between theory and experiment. The theoretical curves for the velocity-dependent neutron flux in a 2.7-cm slab are in good agreement with the measurements in a $5 \times 5 \times 5$ cm cube in the center as well as at the boundary of the media (Fig. 2).

In another experiment[3] the assumption of slab geometry has been fulfilled reasonably well ($2 \times 20 \times 20$ cm and $3 \times 20 \times 20$ cm vessels).

In Figure 3 is shown the flux in the center and at the boundary of a 3-cm slab together with the experimental results. The leakage spectrum is shifted markedly towards higher energies compared with the spectrum in the center. In Figure 4 the calculated and measured spectra at the boundary of a 2-cm slab are compared. The solution at the boundary of the halfspace, which is also drawn in Figure 4, shows that the leakage spectrum of a small geometry is cooler than that of a large one. Both the spectra in the center and at the interface show this influence of the diffusion cooling effect (see e.g. Reference 2). Our calculations indicate that the energy dependence of thermal-neutron spectra belonging to the asymptotic flux in pulsed water geometries may be calculated very well with our formalism. A comparison of our solution of the Milne problem with Conkie's result shows that it is necessary to use an appropriate scattering kernel for water that takes into account the effects of chemical binding.

A detailed description will be published in Nukleonik.

Edgar Kiefhaber

Institut für Neutronenphysik
und Reaktortechnik,
Kernforschungszentrum Karlsruhe,
Germany

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Decomposition of Defects by Neutrons in Reirradiated Graphite*

Identification of reactions causing defect decomposition during neutron irradiation still remains an important problem in radiation studies of graphite. "Radiation annealing" experiments in which a sample originally irradiated at one temperature is reirradiated at a higher temperature show that more damage is removed by this technique than by thermal annealing alone[1]. Since these experiments involve simultaneous heating and reirradiation, it is not clear whether the effect represents defect decomposition, radiation enhancement of the thermal annealing reactions, or a combination of the two. The effects can be separated by performing the reirradiations and anneals alternately. When graphite is thermally annealed after each of a series of exposures at a fixed temperature, damage produced by the first irradiation can be removed, maintained at a constant value, or increased, by varying the exposure between anneals.

Studies were made on AGOT graphite samples subjected to more than twenty cycles of 30 C reirradiations followed by 350 C out-of-pile anneals. When the exposure between anneals was smaller than some critical value (determined by the temperature of the anneal and the total damage) each reirradiation and annealing cycle removed more net damage than the reirradiation produced. For exposures at the critical values, the reirradiation and annealing cycle removed a quantity of damage equal to that produced by the reirradiation. When the exposure between anneals exceeded the critical value, only a fraction of the damage produced by the reirradiation was removed during the following anneal. The observed effects are conclusive evidence that the reirradiations involve competitive reactions between production and decomposition of interstitial complexes. Critical exposure values and details on how the damage changes with exposure between anneals are given in reference[2].

Net changes in damage were determined from radiation-induced property changes, which were assumed to vary monotonically with the total defect concentration. This assumption is valid for c-axis changes and stored-energy changes when simple defects are formed[3,4,5]. If the damage involves several different clusters of interstitials, variations in the proportionality constant per interstitial carbon atom must be considered. It is unlikely, however, that contributions to the c-axis due to changing proportionality constants in different species will parallel proportionality constant changes in the stored energy. Simultaneous measurements of c-axis, stored-energy and dimensional

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[1]A review of "radiation annealing" phenomena is given in *Nuclear Graphite* (Academic Press, New York (1962) 376-382), edited by R. E. NIGHTINGALE.

[2]D. G. SCHWEITZER and R. M. SINGER, "Effect of Irradiation Temperature and Annealing Temperature on Expansions and Contractions in Alternately Irradiated and Annealed Graphite". *ANS Transactions*, Vol 6, No 2, 383 (Nov. 1963).

[3]G. J. DIENES and G. H. VINEYARD, *Radiation Effects in Solids* Interscience Publishers Inc., New York (1957).

[4]D. G. SCHWELTZER, *Phys. Rev.* 128 556 (1962).

[5]D. G. SCHWEITZER, "Determination of the Single Interstitial Migration Energy from Stored Energy and Thermal Resistivity Changes in Irradiated Graphite". Submitted for publication in *Carbon*.

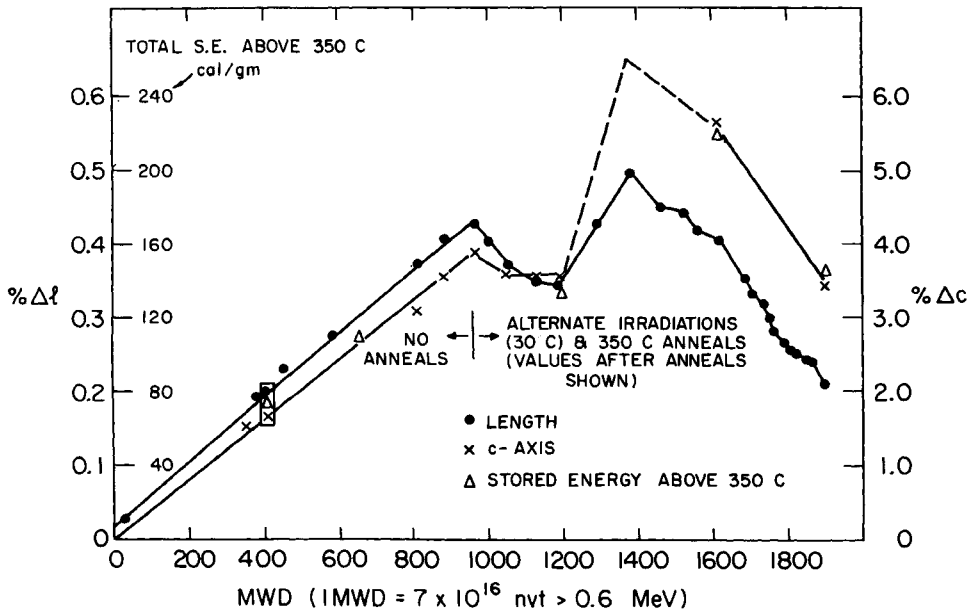

Fig. 1. Property Changes in Graphite Alternately Irradiated at 30 C and Annealed at 350 C.

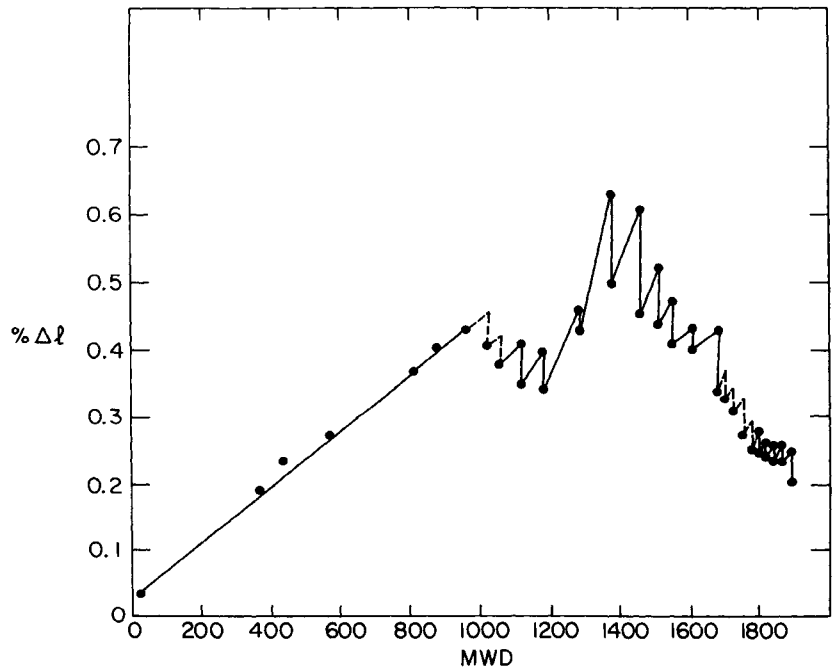

Fig. 2. Length Changes After Irradiation at 30 C and Subsequent Annealing at 350 C.

changes were therefore made on various samples. Data for graphite alternately irradiated at 30 C and annealed to 350 C are given in Figure 1. The correlation between increases and decreases in property changes shown is evidence that the data measure changes in the total damage. The results, with the exception of the values enclosed in the box on the figure, were obtained from a single sample.

The c-axis and dimensional points plotted from 0 to ~1000 MWd (1 MWd = 7×10^{16} nvt for neutrons with energies above 0.6 MeV) denote changes prior to the first anneal. Data between 1000 MWd and 2000 MWd were accumulated over a period of several years. At the end of the program, unirradiated stock material was continuously irradiated to check that irradiation and annealing conditions remained the same. The enclosed points show the data obtained. All values shown for exposures exceeding ~1000 MWd represent measurements after the 350 C anneal. Stored energy was determined from heats of combustion measurements made by the National Bureau of Standards. These measurements consumed appreciable portions of the sample and were made less frequently than those of the other properties. The variation in magnitude of exposures between anneals is shown by the position of the dimensional data points along the abscissa. Length changes were measured before and after most anneals and are shown in Figure 2. The dashed lines are estimates of the values not measured before annealing. The sample and aliquots were annealed at 350 C for 4 hours. No changes outside of experimental error occurred when the time of annealing was varied from 1 hour to 16 hours. C-axis values after irradiation at 30 C and prior to annealing did not change in aliquots that stood at room temperature for six years.

The decomposition during reirradiations demonstrated by these studies involves species formed during the anneals. Stored-energy and c-axis changes during the first irradiation can be quantitatively accounted for by a model which requires that the data reflect properties of one interstitial entity[4,5]. This model does not appear to account for experimental data if radiation decomposition reactions for the interstitial are included. It is therefore concluded that the interstitial formed during the first irradiation is more stable towards irradiation than are the clusters formed during anneals.

The mechanism for cluster break-up during reirradiations cannot be determined from these studies. It is possible that decomposition results from collision effects of energetic neutrons or from "hot atom"[1] reactions.

Donald G. Schweitzer
Robert M. Singer
Seymour Aronson
Jerome Sadofsky
David H. Gurinsky

Brookhaven National Laboratory
Upton, New York

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Decoupling the Energy-Dependent Spherical Harmonics Equations*

We show how to transform the discrete energy approximation of the spherical harmonics equations for the slowing-down of both gamma rays and elastically scattered neutrons to a system of spatially decoupled simultaneous differential equations. The solution of the spherical harmonics moments problem in very high orders of approximation is made feasible by decoupling, since the solution of a large number of decoupled equations requires no more computational effort than the simultaneous solution of a modest number of coupled equations.

This method could be applied advantageously to the study of the deep penetration of gamma rays where the strong forward peaking of the photon distribution necessitates the use of high-order angular expansions. It could also be used to calculate neutron distributions in highly absorbing media and in light moderators in which the mathematically simple age approximation is inadequate.

In the spherical harmonics approximation of order L, the space-angle-energy particle distribution, $\Psi(z, \omega, \phi, u)$, which we restrict to an even function of the azimuthal angle, ϕ, is represented as

$$\Psi(z, \omega, \phi, u) = \sum_{\ell=0}^{L} \sum_{m=0}^{\ell} \sqrt{\frac{2\ell + 1}{4\pi} \frac{(\ell - m)!}{(\ell + m)!}} \cdot \cos m\phi \, (1 - \omega^2)^{\frac{m}{2}} \, P_{\ell m}(\omega) \Psi_{\ell m}(z, u), \qquad (1)$$

where

z is the depth coordinate;

$\hat{\Omega} = \hat{i}\,(1 - \omega^2)^{\frac{1}{2}} \cos\phi + \hat{j}\,(1 - \omega^2)^{\frac{1}{2}} \sin\phi + \hat{k}\omega$ is a unit vector in the direction of particle motion;

$\hat{i}$, $\hat{j}$, and $\hat{k}$ are the unit vectors parallel to the x, y, and z Cartesian axes;

$\cos^{-1}\omega$ and ϕ are the polar and azimuthal angles on the surface of a unit sphere with its axis along the z-direction;

$P_{\ell m}(\omega)$ is the associated Legendre polynomial defined by

$$P_{\ell m}(\omega) = (1 - \omega^2)^{\frac{1}{2}} \frac{d^m P_\ell(\omega)}{d\omega^m} \, ,$$

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