THE THEORY OF IRRADIATION DAMAGE IN GRAPHITE

B T Kelly
UKAEA Reactor Fuel Element Laboratories, Springfields, Salwick, Preston, Lancashire, UK

Introduction

The low atomic weight of carbon and the open crystal structure of graphite simplifies the irradiation damage process in that atomic displacements may be considered to take place at random at a rate G atoms/atom/sec in a neutron flux ϕ. A number of models of the damage process have been based on this feature, but none have attempted to account for detailed crystal dimensional change data at high doses.

In this paper, a full diffusion theory of the growth of interstitial clusters in graphite is developed based on an earlier model due to Kelly et al.(1)

Theory

It is assumed that N_ℓ interstitial dislocation loops are nucleated at a very small dose ($\sim 10^{19}$ n/cm^2(EDN)) which then proceed to grow by two dimensional diffusion of newly created interstitials through a field of single vacancies, concentration C_v. Vacancies disappear by the formation and growth of collapsed vacancy lines, density N_L cm^{-3}. It is postulated that the density of these lines cannot increase beyond a definite value N_m in the crystal lattice which is independent of irradiation temperature and determined by overlap of anharmonic forces. Vacancies are also allowed to diffuse in the basal planes and disappear at collapsed lines or crystallite boundaries defining a crystallite size L_a. These assumptions lead to the following equations for the interstitial concentration C_i and the single vacancy concentration C_v.

$$C_i = (G/\phi)^2 \frac{\Delta^2}{4} \frac{N_\ell}{R} \left[\int_o^\gamma C_v^{-\frac{1}{2}} . d\gamma\right]^2 \qquad \ldots(1)$$

where R is the interstitial-vacancy recombination probability/collision, Δ is a constant and γ is the fast neutron dose.

$$\frac{dC_v}{d\gamma} = \frac{G}{\phi}\left\{\frac{G}{\phi}\frac{\Delta^2}{2R}\frac{N_\ell}{C_v^{\frac{1}{2}}}\left[\int_o^\gamma C_v^{-\frac{1}{2}} . d\gamma\right] - x\,C_v - \frac{2xN_m}{N_o}\right.$$

$$\left[1-\exp\left(-\frac{G}{\phi}\frac{N_o}{N_m}x\int_o^\gamma C_v . d\gamma\right)\right]\Bigg\} - \left\{\frac{8\,D_{ov}\exp(-E_{mv}/kT)}{\phi}\right.$$

$$\left[\frac{Z_v N_m}{a^2 N_o}\left(1-\exp\left(-\frac{G}{\phi}\frac{N_o}{N_m}x\int_o^\gamma C_v . d\gamma\right)\right) + \frac{1}{L_a}\left(\frac{2Z_v}{N_o a^2}N_m\right)^{\frac{1}{2}}\right.$$

$$\left.\left.\left(1-\exp\left(-\frac{G}{\phi}\frac{N_o}{N_m}x\int_o^\gamma C_v . d\gamma\right)\right)^{\frac{1}{2}}\right\}C_v \qquad \ldots(2)$$

where N_o is the number of atoms/cm^3, $x \simeq 3$, a is the defect jump distance, L_a the crystallite size, D_{ov} the vacancy diffusion constant and E_{mv} the vacancy activation energy.

The crystal dimensional changes due to defects are calculated from

$$\left.\begin{aligned} e_{zz} &= C_i \\ e_{xx} &= -\frac{G}{\phi}\frac{x}{N_o}\int_o^\gamma N_L . d\gamma - \frac{1}{2}\int_o^\gamma\left[\left(\frac{dC_v}{d\gamma}\right)_{line}\right. \\ &\left. + \left(\frac{dC_v}{d\gamma}\right)_{boundaries}\right] . d\gamma \end{aligned}\right\} \qquad \ldots(3)$$

where N_L is the number of vacancy lines/cm^3.

$(dC_v/d\gamma)_{line}$ is the rate of vacancy loss at lines due to diffusion.

$(dC_v/d\gamma)_{boundaries}$ is the rate of vacancy loss at boundaries due to diffusion.

Comparison of Theory and Experiment

A detailed comparison of theory and experiment is made over the range 300°C to 1200°C using data obtained in irradiations in DFR and the DIDO-PLUTO Materials Testing Reactors. The theory, using appropriate values of the parameters gives good agreement with the crystal dimensional change up to doses $\sim 10^{22}$ n/cm (EDN) and also gives concentrations of uncollapsed vacancies quite close to experimental measurement. At higher doses it is found that there are far too many uncollapsed vacancies and it is clear that recombination with interstitials with a low probability is necessary to improve agreement at very high doses.

The theory has also been compared with crystal dimensional changes in boron doped (B^{11} to avoid additional displacement damage) polycrystalline and pyrolytic samples irradiated at 430°C and 600°C in DIDO/PLUTO. The general behaviour is predicted quite accurately using loop densities taken from electron microscope studies.

References

(1) Kelly B T, Martin W H, Prince A and Bland J T. Phil. Mag. Vol 14, No. 128, p.343. (1966).