Lattice Parameter and Dimensional Changes in Graphite Irradiated between 300 and 1350°C

by

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Recent studies of irradiation damage in graphite have related the lattice parameter and dimensional changes to the properties of the irradiation defects. (Henson and Reynolds, 1965). This paper extends the work to cover the effect of irradiation at higher temperatures, where the behaviour is complicated by the effects of thermal motion of the vacant lattice sites.

In the low temperature region the problem can be considered in two steps, the effect of the irradiation defects on the lattice parameters and dimensions of the crystals and the relation between the dimensional changes of the crystals and those of polycrystalline aggregates. The second step is dealt with by the growth equations (Perks and Simmons, 1965),

$$g_\perp - g_\parallel = f(x_T) \qquad \ldots\ldots\ldots\ldots (1)$$

$$\frac{\Delta V}{V} = \left(\frac{\Delta V}{V}\right)_x + F(x_T) \qquad \ldots\ldots\ldots\ldots (2)$$

where $g_\parallel$ and $g_\perp$ are the dimensional changes parallel and perpendicular to extrusion, $\frac{\Delta V}{V}$ is the volume change of the graphite aggregate and $\left(\frac{\Delta V}{V}\right)_x$ the volume change of the crystal. $x_T = g_c - g_a$, where g_c and g_a are the dimensional changes of the crystals in the a and c directions. $F(x_T)$ is the volume change due to closure or generation of pore space. A relation between $g_\perp - g_\parallel$ and x_T has been given, (Kelly, Martin and Nettley, 1966). Using a development of the treatment of Henson and Reynolds, an expression for $\left(\frac{\Delta V}{V}\right)_x$ can be obtained

$$\left(\frac{\Delta V}{V}\right)_x = n_v - n_i + \frac{\Delta c}{c} + 2\frac{\Delta a}{a} \qquad \ldots\ldots\ldots\ldots (3)$$

where $\frac{\Delta c}{c}$ and $\frac{\Delta a}{a}$ are the changes in lattice parameters due to concentrations n_i and n_v of interstitial atoms in small clusters and vacant lattice sites.

At higher temperatures the situation is complicated by the effect of the motion of vacancies, which may lead to the formation of vacancy loops. Vacancy loops have been identified in graphite crystals irradiated at 900 and 1350°C; in crystals irradiated at 650°C there is in addition to the interstitial loops, a population of defects which are presumed to be vacancy loops. These observations suggest that vacancy loops are formed in artificial graphite when the temperature is high enough for the vacancies to be mobile. Vacancy movement may also effect the volume change due to the generation of pore space at high crystal strains. To allow for this effect equation 2 is replaced by

$$\frac{\Delta V}{V} = \left(\frac{\Delta V}{V}\right)_x + F(x_T) + \Delta F \qquad \ldots\ldots\ldots\ldots (4)$$

At very high temperatures (>1000°C) the mobility of the vacancies will be sufficient for them to reach crystal boundaries, at which point it is no longer possible to describe the dimensional changes simply in terms of irradiation defects in the lattice.

In the present work, lattice parameter changes have been measured on graphite irradiated in the range 300 to 1350°C in the reactor PLUTO and dimensional changes have been measured at 900°C and 1350°C (data for the other temperatures have been obtained in earlier work). Measurements of the same properties were also made on specimens irradiated in the Dounreay Fast Reactor (D.F.R.) in which the damage is produced about 20 times faster than in PLUTO. In some cases temperature measurements were possible. These enable the changes in the two reactors to be correlated so that the equivalent temperatures in D.F.R. can be evaluated and the appropriate activation energy determined. The activation energy varies from 1.2 eV at low temperatures to about 2.0 eV at 450°C.

The results of the lattice parameter measurements show that the effect of the interstitial clusters is small. It is also observed that the ratio $\frac{\Delta a}{a} / \frac{\Delta c}{c}$ is constant so that equation 3 can be replaced by

$$\left(\frac{\Delta v}{v}\right)_x = A \frac{\Delta a}{a} \qquad \cdots\cdots\cdots (5)$$

The experimental data together with equations 1, 2 and 5 gives $A = -7.5 \pm 0.5$ and enable $F(x_T)$ to be determined. This analysis applies at temperatures below that at which vacancies are mobile (450°C). At higher temperatures it is necessary to make allowances for vacancy loops so that equation 5 becomes

$$\left(\frac{\Delta v}{v}\right) = -7.5 \frac{\Delta a}{a} + v_1 \qquad \cdots\cdots\cdots (6)$$

where v_1 is the number of vacancies in the form of uncollapsed vacancy loops.

Equations 1, 4 and 6 enable the quantity $v_1 + \Delta F$ to be determined as a function of $g_\perp - g_\parallel$ and hence x_T. For small values of x_T it is expected that $\Delta F = 0$ so that v_1 is determined. It is found that at high strains $v_1 + \Delta F$ is greater for the anisotropic PGA than for isotropic graphite. Since it is known (Perks and Simmons, 1965) that pore generation at high strains is more pronounced in anisotropic graphite it is concluded that $\Delta F > 0$ in this case. Probably $\Delta F \sim 0$ for isotropic graphite so that v_1 can be estimated.

Having determined from this analysis the concentrations of the various types of defect present an attempt is made to formulate the kinetics of the various competing processes which take place.

References: Henson, R.W. and Reynolds, W.N., 1965, Carbon, Vol. 3, pp.277-287.
            Perks, A.J. and Simmons, J.H.W., 1966, Carbon, Vol. 4, pp.85-98.
            Kelly, B.T., Martin, W.H. and Nettley, P.T., Phil. Trans. Roy. Soc. A.,
               1966, Vol. 260, p.51.