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BORONATED-GRAPHITE IRRADIATION STUDIES -
FINAL REPORT OF IRRADIATION EXPERIMENTS CONDUCTED
IN A HANFORD REACTOR

By

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BORONATED-GRAPHITE IRRADIATION STUDIES -
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INTRODUCTION

This final report presents data gathered from the boronated graphite study program at Hanford for the Enrico Fermi Reactor. In July 1963, work was started on this study for the Power Reactor Development Company (PRDC) under the aegis of the Chicago Operations Office of the U. S. Atomic Energy Commission. The study involved irradiation tests of two grades of graphite, each containing either 5 or 7 wt% boron carbide (B_4C). Auxiliary tests were conducted on the material before and after irradiation.

Conducted in a Hanford production reactor, the first irradiations were a series of heat-transfer tests in the Snout Facility to establish the capsule design. Long-term tests were then begun in the 2C Facility, which was designed on the basis of initial Snout irradiations and installed before the Snout tests were completed.

Three long-term irradiations were performed in the 2C Facility. Each of the capsules contained three independent sections in a single housing to provide level temperature zones, two at 500°C and one at 370 °C. Generally, samples from the first irradiation were reirradiated in the second and third capsules. Some new samples were added after each irradiation in two sections to replace samples that were destructively tested. Each capsule was fully instrumented with 12 thermocouples to continuously record operating temperatures of the samples and critical points of the capsule.

Supplemental irradiations were performed in the Snout Facility to determine radiation damage to the boronated materials at low neutron exposures. Capsules for these tests were smaller than the 2C capsules and each contained only a single sample-holder section. Thermocouple-monitored tests were conducted at temperatures ranging from 150 to 350 °C.

Other irradiations of boronated graphite performed in thermal-neutron-shielded facilities in the Engineering Test Reactor (ETR) were not a part of this program but are discussed in the pertinent areas of this report.

SUMMARY

Boronated graphites of the type installed in the Enrico Fermi Reactor were irradiated at 370 °C (700 °F) and 500 °C (930 °F) to determine dimensional stability, strength, and a number of other properties. Testing was conducted initially in a Hanford Reactor because:

- (1) No fast-flux testing facilities (which would have been more appropriate) were available.
- (2) At the beginning of the work, the heat-generation rate of the material during irradiation was uncertain and, consequently, rapid development of prototype equipment was needed to assure desired irradiation temperatures.
- (3) Tests in a predominantly thermal-neutron flux were expected to result in at least as much, and probably more, damage than tests in a fast-neutron spectrum, thereby accentuating any problems in the use of the material.

The boronated graphites tested were nominally 5 and 7 wt% boron carbide in a nuclear-graphite matrix. The graphites were designated "black" or "grey" depending on the nature of the boron distribution. Except for the presence of boron-carbide particles, the structure of the black material was similar to that of normal nuclear graphites. The grey material, however, had a shiny, metallic appearance because of extensive crystallization. It is postulated that the grey material, which was located in the central part of the graphitizing furnace, reached the melting point of the boron carbide. This promoted the recrystallization of the graphite and allowed the boron to become more uniformly dispersed in the graphite lattice.

Large differences were found in the behavior of the recrystallized grey material compared to the black material. Irradiation effects were more severe in the grey, probably due to the presence of interlayer and substitutional boron in the graphite lattice. The close proximity of the boron to the graphite allowed energetic fission products of the $B^{10}(n, \alpha) Li^7$ reaction to interact with the graphite lattice, causing many more displacements and consequent damage. The black material was less drastically

affected because most of the fission-product damage was centered in the B_4C particles which are loosely coupled to the lattice.

Length changes induced by neutron irradiation were generally positive, ranging up to 3% for the grey material. Compressive strengths were increased and thermal conductivities were lowered. No generalized fracturing was observed in any sample irradiated, which would indicate that powdering or failure of the material probably would not occur.

In addition to the Hanford irradiation, a capability was developed to perform thermal-neutron-shielded irradiations in the ETR, and data at higher fast-to-thermal ratios were obtained. The results helped to reveal the effects of various neutron spectra.

MATERIALS

Five grades of boronated graphite and one nuclear grade, nonboronated graphite, CSF, were studied. The CSF sample was included to determine the relative effect of boronation on radiation damage. Most of the testing was confined to four types of materials obtained from the production run of boronated graphite for the Fermi Reactor shield. All were made by adding B_4C particles to the petroleum-coke, pitch-binder mix. These four types were designated "D, " "E, " "H, " and "J, " corresponding to 5 wt% boron-black, 5 wt% boron-grey, 7 wt% boron-black, and 7 wt% boron-grey, respectively. One grade tested contained 3 wt% boron as B_4C similar to that installed in the Experimental Breeder Reactor II.

The terms "black" and "grey" were applied by the graphite manufacturer and related to the visual appearance of the material. Less metallic and lustrous than the grey, the black material was microscopically similar to polycrystalline, nuclear-grade graphite. The boron carbide was visible as angular particles with no evidence of melting in the 5 wt% material. The 7 wt% black showed evidence of melted B_4C .

Generalized melting of the B_4C appeared to have occurred in the grey material, as evidenced by conformity of the B_4C particles to the adjacent surfaces of the graphite particles. The grey boronated graphites,

E and J, differ primarily in the degree of recrystallization of the graphite. It is likely that the greater B_4C content of the 7 wt% material was responsible for the more complete alteration.

Photomicrographs of some of the boronated graphites tested (Figures 17 and 18, pp. 32 and 33) show the alteration of the graphite and appearance of the B_4C ; other illustrations referred to later show photomicrographs of samples after irradiation.

The ANL graphite, used in the EBR II, was a black nonrecrystallized material containing 3 wt% boron as B_4C . This material evidently contained more impurities than did the Fermi shield material and consequently was more radioactive after irradiation.

Preliminary phase diagrams⁽¹⁾ indicate that the solubility of boron in graphite is approximately 2.3% at the melting point of boron carbide (2450 °C). The concentration of boron in the graphite crystals for the materials studied is not known, but it is presumed that the amount increases in the order of the samples D, H, E, and J. Boron analyses of the material do not conform exactly to the 5 and 7 wt% description; however, for discussion purposes it is convenient to refer to the nominal percentages. Table I presents the boron concentration measured in the samples tested at Hanford.

The sample blocks were cubes of approximately 6 inches with the furnace number and other manufacturer's designations inscribed on each piece. An arrow mark was used to indicate the direction parallel with extrusion. On receipt, all four specimens were marked with a sampling scheme (Figure 1) to relate the location of the samples in the original block to the observed irradiation effects. Throughout the entire process, strict control was maintained to prevent any error in sample identification.

IRRADIATION TECHNIQUES

Preparation of Samples. For the 2C and Snout experiments, boronated-graphite samples, were machined from rough-cut bars previously sectioned from larger blocks according to the sampling schedule discussed in the preceding section. Because the boronated graphite was very abrasive,

TABLE I
BORON AND LITHIUM CONTENT OF UNIRRADIATED
BORONATED GRAPHITE

Sample	Description	Corner	Boron Concentration, wt%	Lithium Concentration, ppm
D	5 wt% black	1	6.05	12
		2	6.26	1.2
		3	6.33	2.2
		4	6.05	2.6
E	5 wt% grey	1	6.25	< 1.5
		2	6.44	< 1.5
		3	6.54	< 1.5
		4	6.45	< 1.5
H	7 wt% black	1	8.28	2.8
		2	7.96	2.3
		3	8.16	2.1
		4	8.59	2.2
J	7 wt% grey	1	7.74	0.92
		2	7.81	0.90
		3	8.04	1.2
		4	8.75	1.5

FIGURE 1
Sampling Scheme
for Boronated Graphite

diamond-tipped tools were used to machine the samples into cylinders 0.750 ± 0.001 in. long and nominally 0.4500 ± 0.0005 in. in diameter. After each sample was completely machined, the appropriate sample designation was inscribed on the specimen to identify its former location and orientation in the block.

Lengths, diameters, and weights were accurately determined for the samples before irradiation so that radiation-induced changes in the graphite could be measured. A minimum of six length measurements were made for each length determination with the use of a Bausch and Lomb mechanical-optical gage accurate to within 0.000025 in. From these measurements, the average length, standard deviation, and the T-limit at 95% confidence were calculated. The diameters of the samples were measured by a micrometer caliper to the nearest 0.0001 in., primarily for purposes of heat-transfer gas-annulus adjustment and secondarily as an indication of physical distortion of the diameter. Sample weights were determined to 0.0001 g. To determine damage effects, identical measurement techniques were used after irradiations.

Capsule Fabrication. The design of the irradiation capsules used in the long-term 2C experiments and the auxiliary short-term Snout experiments evolved from the preliminary concepts used in the initial heat-transfer studies. Preliminary test irradiations validated the expected high heat-generation rate of the boronated-graphite samples and the variability of heating rate due to differences in boron concentration.

The 2C capsules consisted of three independent sections contained in a continuous aluminum tube (Figure 2). The three sections were designed to provide temperature equalization of the samples at the desired temperatures of irradiation. The holder sections consisted of hollow graphite cylinders, 1.00 in. outside diameter, within which the boronated-graphite samples were located. Integral projections, or "feet," having square cross sections positioned the graphite cylinders within the aluminum can and served as the principal conductance to the water-cooled wall. By regulating the gap between the samples and the graphite tube and the size and number of feet, the desired temperature and temperature distribution of the samples in a

FIGURE 2
2C Capsule

given neutron flux were achieved. Initial design values, established by heat-transfer computer calculations and adjusted by heat-transfer tests, proved quite satisfactory. The first two capsules were geometrically identical, but modification was necessary for the 2C-3 capsule because of decreased heat generation due to burnup of B^{10} in the samples.

The three sections of the 2C capsule were designed to operate at 538 °C (1000 °F), 538 °C (1000 °F), and 343 °C (650 °F). An inert gas blanket for the samples was assured by using an all-welded construction wherever possible and a static gas makeup system which was pressure monitored. Grade "A" U. S. Bureau of Mines helium was introduced to the capsule at a pressure of 18 psia to assure outflow in the case of a leak. Although provision was made to replenish the gas during irradiation, it was not necessary to do so in any of the experiments.

Twelve thermocouples connected to a continuous recording system entered the capsule through a high-pressure, molded-glass hermetic seal. These thermocouples monitored the temperatures of eight samples in the capsule and four positions in the internal graphite sample holders. At the termination of the irradiation, the average temperature of irradiation was determined by integrating the time-temperature curves and dividing by time. The irradiation temperatures calculated by this method were 500 ± 50 °C for high-temperature samples (two sections) and 370 ± 30 °C for the low-temperature samples (one section).

The incident neutron flux was measured with nickel, iron, titanium, and cobalt flux monitors. Integral cross-sections for the fast-neutron monitors were determined from best estimates of the differential cross-sections for the materials and the calculated neutron spectra for the irradiation facility.

Auxiliary test capsules irradiated in the Snout contained only one sample-holder section and temperature monitoring was accomplished with eight thermocouples. Experiments in this short-term series were performed at temperatures of 149 to 343 °C. Other design features of these capsules are like those of the 2C apparatus.

Irradiation Facility. Based on thermal and hydraulic requirements established in the prototype testing phase, a special facility was constructed to irradiate the 2C experiment series in a Hanford reactor.

The facility was a double-walled aluminum tube which penetrated the reactor from side-to-side in a direction perpendicular to the fuel tubes. Water was delivered to the inner tube surrounding the capsule at 150 psig and 25 gpm as established by an orifice plate installed in the exit. Water was also introduced in the annular space of the double tube so that failure of the 2C experiment would not affect reactor operations.

The auxiliary Snout tests were performed in an existing facility like the 2C facility, but one capable of charge-discharge operations while the reactor was at power.

Discharge and Disassembly of Capsules. After completion of each irradiation period, the capsules were discharged into a transfer cask.

Initial disassembly of the capsule was done remotely in a shielded hot cell. Samples holders and the flux-monitor holders were removed from the aluminum can, and all excess radioactive material was stripped from the holders. They were then removed from the hot cell for further disassembly by hand, and the samples were transferred to the graphite laboratories for study. In the 2C experiments, the samples were irradiated, measured and then reirradiated. This allowed measurement of damage to the boronated graphite to be determined throughout a range of exposures on individual samples.

EXPERIMENTAL RESULTS

Length Changes. Length change induced by irradiation was the most important engineering parameter measured in this program. The results of four short-term supplemental irradiations of 1.17×10^{20} n/cm² (thermal)* temperatures of 149 °C (300 °F), 232 °C (450 °F), 336 °C (625 °F), and 343 °C (650 °F) are shown in Tables II through V. The results of the 2C experiments at 370 and 500 °C are given in Tables VI through XV. Mean values of the temperatures were computed from thermocouple readings taken during the irradiations and are reported in each table. For convenience in presentation, the data for irradiation temperatures of 370 and 500 °C have been combined in Figures 3 through 10.

TABLE II
LENGTH CHANGES AT 1.17×10^{20} n/cm² FOR IRRADIATION AT 149 °C (300 °F)

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 8LB	- 0.003 ± 0.026	
D15TD		+ 0.205 ± 0.013
D17TB		+ 0.235 ± 0.028
<u>Grey, 5 wt% Boron</u>		
E12LA	+ 0.006 ± 0.002	
E12LB	+ 0.050 ± 0.019	
E15TD		+ 0.039 ± 0.001
E16TB		+ 0.071 ± 0.012
<u>Black, 7 wt% Boron</u>		
H 8LA	+ 0.015 ± 0.013	
H15TC		+ 0.195 ± 0.015
H16TB		+ 0.149 ± 0.012
<u>Grey, 7 wt% Boron</u>		
J17LA	+ 0.031 ± 0.023	
J17LB	+ 0.017 ± 0.017	
J15TC		+ 0.064 ± 0.020
J15TD		- 0.013 ± 0.012

*All exposures given are in units of thermal neutrons per cm² unless otherwise indicated.

TABLE III
 LENGTH CHANGES AT $1.17 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 232 °C (450 °F)

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 7LB	- 0.035 ± 0.010	
D 8LA	+ 0.015 ± 0.006	
D15TB		+ 0.206 ± 0.025
D17TA		+ 0.134 ± 0.033
<u>Grey, 7 wt% Boron</u>		
E 7LB	- 0.057 ± 0.012	
E10LB	- 0.049 ± 0.006	
E15TB		- 0.007 ± 0.004
<u>Black, 5 wt% Boron</u>		
H 6LB	- 0.014 ± 0.006	
H15TB		+ 0.171 ± 0.055
H16TA		+ 0.142 ± 0.005
<u>Grey, 7 wt% Boron</u>		
J13LB	- 0.052 ± 0.028	
J16LB	- 0.032 ± 0.006	
J15TB		- 0.032 ± 0.019
J16TA		+ 0.004 ± 0.013

TABLE IV
 LENGTH CHANGES AT $1.17 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 336 °C (625 °F)

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D13LA	- 0.041 ± 0.018	
D13LB	- 0.079 ± 0.044	
D17TC		+ 0.039 ± 0.033
D17TD		+ 0.117 ± 0.030
<u>Grey, 5 wt% Boron</u>		
E14LA	+ 0.008 ± 0.046	
E14LB	- 0.106 ± 0.010	
E17TA		- 0.052 ± 0.042
E17TB		- 0.073 ± 0.016
<u>Black, 7 wt% Boron</u>		
H11LA	- 0.060 ± 0.027	
H11LB	- 0.060 ± 0.044	
H17TB		- 0.171 ± 0.016
H17TC		+ 0.096 ± 0.023
<u>Grey, 7 wt% Boron</u>		
J17TA		- 0.078 ± 0.029
J17TB		- 0.091 ± 0.022

TABLE V
 LENGTH CHANGES AT $1.17 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 343 °C (650 °F)

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 7LA	- 0.078 ± 0.011	
D15TA		+ 0.050 ± 0.019
D15 TC		+ 0.032 ± 0.020
<u>Grey, 5 wt% Boron</u>		
E 7LA	- 0.094 ± 0.013	
E10LA	- 0.099 ± 0.068	
E15LA		- 0.065 ± 0.011
E16LA		- 0.056 ± 0.017
<u>Black, 7 wt% Boron</u>		
H 6LA	- 0.049 ± 0.013	
H15TA		+ 0.076 ± 0.015
<u>Grey, 7 wt% Boron</u>		
J13LA	- 0.056 ± 0.013	
J16LA	- 0.069 ± 0.009	
J15TA		- 0.073 ± 0.004

TABLE VI
 LENGTH CHANGES AT $7.2 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 371 °C (700 °F)
 2C-1 EXPERIMENT

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 2TA		+ 0.043 ± 0.006
D 2TB		+ 0.060 ± 0.006
D10LA	- 0.199 ± 0.003	
<u>Grey, 5 wt% Boron</u>		
E10TA		+ 0.058 ± 0.006
E10TB		+ 0.074 ± 0.006
E 9LA	- 0.199 ± 0.005	
E 9LB	- 0.221 ± 0.002	
<u>Black, 7 wt% Boron</u>		
H 1TA		+ 0.103 ± 0.005
H13LA	- 0.169 ± 0.003	
<u>Grey, 7 wt% Boron</u>		
J 1TA		+ 0.086 ± 0.005
J 6LA	+ 0.111 ± 0.004	
<u>Black, 3 wt% Boron (ANL)</u>		
A 5TC		+ 0.026 ± 0.006
A 1LC	- 0.161 ± 0.010	
<u>Black, 0 wt% Boron (CSF)</u>		
142-3A 310 °C (590 °F)		+ 0.004 ± 0.003

TABLE VII
 LENGTH CHANGES AT 7.2×10^{20} n/cm³
 FOR IRRADIATION AT 480 °C (900 °F)
 2C-1 EXPERIMENT

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 3TA		- 0.098 ± 0.004
D 3TB		- 0.050 ± 0.005
D15LA	- 0.253 ± 0.005	
D15LB	- 0.228 ± 0.005	
<u>Grey, 5 wt% Boron</u>		
E11TA		+ 0.056 ± 0.006
E11TB		+ 0.045 ± 0.004
E11TC		+ 0.024 ± 0.007
E11TD		- 0.063 ± 0.006
E 3LA	- 0.215 ± 0.007	
E 3LB	- 0.230 ± 0.006	
E 8LA	- 0.162 ± 0.004	
E 8LB	- 0.206 ± 0.005	
<u>Black, 7 wt% Boron</u>		
H12TA		+ 0.048 ± 0.005
H12TB		+ 0.075 ± 0.002
H 1LA	- 0.145 ± 0.004	
H 1LB	- 0.140 ± 0.002	
<u>Grey, 7 wt% Boron</u>		
J 6TA		+ 0.035 ± 0.007
J 6TB		+ 0.083 ± 0.003
J 6TC		- 0.012 ± 0.003
J 6TD		- 0.086 ± 0.007
J14LA	+ 0.037 ± 0.002	
J14LB	- 0.016 ± 0.005	
J15LA	+ 0.049 ± 0.007	
J15LB	+ 0.019 ± 0.004	
<u>Black, 3 wt% Boron</u>		
A 5TA		+ 0.196 ± 0.005
A 5TB		+ 0.124 ± 0.007
A 1LA	- 0.111 ± 0.002	
A 1LB	- 0.118 ± 0.005	

TABLE VIII
 LENGTH CHANGES AT 9.8×10^{20} n/cm²
 FOR IRRADIATION AT 371 °C (700 °F)
 2C-2 EXPERIMENT

Sample	$\Delta E/E, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 6LA	- 0.148	
D13TC		+ 0.237
D13TD		+ 0.411
<u>Grey, 5 wt% Boron</u>		
E 5LA	+ 0.185	
E13TC		+ 0.493
<u>Grey, 7 wt% Boron</u>		
J12LA	+ 0.920	

TABLE IX
 LENGTH CHANGES AT $9.8 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 496 °C (925 °F)
 2C-2 EXPERIMENT

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D 4LB	- 0.147	
D13TB		+ 0.166
<u>Grey, 5 wt% Boron</u>		
E 4LA	+ 0.088	
E 4LB	+ 0.086	
E13TA		+ 0.426
E13TB		+ 0.429
<u>Black, 7 wt% Boron</u>		
H 5LA	- 0.241	
H13TA		+ 0.189
<u>Grey, 7 wt% Boron</u>		
J 7LA	+ 0.322	
J 7LB	+ 0.299	
J13TA		+ 0.588
J13TB		+ 0.481

TABLE X
 LENGTH CHANGES AT $16.9 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 371 °C (700 °F)
 2C-2 EXPERIMENT

Sample	$\Delta L/L, \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D10LA	- 0.303	
D 2TA		+ 0.271
<u>Grey, 5 wt% Boron</u>		
E 9LB	+ 0.122	
E10TB		+ 1.074
<u>Black, 7 wt% Boron</u>		
H13LA	- 0.341	
H 1TA		+ 0.329
<u>Grey, 7 wt% Boron</u>		
J 1TA		+ 1.539
<u>CSF</u>		
142-3A 310 °C (590 °F)		- 0.169

TABLE XI
 LENGTH CHANGES AT $16.9 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 454°C (850°F)
 2C-2 EXPERIMENT

<u>Sample</u>	<u>$\Delta L/L, \%$</u>	
	<u>Longitudinal</u>	<u>Transverse</u>
<u>Black, 5 wt% Boron</u>		
D15LA	- 0.607	
D 3TA		- 0.297
<u>Grey, 5 wt% Boron</u>		
E 3LA	+ 0.123	
E 8LA	+ 0.125	
E11TA		+ 0.821
E11TC		+ 0.749
<u>Black, 7 wt% Boron</u>		
H 1LA	- 0.359	
H12TA		+ 0.270
<u>Grey, 7 wt% Boron</u>		
J14LA	+ 0.931	
J15LA	+ 0.958	
J 6TA		+ 1.164
J 6TC		+ 0.950
<u>Black, 3 wt% Boron</u>		
A 1LA	- 0.090	
A 5TA		+ 0.389

TABLE XII
 LENGTH CHANGES AT $18.2 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 370°C
 2C-3 EXPERIMENT

<u>Sample</u>	<u>$\Delta L/L, \%$</u>	
	<u>Longitudinal</u>	<u>Transverse</u>
<u>Black, 5 wt% Boron</u>		
D6L-A	- 0.224	
D13T-D		+ 0.975
<u>Grey, 5 wt% Boron</u>		
E5L-A	+ 1.144	
E13T-C		+ 1.978
<u>Grey, 7 wt% Boron</u>		
J12L-A	+ 3.333	

TABLE XIII
 LENGTH CHANGES AT $18.2 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 500°C
 2C-3 EXPERIMENT

Sample	$\Delta L/L \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D4L-B	- 0.181	
D13T-B		+ 0.388
<u>Grey, 5 wt% Boron</u>		
E4L-A	+ 0.426	
E4L-B	+ 0.366	
E13T-A		+ 1.060
E13T-B		+ 1.240
<u>Black, 7 wt% Boron</u>		
H5L-A	- 0.244	
H13T-A		+ 0.385
<u>Grey, 7 wt% Boron</u>		
J7L-A	+ 1.217	
J7L-B	+ 1.124	
J13T-A		+ 1.989

TABLE XIV
 LENGTH CHANGES AT $25.3 \times 10^{20} \text{ n/cm}^2$
 FOR IRRADIATION AT 370°C
 2C-3 EXPERIMENT

Sample	$\Delta L/L \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D10LA	- 0.396	
D2TA		+ 0.449
<u>Grey, 5 wt% Boron</u>		
E9LB	+ 0.461	
E10TB		+ 2.275
<u>Black, 7 wt% Boron</u>		
H13LA	- 0.407	
H1TA		+ 0.737
<u>Grey, 7 wt% Boron</u>		
J1TA		+ 3.112
<u>CSF</u>		
142-3A 310 °C (590 °F)		- 0.222

TABLE XV
LENGTH CHANGES AT $25.3 \times 10^{20} \text{ n/cm}^2$
FOR IRRADIATIONS AT 500°C
2C-3 EXPERIMENT

Sample	$\Delta L/L \%$	
	Longitudinal	Transverse
<u>Black, 5 wt% Boron</u>		
D15LA	- 0.745	
D3TA		- 0.191
<u>Grey, 5 wt% Boron</u>		
E3LA	+ 0.076	
E8LA	+ 0.277	
E11TA		+ 1.111
E11TC		+ 1.004
<u>Black, 7 wt% Boron</u>		
H1LA	- 0.429	
H12TA		+ 0.432
<u>Grey, 7 wt% Boron</u>		
J14LA	+ 1.890	
J15LA	+ 1.672	
J6TA		+ 2.111

FIGURE 3

Effect of Exposure on 5 wt% (Black) Boronated Graphite - Parallel

FIGURE 4

Effect of Exposure on 5 wt% (Black)
Boronated Graphite - Transverse

FIGURE 5

Effect of Exposure on 7 wt% (Black)
Boronated Graphite - Parallel

FIGURE 6

Effect of Exposure on 7 wt% (Black)
Boronated Graphite - Transverse

FIGURE 7

Effect of Exposure on 5 wt% (Grey)
Boronated Graphite - Parallel

FIGURE 8

Effect of Exposure on 5 wt% (Grey)
Boronated Graphite - Transverse

FIGURE 9

Effect of Exposure on 7 wt% (Grey)
Boronated Graphite - Parallel

FIGURE 10

Effect of Exposure on 7 wt% (Grey)
Boronated Graphite - Transverse

It is seen that the length changes to an exposure of $5 \times 10^{20} \text{ n/cm}^2$ are similar for all graphite types tested, but at exposures exceeding this value pronounced effects attributable to the five different types of boronated graphite begin to appear. The grey 7 wt% material expands rapidly without regard to orientation. In the **transverse** direction, the behavior of the other materials is related to the degree of recrystallization indicated by the photomicrographs. Length changes of parallel samples are similarly ranked but differ from the transverse behavior in that, like nuclear graphite, they tend more toward contraction than the transverse samples.

Early in the irradiation testing of boronated graphite, a systematic difference in length changes was noticed which could be attributed wholly

to the positions in the original block. This effect is most effectively shown with the J6T Samples A, B, C, and D in Table VII. The A and B samples, nearest the center in the original block, expanded while progressively greater contraction was exhibited in the C and D samples. On reirradiation in the 2C-2 experiment, the "A" sample expanded more than did the "C" sample. Other samples showed similar behavior. Since this effect of position was noted within a rod length, a survey was made to relate the sample number across the original bar. This was possible, since some samples were taken from the original bar near the edge, and an edge "A" sample has the same qualitative position with respect to the center as does a "C" sample from a center rod. This too indicated a position effect for transverse samples. In most samples studies, the transverse samples near the center of the block changed the most under irradiation when compared with samples near the edge. No analogous position effect was found for parallel samples.

Some of the materials tested exhibited an effect of the irradiation temperature in the range of 370 to 500 °C. In general the lower temperature caused greater changes in properties. This is consistent with the effect of temperature in nuclear graphite. The 7 wt% black material was found to be relatively free of both an effect of temperature and of sample location. This material exhibited the greatest anisotropy.

All of the boronated graphites tested were relatively "black" to thermal neutrons, thereby inducing steep neutron gradients to the center of the samples. This self-shielding would be expected to cause damage gradients and complex internal stress distributions. Because modification of expected dimensional changes in nuclear graphite has been observed when the samples were stressed by an external load, ^(2, 3) it was expected that irradiation-induced internal restraints might modify the dimensional change that the boronated graphite would otherwise undergo. To check this possibility, an irradiation test was performed at 370 °C to an exposure of 1.2×10^{20} n/cm² with several cylindrical cores having 1/16 in. thick annular shields of identical boronated graphite. The external dimensions of the cores with the shields were the same as the solid samples used in

the 2C irradiations. The results of the test were inconclusive at 1.2×10^{20} n/cm², but significant differences in core and shield lengths were obtained for higher exposures. The grey 7 wt% samples irradiated at 500 °C to 8.4×10^{20} n/cm², exhibited a ratio of shield-to-core growth of 8.4. The ratio for 7 wt% black samples irradiated under identical conditions was 2.09, and at an irradiation temperature of 370 °C, the 5 wt% black produced a ratio of 1.34. In all cases, the core-length changes were in good agreement with the changes found for solid samples. Thus, it appears that the relatively unirradiated "cores" of the solid samples restrain dimensional changes until sufficient burnout is achieved in outer regions, thereby lowering the internal restraints.

Early, short-term irradiation tests of boronated graphites indicated that dimensional changes much larger than those observed in nonboronated nuclear graphites might be expected for a given irradiation temperature and neutron exposure. A single sample of nuclear graphite was, therefore, included in the 2C capsules to provide an index of damage to a nonboronated material under irradiation conditions identical to those of the boronated materials. The sample, a CSF transverse specimen, designated 142-3A, was irradiated at a temperature of 310 ± 40 °C (590 ± 70 °F) to a maximum neutron exposure of 3.0×10^{20} n/cm² ($E > 0.18$ MeV), and dimensional changes were measured after each irradiation. After this exposure, the sample contracted 0.222%. A comparison of the length change of this sample with results for PGA, a British graphite having properties similar to CSF, indicates that the CSF sample contracted ten times more than would be predicted from PGA data. ⁽⁴⁾

A satisfactory explanation for the enhanced contraction found in the CSF sample cannot yet be given. One possibility, however, is that boron was transported from adjacent boronated samples to the CSF material, causing increased displacement damage due to the energetic fission fragments of the $B^{10}(n, \alpha) Li^7$ reaction. Tests are being made to determine if boron and/or lithium are present in the sample.

Mechanical Properties. Mechanical-property tests were conducted on unirradiated samples and on samples irradiated in the 2C experiments at 370 and 500 °C to a maximum thermal-neutron exposure of 25.3×10^{20} n/cm².

The samples were cylinders having a nominal diameter of 0.450 in. Unirradiated samples approximately 0.75 in. in length were used. The irradiated 2C samples were shorter (0.563 in.) due to the removal of wafer sections for other tests. Before testing, the lengths and diameters were measured to the nearest 0.0001 in. and the samples were then sealed within individual 0.002 in. thick plastic bags to prevent contamination of the testing equipment. The compressive load was applied to the samples on an Instron Model TT-C-L testing machine at a constant crosshead speed of 0.005 in./min.

Values of compressive strength, strain and strain energy at failure, and Young's modulus were calculated from test data and the results are shown in Table XVI. Compressive strength is shown as a function of thermal-neutron exposure for each of the four types of boronated graphite (Figures 11 and 12) for parallel and transverse samples, respectively.

TABLE XVI
MECHANICAL PROPERTIES OF BORONATED GRAPHITE IRRADIATED IN 2C EXPERIMENTS

Samples	Exposure, n/cm ² x 10 ⁻²⁰	Irradiation Temperature, °C	Maximum Compressive Stress, psi	Strain Energy to Failure, in.-lb/in. ³	Strain to Failure, %	Young's Modulus, psi x 10 ⁻³
<u>Black, 5 wt% Boron</u>						
D-L Parallel	0	---	4470 ± 160*	58 ± 5*	2.29 ± 0.16*	323 ± 54*
D15 LB Parallel	7.2	500	7600	---	---	---
D15 LA Parallel	25.3	500	9660	98	2.31	478
D-T Transverse	0	---	4090 ± 220*	79 ± 11*	3.32 ± 0.37*	245 ± 18*
D2 TB Transverse	7.2	370	6400	80	2.72	296
D13 TC Transverse	9.8	370	6510	---	---	---
D2 TA Transverse	25.3	370	9040	119	2.85	398
D3 TB Transverse	7.2	500	6220	78	2.30	400
D3 TA Transverse	25.3	500	10,000	127	2.72	432
<u>Grey, 5 wt% Boron</u>						
E-L Parallel	0	---	2830 ± 420*	57 ± 5*	2.94 ± 0.23*	235 ± 35*
E9 LA Parallel	7.2	370	5770	70	2.62	249
E3 LB Parallel	7.2	500	5450	69	2.46	292
E8 LB Parallel	7.2	500	4900	55	2.44	230
E3 LA Parallel	25.3	500	6770	---	---	---
E-T Transverse	0	---	2310 ± 570*	63 ± 22*	4.02 ± 0.50*	216 ± 25*
E10 TA Transverse	7.2	370	5240	80	2.89	220
E13 TC Transverse	18.2	370	4850	67	2.63	269
E11 TD Transverse	7.2	500	4740	76	2.77	303
E11 TA Transverse	25.3	500	6060	85	2.98	259
<u>Black, 7 wt% Boron</u>						
H-L Parallel	0	---	4740 ± 440*	56 ± 8*	2.14 ± 0.28*	354 ± 71*
H13 LA Parallel	25.3	370	9230	---	---	---
H1 LB Parallel	7.2	500	5800	87	3.20	207
H1 LA Parallel	25.3	500	8160	92	2.55	381
H-T Transverse	0	---	4790 ± 280*	98 ± 22*	3.40 ± 0.66*	378 ± 18*
H12 TB Transverse	7.2	500	6930	86	2.27	414
H12 TA Transverse	25.3	500	8310	93	2.58	386
<u>Grey, 7 wt% Boron</u>						
J-L Parallel	0	---	2560 ± 130*	56 ± 6*	3.07 ± 0.08*	261 ± 60*
J6 LA Parallel	7.2	370	4620	59	2.72	212
J14 LB Parallel	7.2	500	4430	58	2.52	229
J15 LB Parallel	7.2	500	4590	51	2.20	271
J14 LA Parallel	25.3	500	4490	---	---	---
J-T Transverse	0	---	2430 ± 250*	64 ± 14*	3.89 ± 0.25*	299 ± 38*
J1 TA Transverse	25.3	370	3780	48	2.58	204
J6 TD Transverse	7.2	500	5010	68	2.66	230
J6 TC Transverse	16.9	500	4210	---	---	---
J13 TA Transverse	18.2	500	3700	---	---	---
J6 TA Transverse	25.3	500	4300	47	2.12	284

*95% confidence limit.

FIGURE 11

Compressive Strength of Boronated Graphite (Parallel)
as a Function of Exposure

FIGURE 12

Compressive Strength of Boronated Graphite (Transverse)
as a Function of Exposure

The strength of all samples increases initially on irradiation, but the 5 and 7 wt% grey materials tend to saturation after 7.5×10^{20} n/cm². The 7 wt% grey material begins to weaken in compression after extended exposure but is still a factor of 2.5 stronger than initially.

The general tendency of graphite to become stronger on irradiation is typical, but the decrease of strength as shown for the 7 wt% grey material is not. The decrease in strength may be due to the fracture of the particles as is seen in the postirradiation photomicrographs shown in Figure 18, p. 33).

Thermal Conductivity. The thermal diffusivity of unirradiated and irradiated boronated graphite was measured by a thermal-pulse method.⁽⁵⁾ The thermal conductivity was calculated by assuming a specific heat for boronated graphite equal to that of nuclear-grade graphite. Thermal conductivities measured on selected samples throughout the irradiation program are shown in Table XVII. Since the preparation of specimens for thermal-conductivity measurements destroyed the original samples, not all samples were measured during the progress of the irradiation.

It is seen from Figures 13 and 14 that fractional changes in resistivity of 12 to 16 can be induced by irradiation to 25×10^{20} n/cm². There is a tendency for lower irradiation temperatures to cause lower thermal conductivities in the range of irradiation temperatures used in this experiment.

Coefficient of Thermal Expansion. The coefficient of thermal expansion (CTE) was measured as a function of neutron exposure for the four principal types of boronated graphite irradiated in the 2C experiments. To avoid the risk of oxidizing or annealing the samples scheduled for reirradiation, a limited temperature range of 25 to 200 °C was used for making the CTE determinations. The results are shown in Table XVIII and Figures 15 and 16.

A satisfactory correlation of CTE with length changes could not be made; however, several comparisons of interest were drawn from the results. The principal observation is that the fractional CTE changes of transverse samples for the 5 and 7 wt% grey materials are approximately 2 to 2-1/2

TABLE XVII
ROOM-TEMPERATURE THERMAL CONDUCTIVITY OF BORONATED GRAPHITE
IRRADIATED IN THE 2C EXPERIMENTS

Sample		Irradiation Temperature, °C	Exposure, n/cm ² x 10 ⁻²⁰	Thermal Conductivity, c. g. s. (British)	
Black, 5 wt% Boron					
D 5 L	Parallel	---	0	0.287	(69.4)
D15 LB	Parallel	500	7.2	0.043	(10.4)
D15 LA	Parallel	500	25.3	0.022	(5.32)
D 4 T	Transverse	---	0	0.205	(49.6)
D 2 TB	Transverse	370	7.2	0.023	(5.56)
D13 TC	Transverse	370	9.8	0.022	(5.32)
D 2 TA	Transverse	370	25.3	0.011	(2.66)
D 3 TB	Transverse	500	7.2	0.032	(7.74)
D 3 TA	Transverse	500	25.3	0.013	(3.15)
Grey, 5 wt% Boron					
E11 L	Parallel	---	0	0.216	(52.2)
E 3 LB	Parallel	500	7.2	0.030	(7.25)
E 8 LB	Parallel	500	7.2	0.030	(7.25)
E 3 LA	Parallel	500	25.3	0.018	(4.36)
E 9 LA	Parallel	370	7.2	0.023	(5.56)
E 8 T	Transverse	---	0	0.193	(46.6)
E10 TA	Transverse	370	7.2	0.022	(5.32)
E13 TC	Transverse	370	18.2	0.012	(2.90)
E11 TB	Transverse	500	7.2	0.028	(6.77)
E11 TD	Transverse	500	7.2	0.027	(6.53)
E11 TA	Transverse	500	25.3	0.011	(2.66)
Black, 7 wt% Boron					
H 7 L	Parallel	---	0	0.202	(48.9)
H 1 LB	Parallel	500	7.2	0.036	(8.71)
H 1 LA	Parallel	500	25.3	0.020	(4.84)
H13 LA	Parallel	370	25.3	0.017	(4.11)
H 5 T	Transverse	---	0	0.142	(34.3)
H12 TB	Transverse	500	7.2	0.023	(5.56)
H12 TA	Transverse	500	25.3	0.015	(3.63)
Grey, 7 wt% Boron					
J11 L	Parallel	---	0	0.137	(33.1)
J 6 LA	Parallel	370	7.2	0.017	(4.11)
J14 LB	Parallel	500	7.2	0.029	(7.02)
J15 LB	Parallel	500	7.2	0.024	(5.80)
J14 LB	Parallel	500	25.3	0.0098	(2.37)
J12 T	Transverse	---	0	0.129	(31.2)
J 6 TD	Transverse	500	7.2	0.022	(5.32)
J 6 TC	Transverse	500	16.9	0.015	(3.63)
J13 TA	Transverse	500	18.2	0.016	(3.87)
J 6 TA	Transverse	500	25.3	0.0088	(2.13)
J 1 TA	Transverse	370	25.3	0.0092	(2.23)

FIGURE 13

Fractional Change in Room Temperature Thermal Resistivity
as a Function of Exposure - Parallel Samples

FIGURE 14

Fractional Change in Room Temperature Thermal Resistivity
as a Function of Exposure - Transverse Samples

TABLE XVIII

THERMAL EXPANSION COEFFICIENTS OF BORONATED GRAPHITES
IRRADIATED IN THE 2C EXPERIMENTS

Sample	Orientation	Temperature, °C	Exposure, n/cm ² x 10 ²⁰	CTE, per °C x 10 ⁶ (25 - 200 °C)
<u>Black, 5 wt% Boron</u>				
D18 TA	Transverse	---	0	3.5
D13 TC	Transverse	370	9.8	4.7
D 2 TA	Transverse	370	25.3	6.0
D13 TB	Transverse	500	18.2	5.3
D 3 TA	Transverse	500	25.3	5.4
D16 LA	Parallel	---	0	1.3
D10 LA	Parallel	370	25.3	2.6
<u>Grey, 5 wt% Boron</u>				
E 5 TC	Transverse	---	0	1.2
E10 TB	Transverse	370	16.9	4.1
E13 TA	Transverse	500	9.8	3.0
E11 TC	Transverse	500	16.9	2.9
E11 TC	Transverse	500	25.3	3.2
E11 TA	Transverse	500	25.3	3.2
E15 LA	Parallel	---	0	1.3
E 9 LB	Parallel	370	16.9	2.2
E 4 LB	Parallel	500	9.8	1.7
E 8 LA	Parallel	500	16.9	2.4
E 3 LA	Parallel	500	25.3	3.0
E 8 LA	Parallel	500	25.3	2.8
<u>Black, 7 wt% Boron</u>				
H 7 TC	Transverse	---	0	4.3
H 1 TA	Transverse	370	16.9	5.0
H 1 TA	Transverse	370	25.3	5.9
H13 TA	Transverse	500	18.2	5.0
H12 TA	Transverse	500	25.3	5.3
H12 LA	Parallel	---	0	2.2
H 1 LA	Parallel	500	16.9	1.9
H 1 LA	Parallel	500	25.3	2.1
<u>Grey, 7 wt% Boron</u>				
J 7 TC	Transverse	---	0	1.6
J 1 TA	Transverse	370	25.3	4.2
J13 TB	Transverse	500	9.8	2.3
J 6 TC	Transverse	500	16.9	3.3
J 6 TA	Transverse	500	25.3	3.7
J12 LB	Parallel	---	0	1.7
J 7 LB	Parallel	500	9.8	1.7
J15 LA	Parallel	500	16.9	3.3
J 7 LB	Parallel	500	18.2	2.2
J15 LA	Parallel	500	25.3	3.0
J14 LA	Parallel	500	25.3	3.0

FIGURE 15

Coefficient of Thermal Expansion of Boronated Graphites - Parallel

FIGURE 16

Coefficient of Thermal Expansion of Boronated Graphites - Transverse

times the changes in the black materials. This is generally in agreement with physical distortion measurements made on similarly oriented samples. Also found for CTE was the isotropic behavior of the 7 wt% grey samples in the parallel and transverse dimensions, as indicated by length changes.

Microscopy. Photomicrographs of boronated graphite have been very useful in interpreting structural differences of the four principal materials studied. Figures 17 and 18 show unirradiated and irradiated samples of the 5 and 7 wt% black and grey materials.

Individual difference of structure were often seen within a single specimen, and so it is difficult to compare specific structural characteristics of different samples. However, generalized differences of the several types of boronated graphite studied are easily demonstrated. The unirradiated black material was most like nuclear-grade graphite. Boron carbide was observed as white, angular particles adjacent to the coke particles of the mix. The coke particles were largely unreacted, although some alteration was seen in the 7 wt% black material. The unirradiated grey material differed greatly from the black; almost total transformation of the original structure was seen. Boron carbide was observed to be nonhomogeneously dispersed in the grey material and was always continuous with adjacent recrystallized graphite surfaces. This is clear evidence that it had at one time melted or, in some cases, precipitated from solution in adjacent grains.

The most striking postirradiation condition observed was the extensive intragranular fracturing of the recrystallized particles in the grey material. Fracturing is attributed to irradiation effects because plastic "potting" material can be seen in the interstices of the material, precluding the possibility that polishing operations were responsible. Also, tests were conducted on other grey samples that were deliberately loaded to failure in axial compression, and no intragranular fracture of the sort seen in Figure 18 (lower right) was observed.

It is proposed that the fracturing occurs from intragranular differential stresses in the direction of the long axis (a direction) of the grain. The stress is hypothesized to result from fibers having different rates of

Unirradiated Black, 5 wt% Boron
Neg. 4K1966 75X

Irradiated Black, 5 wt% Boron
Neg. 465391 75X

Unirradiated Black, 7 wt% Boron
Neg. 4K1970 75X

Irradiated Black, 7 wt% Boron
Neg. 465398 75X

FIGURE 17

Pre- and Postirradiation Structure of Boronated Graphite

Unirradiated Grey, 5 wt% Boron
Neg. 4K1968 75X

Irradiated Grey, 5 wt% Boron
Neg. 465396 75X

Unirradiated Grey, 7 wt% Boron
Neg. 4K1972 75X

Irradiated Grey, 7 wt% Boron
Neg. 465400B 75X

FIGURE 18

Pre- and Postirradiation Structure of Boronated Graphite

contraction due to varying crystallite alignment and/or a nonhomogeneous distribution of boron in the direction of the short axis of the particle. In some unirradiated samples, for example, boron carbide is seen lying in cracks which are predominantly oriented along the a axis of the particles. The cracking in the direction of the short axis of the grains is believed to result from column failure at points of weakness.

It is further proposed that the same effect is present in the black irradiated material; however, because the grains of the black type are smaller in size and more randomly oriented than those of the grey materials, the fracture resulting from irradiation is more difficult to see.

Miscellaneous Studies. Crystallite parameters, c, a, L_c and L_a were measured from powder samples. ⁽⁶⁾ At the conclusion of each experiment these measurements were made on samples scheduled for other destructive analyses. Measurements were also made on powders of the unirradiated material for comparison. The results of these measurements, shown in Table XIX, indicate that the four boronated graphites have crystal structures which are well developed and characteristic of quite graphitic materials. The c dimensions for the unirradiated materials are similar to those of natural-flake graphite, but upon irradiation the c spacing expands quite rapidly and exceeds the values obtained from nuclear graphites irradiated to much higher exposures. The average thickness of the crystallite, L_c , for the unirradiated black 5 and 7 wt% samples is similar to nuclear graphites and the irradiation behavior also is much the same. Grey boronated graphites have unirradiated L_c values approximately three times larger; however, upon irradiation, the crystallite thickness is reduced rapidly to a much lower value than is observed in reactor graphites. This would indicate a higher degree of crystal perfection in the unirradiated grey types and also more radiation-induced crystal damage. The parameters a and L_a , measured on some samples, do not deviate much from the values and behavior of nuclear graphites.

TABLE XIX

CRYSTALLITE CHANGES IN BORONATED GRAPHITE
IRRADIATED IN THE 2C EXPERIMENTS

Sample	Irradiation Temperature, °C	Exposure, n/cm ² x 10 ⁻²⁰	c, Å	L _c , Å	a, Å	L _a , Å
<u>Black, 5 wt% Boron</u>						
D 4 LA	---	0	6.704	465	2.4635	1540
D 2 TB	370	7.2	6.784	93	2.4546	550
D 2 TA	370	25.3	6.797	98	----	----
D 3 TB	500	7.2	6.776	135	2.4553	725
D15 LB	500	7.2	6.782	135	2.4584	----
D 3 TA	500	25.3	6.802	119	----	----
D15 LA	500	25.3	6.781	118	----	----
<u>Grey, 5 wt% Boron</u>						
E16 LB	---	0	6.699	1330	2.4659	1395
E 9 LA	370	7.2	6.807	78	2.4646	1380
E10 TA	370	7.2	6.809	70	2.4520	345
E13 TC	370	18.2	6.823	77	----	----
E 3 LB	500	7.2	6.794	85	2.4570	280
E 8 LB	500	7.2	6.779	90	2.4571	945
E11 TB	500	7.2	6.789	88	2.4593	400
E11 TD	500	7.2	6.802	118	2.4585	835
E 3 LA	500	25.3	6.802	95	----	----
E11 TA	500	25.3	6.802	100	----	----
<u>Black, 7 wt% Boron</u>						
H 9 LB	---	0	6.724	485	2.4640	865
H13 LA	370	25.3	6.814	92	----	----
H 1 LA	500	25.3	6.791	108	----	----
H12 TA	500	25.3	6.789	119	----	----
<u>Grey, 7 wt% Boron</u>						
J18 LA	---	0	6.717	1430	2.4668	2000
J 6 LA	370	7.2	6.847(?)	72	----	----
J 1 TA	370	25.3	6.884	62	----	----
J 6 TB	500	7.2	6.809	90	2.4552	975
J15 LB	500	7.2	6.809	89	2.4582	1125
J13 TA	500	18.2	6.820	100	----	----
J 6 TA	500	25.3	6.830	88	----	----
J14 LA	500	25.3	6.848	82	----	----

Boron and lithium analyses of the samples are shown in Table XX. Comparison can be made with original values in Table I. The lithium build-up in the samples is quite uniform as a function of exposure for all samples, whereas the boron decrease is quite variable. This is due to a high initial variability of the boron content which masks the actual changes, while the lithium production is proportional only to the incident thermal-neutron flux for samples of the size used in this program. The lithium content was measured on powdered remnants of the samples by a flame spectrometric technique after mechanical properties were measured. Thus the lithium values are averages for each sample and are proportional

to the burnout of the boron. If no lithium were lost during the irradiation, * B^{10} burnout in the samples was approximately 50% at the conclusion of the 2C-1 experiment.

TABLE XX
BORON AND LITHIUM CONTENT OF SAMPLES IRRADIATED
IN THE 2C EXPERIMENTS

<u>Sample</u>	<u>Irradiation Temperature, °C</u>	<u>Exposure, n/cm² x 10⁻²⁰</u>	<u>wt% Boron</u>	<u>wt% Li⁷</u>
<u>Black, 5 wt% Boron</u>				
D 2 TB	370	7.2	6.8	0.33
D 2 TA	370	25.3	4.7	0.72
D 3 TB	500	7.2	7.8	0.35
D 3 TA	500	25.3	4.3	0.65
D15 LA	500	25.3	4.4	0.65
<u>Grey, 5 wt% Boron</u>				
E 9 LA	370	7.2	5.9	0.35
E10 TA	370	7.2	6.0	0.35
E 8 LB	500	7.2	5.9	0.38
E11 TD	500	7.2	5.7	0.36
E13 TC	370	18.2	5.7	0.78
E 3 LA	500	25.3	4.3	0.78
E11 TA	500	25.3	3.9	0.59
<u>Black, 7 wt% Boron</u>				
H 1 LB	500	7.2	7.4	0.39
H12 TB	500	7.2	7.7	0.36
H13 LA	370	25.3	6.6	0.88
H12 TA	500	25.3	5.9	0.71
H 1 LA	500	25.3	8.2	0.86
<u>Grey, 7 wt% Boron</u>				
J 6 LA	370	7.2	8.2	0.40
J15 LB	500	7.2	7.2	0.40
J14 LB	500	7.2	7.9	0.38
J 6 TD	500	7.2	7.7	0.39
J13 TA	500	18.2	6.7	0.86
J 1 TA	370	25.3	6.3	0.85
J14 LA	500	25.3	6.6	0.88
J 6 TA	500	25.3	6.5	0.85

The weight changes of all samples irradiated in the 2C-1 and 2C-2 experiments appear to be negligibly small. Average values are approximately +0.02 to +0.1%, corresponding to a maximum change of approximately 5 mg per sample. Some evidence points to an enhanced oxidation for those samples irradiated in both the 2C-1 and 2C-2 experiments when compared with the samples first irradiated in the 2C-2 experiment. The

*Test scrapings taken from the graphite holders and aluminum walls of the 2C-1 experiment did not indicate boron or lithium migration had occurred.

full-term samples gained several times the weight that was observed for the new samples. Even though the gas atmosphere and temperature of all samples were nearly identical in the several irradiations. Oxidation of the lithium oxidation would be proportional to exposure. Calculations of weight-gain based on this reaction show it to be a likely possibility.

Nuclear magnetic resonance⁽⁷⁾ and magnetic susceptibility⁽⁸⁾ measurements were attempted on unirradiated boronated graphite to provide information on the dispersion of the boron in the lattice; however, these tests were inconclusive because of a suspected, but not identified, ferromagnetic impurity in the sample.

Helium Retention in Boronated Graphite. An estimate has been made of the average amount of helium retained in boronated graphite samples irradiated during this experiment. The evaluation was based on a calculation of the quantity of helium produced and measurements of helium release from samples during nuclear irradiation. Helium production was calculated from the Li^7 concentration measurements made on samples, assuming that one helium atom was produced for every lithium atom and that no lithium was lost from the boronated materials. Helium release was determined during the irradiation experiments by measuring the pressure of the helium atmosphere in the experimental assembly at intervals when the gas temperature was known to be constant. The volume of the capsule and associated gas system was measured prior to the irradiation and special attention was given to insuring the integrity of the apparatus thus minimizing the possibility of helium loss.

The results of this analysis show that ~60% of the helium generated by the $\text{B}^{10}(\text{n}, \alpha)$ reaction is retained within the boronated-graphite samples tested. It should be emphasized that the figure of 60% helium retention is an average value for the materials irradiated since helium pressure changes were the result of helium released by all samples in the capsule.

ETR Irradiations. An auxiliary program of boronated-graphite testing, designated GEH-23, was conducted in the Engineering Test Reactor (ETR). *

*This work was sponsored by the Division of Reactor Development as part of the Nuclear Graphite Studies program and was carried out by R. Neidner.

Experiments in this series were designed to yield irradiation damage results from a nuclear environment having a neutron energy distribution intermediate to the intense fast energy spectrum of the Fermi Reactor and the thermally-rich spectrum of the Hanford 2C facility, thus providing a basis for correlation of 2C data to fast-reactor conditions. Modification of the ETR spectrum was accomplished by enclosing the test capsule in a removable shroud tube of boron stainless steel. The tube was replaced periodically during the irradiation because of burnout of the boron in the steel.⁽⁹⁾ Using this technique, the thermal flux was reduced by a factor of 15 from the unperturbed level, while the fast flux remained essentially unchanged. The thermal-to-fast neutron ratio for these irradiations was approximately 0.07:1, whereas in the Hanford tests it was 8.3.

Preliminary length-change data from the GEH-23 shielded experiments, GEH-23-8 and GEH-23-13, are shown in Figures 19 and 20. It is seen from these figures that samples irradiated in Capsule GEH-23-8, having thermal-neutron exposures between $3 \text{ and } 6 \times 10^{19} \text{ n/cm}^2$, exhibit considerably greater contractions than 2C samples irradiated to the same exposure. A possible explanation lies in the fact that GEH-23-8 samples were irradiated in a vertical column arrangement with measuring surfaces contacting one another. Axial self-loading and vibration-induced abrasion of the measuring surfaces due to this configuration may have caused a systematic error in the length changes of the GEH-23-8 samples. Further shielded tests are being conducted under conditions where this cannot occur.

Applications of 2C Data to Fermi Conditions. The damage effects discussed in this report are determined by the flux spectra and other conditions to which the samples were exposed. Application of these data to other spectra has been studied theoretically and a method of correlation has been devised based on displacement of carbon atoms both by the high-energy neutron interactions and the thermal-neutron-induced fission in the boron. Since the correlation is extensive, it will be published separately. A conclusion from these auxiliary studies is that the damage produced in the 2C samples is significantly higher than a comparably intense fast spectrum would have produced and that the data should be a valid accelerated test of shield behavior to a high exposure.

FIGURE 19

Effect of Exposure on Boronated Graphite Irradiated in ETR Shielded Facility - Parallel Orientation

FIGURE 20

Effect of Exposure on Boronated Graphite Irradiated in ETR Shielded Facility - Transverse Orientation

CONCLUSIONS

The data presented in this report give a significantly improved understanding of boronated graphite and its application to fast-reactor shields. Also, limitations to the usefulness of the data were better defined. The basic damage mechanism has not been determined nor has the precise rate of change of length during exposure under the Fermi conditions. The latter is complicated by radially nonuniform exposure to thermal neutrons in all available testing facilities and consequent differential stress induced in the specimens tested. The damage produced is estimated to be many times the equivalent fast-exposure conditions in the Fermi shield. Therefore, while no assurance can be given that the accelerated proof-test results obtained in this program are exactly equivalent to the effects of lifetime exposure in the Fermi reactor, it is believed that the physical measurements presented herein are reasonable indicators of damage to boronated graphites after long-term service conditions. Evidence from all the information accumulated during this program and collateral tests indicates that the two "black" grades of boronated graphite used in the Fermi shield are preferable in all respects to the "grey" types. It has been demonstrated that even though the dimensional and physical stability of boronated graphite, and especially grey recrystallized types, is not equal to that of nonboronated reactor-grade graphites, structural integrity of the material is maintained and physical and dimensional changes fall within acceptable limits expressed by APDA even after extended neutron exposure.

Irradiations of boronated graphite are continuing in the ETR to improve understanding of the basic mechanism of damage to the material.

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APPENDIX ALOCATION OF SAMPLES DURING
IRRADIATION IN THE 2C EXPERIMENTS

	<u>2C-1 Samples</u>	<u>2C-2 Samples</u>	<u>2C-3 Samples</u>
Section #1 ~500 °C	D3 T-A D15 L-A E3 L-A E8 L-A E11 T-A E11 T-C H1 L-A H12 T-A J6 T-A J6 T-C J14 L-A J15 L-A A1 L-A A5 T-A	D3 T-A D15 L-A E3 L-A E8 L-A E11 T-A E11 T-C H1 L-A H12 T-A J6 T-A J6 T-C J14 L-A J15 L-A A1 L-A A5 T-A	D3 T-A D15 L-A E3 L-A E8 L-A E11 T-A E11 T-C H1 L-A H12 T-A J6 T-A J18 T-A/B J14 L-A J15 L-A A1 L-A A5 T-A
Section #2 ~500 °C	D3 T-B D15 L-B E3 L-B E8 L-B E11 T-B E11 T-D H1 L-B H12 T-B J6 T-B J6 T-D J14 L-B J15 L-B A1 L-B A5 T-B	D13 T-B D4 L-B E4 L-A E4 L-B E13 T-A E13 T-B H5 L-A H13 T-A J13 T-A J13 T-B J7 L-A J7 L-B H4 L-M J5 L-M	D13 T-B D4 L-B E4 L-A E4 L-B E13 T-A E13 T-B H5 L-A H13 T-A J13 T-A H18 T-A/B J7 L-A J7 L-B H4 L-M J5 L-M
Section #3 ~370 °C	142-3A (CSF) D2 T-A D2 T-B D10 L-A A1 L-C A5 T-C E9 L-A E9 L-B E10 T-B E10 T-A H1 T-A H13 L-A J1 T-A J6 L-A	142-3A D2 T-A D13 T-D D10 L-A D6 L-A D13 T-C E5 L-A E9 L-B E10 T-B E13 T-C H1 T-A H13 L-A J1 T-A J12 L-A	142-3A D2 T-A D13 T-D D10 L-A D6 L-A D18 T-B/C E5 L-A E9 L-B E10 T-B E13 T-C H1 T-A H13 L-A J1 T-A J12 L-A

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