamma ray damage effects for a more complete understanding of radiation embrittlement in light water RPVs has been discussed. Analyses of transport calculations are consistent with simple physical arguments that in certain reactors, the γ-dpa rate can constitute a substantial fraction of the total dpa rate experienced by the RPV. Excluding this component will lead to errors in estimating embrittlement. More importantly however, even in those reactors where the γ-dpa rate is minor, their expected greater efficiency for producing FMDs may significantly enhance the gamma contribution to certain embrittlement mechanisms such as RED-induced Cu precipitation. Experimentally identifying these FMD effects and evaluating their consequences are key to understanding how embrittlement correlates with damage produced in various RPV radiation environments.

While this paper has focused on the damage correlation perspective, as pointed out by Gold elsewhere in this proceedings[18], additional, more accurate RPV gamma flux dosimetry is required. For example, the gamma dpa analyses mentioned above[6,7] relied on results from transport calculations that were optimized to minimize uncertainties in fast neutron flux, without particular regard to gamma flux. Improved modeling as well as experimental efforts to characterize gamma ray spectra are therefore essential to better assess the gamma damage contribution to embrittlement.

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