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A PROPOSED METHOD OF CALCULATING DISPLACEMENT DOSE RATES

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A simple procedure is proposed for calculating the number of atomic displacements produced in a damage cascade by a primary knock-on atom of known energy. The formulae have been chosen to give results in close accord with recent computer simulations of radiation damage phenomena. The proposed new standard is compared with other empirical formulae for estimating the number of atomic displacements in a cascade.

1. Introduction

In analyzing the behavior of reactor materials, it is necessary to know the rate at which the atoms of a solid are displaced from their lattice sites. Recent experiments have demonstrated that a very important effect of displacement damage for reactor technology is the swelling associated with the production of voids in materials irradiated at elevated temperatures. It is not possible to achieve the very high neutron doses required to study these effects in a reactor environment without costly and time-consuming experiments. In consequence, many laboratories are now carrying out so-called simulation experiments using irradiation with energetic ions or electrons. These techniques can be used to produce very high damage rates and hence void production studies may be carried out quickly and more economically.

To compare different reactor experiments and in order that simulation experiments can be used to predict the properties of materials during reactor irradiation, a method of estimating the displacement dose is required. Ideally, this should have some measure of international support. Such an agreed procedure would make it easier to predict the behavior of materials, not only for fission reactors, but also for the more extreme conditions expected in nuclear fusion systems where direct experiments are not presently possible.

In this report, a simple method is outlined for calculating the number of atoms displaced in a solid during irradiation. It is closely related to widely used current procedures, besides which it incorporates recent theoretical and experimental developments in the field. The background of the proposal will be summarized only briefly, as the necessary additional details can be found elsewhere [1,2]. However, an attempt will be made to indicate areas of uncertainty,

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where further experimental or theoretical research are needed.

2. Physical models

Five approaches to the problem of calculating the damage generated in a solid by a primary recoil atom of given initial energy can be identified. The models described are either in common use or else give results which are directly relevant to our purpose of choosing an acceptable standard damage equation.

2.1. The Kinchin–Pease model

Kinchin and Pease [3] estimated the number of displaced atoms N_d generated by a primary knock-on atom (PKA) of energy E as

$$N_d = \begin{cases} 0 & 0 < E < E_d \\ 1 & E_d < E < 2E_d \\ E/2E_d & 2E_d < E < E_1 \\ E_1/2E_d & E_1 < E < \infty \end{cases}, \qquad (1)$$

At energies above E_1, the recoils lose energy only by electron excitation, while below E_1, they slow down entirely by hard-core elastic scattering. An atom receiving an energy greater than the displacement threshold E_d is permanently displaced, while struck atoms receiving energy less than E_d will eventually return to a lattice site. Crystal lattice effects were not considered either in the single atom displacement process or in the cascade development.

2.2. The half-Nelson model

Nelson [4] proposed a semi-empirical modification of the Kinchin–Pease model incorporating a number of corrections believed to be important. His formula is

$$N_d = \alpha \beta(E)\, W(E)\, E/\gamma(E)\, E_f. \qquad (2)$$

Here α is a factor introduced to allow for realistic atomic scattering instead of the hard core approximation. Theoretical estimates [5,6] suggested a value of α around 0.75. The factor $\beta(E)$ was intended to account for defect recombination within the cascade, but this has usually been ignored by setting $\beta(E)$ equal to 1. The factor $W(E)$ is the fraction of the initial PKA energy dissipated in elastic collisions. This was estimated using the stopping power theory of Lindhard et al. [7] but the electronic energy losses were confined to the PKA itself.

Development of the cascade was regarded as terminated by the formation of focused replacement sequences [8]; thus the focusing energy E_f replaces the displacement threshold E_d. It is difficult to justify this step from theoretical considerations or from computer simulations of cascade development. Finally, $\gamma(E)$ which corresponds to the factor 2 in the Kinchin–Pease model, increases at higher energies, a trend that was inferred from cascade calculations of Beeler [9]. This was ascribed to recombination due to overlapping of different branches of a cascade, but no such effect has been observed in more recent calculations. The distinction between the factors $\beta(E)$ and $\gamma(E)$ in this model is obscure.

2.3. Binary collision simulations using the threshold model

Torrens and Robinson [10,11] have constructed computer simulations of collision cascades in which the trajectories of displaced atoms in a crystalline solid are followed through a sequence of isolated binary collisions. The atoms interact according to an explicit and fairly realistic two-body potential. Struck atoms are added to the cascade if their energy exceeds a displacement threshold E_d. Replacement events occur if a target atom receives energy $> E_d$ in a collision and the projectile is left with energy $< E_d$. The sequence of displacements terminates when the energies of all the displaced atoms have fallen below E_d. The effects of temperature can be approximated by giving each atom a random Gaussian displacement based on the Debye model. Inelastic energy losses are included using a modification of a model of Firsov [12].

The results of these calculations may be summarized briefly. The number of Frenkel defect pairs generated in a cascade is given accurately by the modified Kinchin–Pease formula

$$N_d = \frac{\kappa(E - \hat{Q})}{2E_d} = \frac{\kappa \hat{E}}{2E_d}, \qquad \hat{E} > 2E_d/\kappa, \qquad (3)$$

where $\hat{Q}$ is the total energy lost in the cascade by electron excitation and $\hat{E}$, the damage energy, is available for atomic displacement. Eq. (3) is analogous to the half-Nelson equation (2); $\hat{E}$ corresponds to the factor

$W(E)E$ and the term $\alpha\beta(E)/\gamma(E)$ is replaced by $\kappa/2$ where κ is the displacement efficiency. The factor 2 is included explicitly to emphasize the close similarity to the Kinchin–Pease formula (1). Torrens and Robinson find that κ is a constant independent of energy (except for E near $2E_d$) and is insensitive to the target and the temperature for all materials studied (Cu, Fe, Au and W). Its value is approximately 0.8. The values of $\hat{Q}$ calculated using the Firsov model at each collision are similar in magnitude to the predictions of Lindhard et al. [13] using a model in which the projectile slows down continuously.

2.4. Binary collision simulations using the vacancy capture model

Instead of using the initial kinetic energy of a displaced atom as the criterion for producing a stable Frenkel pair, the distance from a vacancy at which the recoil comes to rest may be used. If this distance is less than r_v, the vacancy capture radius, the pair will be mechanically unstable and annihilate; if the separation exceeds r_v, a stable Frenkel pair will result. This model of defect stability has been used in computer simulations by Beeler [9] and by Robinson and Torrens [11]. The latter calculations have shown that with suitable choices of E_d and r_v, the two models of defect stability are completely equivalent at PKA energies below about 0.5 keV. As the PKA energy increases above this, however, the displacement efficiency decreases continuously, at least up to the limit of the available calculations (20 keV [9] or 10 keV [11]). The decreased displacement efficiency is a result of increased point defect annihilation in the highly disturbed tracks of the most energetic cascade particles, not to interferences between distinct cascade branches [11].

2.5. Cascade simulation by molecular dynamics

Vineyard and his co-workers [14–16] integrated the equations of motion of an assembly of atoms interacting by a model potential and simulated the production and annealing of point defects when a single atom was given a sudden impulse. Because of the computation time required, only small crystals can be studied and the PKA energy cannot exceed approximately 1 keV. Also, extensive statistical analysis of results is difficult. However, since it is the only method not requiring the specification of extraneous parameters such as displacement threshold or capture radius, it is especially important in studying the low energy end of the cascade. In fact, the study of Erginsoy et al. [16] showed that the energy needed to displace an atom in bcc Fe varied considerably with its direction of motion, marked minima occurring near the close-packed lattice directions. This prediction has been confirmed experimentally [17].

3. Comparison of the models

Norgett [18] has shown that the binary collision and molecular dynamics calculations are essentially complementary at energies below 1 keV. He made a number of comparisons between the results of Erginsoy et al. [16] and the predictions of the Torrens–Robinson simulations. Although the interatomic potentials differed slightly, it was possible to obtain reasonable agreement between the binary collision and molecular dynamical models by a suitable choice of the displacement threshold. For bcc iron, the only material for which extensive molecular dynamics results are available, the numbers of Frenkel pairs produced by the two models could be matched using an average displacement threshold of 37 eV. It should be stressed that this is the mean energy necessary to produce a stable vacancy-interstitial pair and is considerably greater than the minimum threshold in a preferred direction. The use of a displacement energy which is derived ultimately from the molecular dynamics approach is important because this method considers explicitly such problems as the athermal recombination of unstable near-neighbor point defect pairs.

An alternative approach would be to use a vacancy capture radius based on molecular dynamic or static calculations of defect pair stability, in conjunction with binary collision calculations. This comparison has not been made, but in view of the close similarities between the threshold and vacancy capture models at low energies, it would undoubtedly give similar results. More important would be the extension of the binary collision calculations to the much higher energies characteristic of neutron damage in fission (~50 keV) and fusion (~500 keV) systems. Such calculations are needed to determine whether decreases in the displacement efficiency continue to occur or whether they are limited by the formation of subcascades or otherwise.

4. Proposed method for estimating the number of displaced atoms

With the above discussion in mind, a simple procedure for estimating displacement dose rates is outlined here:

(a) The modified Kinchin–Pease formula of Torrens and Robinson [10] is used to calculate the number of Frenkel pairs N_d generated by a primary knock-on of initial kinetic energy E:

$$N_d = \kappa \hat{E}/2E_d, \tag{4}$$

where $\hat{E}$ is the energy available to generate atomic displacements by elastic collisions.

(b) The displacement efficiency κ is given the value 0.8, independent of the PKA energy, the target material, or its temperature.

(c) The inelastic energy loss is calculated according to the method of Lindhard et al. [13] using a numerical approximation [19] to the universal function $g(\epsilon)$:

$$\hat{E} = \frac{E}{[1 + k\, g(\epsilon)]}, \tag{5}$$

$$g(\epsilon) = 3.4008\, \epsilon^{1/6} + 0.40244\, \epsilon^{3/4} + \epsilon, \tag{6}$$

$$k = 0.1337\, Z_1^{1/6}\, (Z_1/A_1)^{1/2}, \tag{7}$$

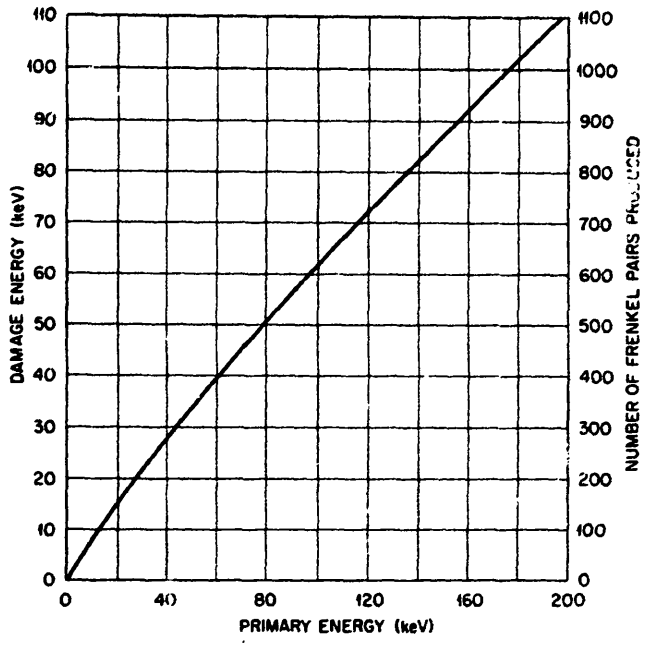

Fig. 1. Plot of damage energy $\hat{E}$ and number of Frenkel pairs N_d produced by a primary knock-on atom of given energy: calculated for iron using the standard method proposed in the text.

$$\epsilon = [A_2 E/(A_1 + A_2)]\, [a/Z_1 Z_2 e^2], \tag{8}$$

$$a = (9\pi^2/128)^{1/3}\, a_0 [Z_1^{2/3} + Z_2^{2/3}]^{-1/2}, \tag{9}$$

where a_0 is the Bohr radius, e the electronic charge, Z_1 and Z_2 are the atomic numbers of the projectile and target and A_1 and A_2 are the mass numbers of the two atoms.

(d) Because of the uncertainty in the value of E_d in eq. (4), it seems appropriate to select a single value to be applied irrespective of the target. If a displacement energy $E_d = 40$ eV is chosen, then the final formula obtained has the particularly simple form

$$N_d = 10\, \hat{E}, \tag{10}$$

where $\hat{E}$ is measured in keV. This value is in good agreement with that deduced for bcc iron (see section 3). Fig. 1 is a plot of both $\hat{E}$ and N_d against PKA energy E for this material.

5. Discussion

The model proposed here has been used in radiation damage studies by Lindhard et al. [20], Piercy [21], Sattler [22], Robinson [19] and Doran [23]. It has also been used by Sigmund [24] in a widely accepted theory of sputtering. A comparison of the present model with the half-Nelson and Kinchin–Pease models is shown in fig. 2.

Some limitations of this model must be pointed out. The Lindhard formulation of eqs (5)–(9) applies strictly only to monatomic systems (i.e. $Z_1 = Z_2$) and to energies less (perhaps much less) than about 25 $Z_1^{4/3} A_1$ keV. The former limitation should not be serious as long as the ratio Z_1/Z_2 does not differ too much from unity. If necessary, it could be relaxed by repeating the Lindhard calculation for other cases. The energy restriction limits the standard in certain possible applications. Neutron damage calculations in light elements such as Be in a fission neutron spectrum or C in a D–T fusion neutron spectrum would be of limited reliability. More seriously, in ion irradiation calculations it is important to generate a suitable target PKA spectrum using experimental stopping power data and the Rutherford scattering cross section. In the same way, it is necessary to include in neutron calculations all appropriate nuclear reactions as well as elastic scattering. It is evident that the cross section data used

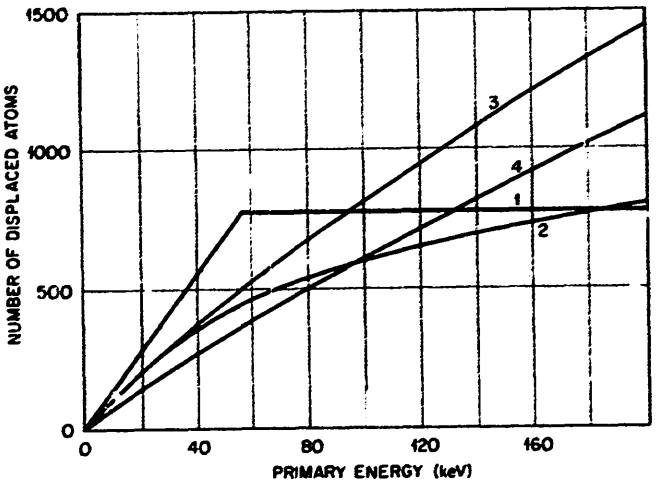

Fig. 2. Comparison of number of displaced atoms generated in bcc iron by a primary knock-on atom. Calculated results correspond to: (1) Kinchin–Pease model with E_d = 40 eV and E_1 = 56 keV; (2) the half-Nelson formula [4]; (3) earlier computer calculations of Norgett [18], using Torrens–Robinson computer simulation program [11]; and (4) the proposed formula, eqs (5)–(10).

should be carefully cited in all radiation damage calculations.

Finally, it is worth mentioning areas needing further study. Possible extensions of the Lindhard energy partition calculation could include not only mass ratio effects, but might also usefully extend the possible energy range, especially for light ions, perhaps through the use of experimental stopping power data. More fundamentally, the whole subject of electronic stopping in the low energy region (i.e. below the limits cited above) still requires satisfactory solution. As noted earlier, extension of cascade simulations to higher energies is needed and these should be combined with studies of cascade structure and its likely importance for point defect annihilation. Another important area for both theoretical and experimental work is that of threshold energies and their dependence, for example, on the crystallographic direction of recoil and on the target temperature. In spite of these limitations and the areas needing further study, it seems to us an appropriate time to put forward the standard proposed above.

6. Conclusion

We have proposed a simple procedure for calculating the number of atomic displacements generated by a primary knock-on atom of given energy. The essential feature of the formula is a linear dependence of the number of displacements on the damage energy. A particularly simple formula, eq. (10), is obtained using a physically reasonable choice of parameters in eq. (4).

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