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Letter to the Editor

ON PRECIPITATE DISSOLUTION USING THE CASCADE SLOWING-DOWN THEORY *

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The dissolution probability for precipitates under irradiation is evaluated using the cascade slowing-down theory. By using a diffusion length calculated for average recoils in a collision cascade and by including the electronic stopping effect in the theory, the results from the cascade slowing-down theory are in agreement with the computational results of Muroga, Kitajima, and Ishino. Also, the theory is consistent with experimental observations by Sekimura et al.

The effect of radiation dissolution on precipitate stability is of considerable concern, since the mechanical properties of nuclear structural materials are mainly determined by precipitate–dislocation interactions. The physics of precipitate dissolution is quite complex and involves synergistic collisional, diffusional and microchemical processes. Collisional processes include implantation, displacement, and energy partitioning. Diffusional processes involve thermal and radiation enhanced atomic diffusion and segregation. Microchemical phenomena are associated with local stoichiometric and chemical changes. The interplay among these three processes complicates a comprehensive investigation of precipitate stability.

Collisional processes that involve energetic ions or atoms occur on a time scale of picoseconds. On the other hand, diffusional and microchemical phenomena, which are thermodynamic in nature, take place on a much longer time scale. We therefore can separate the precipitate dissolution problem into two stages. The first stage involves collisional processes and can be considered to occur instantaneously, while the second stage involves mainly thermodynamic processes and can be best modelled using rate theory. For low- to medium-Z materials, the energy deposition by the primary knock-on atoms (PKAs) is not locally dense enough to result in thermal spikes. Under such situations, precipitate dissolution is dominated by collisional processes.

Wilkes [1] has first theoretically predicted the collisional dissolution of precipitates in steels using the bubble re-solution model, which was originally developed by Hudson and others [2,3] for the treatment of fission gases in uranium fuels. Using the Monte Carlo method, Chou and Ghoniem [4] observed the kinetic dissolution of small precipitates by energetic collision cascade. A convenient way of measuring the collisional dissolution effect is through using the dissolution parameter which is defined as the fraction of precipitate atoms returned to the matrix per dpa (displacement per atom). Chou and Ghoniem [5] developed a cascade diffusion slowing-down theory to evaluate the dissolution parameter. Consequently, Sekimura et al. [6] conducted experiments to investigate the precipitate dissolution phenomenon. Their experimental results indicate that the dissolution parameter is considerably smaller than that predicted by the Chou–Ghoniem theory [5]. Muroga, Kitajima, and Ishino (MKI) [7] incorporated the Monte Carlo ion transport calculation with the Gelles and Garner ion range distribution model [8] to obtain apparently smaller precipitate dissolution parameters than those inferred from the cascade slowing-down theory.

The purpose of the letter is to slow that, by properly choosing the diffusion length and relaxing the assumptions used in the cascade slowing-down theory, good agreements between the results of the MKI model and the Chou–Ghoniem cascade slowing-down theory are obtained. Furthermore, the results are shown to be consistent with the experimental observation by Sekimura et al. [6]. A brief outline of the Chou–Ghoniem cascade slowing-down theory and the MKI model are given below and this is followed by an analysis of the average recoil dissolution rate. Finally, conclusion are drawn in the last section.

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Cascade slowing-down theory

The cascade diffusion slowing-down equation solved by Chou and Ghoniem takes the form [5]:

$$-D\,\nabla^2\Phi(\boldsymbol{r}, E)+\Sigma_t\Phi(\boldsymbol{r}, E) = \int_0^\infty \mathrm{d}E'\,\Sigma_s(E'\to E)\,\Phi(\boldsymbol{r}, E') + Q(\boldsymbol{r}, E)+\frac{\mathrm{d}[S(E)\Phi(\boldsymbol{r}, E)]}{\mathrm{d}E}, \quad (1)$$

where $\Phi(E)$ is the recoil flux at energy E, Q is the displacement rate, D is the diffusion coefficient, Σ_T is the total interaction cross section, $\Sigma_s(E'\to E)$ is the differential scattering cross section from energies E' to E, and $S(E)$ is the electronic stopping. The derivation of this equation requires the use of the concept of particle conservation which balances loss and gain rates in the appropriate phase space. In eq. (1), $-D\,\nabla^2\Phi(\boldsymbol{r}, E)\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ is the net rate at which particles are lost from the $\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ phase volume due to leakage; $\Sigma_t\Phi\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ is the net rate at which particles are lost in $\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ from nuclear (atomic) interactions; $\int\mathrm{d}E'\Sigma_s(\boldsymbol{r}, E'\to E)\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ is the net rate at which particles are produced in $\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ from inscattering; $Q(\boldsymbol{r}, E)\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ is the net rate at which particles are generated in $\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ from sources; and $[\partial S(E)\Phi/\partial E]\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ is the net rate at which particles slow down into $\mathrm{d}\boldsymbol{r}\,\mathrm{d}E$ from interaction with electrons.

To obtain analytical solutions of eq. (1), a number of assumptions have to be used. It is assumed that particle scattering is represented by hard-sphere collisions, that the interaction cross section is constant, and that electronic stopping is negligible. We make the assumption of hard-sphere collisions for two reasons: (1) it is simple to use and (2) most of the recoils in a collision cascade are not energetic enough to penetrate the electronic cloud and their collisional behavior can adequately be treated by a hard sphere potential. Therefore, these hard-sphere-like recoils have constant sizes and hence have constant collision cross sections. Electronic stopping is negligible compared to nuclear stopping for low energy recoils. With these assumptions, Chou and Ghoniem used a Neumann's series expansion of the flux to produce analytical solutions for the dissolution rate. It was found that the dissolution parameter (dissolution rate to displacement damage-rate ratio) can asymptotically be expressed as:

$$b\simeq 1, \qquad r_p\le L/\sqrt{3}, \quad (2)$$

$$\simeq L/\sqrt{3}\,r_p, \qquad r_p\ge L/\sqrt{3}, \quad (3)$$

where r_p is the radius of the precipitate and L is a characteristic diffusion length. The diffusion length can be estimated from the projected range of cascade recoils. In this note, we will estimate the approximate value of L for average recoils such that eqs. (2) and (3) can be directly compared to experiments and to the MKI model. From the random walk theory, the diffusion length may be evaluated from the range of the recoils as

$$R^2=\xi L^2, \quad (4)$$

where ξ is the average number of collisions for a recoil with energy E to be slowed down to the displacement energy E_d, and R is the average range of recoils in a collision cascade. This equation is only valid when the recoils have an isotropic velocity distribution. This is achieved through cascading collisions and isotropic hard-sphere scattering. For planar precipitates, the projected range, which can be measured and calculated easily, should be used for R in the above equation. For spherical precipitates, the chord range should be used. The projected range can be measured and calculated easily. If the value of the chord range is not available, the projected range can be used instead with an error of less than 20%. For like atoms with hard-sphere collisions, the average energy loss for each collision is half the incident atom energy. Accordingly, ξ can be estimated as

$$\xi=\frac{\ln(E/E_d)}{\ln(2)}. \quad (5)$$

The relationship between the recoil projected range R and its energy E obtained by the TRIPOS calculations [4] is fitted as

$$R(\mathrm{nm})=A[E(\mathrm{keV})]^n, \quad (6)$$

where A and n are fitting constants. For an iron atom in iron, $A=0.53$ and $n=0.911$. Also from the cascade slowing-down theory [5], the energy flux spectrum of cascade recoils in an infinite medium is derived as:

$$\Phi(E)=\frac{2QE_0}{\Sigma E^2}+\frac{Q}{\Sigma}\delta(E-E_0), \quad (7)$$

where E_0 is the energy of the primary knock-on atoms of the cascades.

The MKI model

The MKI model [7] describes the dissolution parameter for a recoil in a collision cascade as

$$b=1, \qquad r_p\le 0.5R, \quad (8)$$

$$=\frac{12X-X^3}{16}, \qquad r_p\ge 0.5R, \quad (9)$$

where r_p is the precipitate radius, R is the recoil range, and

$$X=R/r_p. \quad (10)$$

In order to evaluate the average dissolution parameter for a complete cascade, MKI use the Monte Carlo ion transport method to evaluate the range distribution for a cascade. The average dissolution parameter can

then be obtained by integrating the individual dissolution parameter and the recoil distribution over the entire recoil energy distribution range.

Analysis of average recoil dissolution rate

The dissolution of a precipitate by collision cascades results from the interaction with recoils, particularly the higher order recoils generated in the precipitate. As such, those recoil atoms with enough energy to be transported through the precipitate are dissoluted in the matrix. Furthermore, Lindhard's theory [9,10] indicates that none, or only a small fraction, of the PKA energy is consumed in electronic stopping for low-energy collision cascades. However, the fraction of energy in the form of electronic stopping, which does not contribute to the generation of recoils, increases with the PKA energy. Therefore, the energy available for cascade generation is the nuclear stopping energy instead of the primary knock-on energy. The nuclear stopping energy E_n for a PKA with an energy E_0 is given by [10]

$$E_n = \frac{E_0}{1 + 0.13(3.4\epsilon^{0.167} + 0.4\epsilon^{0.75} + \epsilon)}, \tag{11}$$

where the reduced energy ϵ is given as

$$\epsilon = \frac{E_0}{2Z^2e^2/a}, \tag{12}$$

the Thomas–Fermi screening length a is

$$a = \frac{0.88a_B}{Z^{0.333}}, \tag{13}$$

a_B is the Bohr radius (0.053 nm), ϵ is the electron charge, and Z is the atomic number of the recoils.

The average recoil range for recoils in a collision cascade is obtained as

$$\bar{R} = \frac{\int R(E)\Phi(E)\,dE}{\int \Phi(E)\,dE}, \tag{14}$$

where $\Phi(E)$ is the recoil energy flux derived by Chou and Ghoniem [5] as given in eq. (7) with an exception that E_n is used in place of E_0. Likewise, the average number of collisions for precipitate recoils has the form:

$$\bar{\xi} = \frac{\int \xi(E)\Phi(E)\,dE}{\ln(2)\int \Phi(E)\,dE}, \tag{15}$$

$$= \frac{2E_n}{\ln(2)(2E_n - E_d)}\left\{1 - \frac{E_d}{E_n}\left[1 + \ln\left(\frac{E_n}{E_d}\right)\right]\right\}. \tag{16}$$

The value for $\bar{\xi}$ is in a range of 1.30 to 1.45 for PKA energies of 1 keV to 10 MeV. Using random walk theory, the diffusion length for an average recoil can be related to the average recoil range as:

$$\bar{L} = \bar{R}/\sqrt{\bar{\xi}}. \tag{17}$$

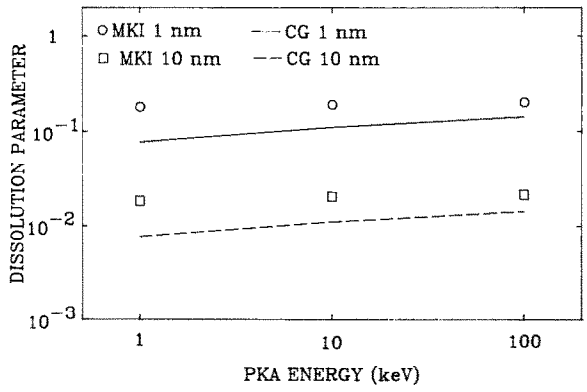

Fig. 1. A comparison of the dissolution parameters between Muroga–Kitajima–Ishino (MKI) model and Chou–Ghoniem (CG) cascade slowing-down model for precipitates with sizes of 1 nm and 10 nm.

The current application of the cascade diffusion theory to average recoils uses barred L and R instead of the originally defined L and R [5]. The average precipitate dissolution parameter has the form:

$$b \simeq 1, \qquad r_p \le 0.48R, \tag{18}$$

$$\simeq 0.48R/r_p, \qquad r_p \ge 0.48R. \tag{19}$$

The coefficient 0.48 in the above equations is for high energy cascades. For low energy cascades, it is about 0.50. Generally speaking, the coefficient is not a sensitive function of the PKA energy. Asymptotically, the cascade slowing-down formula is very similar to the MKI formula for average cascade recoils. However, for very large precipitates, the MKI formula predicts a 50% higher cascade dissolution rate. Fig. 1 shows a comparison of the dissolution parameters between MKI and cascade slowing-down models. The cascade slowing-down results are smaller by less than a factor of 2.0. This is attributed to the differences in the coefficient of the dissolution parameter and the recoil spectrum.

Sekimura et al. [6] performed precipitate dissolution experiments by irradiating titanium modified steels with a 400 keV argon ion beam. The sizes of precipitates investigated range from 25 to about 250 nm, with an average of 93 nm. Their results show that no significant dissolutioning of those precipitates occurs up to an irradiation dose of 100 dpa. The current extension of our cascade dissolution theory yields a 20% reduction in radius for 25 nm precipitates and less than 2% reduction in radius for 250 nm precipitates by 100 dpa. Deviations from experimental observations can be attributed to the thermodynamic recovery processes which may help stabilize these precipitates. Therefore, the average recoil dissolution parameter in the present work is consistent with the experimental results by Sekimura et al.

[6] where precipitate dissolution was found not to be significant up to an irradiation dose of 100 dpa.

Conclusions

This work incorporates the concepts of the nuclear damage energy and the average recoil diffusion length in the cascade slowing-down theory. This relaxes the assumption of neglecting the electronic stopping in the original cascade slowing-down work. Also, adoption of an average recoil diffusion length decreases the characteristic diffusion length for a collision cascade. These allow the cascade slowing-down theory to yield precipitate dissolution parameters which are more consistent with experimental observations on high energy collision cascades by Sekimura et al. [6]. Also, the calculated precipitate dissolution parameter is found to be within 50% of the results of the computational work of Muroga, Kitajima, and Ishino [7].

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