Correlating Fast Fluence to dpa in Atypical Locations

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Abstract. Damage to a nuclear reactor’s materials by high-energy neutrons causes changes in the ductility and fracture toughness of the materials. The reactor vessel and its associated piping’s ability to withstand stress without brittle fracture are paramount to safety. Theoretically, the material damage is directly related to the displacements per atom (dpa) via the residual defects from induced displacements. However, in practice, the material damage is based on a correlation to the high-energy (E > 1.0 MeV) neutron fluence. While the correlated approach is applicable when the material in question has experienced the same neutron spectrum as test specimens which were the basis of the correlation, this approach is not generically acceptable. Using Monte Carlo and discrete ordinates transport codes, the energy dependent neutron flux is determined throughout the reactor structures and the reactor vessel. Results from the models provide the dpa response in addition to the high-energy neutron flux. Ratios of dpa to fast fluence are calculated throughout the models. The comparisons show a constant ratio in the areas of historical concern and thus the validity of the correlated approach to these areas. In regions above and below the fuel however, the flux spectrum has changed significantly. The correlated relationship of material damage to fluence is not valid in these regions without adjustment. An adjustment mechanism is proposed.

1. Background

General Design Criterion 31, “Fracture Prevention of Reactor Coolant Pressure Boundary,” of 10 CRF Part 50 [1] requires that the reactor coolant boundary of a Light Water Reactor (LWR) behaves in a nonbrittle manner. Physically, this means that in an overcooling or overpressure condition the components of the pressure boundary must be able to absorb energy without fracture. Analytically, this means that the neutron embrittlement of pressure boundary components needs to be monitored. Historically, this monitoring is in the form of calculating the E > 1.0 MeV neutron fluence to the pressure vessel in an axial region approximately the same height as the active fuel stack, and correlating it to changes in material properties. While relatively advanced methods exist for the tracking of fast neutron fluence (n(E > 1.0 MeV)/cm²), it is not only fast neutron fluence that causes damage leading to embrittlement.

Microscopically, the damage from irradiation manifests in a few different ways. First, neutron irradiation can produce helium atoms through (n,alpha) reactions. At high temperature, these helium atoms will tend to migrate together and towards grain boundaries creating voids and weakness. A second...
The type of microscopic effect is displacements of atoms within steel. Here, atoms are displaced from one lattice site to either an interstitial site or a vacant lattice site. If displaced to a different site, there is no short term change in material properties. In the long term atoms exchanging sites can cause void swelling and creep. Atoms displaced into interstitial sites produce changes in hardness leading to a material with a greater stress at a significantly lower strain. Furthermore, the failure in these harder materials is brittle compared to the same material with fewer displacements.

Thus, embrittlement is potentially better correlated to displacements per atom (dpa) than fast fluence. It is atoms displaced from their crystal lattice leading to precipitates and point defect tangles that ultimately change the material properties of metals. If dpa were perfectly correlated to fast neutron fluence, then material property changes would also be perfectly correlated to fast neutron fluence and there would be no problem. However, as seen in Fig. 1, the response function for dpa in steel is not a simple step function at 1.0 MeV. This response shows two distinct regions. First from ~1 keV and up, the response represents the fact that while the cross section for Fe-56 is relatively flat, averaging out resonances, the energy in the neutron-induced recoil is enough to displace additional atoms. The scattered points between 10 keV and 100 keV are the groupwise representation of one large resonance, the so-called “iron window” in Fe-56 with minimum at 24.6 keV and maximum at 28 keV. Below about 1 keV, the dpa response increases due to both iron recoil and subsequent gamma induced displacements as the (n, gamma) reaction cross section of iron increases.

Thus, to be able to correlate fast neutron fluence to dpa and then changes in material properties in general, one needs to account for the neutron energy spectrum.

Historically, accounting for energy spectrum has not been necessary. The current simple correlations found in Regulatory Guide 1.99, Revision 2 are acceptable in the regions currently applied. However, as reactors are licensed for 60 or even 80 years monitoring of the areas outside of the traditional area of concern is becoming important. In these regions, the implicit assumption of no spectral variation in the current correlations may not be valid.

The purpose of this study is to confirm the applicability of current correlations for current uses and propose changes to correlations in regions where necessary.
2. Correlating Fast Fluence to Material Property Changes

2.1 Regulatory Requirements

10CFR50 Appendix B defines the Reactor Vessel Beltline as “the region of the reactor vessel (shell material including welds, heat affected zones and plates or forgings) that directly surrounds the effective height of the active core and adjacent regions of the reactor vessel that are predicted to experience sufficient neutron radiation damage to be considered in the selection of the most limiting material with regard to radiation damage.”

This definition means that the physical beltline expands with operation, growing as adjacent regions experience sufficient damage.

2.2 Current Correlations

There are numerous methods for correlating fluence and damage. Some correlate fast fluence to a physical quantity such as change in nil ductility transition, the temperature below which steel behavior is brittle, while others provide a simplistic zero order, with respect to energy, approximation.

2.2.1 US NRC Regulatory Guide 1.99, Revision 2

US NRC Regulatory Guide 1.99, Revision 2 [2] is the manner by which neutron fluence values are correlated to material embrittlement values. This guide was developed with the elevation of active fuel in mind. It does not give a correlation between fast neutron fluence and dpa but does imply one exists by stating a correlation between fast neutron fluence and change in reference temperature for nil ductility transition. It does address the concept of spectral dependence in relation to capsule to vessel differences by stating, “The differences in energy spectra at the surveillance capsule and the vessel inner surface locations do not appear to be great enough to warrant the use of a damage function such as displacements per atom (dpa)”.

Surveillance capsules contain material specimens from the manufacture of the vessel and its welds. These capsules are located inside the vessel such that they receive greater neutron damage for the same operating time. The specimens are periodically removed and tested for properties such as nil ductility transition. It is from these tests that the following equation was developed.

Equation (2) from Regulatory Guide 1.99 states that the change in reference temperature for nil ductility transition \((\Delta RT_{NDT})\) is:

\[
(\text{CF}) f^{0.28-0.1\log(f)}
\]  

where CF is a chemistry factor (not important here) and \(f\) is in terms of \(10^{19} \text{ E > 1.0 MeV neutrons/cm}^2\). Regulatory Guide 1.99 allows \(f\) to be calculated at any depth, of increasing radius through the vessel wall, with a dpa based attenuation (RG1.99 Eq. (3), attenuation coefficient = 0.24 in\(^{-1}\)) but does not suggest a similar adjustment for other locations.

This equation shows no dependence on spectrum whereas Fig. 1 shows that dpa and thus material property changes are dependent on spectrum. In all regions of similar spectrum, and thus similar ratio of dpa to \(\text{E > 1.0 MeV fluence as the material specimen data, this equation should be valid. However, in areas of different neutron spectrum this condition it is not necessarily true. The base data is all from material samples inside of the interior surface of reactor pressure vessels at active fuel elevation [3, 4].}

2.2.2 DPA Correlations

While the goal of this study is to evaluate and improve the procedures from Regulatory Guide 1.99 and 10CFR50 Appendix B, it is instructive to look at a previous correlation between fast neutron fluence and dpa.
3. DPA Correlations

3.1 Correlation with Heavy Reflector

An R0 DORT model of a B&W 177 assembly reactor was altered to approximate the presence of a heavy reflector region such as is in EPR™. The results show that, as the heavy reflector stops some of the E > 1.0 MeV flux from reaching the vessel, the correlation of dpa to E > 1.0 MeV fluence is greater with the heavy reflector. However, the effect is small.

3.2 Variation with Elevation

A 3D Monte Carlo model of a B&W 177 fuel assembly reactor was run to investigate the changes in the dpa to fast fluence correlation with elevation, particularly focusing on the exterior surface of the pressure vessel. At the active fuel elevation, neutron current is generally outward from the fuel. In regions axially above and below the fuel elevation, there is significant contribution from inward reflection from the cavity. From approximately 1 meter above the fuel and upwards, the flux is greater on the outside of the vessel than the inside.

At the inside of the vessel there is agreement with the NUREG-7027 correlation of 15 dpa per $10^{22}$ fast neutrons. Figure 3 shows that in regions far from the active fuel, the dpa to fast fluence correlation can be many times that of the traditional region of concern. This means that areas that had been thought of as having damage too low to be of concern on a cumulative fluence basis may have material damage far greater than expected and should be included in monitoring.
3.3 Variation with Depth

Figure 4 shows a significant change in dpa to fast neutron fluence ratio through the vessel. Figure 5, shows the attenuation of dpa and fast flux through vessel. The absolute values for dpa and fast fluence shown in Fig. 5 are not important. What is important are the attenuation coefficients seen in the fit functions. These show that dpa attenuates more slowly than fast fluence through vessel. For depth effects, Regulatory Guide 1.99, Revision 2 allows attenuation with a coefficient of $0.24 \text{ in}^{-1}$ which is similar to the coefficient of $0.221$ found here. As the fast neutron fluence falls with a coefficient of approximately $0.34$, its direct use would result in a nonconservative estimate of material property change at non-zero depth in the vessel. Thus, the radial effect of changes of dpa to fast fluence is being appropriately treated.

4. Impacts

Since equation 2 of Regulatory Guide 1.99, Revision 2 is an empirical correlation of material property change to fast fluence developed using samples taken from surveillance capsules near the center of the
fuel elevation near the inside surface vessel, it is non-conservative in regions that have a larger dpa to fast neutron fluence correlation than this location. This effect is already considered for depth from the inner surface by use of a dpa based attenuation equation rather than using fast fluence directly. However, this equation is no longer valid when significant flux is coming from reflection outside the vessel which has a different spectrum and is in a different direction than considered in the depth attenuation.
ASTM 1035-08 [6] states that PWR supports need to be evaluated for radiation damage at $1 \times 10^{17} \text{E} > 1.0 \text{MeV n/cm}^2$ or $3 \times 10^{-4} \text{dpa}$. Based on the current study, with a dpa to fast fluence ratio of 60 dpa per $10^{22} \text{E} > 1.0 \text{MeV}$ neutrons, the dpa screening limit will be reached for nozzle supported plants at approximately $5 \times 10^{16} \text{E} > 1.0 \text{MeV n/cm}^2$. If monitoring is done on a fast fluence rather than dpa basis, the dpa criteria will be overshot by approximately 200 percent.

It is proposed that, for areas outside of the active fuel height, a new quantity, the spectral adjusted equivalent $\text{E} > 1.0 \text{MeV}$ fluence, be reported and used with equation 2 of Regulatory Guide 1.99, Revision 2.

The multiplicative spectral adjustment on fast fluence should be:

\begin{equation}
\frac{\text{dpa}}{\text{FastFluence}}_{\text{point of interest}} \cdot \frac{\text{dpa}}{\text{FastFluence}}_{\text{beltline}}.
\end{equation}

For the case presented this could mean an adjusted fluence factor, $f$, of $(60/15) = 4$ for the nozzle region. This adjustment is similar to the one allowed for depth attenuation in Regulatory Guide 1.99, Revision 2. Alternatively, fluence could be calculated in terms of dpa for all regions and new correlations be made between dpa and material properties. This direct dpa method, however, would require new regulatory requirements be expressed in terms of dpa rather than the historical preference for $\text{E} > 1.0 \text{MeV}$ fluence.

5. Conclusions

The correlation in Regulatory Guide 1.99, Revision 2 comes from material testing correlated to fast fluence in the specific spectrum in the region of the pressure vessel near active fuel rather than to dpa. It is appropriate to use this guide as long as the material of interest is irradiated in a region with similar dpa to fast fluence correlation as the material specimens tested. This correlation is reasonably flat throughout the core elevation, from which the material specimens have come, both azimuthally and axially. Thus, this guidance is valid in the region where it has historically been applied.

In regions of analysis outside of historically typical applications, both in space and in terms of material changes, the regulatory guide equation may not be correct and may not be conservative. The assumption in reference 2 that, “[t]he differences in energy spectra at the surveillance capsule and the [locations of interest] do not appear to be great enough to warrant the use of a damage function such as displacements per atom (dpa)” is not valid when the locations of interest are beyond the range of fuel elevation inside vessel surface. In locations and cases of greater dpa to fast fluence correlation, the spectral effects may need to be taken into account.

References

[4] EPRI NP-3319, “Physically Based Regression Correlations of Embrittlement Data from Pressure Vessel Surveillance Programs”