SOME EFFECTS PRODUCED IN GRAPHITE BY NEUTRON IRRADIATION IN THE BNL REACTOR*

W. L. Kosiba, G. J. Dienes and D. H. Gurinsky

Brookhaven National Laboratory, Upton, New York

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The high concentration of lattice defects resulting from neutron bombardment in graphite produces important changes in the physical properties of this material.

The following physical changes produced in graphite by irradiation in the Brookhaven reactor have been studied: 1) stored energy, 2) physical dimensions and 3) c-axis. These studies were made on samples exposed to an integrated neutron flux of 1.2–1.6 $\times$ 10^{20} neutrons/cm^2. The temperature of exposure varied from 50°C to about 130°C.

The results show that: 1) the stored energy (heat content) which was released at 200°C and 400°C ranged from 0 to 50 cal/gm. 2) the physical dimensions increased from 0.09 to 0.18%. 3) the c-axis of the crystallites increased and this expansion was about nine times as great as the gross physical growth. 4) the changes in physical properties are more dependent on the temperature of irradiation than on the integrated flux at these levels of exposure. The effect of increasing the temperature of irradiation from 50°C to 100°C is to decrease the changes in physical growth and c-axis expansion by about a factor of two.

I. INTRODUCTION

When graphite was first considered for use as a moderator in reactors E. P. Wigner pointed out that the bombardment of graphite by neutrons would result in knocking out atoms from their lattice sites and that a change in the physical properties of the material should be expected. The initial theoretical work was extended by F. Seitz[1] whose calculations showed that the number of carbon atoms displaced in graphite in slowing down a single fission neutron (2 Mev) to thermal energies is of the order of 1800.

The earliest experimental work on the effect of radiation on the properties of graphite was performed by a group under the direction of Franck and Burton[2]. The effect of neutron bombardment on the c-axis (distance between hexagonal planes) was first studied by Zachariasen.[2] The work of these investigators confirmed the existence of the so called "Wigner effect". Since graphite is an important moderator, the effect of radiation on its properties has been studied intensively at many of the laboratories in the U.S.A.E.C., in Canada and in the United Kingdom. The list of investigators[2] is so long and their contributions so extensive that the authors of this paper have not attempted to list them.

The present paper is the result of a study of the effect of neutron bombardment on three properties of graphite, i.e. physical dimensions, crystal dimensions (c-axis) and heat content. Since the changes in these properties had been shown by previous investigators to depend on temperature of irradiation as well as on exposure the experiments were carried out over a range of irradiation temperatures.

* This work was done under contract with the U. S. Atomic Energy Commission.

[1] F. Seitz, Disc. Faraday Society No. 5, 271 (1949).

[2] An excellent historical review is given by F. Seitz, Physics Today **5**, 6 (1952).

II. SPECIMENS

Three types of samples were used in this study. The first set consisted of $\frac{1}{2}''$ diameter cylinders approximately 4″ long. These were used primarily for physical expansion measurements. The long axis of the specimen was parallel to the axis of extrusion of the graphite bar. These will be referred to as E samples. These samples were irradiated in experimental holes which are parallel to and between the fuel bearing channels. The second type of sample was used for heat content and crystal expansion measurements and was obtained from the containers in which the E samples were irradiated and were either 1″ diameter by 1″ long cylinders or 1″ cubes. These will be referred to as EC samples. The third set of samples consisted of parts of a solid cylinder (the whole cylinder would be about 1″ diameter and $\frac{1}{2}$ to $\frac{3}{4}''$ in length). These samples (C) which were obtained by means of a coring tool inserted into an empty fuel channel were used for heat content measurements. The first two types of samples differ from the last in one important way, namely, that for the same integrated flux (given in terms of thermal neutrons) the percentage of fast neutrons bombarding the specimen is much greater in the case of the C (cored) samples.

III. METHOD OF MEASUREMENT

A. *Heat Content*

The sample for this measurement, which as indicated above was small, was placed in the center of a cylindrical brass furnace $4\frac{1}{4}''$ I.D. and $29\frac{1}{2}''$ long. The space between the sample and the inner wall was filled with the insulating material "santocel". The brass cylinder was quickly raised to the desired temperature, either 200° or 400°C, and this temperature was maintained constant by means of a thermocouple, attached at the midpoint of the brass cylinder furnace. The temperature of the sample was measured as a function of time by means of a thermocouple imbedded in the specimen. After allowing the furnace and specimen to cool to room temperature, the procedure was repeated. The latter establishes the base heating curve for the specimen in its environs.

B. *Crystal Expansion*

Portions of the EC and C samples were taken before and after the heat content experiments and were polished through 4/0 emery paper. These were then mounted on the sample holder of a North American Philips high angle spectrometer and a trace of the 002 peak was taken. From the angular measurement of the center of the peak the value of the d spacing and therefore the interplanar distance of the 002 planes (c-axis) was calculated.

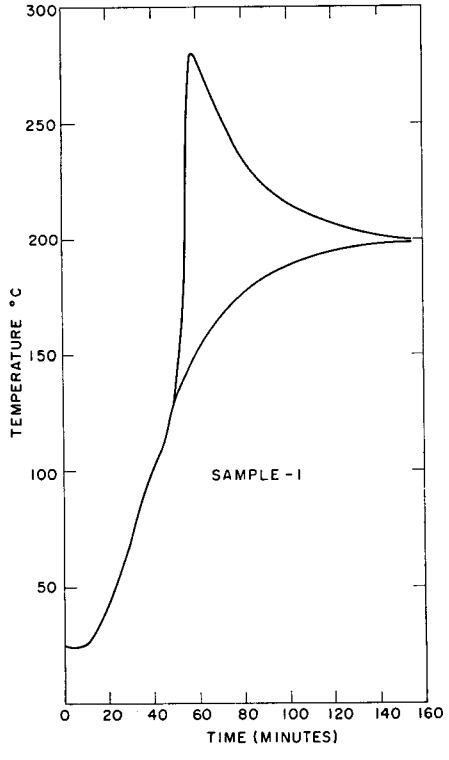

FIG. 1. Time vs temperature plot of a graphite sample in a furnace held at 200°C. Temperature of irradiation—55°C.

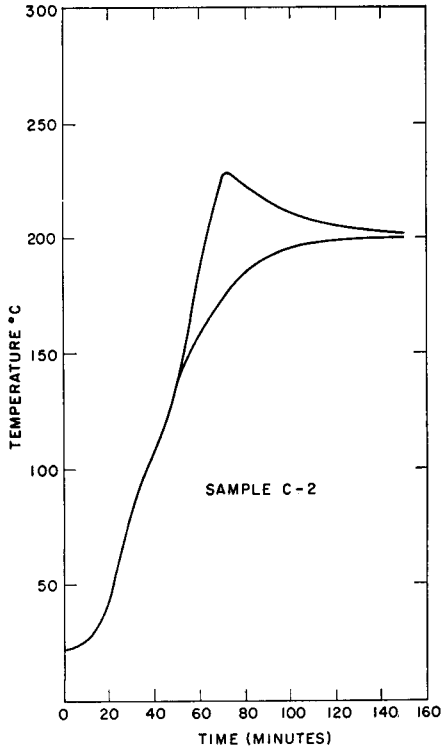

FIG. 2. Time vs temperature plot of a graphite sample in a furnace held at 200°C. Temperature of irradiation—65°C.

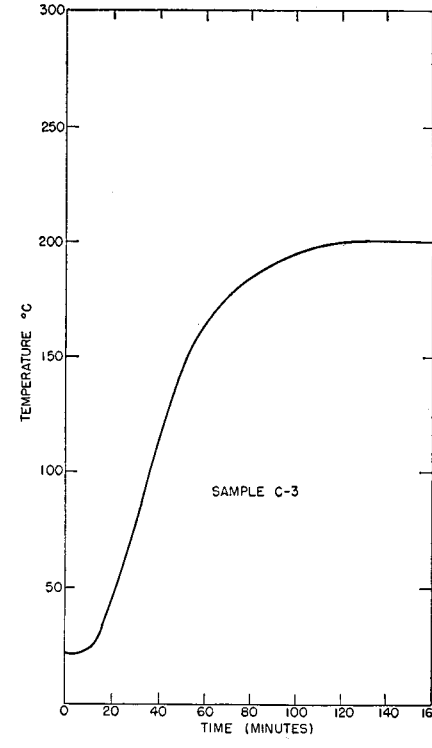

FIG. 3. Time vs temperature plot of a graphite sample in a furnace held at 200°C. Temperature of irradiation—150°C.

C. *Physical Dimensions*

The E samples, which were the only ones used for this measurement, were provided with squared ends. They were measured before and after irradiation by means of a Sheffield Comparator and gauge blocks on a 3.98000″ gauge length. As a rule thirteen individual readings were taken across the ends of the cylinder.

IV. RESULTS

A few representative examples of temperature-time plots for the determination of heat content are presented in Figures 1–6. The upper curve in these figures is obtained on the first heating of the irradiated sample whereas the lower curve is the result of the second heating of the same sample. Most of the irradiated samples showed an increase in heat content. This increase in heat content will be referred to as stored energy. Figures 1 through 3 are the plots obtained on samples which showed a maximum, medium and no stored energy when heated to 200°C. One of the samples which had been heated to 200°C (see Figure 2) was then heated to 400°C to obtain Figure. 4. Figures 5 and 6 were obtained by heating identical samples to 200 and 400°C respectively.

The calculated values of stored energy given in Table I were obtained from the experimental heating curves by a mathematical analysis of the heat flow in this experiment.[3] The stored energy may be roughly calculated as the product of the temperature rise (i.e. the difference between the temperature at which the first heating curve deviates from the second heating curve and the maxi-

[3] We are indebted to J. Chernick of BNL for this calculation.

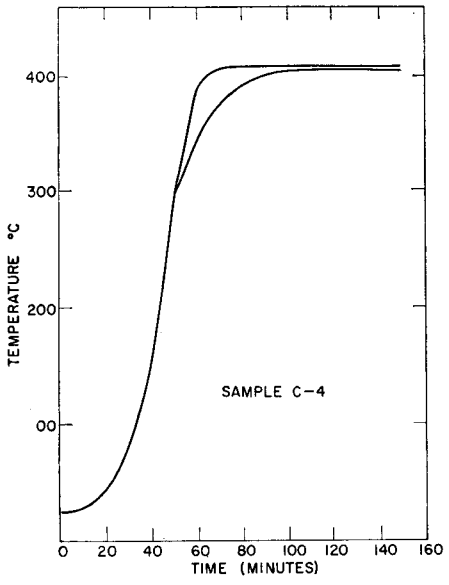

FIG. 4. Time vs temperature plot of a graphite sample in a furnace held at 400°C. Temperature of irradiation—65°C. Some of the stored energy of this sample was released in a furnace held at 200°C. See Figure 2.

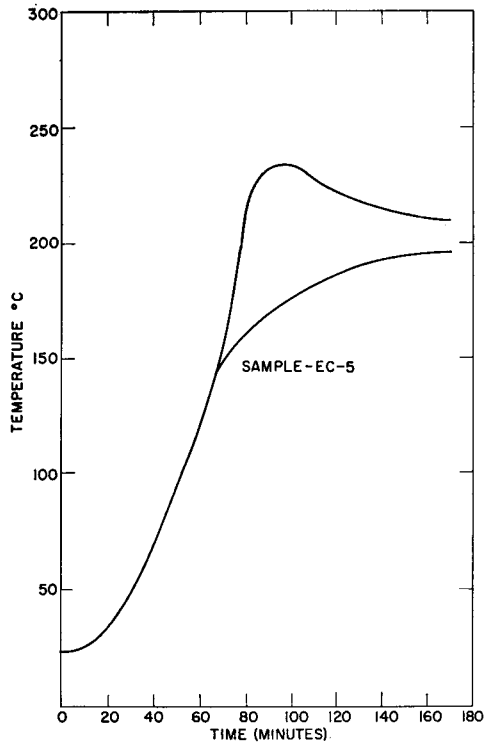

FIG. 5. Time vs temperature plot of a graphite sample in a furnace held at 200°C. Temperature of irradiation—60°C.

mum temperature attained) and the average specific heat of graphite, .25 to .30 cal/g/°C. The more rapid the temperature rise the better is this approximation. Values calculated in this manner agree reasonably well with those calculated by Chernick but are apt to be lower. For example, in Figure 1 the temperature difference is about 152°C. Thus the energy released is approximately 45 cal/g which compares favorably with the calculated value of 53.6 cal/gm.

The results obtained on the physical growth and c-axis measurements are given in Table II and Figure 7 and Table III and Figure 8, respectively.

V. DISCUSSION

The data in the previous section show some of the interesting changes in physical properties which occur upon displacing atoms by means of neutron irradiation. It is well known that radiation effects can be annealed out by heat treatment at elevated

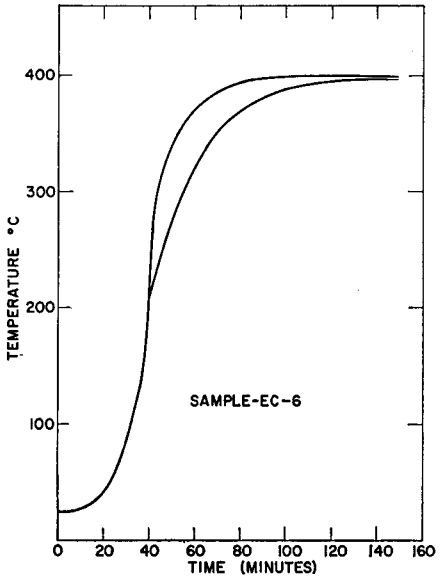

FIG. 6. Time vs temperature plot of a graphite sample in a furnace held at 400°C. Temperature of irradiation—60°C.

TABLE I

Stored Energy (Heat Content)

Sample Number	Average Irradiation Temperature, °C	$nvt \times 10^{20}$	Maximum Temp. Reached, °C	Energy Released on Heating, cal/gram	Furnace Temp., °C
C-1	55	1.13	281.8	53.6*	200
C-2	65	1.61	228.0	24.0	200
C-3	150	1.66	200.0	00.0	200
C-4	65	1.61	400.5	—†	400.5
EC-5	60	1.19	234.0	24.9*	200
EC-6	60	1.19	400.5	43.9*	400.5

* Values of stored energy calculated by J. Chernick. Other values calculated using approximate method outlined previously.

† No calculation done on Sample C-4.

TABLE II

Physical Growth

Sample Number	Average Irradiation Temp., °C	$nvt \times 10^{20}$	Change in Length in 3.98000″ Gauge Length Sample Due to Irradiation (inches)	% Growth
E-1	50	1.18	+.00696	.18
E-2	55	1.19	+.00671	.17
E-3	65	1.20	+.00545	.14
E-4	85	1.21	+.00525	.13
E-5	120	1.20	+.00371	.093

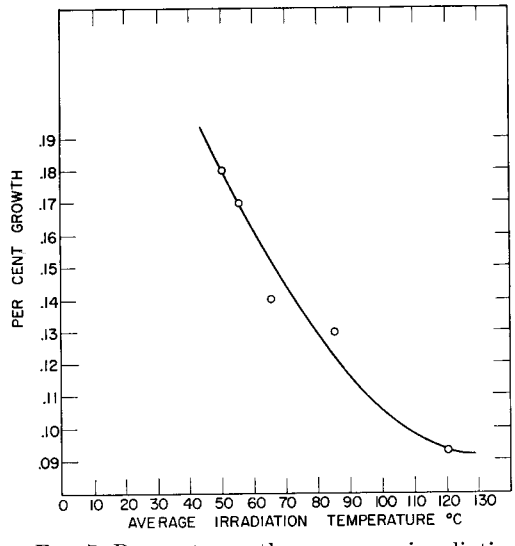

FIG. 7. Per cent growth vs average irradiation temperature of graphite samples. Integrated flux 1.2×10^{20} neutrons/cm².

TABLE III

C-axis Growth

Sample Number	Average Irradiation Temp., °C	$nvt \times 10^{20}$	C-axis After Irradiation (angstroms)	% C-axis Expansion*
EC-4	50	1.18	6.821	1.8
EC-5	60	1.19	6.807	1.6
EC-7	130	1.20	6.735	0.54
EC-8	50	1.18	6.817	1.7
EC-9	60	1.19	6.802	1.5
EC-10	85	1.21	6.775	1.1
EC-11	130	1.20	6.753	0.81

* Calculation based on c-axis = 6.699 angstroms for non-irradiated graphite.

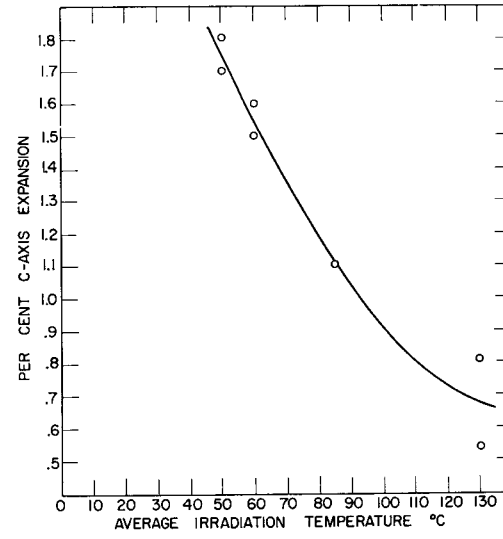

FIG. 8. Per cent c-axis expansion vs average irradiation temperature of graphite samples. Integrated flux 1.2×10^{20} neutrons/cm².

temperatures.[4] The recovery or healing of the lattice is due to the migration of the radiation induced defects out of the crystal, their mutual annihilation or aggregation into less effective clusters. The changes produced by an exposure at a given temperature depend on two competing processes, namely, the rate of damage production and the rate

[4] For general reviews, see J. C. Slater, J. Appl. Phys. **22,** 237 (1951) and G. J. Dienes, Ann. Rev. of Nucl. Sci. **2,** 187 (1953).

of annealing. At low temperature, annealing is slow and defect production depends on the rate of bombardment and is expected to vary linearly with total integrated flux. At temperatures where appreciable annealing takes place a typical growth curve, characterized by a steadily decreasing slope, is expected when defect concentration, or an associated change in physical property, is plotted against integrated flux. Such curves have been published for various metals.[5, 6] Thus, radiation effects are expected to approach a limiting value at any given temperature whenever annealing is important, and since annealing is a thermally activated process the amount of attainable damage will be a more sensitive function of the temperature of exposure than of the integrated flux.

According to the data presented graphite exhibits a behavior which is quite similar to other crystalline materials and follows the above outlined general pattern. Each physical property measured is briefly discussed below.

A. *Stored Energy*

Although the data presented in Table I are meager it is obvious that the extent of damage as determined by the stored energy measurement is not linear with exposure (integrated flux). Rather, as pointed out above, the data suggest that the amount of stored energy is more dependent on the temperature of exposure than on the integrated flux at these levels of exposure. The fact that more energy is released on heating to 400°C shows that annealing is an activated process.

B. *Physical Growth*

Similar trends are shown by the physical growth data of Table II. Expansion decreases with temperature of exposure as shown in Figure 7 again in agreement with the ideas presented above.

C. *C-axis Growth*

C-axis, or crystallite expansion, in general parallels changes in gross physical dimension with one important difference. C-axis expansion is about a factor of 9 larger than gross physical expansion over the range of exposures and temperatures covered in these experiments. Crystallite expansion is probably due to the presence of interstitial atoms since far larger strains are associated with these defects than with vacant lattice sites. The difference between crystallite expansion and physical expansion is probably attributable to the porosity of graphite, i.e. there is considerable room for crystallite expansion without a correspondingly large change in the overall dimensions of the specimen.

ACKNOWLEDGEMENT

The authors are indebted to Otto Kammerer and J. Sadofsky for the *c*-axes measurements and Eugene F. Walsh for the physical growth measurements. Also thanks are due to R. W. Powell for supplying the samples and general aid in performing these experiments.

[5] J. W. Marx, H. G. Cooper, and J. W. Henderson, Phys. Rev. **88**, 106 (1952).

[6] A. W. Overhauser, Phys. Rev. **90**, 393 (1953).