

CHAPTER 1

FUNDAMENTALS OF RADIATION DAMAGE IN GRAPHITE DUE TO ENERGETIC NEUTRONS

Study of the basic processes which produce the property and dimensional changes known as radiation damage due to energetic neutrons is necessary for two distinct reasons:

- (i) The number and distribution of displaced atoms and transmutation products are required for theories of the development of the defect structure and prediction of the consequent property and dimensional changes.
- (ii) The use of graphite components in fission and fusion reactor systems requires predictions of dimensional and physical property changes using data obtained under different conditions of neutron spectrum and temperature.

The source neutrons produced by nuclear fission extend over a wide range of energies, from a small fraction of an eV to ~10MeV, with a mean energy of ~2MeV. The fusion of tritium and deuterium atoms, which is likely to be the basis of fusion power generation systems, produces source neutrons with energies of 14.1MeV. In every practical case the source neutron spectra are degraded by elastic and inelastic scattering in the reactor structure and it is necessary to consider spatially varying neutron spectra. The neutron flux with energies in the range E_n to $E_n + dE_n$ is defined to be $\phi(E_n) dE_n$ at a field point \vec{R} .

In this chapter we consider the simplest model of the process of atomic displacement in graphite in fission neutron spectra, the comparison of calculated displacement rates with experiment, and the extension of these ideas to fusion neutron systems. Transmutation does not play any significant role in property and dimensional changes of graphite in fission neutron systems, but this is not certain for the higher energies of fusion neutrons.

The neutron induced fissions of the isotopes of uranium and plutonium which are of practical importance produce on average ~3 neutrons/fission with an energy distribution which may, for example, be described by the Watt-Cranberg expression:

$$X(E_n)dE_n = K \exp(-AE_n) \sinh(BE_n)^{1/2} dE_n \quad (1.1)$$

where K , A and B are constants. The current values of these parameters are:

$$\begin{aligned} A &= 1.0123 \text{ MeV}^{-1} \\ B &= 2.189 \text{ MeV}^{-1} \\ K &= 0.453 \end{aligned}$$

with a mean energy of

$$E = (3 + B/2A)/2A = 2.0158 \text{ MeV}$$

In both the fission and fusion cases the neutron spectra in particular materials may be calculated using modern Monte Carlo or diffusion theory based computer codes.

The process of creation of crystal lattice defects, which produce the changes in properties and dimensions of graphite, is due to the scattering of the energetic neutrons by the nuclei of the carbon atoms either elastically or inelastically. The binding energy of a carbon atom in the graphite crystal lattice is $\sim 7\text{eV}$ and it is reasonable to suppose that the transferred energy from the neutron to the nucleus which will irreversibly displace the lattice atom from its site (primary displacements) will be a few times this value, and may be expected to depend upon the direction in which the struck atom is propelled. This energy is denoted by E_d and is assumed to be a function of Q , the angle between the direction of motion and the crystallite hexagonal axis. In fact, the two pairs of atoms in the unit cell do not have identical environments (see Fig 1.1) and this may complicate the variation of E_d . The maximum energy which can be transferred to a nucleus of atomic weight A by a neutron of energy E_n in a purely elastic collision is:

$$\Delta E_n = \frac{4A}{(A+1)^2} E_n \quad (1.2)$$

which for $^{12}\text{C}_6$ is:

$$\Delta E_n = 0.284 E_n \quad (1.3)$$

Equation (1.3) shows that a large proportion of neutron energy is transferred in a collision between a neutron and a carbon nucleus (which, together with its low cross-section for neutron absorption, accounts for its use as a reactor moderator). In both fission and fusion systems, neutrons with energies greater than about 100eV produce displacements, and the displaced atoms with greater energies produce further displacements in collisions with other carbon atoms in a cascade. Neutron-nucleus collisions may be treated as elastic for neutron energies up to 5.5MeV in carbon. The cross-section for the (n,a) reaction becomes appreciable for neutron energies above $\sim 9\text{MeV}$, and it is also necessary to account for anisotropic scattering.

The energy E_d required to produce displaced atoms and the consequent vacant lattice sites in graphite can be measured using radiation of well-defined energy and interaction and finding the minimum energy necessary to produce a measurable property change. The first measurement was due to Eggen (1950) using electrons of controlled energy ($\sim 1\text{MeV}$) and finding the minimum energy to give a change in the electrical resistivity in a polycrystalline graphite; he found $E_d = 24.7 \pm 0.9\text{eV}$, presumably the lowest value. The variation of E_d with Q can be obtained using highly oriented graphite samples, such as natural graphite flakes or highly oriented pyrolytic graphite, bombarded at various angles to the hexagonal axis. Lucas and Mitchell (1964) measured the effect of electron irradiation with energies between 0.3 and 2MeV on the electrical resistivity of natural graphite crystals - the displacement energy was found by fitting curves of damage rate as a function of electron energy. The value of 60eV obtained presumably corresponds to the value for $Q = 0$ (i.e. beam parallel to the hexagonal axis). Ohr *et al* (1972) observed the onset of visible damage in transmission electron microscopy and obtained a value of $E_d = 24\text{eV}$ for $Q = 0$, for sample temperatures in the range $300 - 600^\circ\text{C}$. Montet (1967) and Montet and Myers (1971) determined the minimum electron energy to create sites which could subsequently be etched to become visible in the electron microscope. Values of 33eV and 31eV were obtained for the beam parallel to the hexagonal axis. Rotation of the beam about the hexagonal axis gave the same values for E_d until $Q = 60^\circ$, when the value increased reaching a value of 60eV at an

Space Group: $C6/mc(D_{6h}^4)$
Unit cell:
 $\underline{a} = 2.4612 \pm 0.0001 \text{ \AA}$
 $\underline{c} = 6.7079 \pm 0.0007 \text{ \AA}$
Volume = 35.190 \AA^3
Atoms per unit cell = 4
Crystal density = 2.266 g/cm^3

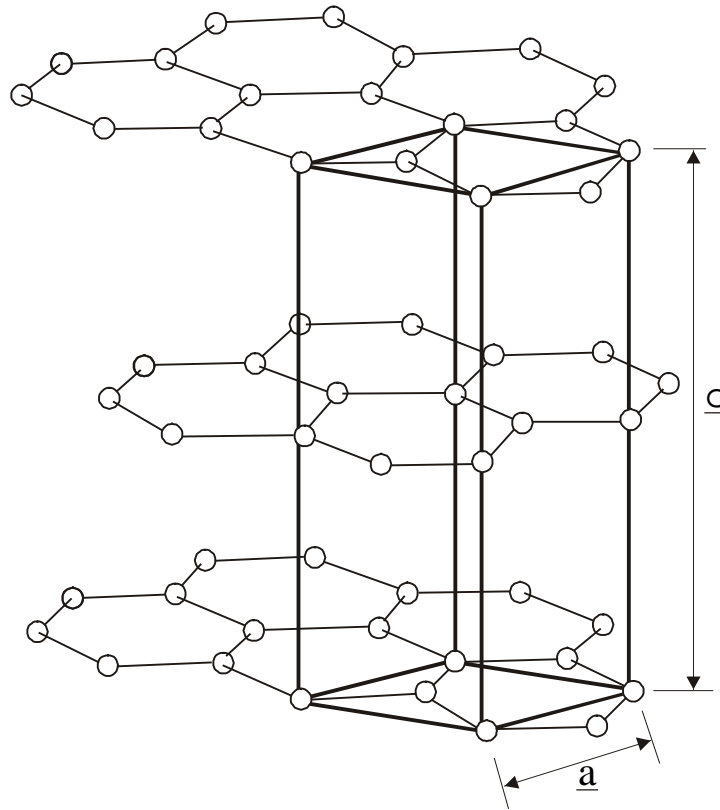


Figure 1.1 Unit cell of Graphite

angle $Q = 80^\circ$. Iwata and Nihira (1966, 1971) bombarded highly oriented pyrolytic graphite with electrons at 6, 80 and 285K. In each case the initial rate of change of electrical resistivity parallel to the deposition plane was measured and the results fitted using a model which allowed for secondary displacements caused by the primary displaced atom. The displacement energy as a function of Q was expressed as:

$$E_d = A \cos^2 Q + B \sin^2 Q + C(1 - \cos 4Q) \quad (1.4)$$

where A , B and C are constants. Values of A varied from 23 - 32eV and B from 30 - 42eV, increasing with irradiation temperature. C varied between 0 and -2eV. Evidence was obtained for different values of E_d for different atoms in the unit cell.

The situation regarding the displacement energy and its variation with crystallographic direction is not satisfactory. Throrer and Mayer (1978) concluded that a value of 40eV is appropriate for irradiation in an isotropic electron flux. The value of E_d currently used in the United Kingdom is 60eV, which is certainly too high. However the calculated atomic displacement rates are essentially inversely dependent on E_d , so that relative damage rates are independent of E_d (Wright, 1962), which is the situation of practical interest where data taken in one neutron spectrum are required to predict effects occurring in a different spectrum. The value of the displacement energy is important in comparison of theoretical models of the radiation damage process with data. It is possible that E_d depends upon the temperature, increasing with temperature, and this needs to be included in theoretical models (Iwata and Nihira, 1966, 1971).

A number of theoretical models have been used to estimate the number of atomic displacements produced by a collision of a neutron of energy E_n and a carbon nucleus, which then permits calculations of the number of atomic displacements in a neutron spectrum, knowing the cross-sections for energy transfer. The most comprehensive calculations, which also consider the spatial distribution of atomic displacements, have been presented by De Halas (1962) and Simmons (1965). We follow the latter in most of the ensuing discussion.

The rate of atomic displacement in a neutron spectrum $\phi(E_n)$ is given by:

$$\frac{dN}{dt} = \phi(E_n) \Omega(E_n, E_p) \nu(E_p) dE_n dE_p \quad (1.5)$$

where N is the fraction of atoms displaced, $\Omega(E_n, E_p)$ is the cross-section for a neutron of energy E_n to produce a carbon atom recoil with energy E_p , and $\nu(E_p)$ is the number of atomic displacements due to a knock-on primary displacement of energy E_p . If $\bar{\nu}(E_n)$ is the average number of displacements produced by a collision with a neutron of energy E_n , then equation (1.5) can be written:

$$\frac{dN}{dt} = \phi(E_n) \sigma(E_n) \bar{\nu}(E_n) dE_n \quad (1.6)$$

where $\sigma(E_n)$ is the elastic scattering cross-section of the carbon nucleus for neutrons, illustrated in Fig 1.2 and listed in Table 1.1. This assumption is satisfactory for fission neutrons, where the mean energy of the source neutrons is ~2MeV and only a very small proportion have energies greater than 10MeV. The source neutrons in deuterium-tritium

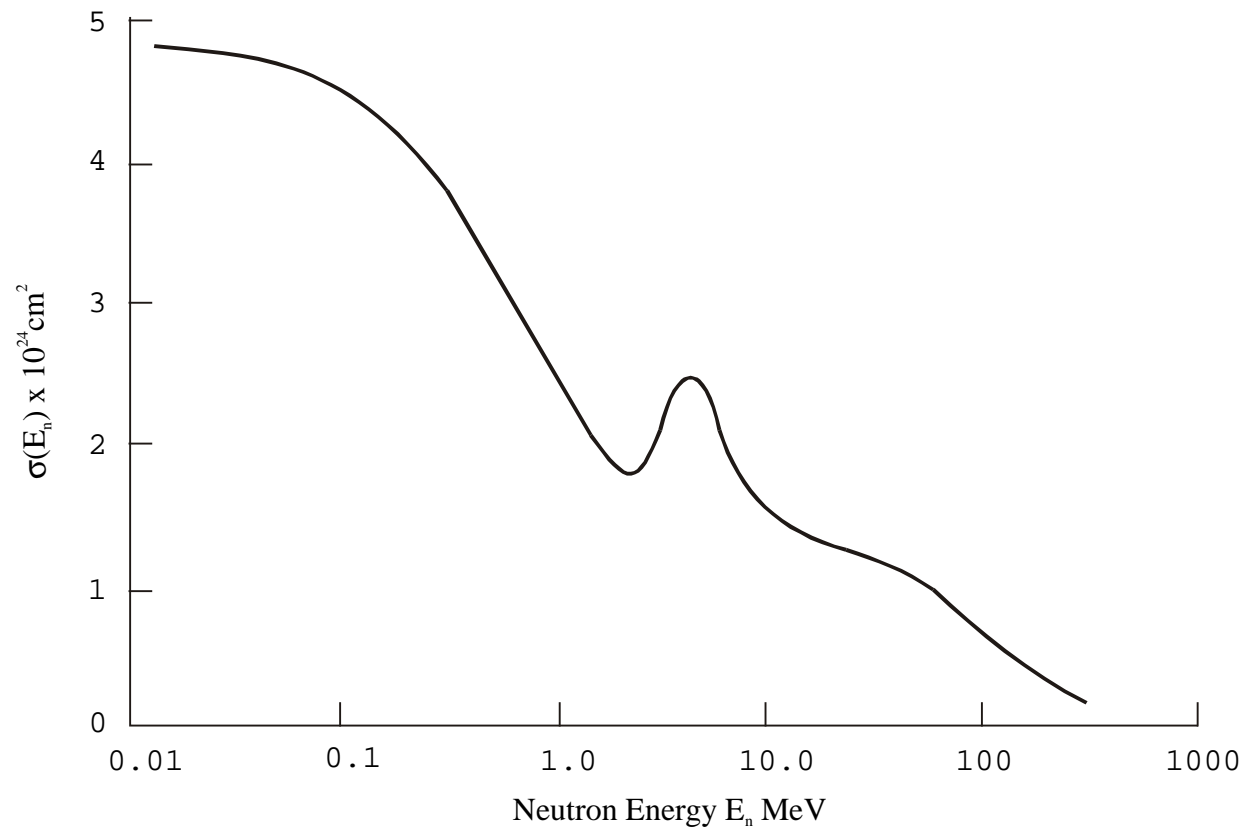


Figure 1.2 Neutron elastic scattering cross-section

systems have an energy of 14.1MeV and thus it is necessary to account for anisotropic and inelastic scattering.

The first model used to estimate the atomic displacement rate in fission systems was due to Kinchin and Pease (1955). It is assumed that the primary neutron-carbon atom collisions are isotropic, each collision transferring the average energy transfer, that is:

$$\Delta E_n = E_p = \frac{1}{2} \left| \frac{4A}{(A+1)^2} \right| E_n = \frac{1}{2} \alpha E_n \quad \text{say,} \quad (1.7)$$

where A is the atomic weight of carbon. It is a good approximation for the energies relevant to fission reactors. The primary knock-on carbon atom loses energy by two separate mechanisms, the first by collisions with other carbon atoms and the second to the electrons. The latter is dominant at the higher knock-on energies because the majority of the knock-on atoms are ionised. The degree of ionisation decreases with the velocity of the atom until the rate of energy loss is dominated by atom-atom collisions. Kinchin and Pease, as an approximation, assume that for moving atom energies greater than L_c , all of the energy loss above L_c is electronic in character, while for lower energies it is solely due to atom-atom collisions. A rough estimate for L_c is A keV. The atom-atom collisions may be treated to a first approximation as between billiard balls with equal masses undergoing isotropic scattering in the centre of mass system for a monatomic lattice. These assumptions lead to:

$$\begin{aligned} \frac{L_c}{2E_d} & \dots\dots\dots E_p \geq L_c \\ \frac{E_p}{2E_d} & \dots\dots\dots 2E_d \leq E_p \leq L_c \\ v(E_p) = & \dots\dots\dots \\ 1 & \dots\dots\dots E_d \leq E_p \leq 2E_d \\ 0 & \dots\dots\dots 0 \leq E_p \leq E_d \end{aligned} \quad (1.8)$$

which in turn leads to:

$$\begin{aligned} \frac{L_c}{E_d} \left| 1 - \frac{L_c}{\alpha E_n} \right| & \dots\dots\dots E_n \geq \frac{2L_c}{\alpha} \\ \frac{\alpha E_n}{4E_d} & \dots\dots\dots \frac{4E_d}{\alpha} \leq E_n \leq \frac{2L_c}{\alpha} \end{aligned}$$

Neutron Energy Group	Upper Energy (MeV)	ENDF-IV Elastic Scattering Cross-Section for Graphite (barns)	Thompson and Wright Displacement Weighting Function
22	1.83	1.85	359.4
23	1.65	1.97	349.1
24	1.50	2.10	338.7
25	1.35	2.23	328.2
26	1.22	2.35	317.6
27	1.11	2.49	306.9
28	1.00	2.62	296.1
29	0.907	2.75	285.2
30	0.821	2.87	274.2
31	0.743	2.99	262.9
32	0.672	3.11	251.5
33	0.608	3.22	240.5
34	0.550	3.32	229.2
35	0.498	3.43	218.1
36	0.450	3.52	207.3
37	0.408	3.62	196.7
38	0.369	3.70	186.3
39	0.334	3.78	176.1
40	0.302	3.86	166.2
41	0.273	3.93	156.5
42	0.247	4.00	147.0
43	0.223	4.06	137.8
44	0.202	4.12	128.8
45	0.183	4.17	120.0
46	0.166	4.22	111.5
47	0.150	4.26	103.2
48	0.136	4.30	95.2
49	0.123	4.34	87.4
50	0.111	4.40	74.8

Neutron Energy Group	Upper Energy (MeV)	ENDF-IV Elastic Scattering Cross-Section for Graphite (barns)	Thompson and Wright Displacement Weighting Function
51	0.0865	4.47	61.2
52	0.0674	4.53	49.7
53	0.0525	4.57	40.2
54	0.0409	4.61	32.3
55	0.0318	4.63	25.9
56	0.0248	4.65	20.7
57	0.0193	4.67	16.5
58	0.0150	4.68	13.1
59	0.0117	4.69	10.3
60	0.00912	4.70	8.0
61	0.00710	4.71	6.2
62	0.00553	4.71	4.8
63	0.00431	4.71	3.7
64	0.00335	4.72	2.9
65	0.00261	4.72	2.3
66	0.00203	4.72	1.8
67	0.00158	4.72	1.4
68	0.00123	4.72	1.1
69	0.000961	4.72	0.8
70	0.000749	4.73	0.6
71	0.000583	4.73	0.5
72	0.000454	4.73	0.4
73	0.000353	4.73	0.3
74	0.000275	4.73	0.2
75	0.000214	4.73	0.1

The Kinchin-Pease formulae were used by Wright (1962) to calculate the atomic displacement rates in a number of reactor neutron spectra, obtained using Monte Carlo methods, for varying values of L_c . A comparison of these results with relative measured rates of change of graphite properties in the reactor locations for which the spectra had been obtained indicated a value of $L_c = 25\text{keV}$, but the relative damage rates are very insensitive to

the value. A complete theory of radiation damage requires knowledge of the spatial distribution of displaced atoms as well as their numbers. This aspect has been considered by Simmons (1965) as follows.

A detailed treatment of a collision between a moving carbon atom and a lattice atom performing thermal vibrations would need to account for the interactions between the electrons and nuclei of the moving atoms and the lattice atoms. As an approximation, collisions between free atoms interacting with a potential $V(r)$, where r is the separation, are considered with a separate treatment of the electronic energy loss. The approximation should be adequate provided that the mean distance between displacement collisions is large compared to the interatomic distances in the crystal lattice, which is satisfied in the case of graphite.

The potential chosen for collisions between similar atoms was that due to Bohr:

$$V(r) = \frac{Z^2 e^2}{r} \exp\left| -\frac{r}{a} \right| \quad (1.10)$$

Z is the atomic number of the atom involved in the collisions, e is the electronic charge and a is the "screening radius". The parameter values for carbon atoms are:

$$Z^2 e^2 = 520 \times 10^{-8} \quad \text{cm. EV}$$

$$a = 0.207 \times 10^{-8} \quad \text{cm}$$

The classical collision diameter is defined as:

$$b = \frac{2Z^2 e^2}{T_1} = \frac{1040 \times 10^{-8}}{T_1} \quad \text{cm} \quad (1.11)$$

where T_1 is the kinetic energy of the moving atom before the collision. The collisions are most conveniently considered in relative co-ordinates where the centre of mass is at rest. If q is the angle of deflection in this co-ordinate system, then the energy transferred to the struck atom is

$$T_2 = T_1 \sin^2 \left| \frac{\theta}{2} \right| \quad (1.12)$$

Classical theory is adequate for the calculation of atomic displacements, and thus the differential collision cross-section may be written

$$d\sigma_a = 2\pi p dp \quad (1.13)$$

where p is the "impact parameter", defined as the perpendicular distance from the centre of the struck atom to the line of motion of the moving atom. A relationship between p and a and hence p and T_2 can be obtained. The differential cross-section can then be expressed as

$$d\sigma_a = W_a(T_2)dT_2 \quad (1.14)$$

At energies $T_1 > \sim 50\text{keV}$ the scattering follows the Rutherford law (valid for $\theta > b/a$)

$$d\sigma_a = \frac{\pi Z^4 e^4}{T_2^2 T_1} dT_2 \quad (1.15)$$

The appropriate value of equation (1.15) for carbon atoms is

$$d\sigma_a = \frac{8.46 \times 10^{-11}}{T_2^2 T_1} dT_2 \quad \text{cm}^2 \quad (1.16)$$

with T_1 and T_2 in electron volts.

The lower energy collisions were treated using the results of Everhart *et al* (1955) who employed classical theory and the Bohr potential (equation (1.10)) to calculate the differential cross-sections. Table I.2 presents Simmons' results based on their calculations.

Equation (1.15) and Table 1.2 show that at high energies the probability is greater that the energy transferred is a small fraction of the initial energy of the moving atom, while at lower energies there is a tendency towards an equal distribution between moving and struck atoms. Simmons (1965) reviewed the theories then available. Qualitatively, the moving atom can be regarded as a charged particle with average charge qZe where q depends upon the relative values of the cross-sections for electron capture and loss (ignoring the loss of energy by charge exchange amounting to $\sim 20\%$ for the slower moving atoms). Knipp and Teller (1941) have estimated q from a statistical model of the atom rather than attempt the computation of the capture and loss electron cross-sections. The work gives a relationship between q and $(v/v_0)Z^{2/3}$ where $v_0 = 2\pi e^2/h$ is the velocity of an electron in the hydrogen atom ground state. It was found that q varies from 0.6 at 10^5eV to ~ 2 at 10^6eV . The energy loss per atom for $v > v_0$ and $v > qZv_0$ may then be obtained using the relationship due to Mott and Massey (1949). However, data were available on the rate of energy loss of carbon ions in graphite due to Porat and Ramavataram (1961) and Ormrod and Duckworth (1963). The two sets of data join smoothly and this was used by Simmons to calculate the ranges of moving atoms of various energies. The energy of the moving atom at which the rate of energy loss due to collisions is equal to that due to the electronic system was found to be $\sim 12\text{keV}$ (which is an estimate for L_c in the Kinchin-Pease model). The number of displaced atoms was assumed to be given by equation (1.8), and the energy lost to lattice vibrations T_L is estimated as that transferred in collisions which transfer energy less than E_d . The results of these calculations are given in Table 1.3.

The calculations permit visualisation of the events following the creation of a primary knock-on. Table 1.3 shows that when the primary energy is between 10^3eV and 10^6eV the mean distance between displacement collisions is large compared to the interatomic spacing,

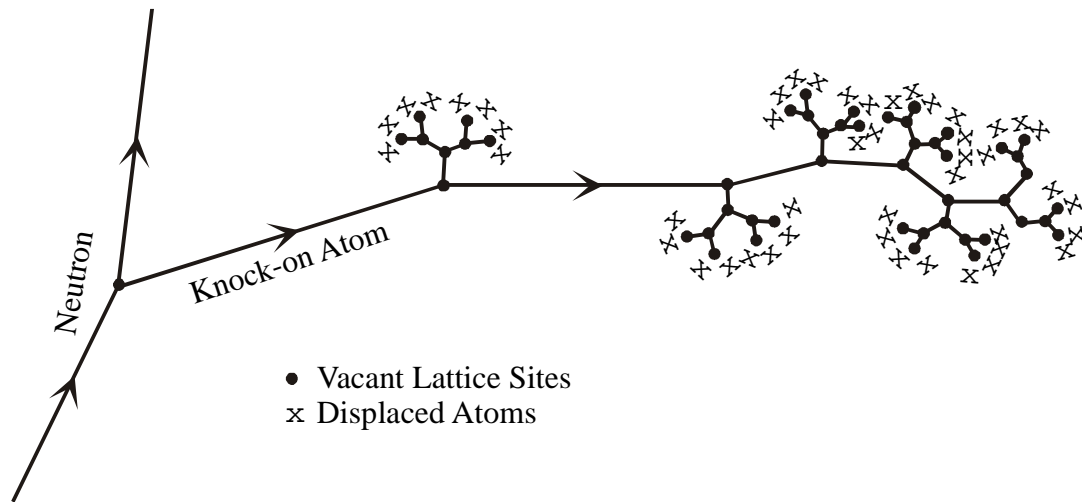


Figure 1.3 Distribution of displaced atoms and vacant lattice sites

TABLE 1.2. LOW AND MEDIUM ENERGY ATOMIC COLLISIONS

Energy of Moving Atom T_1 (eV)	Energy Transferred T_2 (eV)	$W_a(T_2)$ [$\text{cm}^2 \text{eV}^{-1}$]	Energy of Moving Atom T_1 (eV)	Energy Transferred T_2 (eV)	$W_a(T_2)$ [$\text{cm}^2 \text{eV}^{-1}$]
500	25	1.5×10^{-18}	10,000	25	7.2×10^{-19}
	100	3.0×10^{-19}		2000	1.0×10^{-21}
	200	1.3×10^{-19}		4000	2.9×10^{-22}
	300	8.0×10^{-20}		6000	1.4×10^{-22}
	400	5.6×10^{-20}		8000	8.1×10^{-23}
	500	4.2×10^{-20}		10,000	5.4×10^{-23}
1000	25	1.3×10^{-18}	25,000	25	5.2×10^{-19}
	200	1.0×10^{-19}		5000	1.0×10^{-22}
	400	3.9×10^{-20}		10,000	2.6×10^{-23}
	600	2.2×10^{-20}		15,000	1.2×10^{-23}
	800	1.5×10^{-20}		20,000	6.8×10^{-24}
	1000	1.1×10^{-20}		25,000	4.3×10^{-24}
2500	25	1.0×10^{-18}	50,000	25	4.0×10^{-19}
	500	1.9×10^{-20}		10,000	1.5×10^{-23}
	1000	6.8×10^{-21}		20,000	3.8×10^{-24}
	1500	3.6×10^{-21}		30,000	1.7×10^{-24}
	2000	2.2×10^{-21}		40,000	9.4×10^{-25}
	2500	1.6×10^{-21}		50,000	6.0×10^{-25}
5000	25	8.3×10^{-19}			
	1000	4.6×10^{-21}			
	2000	1.5×10^{-21}			
	3000	7.4×10^{-22}			
	4000	4.5×10^{-22}			
	5000	3.0×10^{-22}			

and over this range of energy the average energy of the secondary knock-on is less than 500eV. As a consequence, the damage produced by the primary knock-on consists of separate groups of displaced atoms. This is illustrated in Fig 1.3. It is a reasonable approximation to assume that individual displacements occur at random in theoretical models of the creation of the more complex defects formed from the displacements.

Each of the groups contains less than an average of ten displacements, and is known as "displacement groups". The range of the primary knock-on atom varies from about 10^{-6} cm at 10^3 eV to 10^{-4} cm at 10^6 eV. When the primary energy falls below ~ 500 eV each knock-on produces one displacement group. The collisions within the displacement groups can be regarded as separate events until the energy falls to about 100eV, at which the collision separation becomes comparable with the interatomic spacing. At this point some interstitial atoms will spontaneously recombine with vacancies and some multiple vacancies may be produced. Computer studies (see later) show that multiplication of the total number of

displaced atoms by a constant factor allows for the first of these effects. No attempts have been made to estimate the fraction of multiple vacancies.

The lattice structure of graphite favours channelling of displaced atoms into the large interlayer spacing and this will have the effect of spreading the displacement groups over a larger volume and increase the probability of collisions with atoms in interstitial positions. It has also been suggested that focused collisions may occur in $\{11\bar{2}0\}$ lattice directions. The lack of high quality single crystals of any size has precluded detailed experimental studies of channelling and focusing effects.

TABLE 1.3. DETAILS OF DISPLACEMENT COLLISIONS IN GRAPHITE

Energy T_1 of Moving Atom (eV)	Energy lost/Collision of Primary Knock-on (eV)			Mean Path between Displacement Collisions** (cm) x 10^8	Approximate Range of Primary Knock-on (cm) x 10^8
	Energy lost to lattice vibrations	Energy transferred	R^*		
	T_L	T_2			
500	11	136	-	9.8	-
1000	12	196	-	11.0	67
5000	13	368	200	18.0	210
10,000	14	415	405	22.5	250
50,000	18	424	1600	45.2	1300
100,000	22	382	3100	64.6	2350
500,000	39	297	15,140	173.0	7900
1,000,000	50	282	32,800	285.0	13,650

R^* is the amount of energy lost by electronic excitation between successive displacement collisions.

** The mean free path is calculated from $(N_0 s_d)^{-1}$ where N_0 is the number of carbon atoms/unit volume and s_d is the cross-section for displacement of a moving atom of energy T_1 .

Simmons provided a method of calculating the number of atomic displacements in carbon and their spatial distribution in fission reactor neutron spectra. Calculations of the atomic displacements in fusion reactor spectra are, as already noted, more difficult because of the higher energy source neutrons, but in fact for many realistic spectra in such systems substantial degradation in energy has occurred and the methods may with some care be adequate in these cases.

Examination of the neutron cross-section as a function of energy in the Evaluated Nuclear Data File (ENDF) (see Fig 1.2 and Table 1.1 for values) shows that the elastic scattering cross-section for neutrons in carbon is the same as the total scattering cross-section for neutrons of energies up to 2MeV (the energy of the peak in the fission spectrum) and the

scattering is isotropic in the centre of mass system up to an energy of 0.1MeV. For neutron energies of greater than 2MeV the scattering cross-section shows many resonances and the effects of inelastic and anisotropic scattering become important. It is clear that relative atomic displacement rates in different fission reactor spectra will be quite accurate.

Prior to consideration of improved methods of calculating atomic displacement rates for fission and fusion reactor spectra, it is necessary to compare the results with experiment and decide whether they are useful. The fundamental technological reason for such calculations is, after all, the prediction to acceptable accuracy of the dimensional changes and property changes of graphite in different neutron spectra.

The changes in a graphite property P_i may be written

$$R_i = \frac{1}{P_i} \frac{dP_i}{dt} \propto \int_0^{\infty} \phi(E_n) \psi_i(E_n) dE_n \quad (1.17)$$

where $\psi_i(E_n)$ is a weighting function for the effect of a flux of neutrons of energy E_n on the property P_i . If ψ_i is independent of property then the damaging power, that is the ability to change properties, becomes unique and an extremely useful concept. (In principle ψ_i may include transmutation as well as classical atomic displacement effects.) It can readily be imagined that ψ_i could depend upon the property considered, obvious possibilities being the number of displaced atoms, the number of displacement groups or perhaps the number of closely spaced groups. Fortunately, as we shall see, in fission neutron systems the number of displaced atoms is an adequate approximation to relate the damaging power of different neutron spectra.

If the distribution of the displaced atoms were truly random then equation (1.17) would be sufficient to characterise the damaging power of the spectrum of neutrons. However the distribution of displacements is not truly random and, following Simmons, it is necessary to consider more carefully the meaning of "damaging power". If identical specimens of graphite are irradiated for the same time at the same temperature in two different neutron spectra, and then show identical property changes, then the damaging powers of the two spectra are the same, even if the neutron spectra are different. This definition of damaging power is useful only if the equality of the property changes is maintained whatever the duration or temperature of irradiation.

If the damaging power may be represented by equation (1.17) then it must be possible to write the damage flux

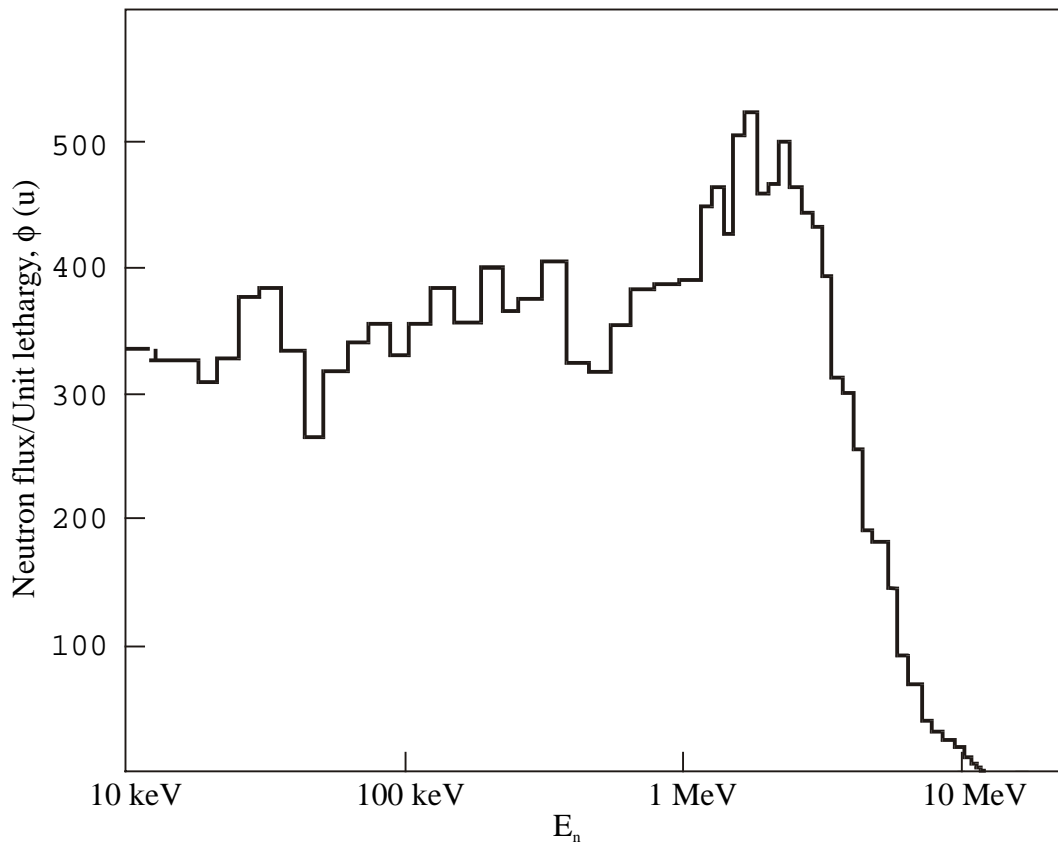
$$\phi_d = \frac{R}{\sigma_d} \quad (1.18)$$

where s_d is a normalising factor, which may be chosen quite arbitrarily. In Simmons (1965) s_d is defined with respect to the source neutron flux due to fission, i.e.

$$\sigma_d \propto \int_0^{\infty} \phi(E_n) \sigma(E_n) \bar{v}(E_n) dE_n \quad (1.19)$$

That is, ϕ_d is the equivalent fission flux for displacement. However the most generally used normalisation is the neutron spectrum in a Mk III hollow fuel element in the DIDO Materials Test Reactor at the Atomic Energy Research Establishment, Harwell, England. (The reactor is no longer operational.) The spectrum is shown in Fig 1.4 and the model used for $\bar{\nu}(E_n)$ is due to Thompson and Wright (1965), discussed later.

The calculated damage ratios in different neutron spectra have been compared with measured rates of property change in many experiments, beginning with the work of Bell *et al* (1962), who used changes in the electrical resistivity of a standard block of graphite irradiated at about room temperature. There is evidence that similar methods were in use in the USA in the late 1940's (Primak, 1956). Bell *et al* noted that in fact the property changes depend on three variables; that is the damage flux ϕ_d , the irradiation temperature T_i K and the irradiation time t sec. Determination of the damaging power of two reactor spectra is then difficult,



**Figure 1.4 Spectrum in Mk. III fuel element in DIDO.
(The standard position for the DIDO equivalent dose)**

because it would be necessary to adjust the reactor powers to give the same property changes after the same irradiation time. It has often been assumed that the property changes just depend upon the irradiation dose and temperature and thus having measured property changes as a function of dose at a particular temperature in one reactor, measurements to a convenient dose at the same temperature yield the damaging power in another reactor. It is known that if two identical graphite samples are irradiated to the same damage dose but taking different times, that is one quickly and the other more slowly, then in general the property changes are not identical; the sample irradiated quickly showing the larger changes. This is a flux level effect - it occurs because the damage depends upon the dose, but the thermal processes to which the defects are subject depend upon the temperature and the time. The slower irradiation allows more time for the thermal processes to occur. These effects can only be avoided by carrying out experiments at similar displacement rates. In practice the concept was introduced of an "Equivalent Temperature" q K which would be required in a flux producing a standard displacement rate $A_s \text{ sec}^{-1}$ to produce the same observed property changes as a function of damage dose as those observed at a temperature T_i K, displacement rate $A \text{ sec}^{-1}$. The two temperatures are related by:

$$\frac{1}{\theta} - \frac{1}{T_i} = \left| \frac{k}{E} \right| \ln \left(\frac{A}{A_s} \right) = \left(\frac{k}{E} \right) \ln \left(\frac{\phi_d}{\phi_s} \right) \quad (1.20)$$

where k is Boltzmann's constant and E is an activation energy.

Equation (1.20) can be obtained using an argument due to G H Kinchin (unpublished) as follows. During irradiation the displaced atoms pass through a sequence of configurations which depend upon the rate of displacement and the diffusion rate (thermally activated). If the damage flux is increased then it is necessary, to obtain the same final damage state, to increase the diffusion rate in the same ratio by increasing the temperature as given by equation (1.20). The equation may be derived from a wide range of kinetic models of damage accumulation. The argument is not entirely satisfactory and begins to break down for ratios of A/A_s greater than about 20. The activation energy E must be determined experimentally. Given this value all data can be reduced to a function of two variables only; that is the damage dose and the temperature obtained from equation (1.20) appropriate to irradiation at the standard rate.

Bell *et al* (1962) made the first direct determination of the activation energy E using electrical resistivity changes in a standard block of graphite. The dose and temperature dependence of the electrical resistance changes was determined using a temperature controlled irradiation facility in the graphite moderated reactor BEPO located at the Atomic Energy Research Establishment, Harwell. The dose was measured using the activation reaction $^{58}\text{Ni} (n,p) ^{58}\text{Co}$, which is sensitive to neutrons with energies greater than $\sim 1\text{MeV}$. The changes in the electrical resistivity were expressed as

$$\frac{\Delta\rho}{\rho} = 0.162[1 - 0.00514(T - 273)] \times [\gamma_{Ni} \times 10^{-16}]^{0.75} \quad (1.21)$$

where γ_{Ni} is the nickel dose. Electrical resistivity changes were then measured in a single reactor position at high and low reactor power in the PLUTO Materials Testing Reactor, also

at Harwell. (Actually reactor powers of 300 kW and 7.5 MW were employed.) The resulting data permitted the apparent irradiation temperatures for identical real irradiations to be determined and hence a value of E , as well as the relative damaging powers of the two spectra. The value obtained for E was 1.58eV. Bridge *et al* (1963) using the DR-3 Materials Testing Reactor at Risø, Denmark, measured changes in dimensions, crystal lattice parameter changes, Young's modulus, electrical resistivity and thermal conductivity as a function of dose for irradiation temperatures between 150 and 220°C (dose measured using the ^{58}Ni (n,p) ^{58}Co reaction) in a hollow fuel element with reactor powers of 1MW and 10MW. The flux level effect was clearly visible and yielded a value of $E = 1.2\text{eV} \pm 0.1\text{eV}$ for all properties.

The value of E would be expected, on very general grounds, to increase with irradiation temperature. Two attempts have been made to determine the value of E appropriate to higher temperatures (Brocklehurst, Kelly and Gilchrist, 1981; Kennedy and Eatherly, 1981). The first authors found that different values apparently applied to different properties, which strictly contradicts the theory, while the latter authors essentially found no effect - that is the work was inconclusive. This probably reflects the difficulties of close temperature control in high temperature irradiations (see later).

Given knowledge of the effects of flux level (displacement rate), the relative damaging powers of different reactor spectra can be determined and compared with the calculations of relative damage rate. Table I.4 shows calculations due to Simmons, using the Kinchin-Pease model with $L_c = 25\text{keV}$ and neutron spectra calculated by Monte Carlo methods for the reactor facilities in which electrical resistivity data had been obtained. (The variation of the calculated displacement rate to nickel activation ratio relative to the standard position is only weakly dependent on L_c in the Kinchin-Pease model, but comparison of calculated ratios with the experimental data for the BEPO and PLUTO data showed that the best value for L_c was 25keV.) The data are normalised to a value of unity for the ratio of ϕ_d/ϕ_{Ni} in the TE-10 position in the moderator of BEPO. ϕ_{Ni} is the flux measured using the reaction ^{58}Ni (n,p) ^{58}Co with a cross-section of $0.107 \times 10^{-24} \text{ cm}^2$.

The studies of relative damage rate so far described showed that calculated relative displacement rates were in good agreement with measured relative rates of property change, and were therefore of practical use. The first proposals for an improved model were made by Thompson and Wright (1965). The energy E_D out of an initial energy E_p which produces displacements was calculated in the form

$$E_D = \int_0^{E_p} \frac{\left| \frac{dE}{dx} \right|_c}{\left\{ \left(\frac{dE}{dx} \right)_c + \left(\frac{dE}{dx} \right)_e \right\}} dE \quad (1.22)$$

TABLE 1.4. VARIATION OF DAMAGE FLUX/NICKEL FLUX RATIONORMALISED TO UNITY IN BEPO TE-10

Experimental Position	Reactor Moderator	(ϕ_d/ϕ_{Ni}) Experimental	(ϕ_d/ϕ_{Ni}) Calculated
BEPO TE -10 Exp. Hole	Graphite	1.00 (Definition)	1.00
BEPO HFE	Graphite	0.43	0.43
BEPO Empty Fuel Channel	Graphite	1.00	1.00
PLUTO RFE*	D ₂ O	0.59	0.52
PLUTO HFE**	D ₂ O	0.49	0.49
DR-3 RFE*	D ₂ O	0.51	0.52
DR-3 HFE**	D ₂ O	0.49	0.49
Herald RFE	H ₂ O	0.39	-

* Denotes similar positions - results should be identical.

** Denotes similar positions.

HFE denotes Hollow Fuel Element and RFE denotes Replacement Fuel Element.

where

$(dE/dx)_c$ is the rate of energy loss/unit path length due to collisions.

$(dE/dx)_e$ is the rate of energy loss/unit path length due to electronic processes.

The number of displaced atoms produced on average by a knock-on of energy E_p is now given by

$$\frac{E_D}{2E_d} \quad \text{for } E_D > 2E_d$$

$$\bar{\nu}(E_p) = \quad (1.23)$$

$$1 \quad \text{for } E_d \leq E_D \leq 2E_d$$

with $E_d = 60\text{eV}$, following Lucas and Mitchell (1964). The electronic energy losses were taken from the experimental data due to Ormrod and Duckworth (1963) and Porat and Ramavataram (1961).

The average number of displacements for a primary knock-on of energy E_p , $\bar{\nu} 1(E_p)$, is shown in Fig 1.5 for the Kinchin-Pease model with two values of L_c , 12keV and 25keV, together with the Thompson-Wright model, all using $E_d = 60\text{eV}$.

The new method did not make any significant difference to the calculated ratios of displacement rate to activation rate normalised to a standard position; the values are shown in Table 1.5, now normalised to unity in the spectrum associated with the DIDO hollow fuel element shown

TABLE 1.5. COMPARISON OF CALCULATED AND MEASURED GRAPHITE DAMAGE RATES USING THE THOMPSON-WRIGHT MODEL

Location	Calculated	Measured/Standard	Reference
DIDO hollow fuel element	1.00	1.00	By definition
PLUTO empty lattice position	0.975	1.22	Bell <i>et al</i> (1962)
DR-3 empty lattice position	0.975	0.90	Bridge <i>et al</i> (1963)
BR-2, Mol, hollow fuel element	1.00	0.90	
HFR-Petten core	1.02	1.0	Zijp and Rieffe (1972)
BEPO TE-10 hole	2.31	2.04	
1. Empty fuel channel	2.36	2.04	Bell <i>et al</i> (1962)
2. Hollow fuel channel	0.98	0.87	
Windscale AGR			
1. Replaced fuel stringer B	2.70	2.28	
Replaced fuel stringer D	2.71	2.03	
2. Loop stringer	2.60	2.08	
3. Loop control stringer	2.60	2.51	Gray and Thorne (1968)
4. Fuel element - inner ring	1.18	1.06	
- outer ring	1.39	1.06	
Calder x-hole	2.12	2.10	
Dounreay Fast Reactor core	0.46	0.50	Martin and Price (1967)

in Fig 1.4. All doses can be expressed as "DIDO Equivalent Dose"; that is the dose measured by the ^{58}Ni (n,p) ^{58}Co reaction in the hollow fuel element spectrum, using a cross-section of $0.107 \times 10^{-24} \text{ cm}^2$, which would give the same number of displacements as the sample spectrum. A number of authors have calculated the hollow fuel element spectrum and the ratio of displacement rate to nickel activation rate for the spectrum. The original Thompson-Wright calculation gave $1.313 \times 10^{-21} \text{ atoms/atom/n.cm}^{-2}$ which is normally used, but other authors have obtained values ranging from 1.178×10^{-21} to $1.136 \times 10^{-21} \text{ atoms/atom/n.cm}^{-2}$. A DIDO Equivalent Dose of $7.62 \times 10^{20} \text{ n.cm}^{-2}$ corresponds to every atom being displaced once (in modern notation 1dpa). The calculations of DIDO Equivalent Temperature for this facility assume a flux measured by the nickel activation reaction of $4 \times 10^{13} \text{ n.cm}^{-2}.\text{s}^{-1}$, corresponding to a displacement rate $A_s = 5.25 \times 10^{-8} \text{ atoms/atom/s}$. The damage doses are generally expressed in these units for experiments carried out in the UK, Japan and Germany, but in the USA it was noted that for thermal reactor spectra the relative damage rates could be accurately

estimated by taking the neutron flux with energy above some value E_n . Two values have been used: $E_n = 0.18\text{MeV}$ and $E_n = 0.05\text{MeV}$ (sometimes written as 50keV).

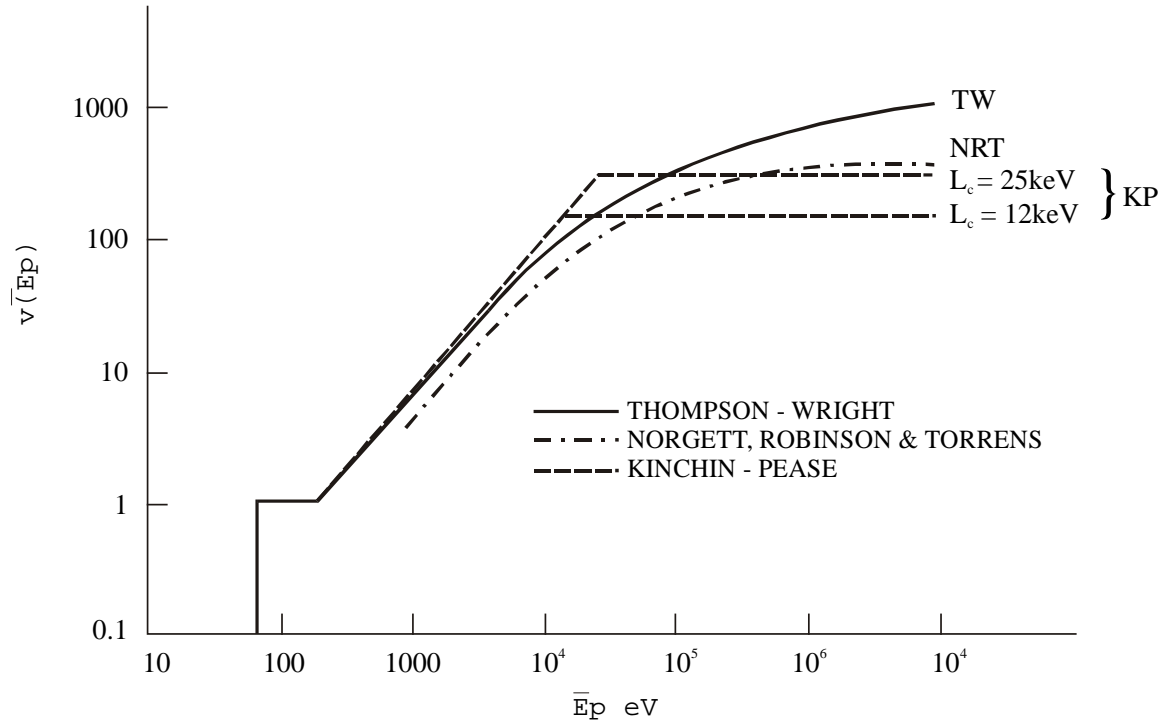


Figure 1.5 Comparison of primary displacement functions

Direct comparison of data shows that

$$1 \text{ n/cm}^2 \text{ (DIDO Equivalent Dose)} = 2.0 \text{ n/cm}^2 \text{ (} E_n > 0.05\text{MeV)}$$

$$1 \text{ n/cm}^2 \text{ (DIDO Equivalent Dose)} = 1.5 \text{ n/cm}^2 \text{ (} E_n > 0.18\text{MeV)}$$

A variety of damage dose units were used before the theoretical studies and experimental studies described here. Table 1.6 shows the conversion factors between these units.

TABLE 1.6. CONVERSION OF VARIOUS GRAPHITE DAMAGE DOSE UNITS TO EQUIVALENT DIDO NICKEL DOSE

Dose or Flux Units and Original Source	Multiply by
Equivalent DIDO Nickel Dose (EDND or DNE) UKAEA	1.0
Equivalent Fission Dose (33) UKAEA	0.547
Calder Equivalent Dose (MWd/Ate) UKAEA	1.0887×10^{17}
¹ BEPO Equivalent Dose UKAEA	0.123
	(Morgan gives 0.0962 ± 0.01)
Neutron dose n.cm^{-2} ($E_n > 0.05\text{MeV}$) USA	0.5
Neutron dose n.cm^{-2} ($E_n > 0.18\text{MeV}$) USA	0.67
Neutron dose n.cm^{-2} ($E_n > 1.0\text{MeV}$) USA	0.9
MWd/Ate(USA) - Hanford Irradiations, USA. Morgan (1974a) gives values for 1MWd/Ate of $4.56 \times 10^{16} \text{ n.cm}^{-2}$ ($E_n > 0.18\text{MeV}$) originally due to Nightingale (1962) and $5.18 \times 10^{16} \text{ n.cm}^{-2}$ ($E_n > 0.18\text{MeV}$) due to De Halas; that is multiply by 3.26×10^{16} to give EDND.	

Note that 1 n.cm^{-2} (EDND) = 1.313×10^{-21} displacements/atom = 148.4 displacements/cm³ for 1.13×10^{23} atoms/cm³.

Early design studies of graphite moderated reactors required estimates of the variation of damage dose distribution in the structure, and these were obtained experimentally. In the United Kingdom, Kinchin determined the relative damage rates at a variety of positions in the core and reflector of BEPO (using electrical resistivity measurements). The results were analysed to give the relative damage as a function of distance from a line source of fission neutrons in a graphite medium of fixed density. The curve obtained is illustrated in Fig 1.6. The damage rate in a reactor could then be obtained by summing the contributions from each fuel channel, allowing for the power distributions (see Bell *et al*, 1962). Similar methods were developed in the USA, but have not been reported in detail (Primak, 1956). Wright (1965) showed that the empirical curve due to Kinchin was well reproduced by calculations of the variation of the neutron spectrum with distance from a line source of fission neutrons in graphite combined with the Thompson-Wright model. The results can be fitted by

$$\phi_d \propto \frac{1}{r} \{A_1 \exp(-\lambda_1 r) - A_2 \exp(-\lambda_2 r)\} \quad (1.24)$$

where

$$A_1 = 1090 \pm 29 \text{ cm}, \quad l_1 = 0.0822 \pm 0.0004 \text{ cm}^{-1}$$

$$A_2 = -918 \pm 27 \text{ cm}, \quad l_2 = 0.208 \pm 0.010 \text{ cm}^{-1}$$

¹ Note that early UK studies were reported using BEPO Equivalent Doses (BED) rather than the Equivalent DIDO Nickel Dose now recommended for use. The BEPO Equivalent Dose was the thermal neutron dose in a standard position in BEPO which gives the observed damage. The thermal flux is readily perturbed and is not really suitable for the purpose.

for a graphite of density 1.6 g.cm^{-3} , valid for $2 \text{ cm} < r < 115 \text{ cm}$. Different graphite densities may be allowed for by scaling the radial distance with the relative density, i.e.

$$r_g = 1.6 \frac{r}{\rho_g} \quad (1.25)$$

where r_g is the graphite density. These methods are still useful for initial estimates of local damage, but are now superseded by more modern computer based methods.

Morgan (1974b) compared (see Fig 1.7) the calculated ratios of displacement rate to nickel activation rate normalised to unity in the DIDO Mk III hollow fuel element neutron spectrum, using the Thompson-Wright model, with the experimental measurements. It is clear from Fig 1.7 that there is a deviation from the theoretical values, the calculated values being too high, particularly for values close to 2. This was interpreted by Morgan as showing that a larger fraction of the displacement damage is produced by higher energy neutrons than is implied in the Thompson-Wright model. Morgan proceeded to recalculate the damage function using the neutron cross-sections taken from the third version of the Evaluated Nuclear Data File (ENDF/B III), including the effects of inelastic (n,a) scattering and anisotropic scattering, together with a theoretical treatment of the stopping cross-section of carbon due to Lindhard *et al* (1963). Fastrup *et al* (1966) demonstrated that the experimentally measured stopping cross-sections (Porat and Ramavataram, 1961; Ormrod and Duckworth, 1963) were in reasonable agreement with the Lindhard theory. The Thompson-Wright model and the Kinchin-Pease model allowed for electronic energy loss in the primary knock-on only, but the Lindhard *et al* model more realistically allows for such losses in the secondary and later knock-ons. Lindhard *et al* show that, to a good approximation

$$\bar{v}(E_p) = \frac{E_p}{2E_d} \frac{1}{[1 + K g(\epsilon)]} \quad (1.26)$$

where

$$e = E_p/E_l, \text{ where } E_l \text{ is a constant}$$

and for carbon atoms

$$K = 0.12748$$

$$E_l = 5686.7 \text{ eV}$$

Robinson (1969) parameterised $g(e)$ in the form

$$g(\epsilon) = 3.4008\epsilon^{1/6} + 0.40244\epsilon^{3/4} + \epsilon \quad (1.27)$$

Morgan compared the normalised displacement cross-sections over the neutron energy range 0.15MeV to 5MeV and showed that, as required, the new displacement cross-sections increased more rapidly with energy than in the Thompson-Wright model. Calculations due to Jenkins (1970), who used the ENDF/B nuclear data file and the Thompson-Wright model,

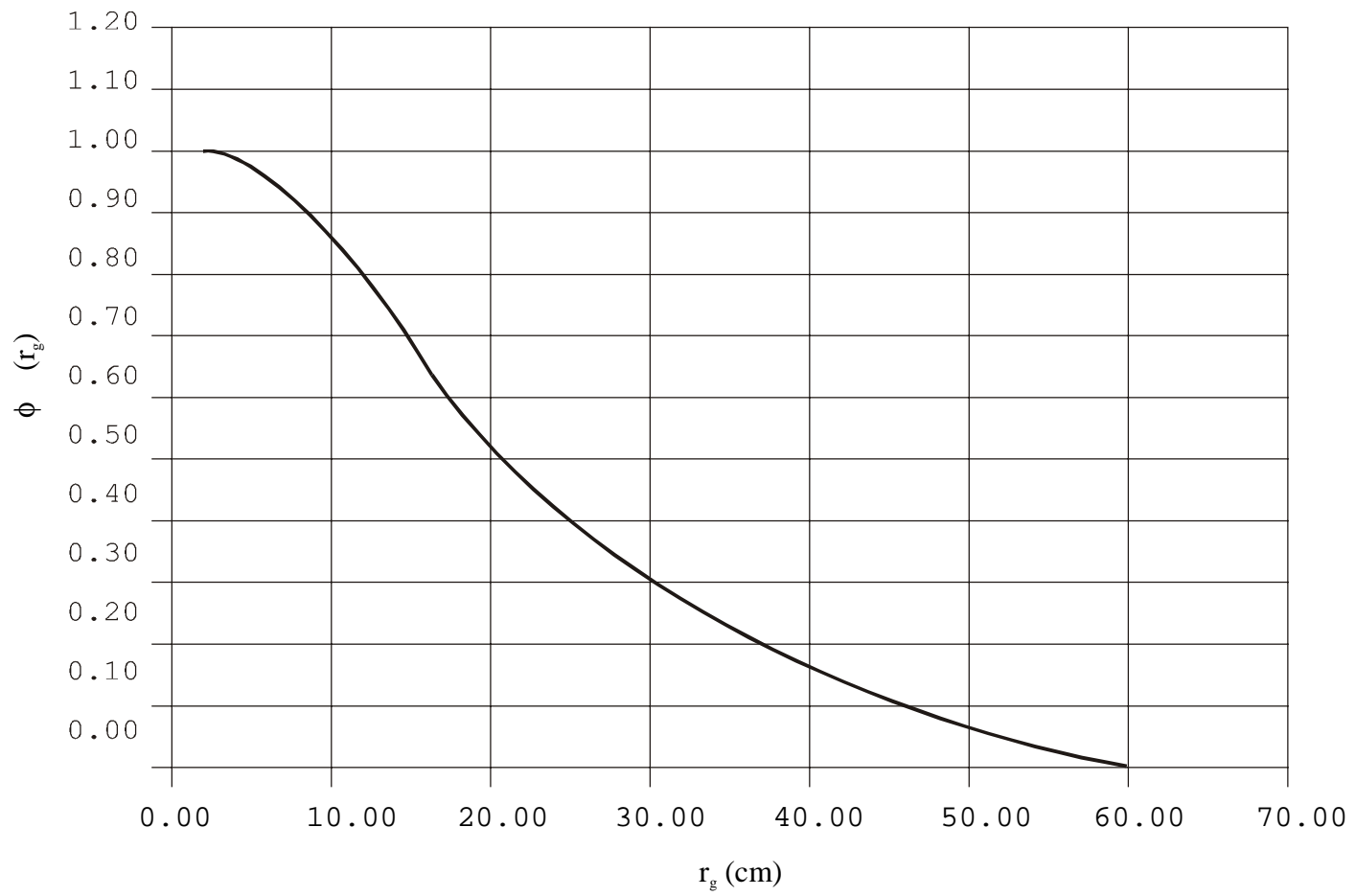


Figure 1.6 Variation of graphite damage flux with distance through graphite

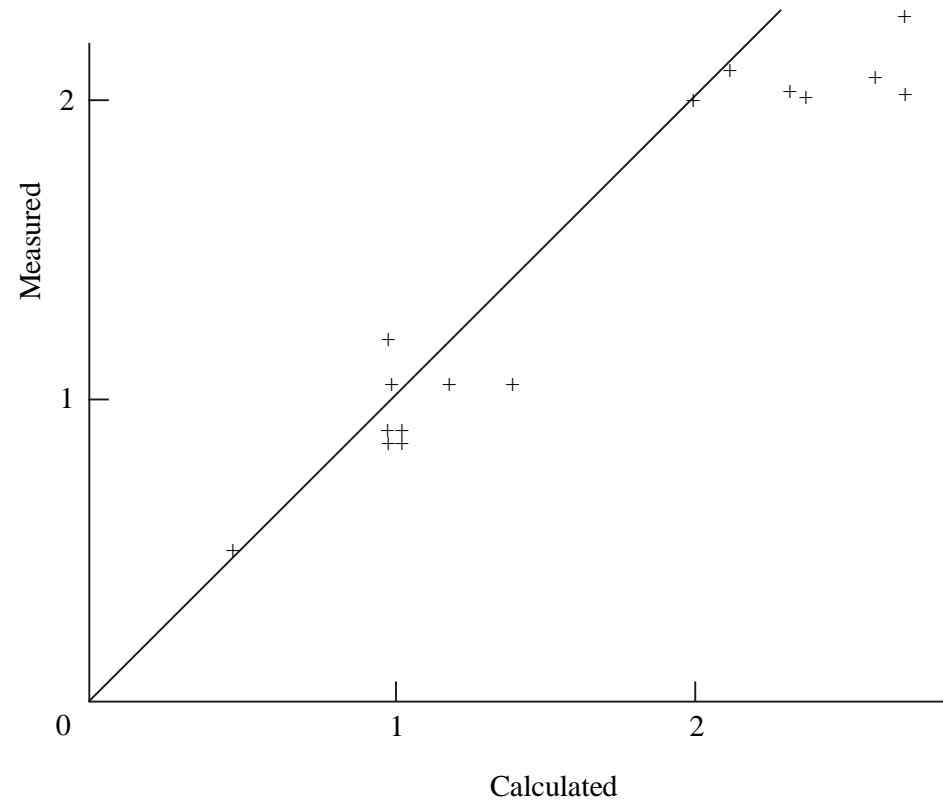


Figure 1.7 Calculated ratio of damage flux to nickel flux compared with measured rates for graphite damage calibration experiments (Thompson-Wright Model)

apparently showed the opposite effect in this neutron energy range, the lower energy neutrons being relatively more damaging and the high energy neutrons less damaging than the original Thompson-Wright model. The source of the discrepancy must lie in the nuclear data and its treatment. All of the ratios lie within the range 0.9 to 1.2. The displacement cross-section for 14.1MeV neutrons, the source energy for D-T reactions, obtained by Morgan is $254 \times 10^{-24} \text{ cm}^2$.

It has been demonstrated that for a very general class of scattering potentials for atom-collisions, the mean number of displacements is given by

$$\bar{\nu}(E_p) = \beta \left| \frac{E_D}{2E_d} \right| \text{ for } E_D \gg 2E_d \quad (1.28)$$

where β is known as the "displacement efficiency" and which allows for deviations from hard-sphere scattering and rapid local interstitial-vacancy recombination. Sigmund (1969) demonstrated that the displacement rate is largely determined by the collision cross-section at low energies and that an appropriate value for β is 0.8, independent of material or primary knock-on energy. This factor obviously does not affect relative damage rates. Inclusion of the displacement efficiency in the Lindhard model is known as the Norgett/Robinson/Torrens or NRT model. $\bar{\nu}(E_p)$ calculated using this model is shown in Fig 1.5. Genthon (1972) showed that the relative damage rates in fission based systems calculated using the Lindhard model were very little different from those calculated using the Thompson-Wright model, the latter being in slightly better agreement with experiment.

The information available on energy loss and the comparison of experimental and theoretical damage rates show that the Lindhard model overestimates the electronic energy loss (and hence underestimates the energy loss due to collisions) at high primary energies. This is apparently true for the Thompson-Wright model also.

The Lindhard model and its developments are clearly more suitable for damage calculations on fusion systems and have been applied by Robinson (1969), Morgan (1974b) and Gabriel *et al* (1976). Strictly speaking it is unsatisfactory for carbon recoils with energies greater than 3.2MeV (or collisions with neutrons of energy greater than ~11MeV) due to inelastic effects.

Adamson (1987) has examined application of the Lindhard model to 14.1MeV neutrons and a typical fusion reactor spectrum. The neutron cross-sections for elastic, inelastic and (n, α) reactions were processed from the Fine Group Library (FGL5) at the Atomic Energy Establishment, Winfrith Heath, England for the 14.1MeV neutrons. (Actually three energy groups in the range of neutron energies 13.78 - 14.44 MeV of equal lethargy interval.) The average carbon atom recoil energies were calculated as well as the displacement cross-sections. It was found the (n, α) reaction contribution was ~25% of the total cross-section. This is because

- (i) The recoil energy from the (n, α) reaction is large in relation to that from the scattering reactions (~3MeV as against 1MeV) and the fraction of the energy lost in inelastic collisions in the lattice is consequently large.

- (ii) The recoil energy from the (n,a) reactions has a significantly smaller mass than the C_6 nucleus, leading to an increased energy loss to electronic processes.

The existence of other reactions such as (n,d), (n,2n) and (n,p) were ignored. Adamson (1987) found that the displacement cross-section for neutrons of average energy 14.1MeV was $271.1 \times 10^{-24} \text{cm}^2$, using the same displacement energy of 60eV. Table 1.7 compares the calculations of Adamson, Robinson, Gabriel *et al*, Morgan, and Huang and Ghoniem (see later).

TABLE 1.7. COMPARISON OF THE DISPLACEMENT CROSS-SECTION FOR 14.1MeV NEUTRONS IN GRAPHITE

Source	Reactions included	dpa/n.cm ⁻²
Robinson (1969)	(n,2n)	2.34×10^{-22}
Morgan (1974b)	(n,a)	5.18×10^{-22}
Gabriel <i>et al</i> (1976)	(n,a)	3.76×10^{-22}
Adamson (1987)	(n,a),(n,n')	2.71×10^{-22}
Huang and Ghoniem (1993)	(n,a),(n,p),(n, γ)	4.9×10^{-22}

Three of the results are similar, but both Morgan's and Huang and Ghoniem's are significantly higher, even when, as here, they have been corrected for the difference in E_d and the displacement efficiency. The reasons for these differences are not clear but must lie in the treatment of the cross-sections. The degradation of the spectrum in real fusion systems due to scattering almost certainly diminishes the differences in comparable dpa calculations.

Huang and Ghoniem (1993) developed a model in which the electronic energy loss treatment was improved. Comparison of the energy losses of carbon ions in carbon with the Lindhard model showed that the losses were overestimated for moving atom energies $> \text{MeV}$. For substantially greater energies ($> 8\text{MeV}$) the Bethe-Bloch theory is appropriate (Bethe, 1930; Bloch, 1933), for which energy loss rates decrease with increasing energy. Huang and Ghoniem show that a good fit to the energy loss data given by Northcliffe and Schilling (1970) may be obtained using an empirical method due to Biersack and Haggmark (1980). These authors propose that the energy loss rate S_e is given by:

$$\frac{1}{S_e} = \left(\frac{1}{S_e} \right)_L + \left(\frac{1}{S_e} \right)_{B-B} \quad (1.29)$$

where $(1/S_e)_L$ is the reciprocal of the energy loss rate obtained from the Lindhard model and $(1/S_e)_{B-B}$ is the reciprocal of the energy loss rate obtained from the Bethe-Bloch treatment.

Huang and Ghoniem compare the primary carbon recoil spectra in a number of neutron spectra obtained from both fission and fusion systems allowing for (n,a),(n,p) and (n, γ) reactions. There are considerable differences in the spectra for primary energies greater than 1MeV. The fraction of the primary knock-on energy dissipated in atomic displacements is found to fall smoothly from unity at an energy of 10^{-4}MeV to 0.03 at an energy of 10MeV. The displacement cross-section for 14MeV neutrons estimated from Fig 8 of the paper is

found to be $18 \times 10^{-22} \text{ cm}^2$ for $E_d = 16.3\text{eV}$, or $4.9 \times 10^{-22} \text{ dpa/n.cm}^{-2}$ corrected to $E_d = 60\text{eV}$ and $\beta = 0.8$ (the latter correction may not be appropriate), roughly in line with the other estimates. The differences merit a careful examination of cross-section data at the higher energies.

The relative damaging power of the high-energy neutrons must eventually be determined experimentally. The only attempt known to the author was made by Gray and Morgan (1979). The problem is difficult because only very low fluxes of mono-energetic neutrons are available and thus it is necessary to use properties which are very sensitive to radiation damage. It was known (see Chapter 5) that the basal shear elastic modulus of highly oriented pyrolytic graphite was extremely sensitive to radiation damage, changes being detectable at neutron doses of $\sim 10^{14} \text{ n.cm}^{-2}$, which can be measured as changes in the sonic velocities in the graphite, with appropriate polarisation and direction. Gray and Morgan irradiated samples in three very different neutron spectra:

- (i) The Medical Research Reactor at Brookhaven National Laboratory (USA) which has a typical, well-thermalised fission neutron spectrum, with a peak at $\sim 1.5\text{MeV}$.
- (ii) The rotating target neutron source at the Lawrence Radiation Laboratory which has a D-T fusion neutron source spectrum which is practically constant at $\sim 15\text{MeV}$.
- (iii) The deuterium-beryllium neutron source at the University of California (Davis) which produces a neutron spectrum intermediate between (i) and (ii), peaking at about 5.5MeV , but with a distribution of energies of about one order of magnitude.

Three grades of highly oriented pyrolytic graphite were exposed in each facility at temperatures between 15 and 30°C , together with appropriate flux monitors to measure the neutron dose. Changes in the measured shear modulus as a function of neutron dose with energy $> 50\text{keV}$ showed changes increasing in the order $15\text{MeV} > 5.5\text{MeV} > 1.5\text{MeV}$, apparently demonstrating the greater damaging power of the higher energy neutrons.

The relative changes in shear modulus/unit dose were averaged for the three grades of highly oriented material up to fractional changes less than 1.6 in the shear modulus, where changes were linear with dose. The SAND-II computer program is able, given an initial guess, to estimate a neutron spectrum from the activation of an appropriate series of flux monitors or, given relative damage rates in well-differentiated spectra, the damage cross-section as a function of neutron energy. (Note: because this is based on property changes, it need not be the same as the displacement cross-section.) Given atomic displacement models as trial guesses, the damage function generated apparently shows that much greater damage is generated by the high energy neutrons than predicted by the atomic displacement models.

There are therefore two separate pieces of evidence that high-energy neutrons are more damaging than anticipated from displacement models. The Morgan-Gray experiments are however open to three criticisms:

- (i) The rate of change of the elastic modulus of graphite is very sensitive to the irradiation temperature (Simmons, 1965) for temperatures close to ambient and no correction has apparently been made for this.

- (ii) The damage obtained in irradiation experiments around room temperature anneals significantly at ambient and the observed changes should be corrected to the end of the experiment in each case.
- (iii) A very significant displacement rate effect would be expected because of the strong dependence on irradiation temperature.

In a subsequent paper by Gray and Thrower (1979) the annealing of the modulus changes was discussed in detail. It was assumed that a sample with an initial shear modulus C_0 was irradiated to give a new value C_i , which after a long period of time tended to a value C_∞ asymptotically. The decay was assumed to be given by

$$C = (C_i - C_\infty)\exp[-\alpha t] + C_\infty \quad (1.30)$$

where a may depend on the irradiation temperature.

The three grades of graphite examined show different values of C_0 and different values of C_i and C_∞ , the latter because of exposure to different doses. However, all samples irradiated in the same facility fall on a single curve if plotted as $C/(C_\infty - C_0)$ - this is accounted for by noting that a fixed fraction of the shear modulus changes can be annealed at room temperature, independent of the dose or grade of graphite. The value of C_∞ was taken to be that at the largest value of the time, always greater than 190 days. (This was confirmed by additional annealing studies.) It was found that the fraction of the modulus change which could be annealed at room temperature varied between the three neutron energies - that is 0.40, 0.25 and 0.22 for the 14.8, 5.5 and 1.5MeV irradiations, corresponding to a values of 3×10^{-3} , 14×10^{-3} and 37×10^{-3} /day.

The possibility was examined that mechanical handling of the samples could be a source of changes in the shear modulus and some effects were found. Transmission electron microscopy showed defects, in the form of small interstitial atom clusters $< 15 \times 10^{-8}$ cm in diameter, which did not show any change in the process of annealing, thus demonstrating that even smaller defects are involved in the shear modulus changes and the annealing.

The current situation appears to be that it is possible in fission reactor systems to estimate the relative damage rates in different reactor spectra with an accuracy adequate for design - that is data taken in one spectrum can be used to predict behaviour in another, provided that proper consideration is given to the existence of rate effects. The situation with respect to fusion reactor spectra is more complex - quite large differences exist in displacement cross-sections for high energy neutrons estimated by different authors, and thus damage estimates in spectra dominated by source neutrons are not likely to be very accurate. The situation is much better in the substantially degraded spectra which are likely to be found in most of the components of a fusion reactor.

The work of Gray and Morgan (1979) has demonstrated that it is possible to carry out experimental studies of the damage function using available high-energy neutron sources. It is clear that further experimental and theoretical studies are necessary, taking care to control the

experimental temperatures and to either control the post-irradiation temperatures to prevent annealing or make post-irradiation measurements which permit correction of properties to the end of the irradiation. This work needs to be undertaken and should include measurements of the transmutation levels.

The current recommendations for the expression of damage dose, due to the IAEA (1972), are to give the fission neutron dose which will produce the number of displacements estimated for the experiment using the Thompson-Wright damage model. This dose bears a constant relationship to the DIDO Equivalent Dose, which is most generally used (see Table 1.6). It is clear that while the use of dose units based on neutron flux with energies greater than a particular value is not likely to be correct in fusion systems, particularly near the sources, it would be a more modern approach in line with other materials to use a scale expressed in displacements/atom. (It should also be noted that it is quite common in discussion of damage rates in fusion reactor systems to express the neutron flux in terms of the thermal loading, that is in MW/m², one unit corresponding to a flux of 4.4 x 10¹³ n.cm⁻² of 14.1MeV neutrons, which using the calculations by Adamson (1987) corresponds to 0.4 displacements/atom/yr.)

At this point we turn to the question of the data which are used to evaluate displacement models, and changes which have occurred since the formulation of the Thompson-Wright model and the Lindhard model.

A considerable amount of information has been obtained since the formulation of the Thompson-Wright model on the rate of energy loss of ions in solids. The electronic energy loss data have been summarised by Garnir-Monjoie *et al* (1980) in the formula

$$S_e = \left| \frac{dE}{dx} \right|_e = k_2 \left(\frac{E}{M} \right)^{k_1} \left[1 - \exp \left(-k_3 \left(\frac{E}{M} \right)^{-1} \right) \right]^{k_4} \quad (1.31)$$

where for $(dE/dx)_e$ in MeV/(mg.cm⁻²) and (E/M) in MeV/amu, the constants k_1 to k_4 are given by

$$\begin{aligned} k_1 &= 0.5 + 1.8 \times 10^{-3}Z \\ k_2 &= 0.8Z + 7.8Z^{1/2} - 3.9 \\ k_3 &= -2.3 \times 10^{-2}Z + 0.42Z^{1/2} - 0.37 \\ k_4 &= (2.4 \times 10^{-3}Z^2 + 1.12Z + 0.88)/Z \end{aligned}$$

Insertion of $Z = 6$ (appropriate to carbon) into equation (1.31) leads to

$$k_1 = 0.51, k_2 = 20, k_3 = 0.521, k_4 = 1.28$$

and thence on conversion to units of eV/Å (1Å = 10⁻⁸cm) leads to

$$\left| \frac{dE}{dx} \right|_e = 452 \left(\frac{E}{12 \times 10^6} \right)^{0.51} \left[1 - \exp \left(-\frac{6.25 \times 10^6}{E} \right) \right]^{1.28} \quad (1.32)$$

Comparison of the energy loss given by equation (1.32) with the experimental data used in the Thompson-Wright model shows that it gives lower loss rates, but with a similar energy dependence, while at high energies it is in better agreement with experiment than the Lindhard model. The differences would increase the energy going into atomic displacement, particularly at high energies where the effect is similar to the analysis of Huang and Ghoniem. However, no detailed evaluation has been made.

The high-energy neutrons characteristic of fusion sources produce transmutation effects to a much greater degree than in fission systems. In particular, transmutation to helium or other elements may have important effects on property changes. The effects of helium generation have been observed directly by Kelly and Mayer (1969), who studied the effects of substitutional doping with ^{10}B and ^{11}B on irradiation damage in a Materials Test Reactor facility with roughly equal thermal and fast neutron flux. In the samples doped with ^{10}B the thermal neutrons produce helium by the $^{10}\text{B} (n,\alpha) ^7\text{Li}$ reaction and displacement damage due to the helium and lithium recoils. The helium particle production mimics the helium generation due to $^{12}\text{C} (n,n')^3\alpha$ reactions (but also produces lithium). Samples of Ticonderoga flake doped with ^{10}B and ^{11}B concentrations in the range 30ppm to 10^4 ppm were irradiated at temperatures of 650°C and 900°C. Electron microscope studies of the flakes did not reveal any helium bubbles, but samples containing 1.4% ^{10}B irradiated at 650°C showed a huge expansion parallel to the hexagonal axis direction due to the exfoliation of packets of layer planes rather similar to that produced by heating the bromine residue compounds (Martin and Brocklehurst, 1964). These effects were not observed in the ^{11}B samples or the 0.5% ^{10}B samples under the same conditions. The implication of this result may well be that if the helium production rate is high enough and its diffusion rate (presumably parallel to the basal planes) low enough, bubbles will nucleate homogeneously and grow as helium is added, producing eventual exfoliation. Kelly and Mayer estimate that the helium and lithium products of the (n, α) reaction produce 69 and 121 displacements ($E_d = 60\text{eV}$) for recoil energies of 1.53MeV and 0.87MeV respectively.

It is clear that neutron irradiation effects are actually characterised by more than one number, that is at least displacements and helium generation, while currently only the first is considered, and there is no experimental evidence for effects due to the latter. The calculation of displacement rate is less accurate for high-energy neutrons and further experimental studies are desirable.

Recently, experimental data obtained from irradiation of materials for possible use in fusion reactors have had their doses expressed as displacements/atom, calculated using the Thompson-Wright method (Burchell and Eatherly, 1991). The relationship with Equivalent DIDO Nickel Dose is

$$1 \text{ dpa} = 7.62 \times 10^{20} \text{ n/cm}^2 \text{ (EDN)}$$

It is highly desirable that a better absolute displacement rate scale should be achieved, which requires further studies of the displacement energies, particularly the angular and temperature dependencies. The best representation of the electronic energy loss data and

neutron cross-sections requires agreement and proper allowance made for inelastic and anisotropic scattering. Calculations for a particular irradiation should also give best estimates of the transmutation rates, particularly the production of helium. It would be sensible for all future data to be expressed on a displacement scale using an agreed set of input data (conditional on transmutation effects being unimportant).

REFERENCES

- [1.1] ADAMSON J. Atomic Displacement Cross-Sections in Graphite for Neutrons of 14MeV, UKAEA Report AEEW-R 2132 (1987).
- [1.2] BELL J.C, BRIDGE H., COTTRELL A.H., GREENOUGH G.B., REYNOLDS W.N. and SIMMONS J.H.W. PHIL. TRANS. ROY. SOC. A, 254, 361 (1962).
- [1.3] BETHE H. ANN. PHYSIK, 5, 325 (1930).
- [1.4] BIERSACK J.P. and HAGGMARK L.G. Nuclear Instruments and Methods, 174, 257 (1980).
- [1.5] BIRCH M. AND BROCKLEHURST J.E. A Review of the behaviour of graphite under the conditions appropriate for protection of the first wall of a fusion reactor, UKAEA Report ND-R-1434(S) (1987).
- [1.6] BLOCH F. ANN. PHYSIK, 16, 285 (1933).
- [1.7] BRIDGE H., GRAY B. S., KELLY B.T. and SØRENSEN H. Proc. Int. Conf. on Radiation Damage in Reactor Materials, Venice 1962, 3, 531. IAEA Vienna (1963).
- [1.8] BROCKLEHURST J.E, KELLY B.T. AND GILCHRIST K.E. Extended Abstracts 15th Biennial Conference on Carbon, p546 (1981).
- [1.9] BURCHELL T.D. AND EATHERLY W.P. J. Nuclear Materials, 179, 205 (1991).
- [1.10] DE HALAS D. See Nuclear Graphite (Ed. R.E. Nightingale), Academic Press (1962).
- [1.11] EGGEN D.T. Report NAA-SR-69 (1950).
- [1.12] EVERHART E., STONE G. and CARBONE R.J. Phys. Rev., 99, 1287 (1955).
- [1.13] FASTRUP B., HVELPLUND P. AND SAUTTER C.A. MAT. FYS. MEDD. DAN.VID.SELSK.,35,No.10(1966).
- [1.14] GABRIEL T.A., AMBURGEY J.D. AND GREENE N.M. Nucl. Sci. & Eng., 61, 21 (1976).
- [1.15] GARNIR-MONJOIE F.S., GARNIR H.P., BAUDINET-ROBINET Y. AND DUMON P.D.J.Physique,41,599 (1980).
- [1.16] GENTHON J.P. EUR 4867f, Joint Nuclear Research Centre (1972).
- [1.17] GRAY B.S. and THORNE R.P. J. BRIT. Nuclear Energy Soc., 7, 91 (1968).
- [1.18] GRAY W.J. and MORGAN W.C. J. Nuclear Materials, 85 & 86, 237 (1979).
- [1.19] GRAY W.J. and THROWER P.A. J. Nuclear Materials, 87, 200 (1979).
- [1.20] HUANG H. and GHONIEM N. J. Nuclear Materials, 199, 221 (1993).
- [1.21] IAEA. Specialists' Meeting on Radiation Damage Units for Graphite, Seattle(1972).(See Morgan, 1974a.)
- [1.22] IWATA T. and NIHIRA T. Physics Letters, 23, 631 (1966).
- [1.23] IWATA T. and NIHIRA T. J. Phys. Soc. Japan, 31, 1761 (1971).
- [1.24] JENKINS J.D. Nucl. Sci. & Eng., 41, 155 (1970).
- [1.25] KELLY A. and MAYER R.M. PHIL. MAG., 19, 701 (1969).
- [1.26] KENNEDY C.R. and EATHERLY W.P. Extended Abstracts 15th Biennial Conference on Carbon, p552 (1981).
- [1.27] KINCHIN G.H. and PEASE R.S. Rep. Progress in Physics, 18, 1 (1955).
- [1.28] KNIPP J. and TELLER E. PHYS. Rev., 59, 659 (1941).

- [1.29] LINDHARD J., NIELSEN V., SCHARFF M. and THOMSEN P.V. MAT. FYS. MEDD.DAN. Vid. Selsk., 33, No.10 (1963).
- [1.30] LUCAS M.W. and MITCHELL E.W.J. Carbon, 1, 345 (1964).
- [1.31] MARTIN W.H. and BROCKLEHURST J.E. Carbon, 1, 133 (1964).
- [1.32] MARTIN W.H. and PRICE A.M. J. Nuclear Energy, 21, 359 (1967).
- [1.33] MONTET G.L. Carbon, 5, 19 (1967).
- [1.34] MONTET G.L. and MYERS G.E. Carbon, 9, 179 (1971).
- [1.35] MORGAN W.C. Nuclear Technology, 21, 50 (1974a).
- [1.36] MORGAN W.C. J. Nucl. Mater., 51, 209 (1974b).
- [1.37] MOTT N.F. and MASSEY H.S.W. The Theory of Atomic Collisions. Oxford, p271 (1949).
- [1.38] NIGHTINGALE R.E. Nuclear Graphite. Academic Press (1962).
- [1.39] NORTHCLIFFE L.C. and SCHILLING R.F. Nuclear Data Tables A7, p233 (1970).
- [1.40] OHR S.M., WOLFENDEN A. and NOGGLE T.S. Electron Microscopy and the Structure of Materials (Ed. G. Thomas). Univ. of California Press (1972).
- [1.41] ORMROD J.H. and DUCKWORTH H.E. Can. J. Phys., 41, 1424 (1963).
- [1.42] PORAT D.I. and RAMAVATARAM K. Proc. Phys. Soc., 77, 97 (1961).
- [1.43] PRIMAK W. Phys. Rev., 103, 1681 (1956).
- [1.44] ROBINSON M.T. BNES Nuclear Fusion Reactor Conference, Culham. Paper 4.3, p364 (1969).
- [1.45] SIGMUND P. Radiation Effects, 1, 15 (1969).
- [1.46] SIMMONS J.H.W. Radiation Damage in Graphite. Pergamon Press (1965).
- [1.47] THOMPSON M.W. and WRIGHT S.B. J. Nuclear Materials, 16, 146 (1965).
- [1.48] THROWER P.A. and MAYER R.M. Phys. Stat. Sol.(a), 47, 11 (1978).
- [1.49] WRIGHT S.B. Radiation Damage in Solids, 2, 239. IAEA Vienna. (1962).
- [1.50] WRIGHT S.B. A Calculation of the Carbon-Atom Displacement Rate in the BEPO Core and Reflector, UKAEA Report AERE-M 1548 (1965).
- [1.51] ZIJP W.L. and RIEFFE H.CH. RCN-161. Reactor Centrum Nederland, The Hague, Netherlands (1972).