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ABSTRACT

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.

*Operated by the Union Carbide Corporation for the U. S. Atomic Energy Commission.

†This work performed while M. T. Robinson was on assignment at Kernforschungsanlage, Jülich, West Germany.

1. Introduction

In analysing the behaviour of reactor materials, it is important to know the rate at which the atoms of a solid are displaced. 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 impossible 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 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.

In order 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 behaviour 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 standard 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 the most recent theoretical and experimental developments in the field. The background of a proposed standard will be summarised only briefly,

as the necessary additional details can be found elsewhere (Robinson 1972, Sigmund 1972). However, we shall attempt to indicate areas of uncertainty, where further experimental or theoretical research would be helpful.

2. Physical Models

Four approaches to the problem of calculating the damage generated in a solid by a primary recoil atom of given initial energy will be described briefly in this section. 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.

(a) The Kinchin-Pease Model

Kinchin and Pease (1955) estimated the number of displaced atoms N_d generated by a primary knock-on atom (PKA) of energy E as

$$\begin{aligned} N_d &= E/2\,E_d \qquad 2\,E_d < E < E_I \\ &= E_I/2\,E_d \qquad E_I < E < \infty \end{aligned} \tag{1}$$

where E_d is a sharp threshold energy for displacing an atom from a lattice site. At energies below E_I, the recoils slow down only by elastic hard-core scattering, while above E_I, they lose energy only by electron excitation. Once produced, the defects are permanent, no recombination being allowed. Crystal lattice effects were not considered either in the single atom displacement process or in the cascade development.

(b) The Half-Nelson Model

Nelson (1969) proposed a semi-empirical modification of the Kinchin-Pease model incorporating a number of corrections believed to be important. His formula is

$$N_d = \frac{\alpha \cdot \beta(E) \cdot W(E) \cdot E}{\gamma(E) \cdot E_f} \qquad (2)$$

Here α is a factor introduced to allow for realistic atomic scattering instead of the hard core approximation. Theoretical estimates (Robinson 1965, Sigmund 1969a) suggested a value of α about 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.* (1963a), but the electronic energy losses were confined to the PKA itself.

Development of the cascade was regarded as terminated by the formation of focussed collision sequences (Silsbee 1957); thus the focussing 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 a cascade simulation of Beeler (1966). 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.

(c) Cascade Simulation in the Binary Collision Approximation

Torrens and Robinson (1972a,b) have constructed a computer simulation of collision cascades in which the trajectories of displaced atoms in a given crystal lattice are followed through a series 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 simulated 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 (1959).

The results of Torrens and Robinson may be briefly summarized. The number of defect pairs generated in a cascade is given very accurately by the modified Kinchin-Pease formula

$$N_d = \frac{\kappa(E-E_{Inelas})}{2E_d} = \frac{\kappa E_{Damage}}{2E_d} \qquad (3)$$

where E_{Inelas} is the total energy lost in the cascade by electron excitation and E_{Damage} is the energy available for atomic displacement.

The formula (3) is analogous to the half-Nelson equation (2); E_{Damage} 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 emphasise 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 very insensitive to the target and temperature for materials studied to date (Cu, Fe, Au, W), having a value approximately equal to 0.8. The values of E_{Inelas} calculated using the Firsov model at each collision are similar in magnitude to the predictions of Lindhard *et al.* (1963b) using a model in which the projectile slows down continuously. Equation (3) assumes no close pair recombination within the cascade. The effect of recombination is to reduce the number of defects remaining, although N_d remains directly proportional to E_{Damage}. It is not possible to use the binary collision model to predict absolute numbers of displacements because this would require specification of a displacement threshold and a recombination radius. These parameters are complementary as a small value of E_d coupled with a large recombination radius is in many respects equivalent to a larger value of E_d with no recombination.

(d) Cascade Simulation by Molecular Dynamics

Vineyard and his co-workers (Gibson *et al.* 1960, Erginsoy, *et al.* 1964, 1965) integrated the equations of motion for an assembly of atoms interacting by a model interatomic 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, the size of block of crystal which can be studied is small 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 recombination radius, it is especially important in studying the low energy end of the cascade. In fact, the study of Erginsoy *et al.* (1965), showed that the energy needed to displace an atom in bcc iron varied considerably with its direction of motion, marked minima occurring near the close-packed lattice directions. This prediction has been confirmed experimentally (see, for example, Jung and Schilling, 1972).

3. Comparison of the Models

There is a basic discrepancy between the half-Nelson model and the results of Torrens and Robinson. The recent binary collision calculations show that the displacement efficiency does not decrease at high energy. This view is supported by analytical treatments of cascade theory (Sigmund 1969a, Robinson 1972). The origins of the discrepancy between the calculations of Torrens and Robinson and of Beeler are not known. The absence of a sufficiently detailed description of Beeler's programme makes comparisons difficult. The evidence of both analytical theory and a comprehensive computer simulation supports the conclusion that the displacement efficiency is independent of energy.

Norgett (1971) has recently shown that the binary collision and molecular dinamics calculations are essentially complementary. He made a number of comparisons between the results of Erginsoy *et al.* (1965), 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 choice of a suitable displacement threshold. For bcc iron, the only material for which extensive molecular dynamics results are currently available, the numbers of Frenkel pairs produced by the two models could be matched using an average displacement threshold of 37 eV without any recombination. 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-neighbour point defect pairs. An alternative approach would be to use a displacement energy as low as or lower than the minimum threshold, and to couple this with a recombination radius based on molecular dynamic or static calculations of defect pair stability. In any event, the comparison of the binary collision calculation and the dynamical model provides considerable additional justification for the validity of the binary model even at low energy.

4. A Proposed Method for Estimating the Number of Displaced Atoms

We incorporate the above conclusions into a simple procedure for estimating displacement dose rates.

(a) The modified Kinchin-Pease formula of Torrens and Robinson (1972) is used to calculate the number of Frenkel pairs N_d generated by a primary knock-on of initial kinetic energy E;

$$N_d = \frac{\kappa E_{Damage}}{2E_d} \qquad (4)$$

where E_{Damage} 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.* (1963b), using a numerical approximation to the universal function $g(\varepsilon)$ (Robinson 1970)

$$E_{Damage} = \frac{E}{[1 + k\, g(\varepsilon)]} \tag{5}$$

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

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

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

$$a = \left(\frac{9\pi^2}{128}\right)^{1/3} a_o \left[Z_1^{2/3} + Z_2^{2/3}\right]^{-1/2} \tag{9}$$

where a_o 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 equation (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

$$N_d = 10\, E_{Damage} \tag{10}$$

where E_{Damage} is measured in keV. This value is in good agreement with that deduced for bcc iron (see section 3). Figure 1 is a plot of both E_{Damage} 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.* (1962), Piercy (1968), Sattler (1968), Robinson (1970) and Doran (1972). It has also been used by Sigmund (1969b) in a widely accepted theory of sputtering. A comparison of the present model with the half-Nelson and Kinchin-Pease models is shown in Figure 2.

We should also point out the limitations of the model. The Lindhard formulation of equations (5) to (9) applies strictly only to monatomic systems (i.e. $Z_1 = Z_2$) and to energies 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 either by extensive calculations with the Torrens-Robinson programme, or perhaps more efficiently, 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 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. 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, equation (10), is obtained using a physically reasonable choice of parameters in equation (4).

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Captions to Figures

Figure 1

Plot of Damage energy and number of Frenkel pairs produced by a primary knock-on atom of given energy; calculated for iron using the standard method proposed in the text.

Figure 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_l = 56 keV

(2) The half-Nelson formula

(3) Earlier computer calculations of Norgett (1971), using Torrens-Robinson computer simulation program MARLOWE

(4) The new proposed standard formula.