

22.14: Nuclear Materials, Spring 2015

Problem Set 4: Due March 20, 11:59PM

March 23, 2015

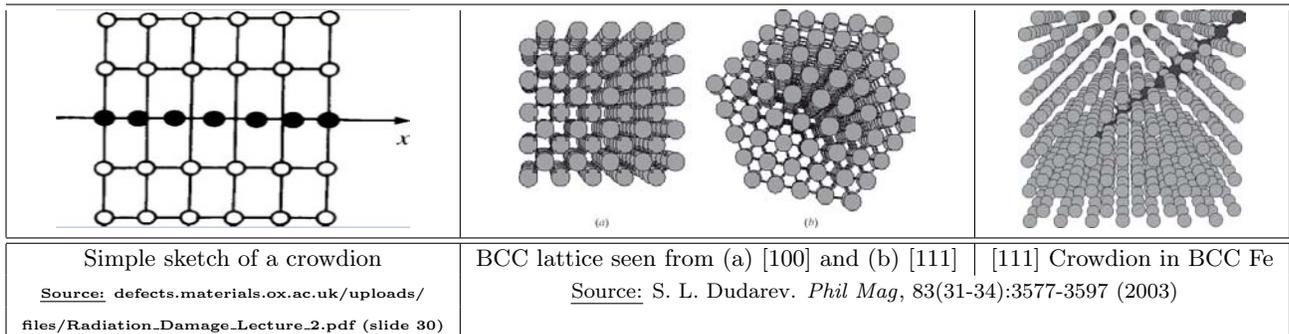
Topic: Radiation Damage

Purpose

An understanding of the damage left behind by radiation is the key to understanding how it affects material properties. In addition to simple vacancies and interstitials, a complex zoo of defect clusters can be left behind by a radiation damage cascade. In this problem set, you will explore the predicted effects of accounting for a third type of defect, besides vacancies and interstitials.

Assignment

We will consider the formation of a *crowdion*, or a one-dimensional defect made of self-interstitial atoms (SIAs). We will specifically consider the formation of crowdions in iron below its phase transformation temperature (BCC Fe) on the [111] directions. Some images to help explain what a crowdion is, along with a view of the BCC lattice from two angles, are shown below:



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These crowdions can diffuse very quickly (low migration energy) in one dimension. The formation of crowdions, being interstitial in nature, must compete with the formation of single SIAs.

1 (30 points) New Point Defect Equations

- (10 points) Assuming that $D_{\text{vacancy}} < D_{\text{interstitial}} \ll D_{\text{crowdion}}$ at all temperatures, develop a new set of point defect balance equations based on those from the reading (Was, "Fundamentals of Radiation Materials Science," p. 192) by introducing a fraction f_c of interstitial defects that become crowdions. Estimate the relative strength of any new rate constants to those that exist in the standard vacancy-interstitial point defect balance equations.
- (20 points) Using your new balance equations, draw new *qualitative* curves showing the time evolution of the three types of defects as functions of temperature and sink density (four cases in total), as in the reading (Was, pp. 194-200).

Solution

Using the two-defect equations as a template, we arrive at the following set of three equations:

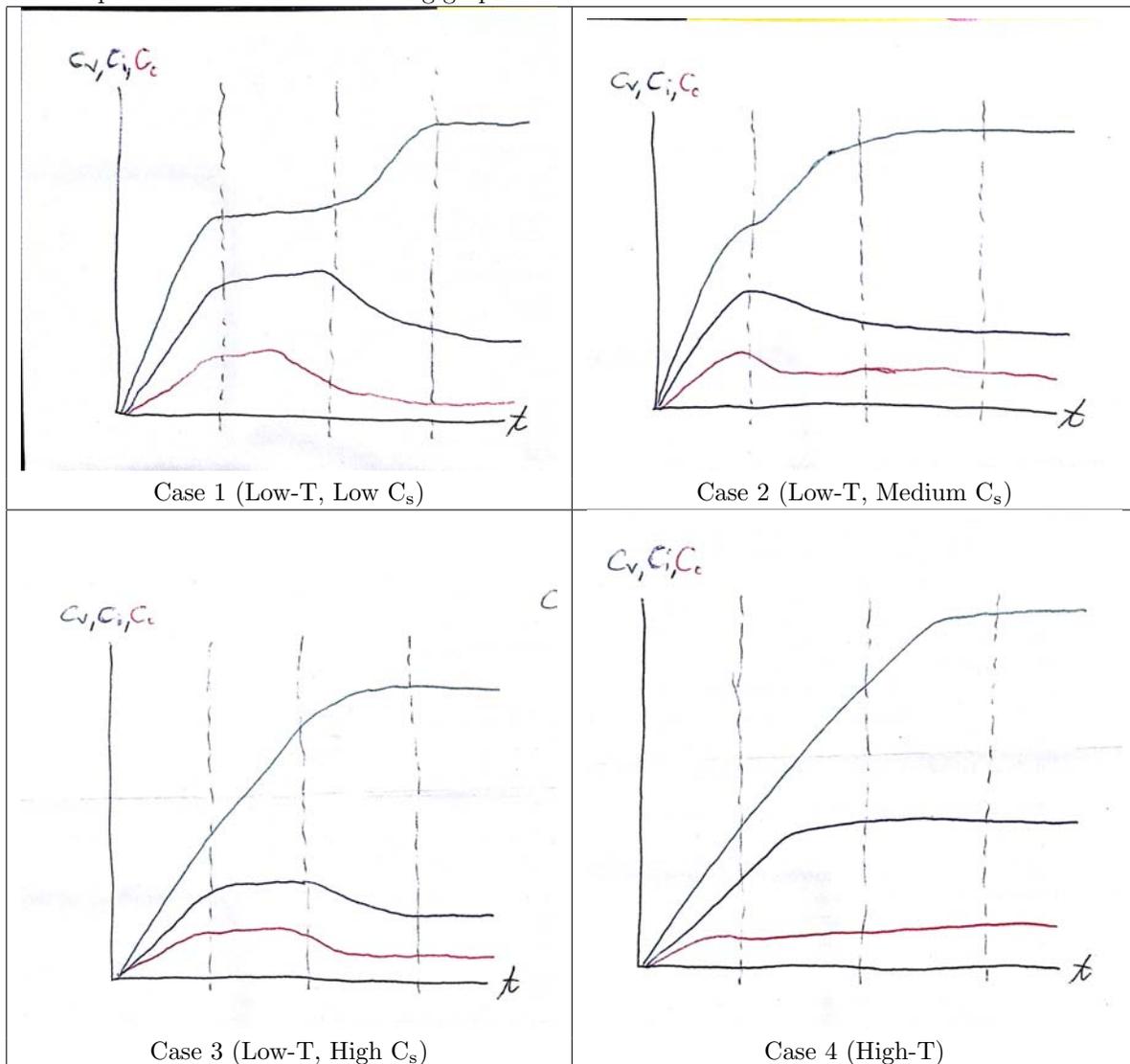
$$\frac{\partial C_v}{\partial t} = K_0 - K_{iv}C_vC_i - K_{cv}C_cC_v - K_{s,v}C_sC_v + D_v\nabla^2C_v \quad (1)$$

$$\frac{\partial C_i}{\partial t} = (1 - f_c)K_0 - K_{iv}C_iC_v - K_{ci}C_cC_i - K_{s,i}C_sC_i + D_i\nabla^2C_i \quad (2)$$

$$\frac{\partial C_c}{\partial t} = f_cK_0 - K_{cv}C_cC_v - K_{ci}C_cC_i - K_{s,c}C_sC_c + D_c\nabla^2C_c \quad (3)$$

Note here that we have omitted all spatially dependent terms, in this case the migration due to diffusion, as well as the interaction term between interstitials and crowdions. We assume that interstitials and crowdions are created within the damage cascade, and don't turn into each other afterwards. Because we assume that $D_{\text{vacancy}} < D_{\text{interstitial}} \ll D_{\text{crowdion}}$, we must also assume that $K_{iv} < K_{cv}$ and $K_{s,v} < K_{s,i} \ll K_{s,c}$, as diffusivities (mobilities) are the largest contributor to these terms.

These equations result in the following graphs:



Here, note that in all cases, the crowdions reach their eventual steady state much more quickly than the interstitials, remember that these are log-log plots. We have shown a fraction of crowdions well below 50%, for the sake of illustration. It is important that the steady state concentration of crowdions is always far lower, as their far higher mobility means that they will reach sinks at a disproportionately higher rate during steady state.

2 (40 points) Improving the Kinchin-Pease Model with Stopping Powers

Let's assume that the Kinchin-Pease model is too simplistic (it really is simple, isn't it?). We would like to develop a more comprehensive model, which accounts for the continuously changing interplay between nuclear and electronic stopping power.

2.1 (20 points) Develop a new expression for $v(T)$, removing the assumption of a linear increase in displacements with any cutoff value. Start by looking at how much energy is lost by a PKA to nuclear displacing collisions, compared to the total amount of energy lost by the PKA over its whole recoil energy range. Use the Ziegler-Biersack-Littmark (ZBL) universal nuclear stopping power correlation in Was' book, equations 1.149-1.151 for the nuclear stopping power, and equations 1.182-1.183 for the electronic stopping power. See the stopping power reading on the Stellar site, and assume self-ion irradiation.

For this problem, we must find expressions for nuclear and electronic stopping power over the full relevant energy range. Looking at the integrated stopping power from a maximum energy down to the displacement threshold energy E_d (NOT zero) will give the proportion of energy lost to nuclear collisions, and therefore displacements. Dividing this energy lost to nuclear stopping power by twice the displacement threshold energy ($2E_d$) will give the number of displacements created. This expression can be found in Equation 2.26 of Was' book:

$$E_{elastic\ collisions} = \int_{E_d}^{E_i} \frac{\left(\frac{dE}{dx}\right)_{nucl.}}{\left(\frac{dE}{dx}\right)_{nucl.} + \left(\frac{dE}{dx}\right)_{elec.}} dE \quad (4)$$

where E_i is the incoming particle's initial energy. Here we are neglecting radiative stopping power, as before. We will use the ZBL correlation (Ziegler-Biersack-Littmark, Was Eqns. 1.149-1.151), as it seems to be the easiest to implement. First, we start by defining a reduced ZBL energy (ϵ):

$$\epsilon = \frac{32.53 M_2}{Z_1 Z_2 (M_1 + M_2) (Z_1^{0.23} + Z_2^{0.23})} E_i \quad (5)$$

Note that in the case of self ions (which we are studying), $Z_1 = Z_2$ and $M_1 = M_2$. This reduces the ZBL energy to:

$$\epsilon = \frac{32.53 M}{Z^2 (2M) (2(Z^{0.23}))} E_i = \frac{32.53}{4Z^{2.23}} E_i \quad (6)$$

Next, we defined a reduced nuclear cross section based on this energy:

$$S_n(\epsilon) = \frac{0.5 \ln(1 + 1.1383\epsilon)}{(\epsilon + 0.01321\epsilon^{0.21226} + 0.19593\epsilon^{0.5})} \quad (7)$$

Finally, we use this to define a nuclear stopping cross section:

$$S_n(E_i) = \frac{8.462 \cdot 10^{-15} Z_1 Z_2 M_1}{(M_1 + M_2) (Z_1^{0.23} + Z_2^{0.23})} S_n(\epsilon) = \frac{8.462 \cdot 10^{-15} Z^2 M}{(2M) (2Z^{0.23})} S_n(\epsilon) = 2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon) \quad (8)$$

For the electronic stopping power, we will use a simplified expression (equation 1.182 in Was' book) valid over the range $0 < E$ (keV) $< 37Z^{\frac{7}{3}}$, which for the case of aluminum brings us just over 1 MeV:

$$S_e(E_i) = 3 \cdot 10^{-15} Z^{\frac{2}{3}} \sqrt{E_i} \quad (9)$$

Note that both stopping cross sections are in units of $[cm^2]$, and that both stopping power terms contain the number density, so it can be eliminated from our final expression:

$$E_{elastic\ collisions} = \int_{E_d}^{E_i} \frac{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon)}{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon) + 3 \cdot 10^{-15} Z^{\frac{2}{3}} \sqrt{E_i}} dE \quad (10)$$

In the attached spreadsheet, these values were entered for logarithmically increasing values of E_i , and the integral was performed numerically as follows:

$$\int_{E_d}^{E_{cell_n}} [Ratio] = \int_{E_d}^{E_{cell_{n-1}}} [Ratio] + \frac{\frac{1}{2} (Ratio_n + Ratio_{n-1})}{E_n - E_{n-1}} \quad (11)$$

This scheme adds the previous ratio's value to a rectangular estimate of the area under the ratio-energy curve between the previous and current cells. The accuracy of this method is dependent on the energy step size, and perhaps better numerical integration schemes (trapezoidal rule, Simpson's rule...) would yield more accurate results. No matter though, the qualitative result is the same.

This scheme has the benefit of only requiring two parameters unique to the material: the atomic number (Z) and the displacement threshold energy (E_d). A logarithmic energy step of $\log(\Delta E) = 0.1$ was used to refine the data and increase accuracy.

Finally, we arrive at our new expression for $\nu(T)$, by dividing by $2E_d$ to account for the number of possible collisions with each energy transfer:

$$\nu(T) = \frac{\int_{E_d}^{E_i} \frac{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon)}{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon) + 3 \cdot 10^{-15} Z^{\frac{2}{3}} \sqrt{E_i}} dE}{2E_d} \quad (12)$$

2.2 (10 points) In many atomistic simulations (such as DFT, MD, etc.), a “cut-off energy” and/or “cutoff radius” is often employed, whereby very small variations in calculations over large values of distance or small values of energy are neglected to save time. Here, a maximum energy cutoff value is in order. Devise an expression for this cutoff energy in your expression in (2.1), assuming that some fraction of displacements is enough to model the total number of displacements caused by a PKA. What is this cutoff energy value for aluminum (Al) and lead (Pb)?

Let's assume that once we reach an energy that would create 99% of the possible damage that an infinite particle energy would create, we can define a cutoff energy accordingly:

$$E_c = 0.99 \frac{\int_{E_d}^{\infty} \frac{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon)}{2.1155 \cdot 10^{-15} Z^{1.77} S_n(\epsilon) + 3 \cdot 10^{-15} Z^{\frac{2}{3}} \sqrt{E_i}} dE}{2E_d} \quad (13)$$

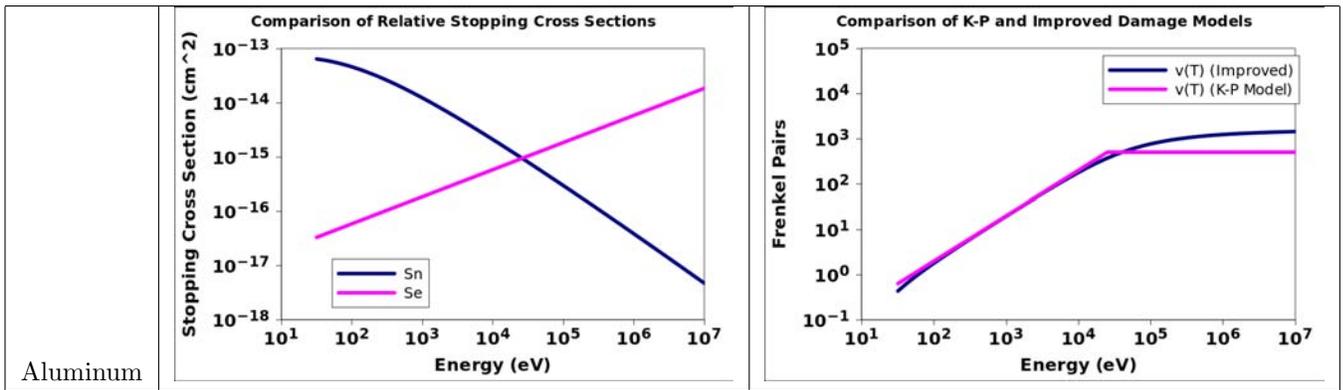
Using our spreadsheet, we examined the maximum number of collisions possible by setting the maximum energy to 1 TeV, a ridiculously high number. The table below summarizes relevant parameters for the four materials in question (C, Al, Fe, Pb), including some which are needed for question 2.3 for brevity:

Material	Z	E_d (Was Book)	E ($S_e \simeq S_n$)	$\nu(1\text{ TeV})$	$0.99 \cdot \nu(1\text{ TeV})$	E_c
Al	13	25 eV	25 keV	77,678	76,901	1 GeV
Pb	82	25 eV	1 MeV	2,974,410	2,944,666	25 GeV

Clearly these cutoff values are insane... but also the K-P model greatly underestimates the amount of damage done at high energies!

2.3 (10 points) Graph your expression for $\nu(T)$ on top of that for the Kinchin-Pease model for aluminum. How does your model improve upon the K-P model, and why? What shortcomings does your new model still have?

Graphs comparing stopping powers and $\nu(T)$ curves are shown below for each of the four materials:



This new model still neglects many features, such as energy focusing through close elastic collisions, channeling as additional electronic stopping power, any high-energy reactions, and radiative stopping power. In addition, the stopping powers themselves are semi-empirical, with accuracies between 0-18% over the energy range given.

3 (30 points) Crowdions to the Rescue

1. The presence of crowdions has recently been suggested as *the* reason that nuclear structural materials can even exist. Here, you will read through a recent paper concerning crowdion energetics, and try to understand why crowdion stability can lead to decreased swelling.

Read through the attached paper by Golubov et al., and explain in no more than one page why you think that crowdions are responsible for the far smaller observed void swelling compared to predicted levels. You may want to consider the following in your response:

- (a) What is the major issue being addressed, and why was this study necessary?
- (b) What assumptions were made in performing the simulations in this study?
- (c) What do the authors mean by “interaction energy landscapes?”
- (d) Why does the fact that crowdions can move in 1-D (as opposed to 3-D) help them escape the “pull” of dislocations?
- (e) Void swelling is typically caused by the coalescence of vacancies formed from radiation. Why would surviving crowdions help reduce void swelling?

The issue being addressed is that the presence of interstitials, which can diffuse in 3D, at assumed concentrations fails to explain the slow nucleation and growth of void swelling in structural nuclear materials. The assumptions made in the study include that the split dumbbell is the stable defect, and that it can convert into a crowdion in the presence of the stress field of a dislocation. These crowdions then only are assumed to diffuse in 1D, in directions parallel to, but not intersecting, the dislocation cores, meaning that then the crowdions are disproportionately available to recombine with vacancies and annihilate at other sinks. The interaction energy landscape shows the energy potential gained by either type of defect as it approaches the dislocation core, shown in a graphical manner. The reason that crowdions can help reduce void swelling is that once they convert into 1D diffusing defects (from the split dumbbell), the dislocation nearby effectively “turns off” as a sink, meaning that it is free to find the nearest other defect, which may be a vacancy or a nucleating void. This reduces the number and size of nucleating voids, delaying the incubation period of void swelling.

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