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The Effect of Neutron Flux on Radiation-Induced Embrittlement in Reactor Pressure Vessel Steels

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ABSTRACT: The effect of neutron flux level is a longstanding concern for determining the applicability of test reactor data or high lead-factor surveillance data to the prediction of embrittlement in commercial reactor pressure vessels (RPV). However, as operating reactors reach higher fluences, the question of flux effects is becoming increasingly relevant for situations such as embrittlement attenuation through the (RPV) and the comparison of boiling and pressurized reactor RPVs. In spite of its technological importance and extensive experimental and theoretical investigation, the issue of neutron flux effects on radiation-induced embrittlement in RPV steels remains unresolved. For neutron fluxes much greater than RPV operating conditions (>100 times higher), a regime exists in which the effects of flux are well defined experimentally and are well predicted by current models. These same models predict that the effect of flux should be relatively weak for values near those obtained in commercial RPVs, but may increase at lower fluxes. However, it is difficult to obtain sufficient data at low to intermediate fluxes to unambiguously determine the effects of flux since the time required to reach the desired fluences is necessarily long. In order to demonstrate the potential effects of flux on RPV embrittlement, and to help interpret the available experiments, a kinetic embrittlement model has been used in an analysis of the primary variables involved. The results of the analysis are consistent with a modest effect of flux on embrittlement for fluxes near those typical of commercial RPVs.

KEYWORDS: neutron flux, embrittlement, ferritic steels, modeling, point defects, pressure vessels, radiation damage

Introduction

A concern about potential effects of neutron flux level on radiation-induced embrittlement in commercial reactor pressure vessel (RPV) steels arises from the range of neutron exposure conditions that are relevant to this problem. The RPVs are exposed to fast neutron fluxes (E>1.0 MeV) in the range of 10^{12} to 10^{15} n/m^2/s, corresponding to

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atomic displacement rates in the range of 10^{-9} to 10^{-11} displacements per atom (dpa)/s. Higher fluxes are typical of pressurized water reactors (PWR), and lower fluxes of boiling water reactors (BWR). The fast flux is reduced by about a factor of 20 from the inside to the outside of an 8-inch-thick PWR RPV and by about a factor of 12 for the thinner (~6 inch) BWR [*1*]. The specimens employed in commercial reactor surveillance programs are normally exposed at a a flux that is 3 to 5 times greater than the maximum RPV value to provide an early indication of the expected effect of irradiation. Finally, much of the data used to determine the radiation sensitivity of RPV materials has been obtained from irradiations conducted in materials test reactors at fluxes between 10 and 1000 times greater than the in-service exposure conditions. As a result, the effect of neutron flux level is a longstanding concern for determining the applicability of test reactor data or high lead-factor surveillance data to the prediction of embrittlement in commercial RPVs. Moreover, as operating reactors reach higher fluences, the question of flux effects is becoming increasingly relevant for situations such as embrittlement attenuation through the RPV and the comparison of PWR and BWR vessels.

In spite of its technological importance, and extensive experimental and theoretical investigation, the issue of neutron flux effects on radiation-induced embrittlement in RPV steels remains unresolved. This is witnessed by differences between the two alternate forms for an embrittlement correlation [*2,3*] that have been proposed to replace the current version in NRC Regulatory Guide 1.99 [*4*]. It has been difficult to unambiguously determine the effects of flux in the relevant range because the irradiation time required to reach the desired fluences is necessarily long. The experimental data are further confounded by the variations in other irradiation and material parameters such as temperature, neutron energy spectrum, fluence, and chemical composition to mask the effect of flux. In order to demonstrate the potential range of flux effects on RPV embrittlement, and to help interpret the available experiments, a kinetic embrittlement model has been used to analyze the primary variables involved. As discussed below, this analysis and the model predictions indicate that the effects of neutron flux level could be significant in the flux range typical of commercial RPV exposure conditions.

Model Summary

The model employed in this work has been under development for several years, and only a brief summary of its features will be presented here. A detailed description of the model can be found in References [*5-7*]. The model applies the reaction rate theory method to describe the time-dependent evolution of point defects, point defect clusters, and copper precipitates. The results of molecular dynamics simulation studies [*8,9*] were used to provide guidance for the choice of the primary damage source term in the model, i.e. the displacement cascade survival efficiency and in-cascade vacancy and interstitial clustering fractions. Interstitial cluster formation can occur in the model by essentially classical nucleation (random collisions between single interstitials) or directly in the displacement cascade. Because vacancy clusters are thermally unstable, a simpler treatment of vacancy clustering is used. A single-size population of small vacancy clusters is assumed to form within the cascades and their mean lifetime is calculated based on the instantaneous point defect fluxes and the temperature-dependent rate at which vacancies

are evaporated from the clusters. In this case, the vacancy cluster is a transient sink that acts primarily as a recombination site.

The major features of the copper precipitate model are consistent with experimental observations of these features in RPV steels [*10*]. These include the observation that the precipitate nuclei are found at very low doses and the number of precipitates is only weakly dependent on dose for the neutron fluences of interest. Thus, the details of precipitate nucleation not modeled. The assumed precipitate number density is a fluence-independent function of irradiation temperature and copper content. The initial precipitate radius is set equal to the magnitude of the Burgers vector, which is small enough to result in essentially no hardening at zero dose. Subsequent precipitate growth under irradiation is calculated assuming diffusion limited kinetics with radiation-enhanced diffusion of copper by a vacancy mechanism. The influence of other solutes such as nickel and manganese that may also be incorporated into the precipitates is not accounted for.

A simple dislocation barrier hardening model is used to calculate the hardening due to point defect clusters. Both vacancy and interstitial clusters are treated as weak dislocation barriers with a barrier strength of 0.25. The Russell-Brown model [*11*], which is based on the modulus difference between precipitate and matrix, is used to compute hardening due to copper precipitates. A root-sum-square combination of the shear strength increment arising from the individual defect contributions used to obtain total hardening. The total shear strength increments is converted to a corresponding change in the uniaxial yield strength using the Taylor factor ($\Delta\sigma_y$=3.06*$\Delta\tau$) [*12*], and yield strength changes can be related to Charpy shifts using correlations from the literature, ΔT_{41}~0.65*$\Delta\sigma_y$ [*13*].

Typical Model Predictions

The predictions of this kinetic embrittlement model have been shown to be in reasonable agreement with surveillance data from the Power Reactor Embrittlement Database (PR-EDB) which is maintained by the U. S. Nuclear Regulatory Commission *14,15*]. A typical model comparison between the model's predictions and Charpy data from the PR-EDB is shown in Fig. 1 which is reproduced from Ref. [*6*]. In Fig. 1 and throughout this paper, the calculated dpa are converted to neutron fluence using a representative displacement cross section of 1500 barn (0.015 dpa/1x10^{19} n/cm^2, E>1.0 MeV). Although there is considerable scatter, the mean behavior of the data (e.g. the dependence on fluence and copper level) is well represented be the model. The model exhibits a stronger dependence on bulk copper concentration than is seen in the data. This difference arises because all the copper is available for precipitation in the model, whereas it is known that the matrix copper content is reduced in real materials as a result of the formation of coarse copper precipitates during thermal treatment. The transition from low to high Charpy shift is more abrupt in the model predictions due to simplifications in the copper precipitation model, i.e. precipitate nucleation is not well simulated.

Optimization of material parameters within a reasonable range can certainly be used to improve model predictions, but results obtained with the basic parameter set used in Ref. [*6*] are adequate to illustrate the impact of displacement rate in the model. The primary material parameters are shown in Table 1. Specific parameter variations from this basis set will be discussed below to illustrate their affect on the model predictions of

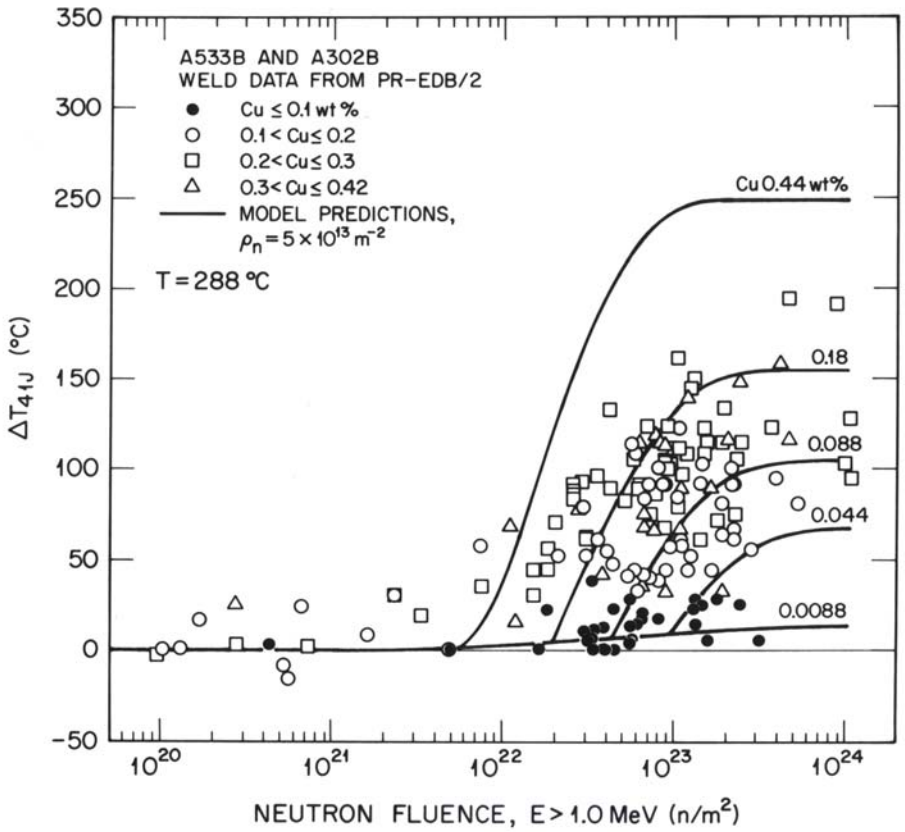

FIG. 1 — *Comparison of model predictions and Charpy data from PR-EDB.*

TABLE 1 - *Basis set of material parameters for RPV steels.*

Parameter (units)	Value
Cascade survival efficiency	0.25
Total in-cascade interstitial clustering fraction	0.0 to 0.5
Individual interstitial clustering fractions	0.0 to [0.15, 0.1, 0.05]
Total vacancy clustering fraction	0.0 to 0.5
In-cascade vacancy cluster radius (nm)	0.5
Interstitial migration energy (eV)	0.25
Vacancy migration energy (eV)	1.25
Vacancy formation energy (eV)	1.55
Free surface energy (J/m^2)	2.8
Network dislocation density (m^{-2})	2.0 x10^{14}
Dislocation capture efficiency: interstitials vacancies	1.25 1.00
Dislocation barrier strength: point defect clusters	0.25

displacement rate affects. These parameters are primarily those that influence the point defect cluster population: in-cascade clustering fractions for interstitials and vacancies, migration energy of interstitial clusters, and surface energy.

Basic Mechanisms Responsible for a Displacement Rate Effect

Radiation-induced property changes are driven by the excess point defect (vacancy and interstitial) fluxes created by displacive irradiation. The nature of the microstructural evolution that occurs is determined by the transport and fate of mobile point defects and solute atoms. In the case of RPV steels, displacement rate effects arise primarily from two processes: (1) the competition between formation and dissolution of defects that are unstable at the irradiation temperature (this component is nearly inseparable from the effects of irradiation temperature) and (2) the influence of the displacement rate on radiation-enhanced diffusion. Hardening from point defect clusters is most strongly influenced by the first process, and from copper precipitation by the second.

At relatively low fluxes, an increase in flux will increase the quasi-steady-state density of both mobile point defects and point defect/solute clusters. The former will lead to more radiation enhanced diffusion and an increase in precipitate hardening, and the higher cluster density will lead to greater hardening from this component. However, at still higher fluxes, the density of unstable point defect clusters may reach a high enough level to provide a significant sink for mobile point defects and thereby begin to reduce their concentration. At this point, higher fluxes will reduce radiation enhanced diffusion. Thus, an increase in flux (displacement rate) may lead to either an increase or decrease in the observed hardening [*5,6*].

The influence of flux as mediated by the point defect clusters at 300°C and 0.01 dpa is illustrated in Fig. 2. Results are included for displacement rates and total doses higher than those typically obtained in commercial RPVs to illustrate a broader range of parameters. The higher displacement rates have been used in some accelerated irradiation experiments, and the dose of 0.1 dpa is somewhat beyond the maximum for commercial RPVs that have plans for a 20-year life extension. In Fig. 2a, the ratio of the vacancy concentration to the thermal equilibrium value is plotted as a function of displacement rate for two cases. The ratio shown by solid line in Fig. 2a is obtained if there is no in-cascade clustering of either vacancies or interstitials, and the dashed line is for the case where 40% of the vacancies and 50% of the interstitials produced are in such clusters. The deviation from a linear flux dependence which is associated with sink-dominated absorption of point defects occurs at about an order of magnitude lower flux for the case in which in-cascade clustering leads to a much higher defect cluster density. The ratio of point defects lost to matrix recombination to those lost absorption at sinks can also be used to illustrate the impact of point defect clusters as shown in Fig. 2b for the same two cases as in Fig. 2a.

Model Predictions of Displacement Rate Effects

The predicted effect of displacement rate on the fluence dependence of hardening (yield strength change) is shown in Fig. 3, which was obtained using the same basis set of model parameters as in Fig. 1. For a relatively high-copper steel, the primary effect of lower displacement rate is a reduced fluence to initiate hardening from copper-rich precipitates. Little effect on the peak hardening is observed since this value is determined

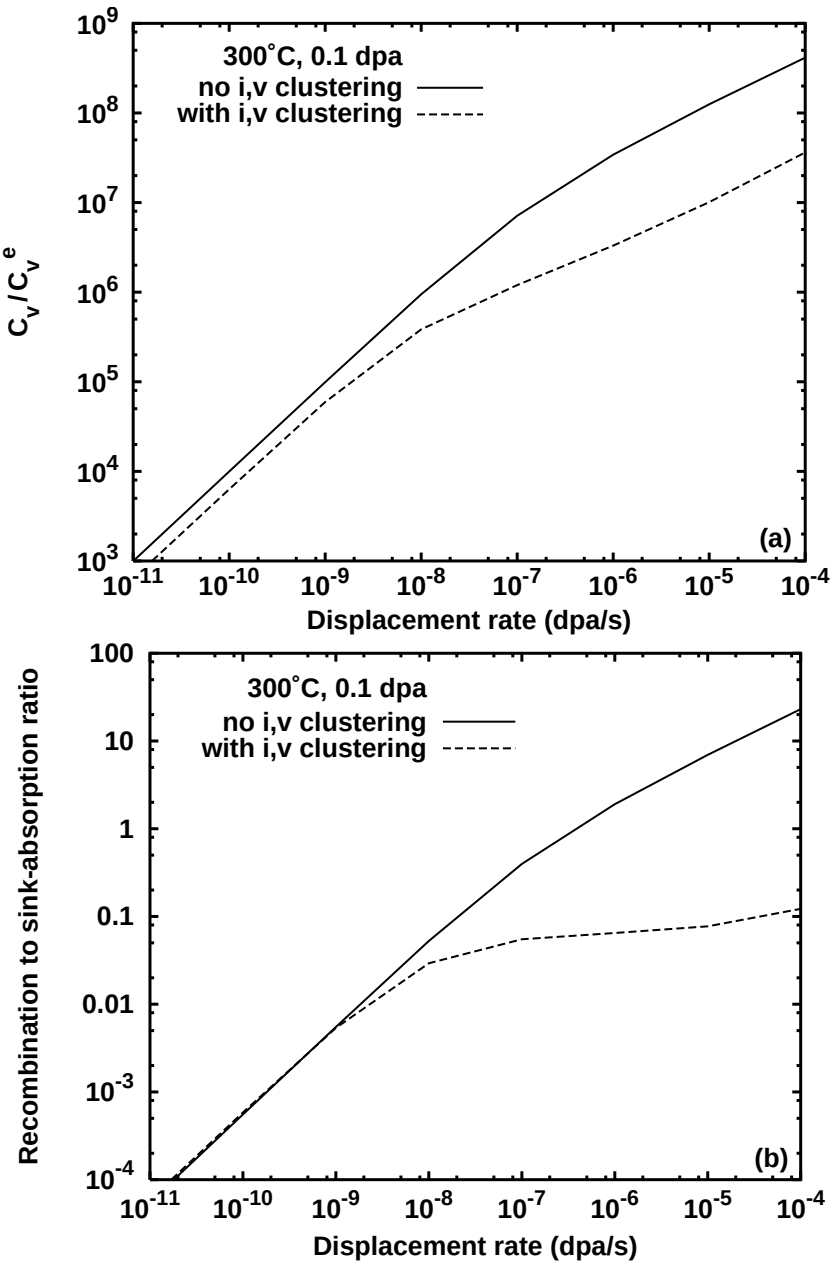

FIG. 2 — *Impact of displacement rate on vacancy supersaturation (a) and relative ratio of point defect lost to matrix recombination and absorbed at sinks (b).*

by the amount of copper available to precipitate. At low fluences, hardening is reduced at lower displacement rates which can lead to a crossover at intermediate fluences. This effect on precipitation is more clearly shown in Fig. 4, where the dose required to obtain a given level of copper precipitation is shown as a function of displacement rate. The increase in the dose required to reach a fixed fraction of copper precipitated increases sharply above a threshold value of the displacement rate. The boundary between dose-rate independent and dose-rate dependent behavior decreases for higher levels of precipitation. The asterisk placed at the end of the 50% and 95% precipitation curves indicate that this level was not reached by 0.1 dpa.

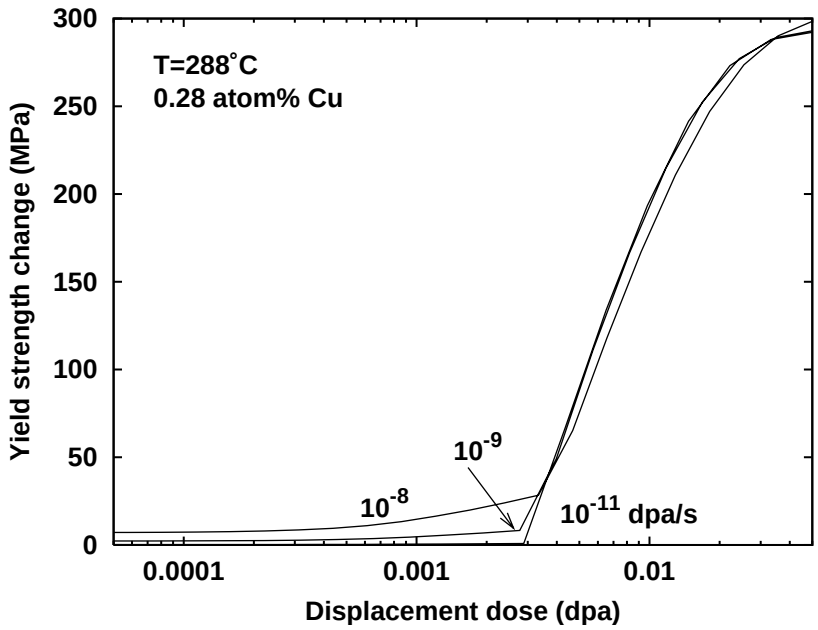

FIG. 3 — *Effect of displacement rate on fluence dependence of hardening in a high-copper steel.*

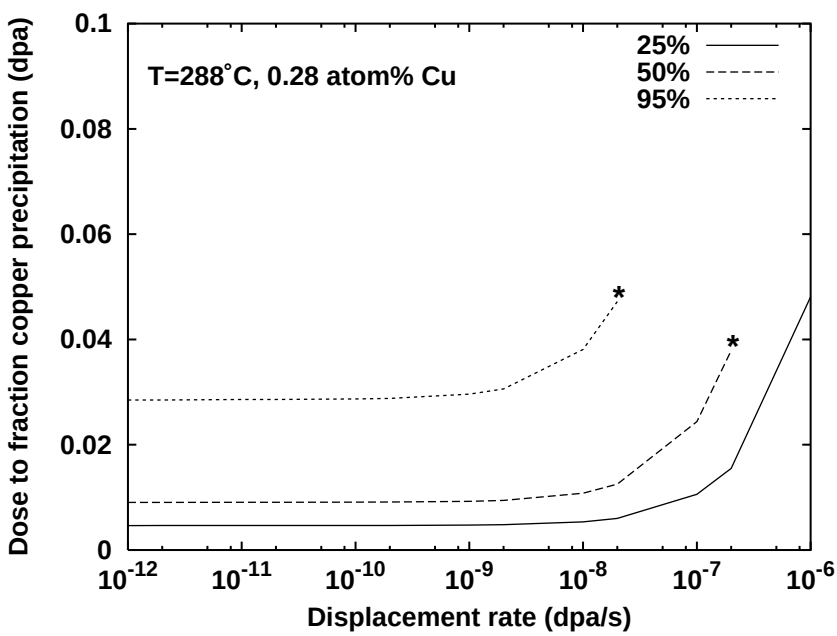

FIG. 4 — *Effect of displacement rate on dose required to reach a specified fraction of copper precipitation (* - indicated level of precipitation not reached by 0.1 dpa).*

The predicted hardening in a low copper steel is shown in Fig. 5 for the same range of doses and displacement rates as in Fig. 3. In contrast to the high copper steel, hardening is reduced at all doses for lower displacement rates. This is because hardening is dominated by the point defect cluster population in the low copper steel. It should be pointed out that the predicted effect of displacement rate in low copper steels is much

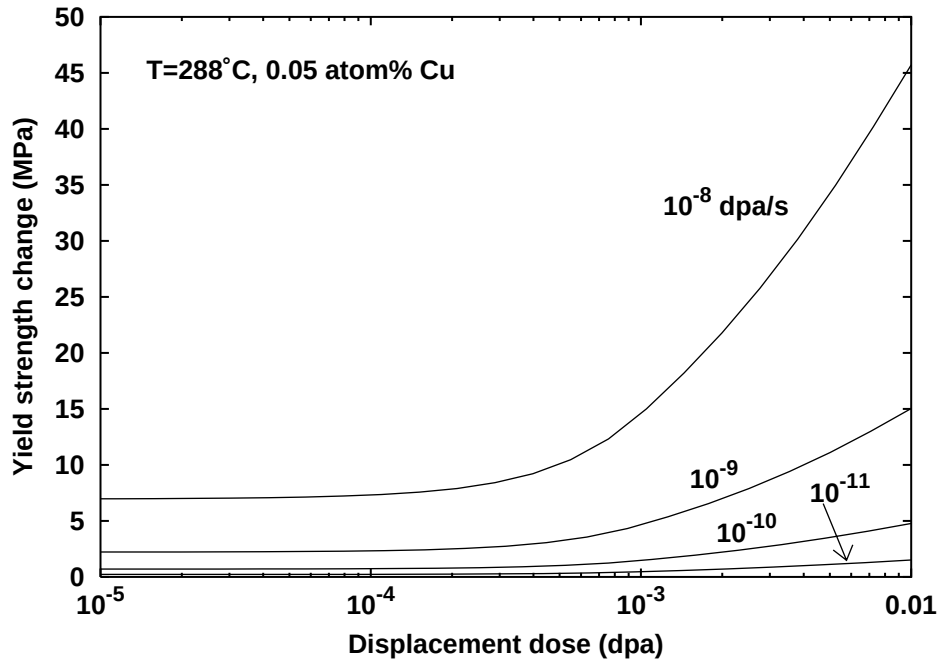

FIG. 5 — *Effect of displacement rate on fluence dependence of hardening in a low-copper steel at 288°C.*

stronger than that which is generally reported for these materials [*15*-17]. One potential explanation for this discrepancy will be discussed below.

Two final examples will be used to illustrate the displacement rate dependence of the model: (1) interstitial cluster mobility, which strongly impacts the interstitial cluster population, and (2) the free surface energy, which affects the vacancy cluster population. The possibility that small interstitial clusters may be mobile is supported by the results of molecular dynamics simulations of the evolution of displacement cascades in iron [*8,9,18,19*]. The interstitial clusters were observed to have a very low activation energy for migration, nearly the same as that for the single interstitial. In addition, their migration appeared to be preferentially 1-dimensional, rather than 3-dimensional. The impact of this mobility on embrittlement predictions from this kinetic radiation damage models has been discussed previously [*7*]; the focus of this paper is how mobility alters the model's dependence on displacement rate. As discussed in Ref. [*7*], including only the mobility of interstitial clusters of 2, 3, and 4 is considered adequate to evaluate the phenomenon. They are assumed to migrate in 3-D, which is consistent with recent Monte Carlo simulations which indicate that 1-D diffusion with frequent direction changes is nearly equivalent to 3-D migration. [*20*].

The predicted hardening at a dose of 0.01 dpa is shown as a function of displacement rate in Fig. 6 for both high and low copper steels. Predictions obtained with immobile interstitial clusters are shown in Fig. 6a and those for which the di-, tri-, and tetra-interstitial clusters are mobile are shown in Fig. 6b. The single interstitial migration energy of 0.25 eV was used for the small interstitial clusters. High interstitial cluster mobility greatly reduces the concentration of larger interstitial defects and their contribution to hardening decreases accordingly. This leads to a significant reduction in

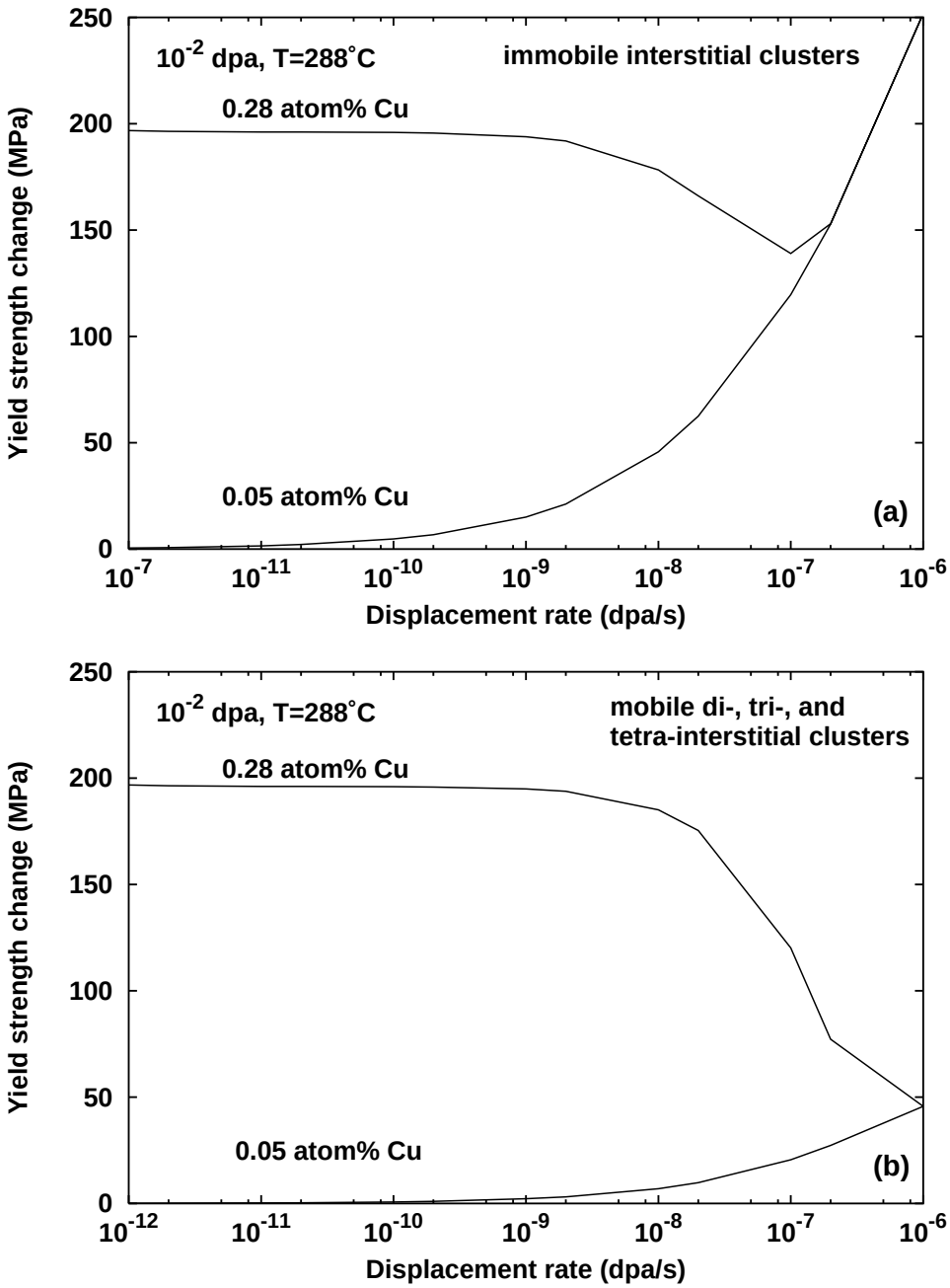

FIG. 6 — *Influence of interstitial cluster mobility on displacement rate dependence of hardening predicted at 0.01 dpa for low and high copper steels 288°C. Interstitial clusters are immobile in (a) and mobile in (b).*

the effect of displacement rate for low copper steels. Although this dependence is more consistent with the data mentioned above, to absolute level of hardening is now lower than the data would indicate. For high copper steels, the effect of displacement rate is increased at intermediate to high values because the interstitial cluster contribution to hardening does not increase significantly.

The free surface energy plays a critical role in determining the lifetime of vacancy clusters, with higher surface energies reducing the lifetime by promoting vacancy

emission. The value shown in Table 1 is taken from the temperature-dependent expression used in Ref. [*5*] and derived from Ref. [*21*]. This value is higher than that typically used in radiation damage modeling, with the argument for lower values based on the role of impurities and surface active elements such as oxygen. The effect of reducing the surface energy from 2.8 to 1.4 J/m^2 is shown in Fig. 7, which may be compared with Fig. 6b as a basis. The lower surface energy increases the vacancy cluster lifetime from about 100 s to 10^5 s, a value that is more consistent with lifetimes obtained from post-irradiation annealing studies [*22*]. The longer lifetime leads to higher vacancy cluster concentrations, which results in increased hardening levels and a stronger dependence on displacement rate in low copper steels. Two related effects are observed in high copper steels. Hardening is increased at high displacement rates, and the minimum in the hardness vs. displacement rate curve is moved to a lower value.

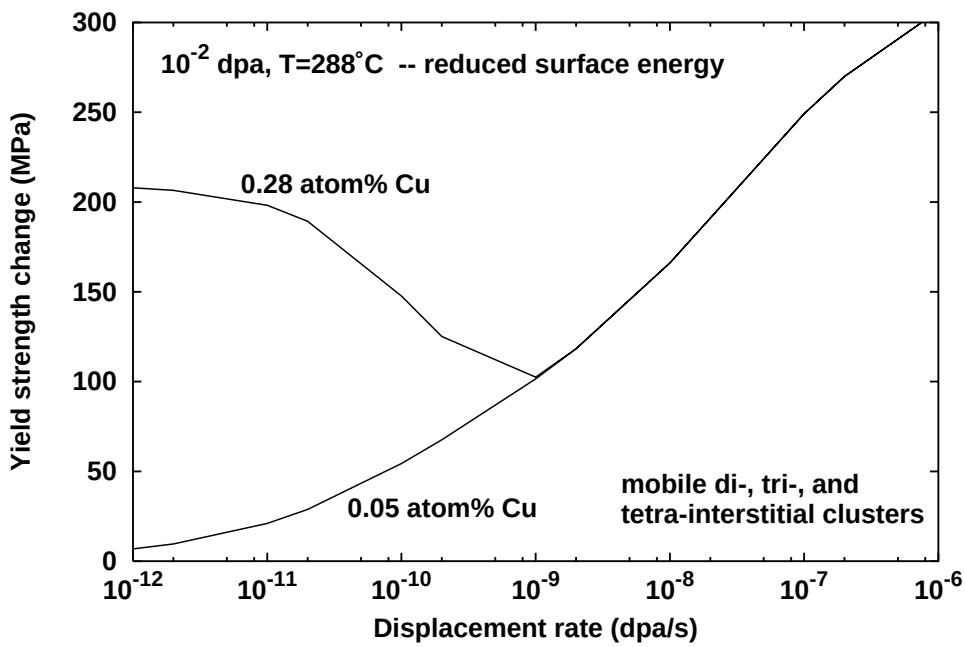

FIG. 7 — *Predicted displacement rate dependence of hardening at 288°C when a reduced value is used for the surface energy, c.f. Fig. 6b.*

The comparison of model predictions for low and high copper steels, and the fact that the displacement rate dependence is generally too high for the low copper steels raises an additional issue. This is the assumed dislocation barrier strength of vacancy and interstitial clusters. The dose-rate effect observed in high copper steels implies that the cluster sink strength must be a strong function of displacement rate to provide the required influence on radiation-enhanced diffusion. However, in low copper steels, these high cluster sink strengths give greater than expected hardening, and are responsible for the higher than expected displacement rate dependence. This implies that even the relatively moderate value of 0.25 that has been assumed for the dislocation barrier strength may be too high. Lower barrier strengths should be examined as a way to decouple the hardening effect from the sink strength effect on radiation-enhanced diffusion.

Summary

A simple kinetic model has been used to investigate the effect of variations in flux on radiation-induced hardening and embrittlement. Within the constraints of the model, the predicted influence of damage rate on embrittlement is not simple or monotonic since the response of the material to changes in flux depends on several other material and irradiation variables, including copper content, the total neutron fluence, the range over which flux is changed, and the irradiation temperature. Depending on the value of other material and irradiation variables, 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 also depends on the details of the model and the material parameters.

Overall, the model predictions are consistent with a modest effect of neutron flux on embrittlement for the range of fluxes of interest to commercial LWR pressure vessels at ~290°C. This effect has significant implications for the prediction of embrittlement variation through the thickness of the RPV, high fluence exposure in BWR pressure vessels, and reactor plant life extension. Since it is difficult to obtain the low-flux, high-fluence experimental data needed to unambiguously resolve this question, more detailed modeling work and additional analysis of available data are needed.

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