Neutron Flux Effects on Embrittlement in Reactor Pressure Vessel Steels

R. E. Stoller
Metals and Ceramics Division
Oak Ridge National Laboratory
Oak Ridge, TN 37831-6151 USA

International Workshop
Influence of Atomic Displacement Rate on Radiation-induced Aging of Power Reactor Components: Experimental And Modeling
3-7 October 2005
Ulyanovsk State University
Ulyanovsk, Russia
INTRODUCTION

• issue of displacement rate effects on radiation-induced embrittlement of reactor pressure vessel steels remains unresolved
  - witness the differences between recent alternate forms of RPV embrittlement correlation, ASTM E10 and NRC
• issue is relevant to:
  - difference between RPV surveillance positions and test reactor irradiations
  - BWR vs PWR vessels
  - damage attenuation through the RPV, simultaneous spectrum changes
• present a short summary of model-based predictions of flux or displacement rate effects on radiation-induced hardening (yield strength changes)
• illustrate the range of possible/plausible effects in both low and high copper steels
• briefly describe model, key parameters, relevant mechanisms, and model predictions
MODEL SUMMARY

A model using the kinetic rate theory has been under development to describe the time-dependent evolution of point defects, point defect clusters, and copper precipitates (Stoller in ASTM STPs 1175, 1270, and 1325)

- Interstitial cluster formation can occur in the model by essentially classical nucleation (random collisions between single interstitials) or directly in the displacement cascade

- Vacancy clusters were treated as forming at a single size due to cascade collapse

- The results of molecular dynamics simulation studies were used to provide guidance for in-cascade clustering fractions and defect survival fractions.

- Copper precipitate model follows general approach of Odette et al. and Fisher, et al.

  - precipitate nucleation not modeled, \( N_{ppt} = f(Cu, T) \) and \( r_{ppt}(0) = 0.25 \text{ nm} \)
MODEL SUMMARY, cont.

- precipitate growth assuming diffusion limited kinetics

\[
\frac{dr_{ppt}}{dt} = 4\pi r_{ppt} D_{Cu}' \left( C_{u}^0 - f_{Cu} \right)
\]

- where: \( C_{u}^0 \) is the initially available copper in the matrix and \( f_{Cu} \) is the amount of copper in CRP formed under irradiation

- \( D_{Cu}' \) is the radiation-enhanced copper diffusion coefficient, \( = D_{Cu} \left( C_v/C_v^e \right) \)

- Strength change due to point defect clusters and copper precipitates

- A simple dislocation barrier hardening model is used to calculate the shear stress increment required to cut through interstitial and vacancy type PDCs

- Based on recent TEM/yield strength correlations (e.g. Kojima, et. al, 1991), both types of PDCs are treated as relatively weak barriers, barrier strength = 0.25

- Russell-Brown model (1972) used to compute hardening due to copper precipitates, based on modulus difference between precipitate and matrix
MODEL SUMMARY, cont.

- Root-sum-square combination of individual contributions used to obtain total hardening:

\[ \Delta \tau_{total} = \left( \Delta \tau_{icl}^2 + \Delta \tau_{vcl}^2 + \Delta \tau_{ppt}^2 \right)^{0.5} \]

- Shear strength increments can be converted to corresponding changes in the uniaxial yield strength using the Taylor factor \((\Delta \sigma_y = 3.06 \Delta \tau)\), and yield strength changes can be related to Charpy shifts using correlations from the literature (e.g. Odette, et al; Williams, et al.) \(\Delta T_{41} \approx [0.5-0.65] \Delta \sigma_y\)

- Model has been applied to evaluate other material and irradiation variables, e.g. the influence of interstitial cluster mobility as seen in molecular dynamics simulations of displacement cascades
TYPICAL MODEL PREDICTIONS

- with original baseline model and parameters, obtain reasonably good agreement with surveillance data from PR-EDB, e.g. copper and fluence dependence
  - considerable data scatter, but mean behavior, e.g. copper dependence of data is similar to data
  - copper dependence stronger in model since all copper is available for precipitation
- rather abrupt transition due to simple copper precipitate model, i.e. no nucleation component
BASIC MECHANISMS RESPONSIBLE FOR A DISPLACEMENT RATE EFFECT

Radiation-induced property changes are driven by the excess point defects fluxes created by displacive irradiation.

- Details of microstructural evolution and effects such as solute segregation are determined by the transport and fate of mobile point defects and solutes.
- Displacement rate effects in RPV steels arise primarily from two processes:
  - the competition between formation and dissolution of unstable defects (this component is nearly inseparable from the effects of irradiation temperature)
  - the influence of the displacement rate on radiation-enhanced diffusion
- Hardening from point defect clusters is most strongly influenced by the first process, and that from copper precipitates by the second.
- However, when the unstable defects provide a significant sink for mobile point defects, they will have an impact on radiation-enhanced diffusion.
- Thus, an increase in displacement rate may lead to either an increase or decrease in hardening.
Effect of Displacement Rate on Vacancy Supersaturation and Matrix Recombination

![Graph](image)

- **300°C, 0.1 dpa**
  - **No i,v clustering**
  - **With i,v clustering**

(a) Vacancy supersaturation ratio ($C_v/C_{v^e}$) vs. Displacement rate (dpa/s)

(b) Recombination to sink-absorption ratio vs. Displacement rate (dpa/s)
EXAMPLES OF MODEL PREDICTIONS

Effect of displacement rate on fluence dependence of hardening (base-case model parameters)

- In high-copper steels, primary effect of lower displacement rate is a reduced fluence to initiate hardening from copper-rich precipitates. Little effect on peak hardening.
- At low fluences, hardening is reduced at lower displacement rates. Can lead to a crossover at intermediate fluences.
- Note: for typical displacement cross section of 1500 barn, $1 \times 10^{19}$ n/cm$^2$ (E$>$1.0 MeV) = 0.015 dpa
EXAMPLES OF MODEL PREDICTIONS

Effect of displacement rate on dose required to obtain a given level of copper precipitation in high-Cu steel, “base-case” model parameters

- threshold dose reduced at lower displacement rates
- dose-independent region at lowest displacement rates
- transition to dose-rate independence is moved to lower displacement rates for higher fractional precipitation

*50 and 95% precipitation not achieved by 0.5 dpa at the highest displacement rates
EXAMPLES OF MODEL PREDICTIONS

Effect of displacement rate on fluence dependence of hardening in low-Cu steel, base-case parameters, interstitial clusters not mobile

- in low-copper steels, hardening is reduced at lower displacement rates
- effect of displacement rate stronger than observed in most low-copper data
Influence of interstitial cluster mobility on rate effect: predicted yield strength change at $10^{-2}$ dpa for low and high copper steel

- **Immobile interstitial clusters, high cluster sink strength:**
  - Cu-ppt dominated below $\sim 10^{-7}$ dpa/s
  - Point defect clusters dominate at high fluxes

- **Mobile interstitial clusters, reduced sink strength:**
  - Greater radiation-enhanced diffusion
  - No interstitial cluster contribution at high fluxes

\[ \text{ornl} \]
EXAMPLES OF MODEL PREDICTIONS

Effect of displacement rate on fluence dependence of hardening, mobile di-, tri-, and tetra-interstitial clusters

- c.f. base case above, little change for high-Cu steel
- rate effect eliminated in low-Cu steels, but level of hardening now much too low
EXAMPLES OF MODEL PREDICTIONS

Effect of reducing surface energy (2800 to 1400 erg/cm²) on fluence dependence of hardening, mobile di-, tri-, and tetra-interstitial clusters (little effect of interstitial cluster mobility as shown below)

- low-Cu steels, increased hardening due to greater vacancy cluster stability, too strong rate dependence
- high-Cu steel, greater vacancy cluster sink strength reduces radiation-enhanced diffusion, delays onset of copper precipitation
• Note that reducing the surface energy to by a factor of two increases the vacancy cluster lifetime from about 100 sec to $\sim 10^5$ sec

• longer cluster lifetime is in better agreement with estimates based on Monte Carlo cascade aging studies and post-irradiation annealing studies which indicate that the unstable matrix defect lifetime is $\sim 3-5 \times 10^5$ sec

• the difference in dose rate dependence of hardening for low- and high-copper steels may be influenced by the assumed barrier strength of vacancy and interstitial clusters

  - the dose-rate effect in high copper steels implies that the cluster sink strength must be a strong function dose rate to affect radiation-enhanced diffusion
  - high cluster sink strengths give greater than expected hardening, and more than the expected dose rate effect on hardening if moderate values such as $\sim 0.25$ are assumed for the cluster dislocation barrier strength
EXAMPLES OF MODEL PREDICTIONS

Effect of reducing surface energy (2800 to 1400 erg/cm²) on dose rate dependence of hardening, 0.01 dpa

- little influence of interstitial cluster mobility since vacancy clusters dominate point defect component, slight reduction in hardening at highest dose rates (note change in Y-axis scale)
EXAMPLES OF MODEL PREDICTIONS

Reduced surface energy moves minimum in dose-rate curve to lower dose rate, increases dose rate dependence at lower values (case shown is immobile interstitial clusters)

• reduces dose-rate independent plateau region
SUMMARY

A relatively simple kinetic model has been used to investigate the effect of variations in flux on radiation-induced hardening

• the predicted influence of damage rate on radiation-induced hardening depends on several other material and irradiation variables:
  - copper content
  - neutron fluence
  - flux range
  - temperature (not discussed here)

• depending on which values apply, a change in flux may lead to either an increase or a decrease in hardening

• the absolute magnitude of the flux effect, and the flux range with the greatest predicted sensitivity depends on the details of the model and the chosen parameters

• model predictions are consistent with a modest effect of neutron flux for the range of fluxes of interest to LWR RPV at ~290°C