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GULF GENERAL ATOMIC

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IRRADIATION-INDUCED DIMENSIONAL CHANGE
IN HTGR CONTROL MATERIALS

MASTER

by
O. M. Stansfield

April 28, 1972

GULF GENERAL ATOMIC COMPANY
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ABSTRACT

Boronated graphites containing 22 to 43 wt-% boron as B_4C and representing current warm-pressing or extrusion technology were irradiated at $650^\circ \pm 100^\circ C$ to fast neutron fluences up to $7 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$). The irradiation caused an increase in thermal expansivity, a decrease in thermal conductivity, and an anisotropic dimensional change related to the preferred orientation of the graphite crystallites in the graphite matrix.

Although irradiation-induced B_4C swelling has been reported, the dimensional change in boronated graphite is not significantly influenced by that effect. Dimensional change in boronated graphite is controlled by the fast neutron and ^{10}B fission damage irradiation properties of the graphite-binder matrix. At a constant ^{10}B -isotope concentration, the irradiation-induced dimensional change of the boronated graphite increases with increasing ^{10}B -isotope enrichment of the boron in the B_4C addition. This effect, which may result from more severe ^{10}B fission fragment damage in the matrix surrounding ^{10}B -enriched B_4C , leads to distortion directly related to the ^{10}B burnup gradient. The use of natural boron in the fabrication of boronated graphite results in dimensional changes independent of the ^{10}B burnup gradient and correlated with fast neutron fluence under HTGR service conditions.

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INTRODUCTION

Boronated graphite is an economical refractory control material for the High-Temperature Gas-Cooled Reactor (HTGR). Boron, which captures neutrons by the $^{10}\text{B}(\text{n},\alpha)^7\text{Li}$ reaction, is added in the form of B_4C granules to a graphite-binder mixture prior to the forming and heat treatment process. Irradiation-induced dimensional change is one of the most important parameters in the use of boronated graphite in reactor design.

Graphites of high boron concentration (20 to 40 wt-% boron) have been irradiated at 300° to 750°C to fast neutron exposures of $2.4 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$) (1,2). Irradiation experiments with boronated graphite containing 5 to 7 wt-% boron have also been conducted at 300° to 500°C and at relatively low ($< 1.0 \times 10^{21} \text{ n/cm}^2$, $E > 0.18 \text{ MeV}$) fast neutron fluence (3,4). The dimensional change observed in the latter work was correlated with a calculated value of carbon atom displacement in the graphite matrix resulting from damage caused by fast neutrons and the fission fragments of the $^{10}\text{B}(\text{n},\alpha)^7\text{Li}$ reaction (5). While these data and analyses provided an estimate of potential dimensional change in boronated graphite, irradiation data at a temperature of $\sim 700^\circ\text{C}$ and fast neutron exposures of $> 6 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$) were required to provide greater confidence in control rod design. Consequently, an irradiation program was conducted in which dimensional change data were gathered from two irradiation capsules and an HTGR control rod (6,7).

MATERIALS

Warm-pressed and heat-treated specimens of boronated graphite were fabricated by addition of an appropriate amount of B_4C granules to a dry blended mixture of one of two types of graphite flour containing 15 wt-% coal-tar-pitch binder. The raw materials are described in Table I.

The materials were warm pressed as 1.5-cm-dia. x 1.5-cm-long or 1-cm-dia. x 1.5-cm-long cylinders at 750°C in a graphite mold at 5000 psi. After 5 min at 750°C, the specimens were ejected hot into a container purged with argon. Prismatic specimens 0.5 x 0.5 x 1.5 cm or 0.5 x 1.0 x 1.5 cm were cut from the carbonized 1.5-cm-dia. cylinders with the 0.5- and 1.0-cm dimensions perpendicular to the forming direction. The specimens were then heated in helium for 2 hr at 1000° or 2000°C.

Additional specimens were taken from a vendor-produced hot-pressed (2000°C) boronated graphite compact 8.3-cm-OD x 5.0-cm-ID x 7.6 cm long made from GP-38 and B_4C with coal tar pitch for the Fort St. Vrain HTGR. These specimens were made by diamond core drilling along the axis of the absorber to produce a solid cylinder within the core drill. The cylinders of boronated graphite were then machined to 1.0 cm in diameter by 1.5 cm long and heat treated 2 hr at 1000°C in helium.

Extruded boronated graphite was obtained from the Peach Bottom HTGR control rods. This material was fabricated by blending B_4C granules (100% < 100 mesh, 5% < 325 mesh), graphite filler, and pitch followed by

TABLE I
Raw Materials Used in Preparation of Warm-Pressed Boronated Graphite

Description	Preparation	Finished Product Chemical Analysis
Graphitized Wilmington petroleum coke	Crushed and sieved into U.S. mesh size fraction -50 +200. Graphitized for 1 hr at $2700^{\circ} \pm 100^{\circ}\text{C}$ in a graphite resistance furnace filled with helium. (Sponge type coke.)	B < 0.5, Ba < 0.5, Mg < 0.5, Si < 10, Ti 2, V 10, Zr 60 (ppm) ^a Ash content None detected, <0.02 wt-% Apparent crystallite size, L_c 730 to 770 Å Layer plane spacing, C_o 6.702 to 6.694 Å
GP-38 graphite flour	Used as received from vendor (<48 μm). (Needle-type coke.)	Fe 400, V 100, Si 400 (ppm) ^a Ash content 0.10 wt-% Apparent crystallite size. 625 Å Layer plane spacing. 6.717 Å
Pitch - Barrett No. 30	Sieved and retained -30 U.S. mesh particles.	Mn < 10, Pb 60, Si 40, Al 40, Bi 6, Mo 6, Ni < 4, Ca 60, Fe 20, Na 200, Ti 4 (ppm) ^a
Technical grade B_4C (commercial)	Sieved size range. U.S. mesh -40 +140.	B 69, B_2O_3 1 to 3, C Bal, Fe 0.80 (wt-%)
Leached technical grade B_4C	Leached several hr in warm distilled water to remove B_2O_3 . Water leach followed by 48-hr leach in 1 l HCl/H ₂ O solution to remove Fe. Distilled water rinse and dried in vacuum oven (25 mm Hg) at R.T.	B 69.6, B_2O_3 0.07, Fe 0.1, C Bal (wt-%)
$B_4^{nat}C$ made in laboratory	Blended 99.5% B^{nat} powder and Wilmington graphite in 75 25 ratio by weight. Heated in closed graphite crucible 2 min at $2500^{\circ} \pm 50^{\circ}\text{C}$ in 1 atm He to fuse. Crushed and sieved to obtain -40 +140. Leached in 1 l HCl/H ₂ O solution 48 hr at R.T. Rinsed in distilled water and dried in vacuum oven (25 mm Hg) at R.T.	B 69.2, B_2O_3 0.1, Fe 0.2, C Bal (wt-%)
$^{11}B_4C$ made in laboratory from ^{10}B -depleted B	Blended 98.9% B powder from ORNL containing 1.45 a/o ^{10}B with Wilmington graphite in 75 25 ratio by weight. Processed material the same as above.	B 72.3, B_2O_3 0.02, Fe 0.1, C Bal (wt-%)
$^{10}B_4C$ made in laboratory from 91%-enriched B	Blended amorphous (<10 μm) boron powder, 91% ^{10}B , with GP-38 graphite flour in 75 25 ratio by weight. Processed into B_4C powder as above. Sieved into U.S. mesh size fraction -48 +150.	B 69, B_2O_3 < 0.01, Fe 0.02, C Bal (wt-%)

^a Elements detected by emission spectrograph.

extrusion to form hollow cylinders approximately 5.7-cm OD x 3.2-cm ID x 42 cm long. The extrusions were heat treated at 2200°C for 30 min in nitrogen and then machined to final shape. Prismatic specimens (0.5 x 1.0 x 1.5 cm) were cut from the wall of an extrusion to form specimens for the irradiation experiments. All specimens were heat treated 2 hr in helium at 1000°C to initiate any dimensional change that might result from thermal stress relaxation.

A description of the specimens is given in Table II, which also lists the irradiation capsules containing the materials. Other materials which were irradiated but do not contribute significantly to the following discussion have been described previously (6) and are not included in the table.

IRRADIATION EXPERIMENTS

Capsule BG-1

The prismatic specimens were held in 28 crucibles of 780-S graphite. There was 0.076 cm clearance at the top and ~0.010 cm side clearance between crucible and specimen. In the capsule assembly the specimens were purged with an argon-helium mixture during irradiation to provide inert gas temperature control at $650^{\circ} \pm 100^{\circ}\text{C}$.

Disks of 0.07-cm-thick gadolinium were placed at the top and bottom of each crucible. Cadmium (0.05 cm thick) and gadolinium (0.07 cm) were placed around the crucibles in a manner that would produce a thermal neutron window

TABLE II
Description of Boronated Graphite Specimens Irradiated in
BG-1 and BG-2 Experiments

Material No.	Description	Heat Treatment Temp (°C)	Capsule No.	No. of Specimens in Capsule (BG-1, BG-2)	Boron Content (wt-%)	B ₄ C Content (vol-%)	Bulk Density (g/cm ³)	Anisotropy ^a $\frac{CTE(\parallel)}{CTE(\perp)}$
3912-34	Extruded and heat-treated Peach Bottom HTGR control rod material	2200	BG-1, BG-2	(3,2)	28	26	1.60	0.78
3685-111-1000	High-B ₂ O ₃ , tech. grade commercial B ₄ C	1000	BG-1, BG-2	(12,4)	22	23	1.77	2.2
3685-111-2000		2000	BG-1	(12,0)	22	23	1.79	2.2
3685-117-1000	Low-B ₂ O ₃ , tech. grade commercial B ₄ C	1000	BG-1	(3,0)	21	22	1.71	2.3
3685-117-2000		2000	BG-1	(3,0)	21	22	1.72	1.7
3685-118-1000	Non-boronated graphite made from Wilmington coke	1000	BG-1, BG-2	(9,3)	0	0	1.77	2.2
3685-120-1000	¹⁰ B-depleted B ₄ C (laboratory made)	1000	BG-1, BG-2	(4,2)	22	20	1.72	1.6
3685-122-1000	Low-B ₂ O ₃ , B ₄ C (laboratory made)	1000	BG-1	(3,0)	23	23	1.73	1.7
3685-122-2000		2000	BG-1	(3,0)	23	23	1.72	1.6
3685-124-1000	Low-B ₂ O ₃ , tech. grade commercial B ₄ C	1000	BG-1, BG-2	(4,2)	34	33	1.68	1.3
3685-124-2000		2000	BG-1, BG-2	(4,3)	34	33	1.68	1.4
4465-26	¹⁰ B-enriched B ₄ C (laboratory made)	2000	BG-2	(0,3)	7.5	8.8	1.70	3.5
4465-42	Warm-pressed and heat-treated Fort St. Vrain control rod material	2000	BG-2	(0,6 ^b)	43	40	1.60	1.1

^aRatio of preirradiation mean coefficient of thermal expansion (CTE) determined between 22° and 700°C parallel and perpendicular to the forming direction.

^bDuring irradiation, 5 of 6 specimens became restrained, and useful data were obtained from only 1 unrestrained specimen.

around 90° of the crucible circumference. Specimens were oriented in the crucibles so as to produce a steep ^{10}B burnup gradient across the width which would simulate the conditions which develop in the wall of a control rod absorber compact in reactor service. The 0.05 x 0.05 x 1.5 cm specimens cut from the same compact were positioned to generate significantly different ^{10}B burnup profiles at the same fast neutron exposure so that dimensional changes due only to ^{10}B burnup could be determined. Temperature was monitored in the specimens by Chromel/Alumel thermocouples inserted into 0.2-cm-dia. holes drilled in the specimens along the crucible center line and in the crucible wall. The relative positions of shielding and thermocouple holes in the specimens are shown in Fig. 1.

Capsule BG-2

The design of capsule BG-2 was similar to that of BG-1. However, the cadmium thermal neutron shield was 0.150 cm rather than 0.050 cm thick, and no gadolinium was employed in the circumferential shield. These design changes in BG-2 were made to increase thermal neutron shielding for 270° of the specimen circumference and achieve a greater ^{10}B burnup gradient than in capsule BG-1. The experiment included 16 prismatic specimens previously irradiated in capsule BG-1 and 21 unirradiated, 1.0-cm-dia. cylindrical specimens.

Control Rod C08-01

The control rods of the Peach Bottom HTGR contain six extruded and heat-treated cylinders of boronated graphite (previously described) which are

Fig. 1. Position of prismatic boronated graphite specimens in graphite crucible relative to thermal neutron window. The hole in the center of the specimen and the holes in the graphite contained Chromel/Alumel thermocouples during irradiation.

supported on a metal spine. Control rod C08-01 was removed from the reactor after being fully inserted for 300 effective full power days (EFPD) at 40 MW(e) and examined to determine dimensional changes in the boronated graphite. The details of that work have been reported previously (7).

RESULTS AND DISCUSSION

Capsules BG-1 and BG-2 were irradiated for four cycles (2736 hr and 4332 hr, respectively, at >165 MW) in the Engineering Test Reactor (ETR). A temperature gradient of $\sim 100^{\circ}\text{C}$ existed between the center line and the surface of specimens, with volume and time-averaged specimen temperatures for the two capsules being approximated at $650^{\circ} \pm 100^{\circ}\text{C}$. The ^{10}B -isotope depletion profile which developed across the specimen during irradiation was determined from mass spectrometer analyses of powder samples taken after irradiation. The results of that work are shown correlated with fast neutron fluence in Fig. 2.

Thermal and physical properties were determined on selected materials before and after irradiation and have been reported previously (7). Results for materials of greater current interest for HTGR technology are shown in Table III. There was an increase in the coefficient of thermal expansion and a decrease in thermal conductivity after irradiation in all specimens examined. The compressive strength was decreased somewhat by irradiation.

Fig. 2. Typical ^{10}B -isotope depletion profile across boronated graphite specimens in irradiation experiments BG-1 and BG-2 and Peach Bottom HTGR control rod C08-01 at various fast neutron fluences. An attempt was made in small specimens to simulate the ^{10}B burnup profile in the walls of an HTGR control rod compact.

TABLE III
Thermal and Physical Properties of Boronated Graphite Materials
Irradiated at $650^\circ \pm 100^\circ\text{C}$

Property	Material 3912-34 (Extruded and Heat-Treated Peach Bottom HTGR Control Rod Material) Irradiated at $6 \times 10^{21} \text{ n/cm}^2$ ^a	Material 4465-42 (Hot-Pressed Fort St. Vrain Control Rod Material) Irradiated at $5.3 \times 10^{21} \text{ n/cm}^2$ ^a
Coef. of thermal exp. relative to forming direction (22° to 700°C), $10^{-6} \text{ in./in. } ^\circ\text{C}$:		
Preirradiation parallel (α_{pa})	3.68	4.83
Preirradiation perpendicular (α_{pe})	4.70	4.58
Ratio (α_{pa}/α_{pe})	0.78	1.1
Postirradiation parallel (α_{pa})	5.34	6.78
Postirradiation perpendicular (α_{pe})	5.70	6.56
Ratio (α_{pa}/α_{pe})	0.93	1.0
Thermal conduct. perpendicular to forming direction, $\text{cal/cm-sec-}^\circ\text{C}$:		
Preirradiation 25°C	0.120	0.054
Preirradiation 400°C	0.090	0.033
Preirradiation 800°C	0.065	0.020
Postirradiation 25°C	0.009	0.008
Postirradiation 400°C	0.012	0.012
Postirradiation 800°C	0.012	0.012
Room-temperature crush strength, loaded parallel to forming direction, psi:		
Preirradiation	5500	6000
Postirradiation	5120	3807

^aTotal fast neutron fluence ($E > 0.18 \text{ MeV}$).

The relative dependence of irradiation-induced dimensional change upon B_4C loading, ^{10}B burnup, and heat treatment temperature for warm-pressed boronated graphite was demonstrated by the data obtained for materials made with graphitized Wilmington coke, as shown in Figs. 3 and 4. The data points on these figures represent the average for 2 to 12 specimens (see Table II) of each material, and the bars represent the range of data for individual specimens. The repeatability of dimensional measurement was ± 0.007 cm, and the fast neutron fluence was assumed to be known to $\pm 10\%$.

Specimens $0.5 \times 0.5 \times 1.5$ cm in dimension showed no bowing or systematic difference in irradiation induced-dimensional change. This was true even though the shielding arrangement (see Fig. 1) produced a high ^{10}B burnup and a large burnup gradient in the specimens near the thermal neutron window while adjacent shielded specimens developed essentially no gradient at a lower level of ^{10}B burnup.

The irradiation-induced dimensional change in warm-pressed boronated graphite, based upon the lines drawn through the data points in Figs. 3 and 4, shows a decrease with increasing heat treatment temperature. The observed performance may have been caused by an increase in the ordering of the binder carbon of the matrix after heat treatment at $2000^\circ C$. The tendency toward increased irradiation stability at higher heat treatment temperatures of the binder is similar to that of non-boronated graphite as observed by other investigators (8). However, in boronated graphite the maximum heat treatment temperature for irradiation stability will optimize at a temperature below the melting point of B_4C ($\sim 2450^\circ C$). Boronated graphite heat

Fig. 3. Irradiation-induced dimensional change in boronated graphite made with graphitized Wilmington coke and varying B₄C loading. Specimens were warm-pressed and heat-treated at 1000°C and irradiated at 650° ± 100°C. The dimensional change was independent of B₄C loading and anisotropy in the range of conditions examined.

Fig. 4. Irradiation-induced dimensional change in boronated graphite made with graphitized Wilmington coke and varying B₄C loading. Specimens were warm-pressed and heat-treated at 2000°C and irradiated at 650° ± 100°C. The dimensional change was independent of B₄C loading and anisotropy in the range of conditions examined.

treated near the melting point of B_4C has exhibited greatly accelerated dimensional change, probably as a result of boron taken into solution in the graphite matrix (3,4). Boron has significant solubility in graphite (2 a/o at 2500°C), and enhancement of irradiation-induced dimensional change takes place in graphite crystals doped with boron (9,10).

The irradiation-induced dimensional change of the hot-pressed boronated graphite made with GP-38 graphite flour and natural B_4C is shown in Fig. 5. This material had a higher B_4C loading and was more isotropic than the warm-pressed boronated graphites discussed previously. The irradiation-induced volume changes of all the boronated graphites heat treated at 2000°C exhibited a similar dependence upon fast neutron exposure, as shown in Fig. 6. In contrast, the boronated graphites heat treated at 1000°C showed about an 80% greater volume swelling at neutron fluences of 6 to 7×10^{21} n/cm² ($E > 0.18$ MeV). These data were taken as further evidence of the importance of heat treatment temperature in dimensional change effects and a lack of strong dependence on variations in the concentration, purity, or ^{10}B burnup in the B_4C component.

The influence of the fabrication process upon irradiation-induced dimensional change was examined by comparing the performance of warm-pressed and extruded boronated graphites. The dimensional change in the extruded boronated graphite material as a function of fast neutron exposure is shown in Fig. 7. Data are shown for both the irradiation experiments and the control rod absorbers taken from the Peach Bottom HTGR. Dimensional changes in the perpendicular direction of the control rod absorbers were

Fig. 5. Irradiation-induced dimensional change in boronated graphite made with GP-38 graphite flour ($< 25 \mu\text{m}$) and hot-pressed at 2000°C . Specimens were taken from a Fort St. Vrain HTGR control rod absorber. Irradiation temperature was $650^\circ \pm 100^\circ\text{C}$.

Fig. 6. Fast neutron fluence dependence of volume change in warm-pressed boronated graphite heat treated at 2000°C and irradiated at 650° ± 100°C.

Fig. 7. Irradiation-induced dimensional change in extruded boronated graphite containing 27 vol-% B_4C . Specimens were heat treated at 2200°C .

not detectable within manufacturing tolerances (± 0.005 cm), but significant parallel length changes were detected which agreed well with predictions based upon the irradiation capsule data. Expansion was observed in the direction perpendicular to the preferred orientation of the basal planes in graphite crystallites of the matrix, as determined from the forming technique and the thermal expansivity in the two principal directions. Initial contraction, as well as a tendency for later expansion, was exhibited parallel to the preferred graphite crystallite basal plane orientation. Although the extruded boronated graphite contained a greater B_4C loading and developed similar ^{10}B burnup profiles across the specimens, the irradiation-induced dimensional change was significantly less than in the warm-pressed material heat treated at either 1000° or $2000^\circ C$. These observations are consistent with the existence of a more stable graphite matrix in the extruded material as a result of higher heat treatment temperature ($2200^\circ C$) and other process variables.

There was no ceramographic evidence of matrix disruption or expansion caused by swelling of the B_4C , but evidence of cracking in the B_4C granules independent of ^{10}B burnup gradient was commonly observed, as shown in Fig. 8. Swelling of B_4C has been reported previously (6,11,12), but the uncertainty in the magnitude of expected B_4C swelling is great, and the uncertainty is made greater by the presence of the graphite matrix, which can restrain the B_4C . Restraint of B_4C swelling during irradiation has been achieved with PyC coatings (11). In view of the uncertainties, a reasonable estimate for swelling in the natural B_4C is 5 to 30 vol-% at 70% to 80% ^{10}B burnup. The lower values of swelling are more probable under the conditions existing in

M367-142

(a)

(75x)

M367-143

(b)

(500x)

Fig. 8. Microstructure of hot-pressed (2000°C) boronated graphite (B_4C in graphite) manufactured for Fort St. Vrain HTGR control rod absorbers (material 4465-42). (a) After irradiation to 4.80 to 5.87×10^{21} n/cm^2 ($E > 0.18$ MeV) at $650^\circ \pm 100^\circ C$ and a local ^{10}B burnup of $\sim 80\%$. (b) Typical B_4C fracture pattern in irradiated boronated graphite under the fast neutron conditions described above; porosity was present prior to irradiation, but B_4C fracture was observed throughout the material only after irradiation.

boronated graphite in view of the lack of dependence of dimensional change upon B_4C loading and ^{10}B burnup.

Although B_4C swelling is not a major contributor to dimensional change, the presence of B_4C granules in the graphite matrix causes the irradiation performance of the composite to differ from the nonboronated graphite. The magnitude of this effect was investigated by irradiating three warm-pressed graphites containing either no B_4C or ~ 20 vol-% B_4C made with natural boron or ^{10}B -depleted boron. The ^{10}B -depleted B_4C was relatively inert with regard to ^{10}B fission damage; and in contrast with the natural B_4C , measurement of the liquid displacement density of loose granules of the ^{10}B -depleted B_4C irradiated at $500^\circ \pm 50^\circ C$ to 6×10^{21} n/cm² ($E > 0.18$ MeV) has shown no apparent swelling (6). The volume change resulting from irradiation was least for the non-boronated graphite and greatest for the graphite containing natural boron in the B_4C as shown in Fig. 9. The intermediate swelling of the graphite containing the relatively inert ^{10}B -depleted B_4C could be ascribed to microstructural effects caused by a complex interaction of the matrix attempting to change dimensions around the inert and unyielding B_4C . The results shown in Fig. 9 suggest that the contribution of the proposed microstructural effects, as exhibited in ^{10}B -depleted boronated graphite, can account for $\sim 25\%$ of the irradiation-induced volume change in a boronated graphite made with natural boron in B_4C .

The strong influence of ^{10}B fission damage upon dimensional change was demonstrated by work in which the irradiation-induced dimensional change in boronated graphite made with ^{10}B -enriched B_4C was determined. The

Fig. 9. Effect of inert and active B₄C addition on irradiation-induced volume change in boronated graphite warm-pressed and heat-treated at 1000°C and irradiated at 650° ± 100°C

boronated graphite was warm-pressed and heat treated at 2000°C with a ^{10}B -enriched B_4C loading. After irradiation in capsule BG-2 to a fast neutron fluence of $4 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$), the ^{10}B -enriched specimens were bowed by a greater length increase on the side exposed to the thermal neutron window. In contrast, the specimens made with B_4C containing natural boron and experiencing similar ^{10}B fission density gradient (material 4465-42) showed no evidence of bowing and considerably less dimensional change, as shown in Fig. 10. The volume loading of B_4C was 4.5 times greater in the graphite containing natural boron than in the ^{10}B -enriched material. However, under the similar ^{10}B fission density and fast neutron fluence which developed, the dimensional change was much greater in the graphite containing ^{10}B -enriched B_4C .

Ceramographic examination of the two materials after irradiation revealed cracks in the B_4C in both, but no disruption of the adjacent matrix. Irradiation-induced swelling of the ^{10}B -enriched B_4C granules determined from liquid displacement measurements on loose granules was similar to natural B_4C (6). Therefore, the enhanced irradiation-induced swelling and bowing in the material made with ^{10}B -enriched boron was attributed to accelerated dimensional change in the matrix surrounding B_4C granules. A similar correlation of dimensional change and ^{10}B burnup profile was not observed in boronated graphites containing natural boron in which the fission fragments (He, Li) ejected from the B_4C into the matrix were approximately one-fifth of the number ejected from ^{10}B -enriched B_4C .

Fig. 10. Effect of ^{10}B enrichment of B_4C on irradiation-induced dimensional change in boronated graphite warm-pressed and heat-treated at 2000°C . Specimens were irradiated at $650^\circ \pm 100^\circ\text{C}$ to a fast neutron fluence of 4.0 to 4.8×10^{21} n/cm^2 ($E > 0.18$ MeV). Nominal specimen dimensions were 1.5 cm $\times$ 1.0 cm dia.

CONCLUSIONS

Irradiation-induced dimensional change in warm-pressed and extruded boronated graphites used in HTGR control rods has been characterized at $650^{\circ} \pm 100^{\circ}\text{C}$ to fast neutron fluences and ^{10}B burnups exceeding expected reactor conditions. There is a tendency for irradiation-induced expansion both parallel and perpendicular to the pressing direction in warm-pressed boronated graphite, with less dimensional change after heat treatment at 2000°C than at 1000°C . Extruded boronated graphite heated at 2200°C exhibited less dimensional change than warm-pressed material heated at 2000°C under similar irradiation conditions.

The anisotropy in dimensional change in both extruded and warm-pressed boronated graphites is consistent with the expected irradiation performance of a graphite matrix showing preferred orientation of graphite crystallites as determined by the forming process. The dimensional change is not significantly influenced by swelling of B_4C during irradiation. The dimensional change in boronated graphite is controlled by fast neutron damage and ^{10}B fission fragment damage in the graphite-binder matrix. The use of natural boron in the B_4C of boronated graphite results in dimensional changes independent of ^{10}B burnup gradient and correlated with fast neutron fluence under anticipated HTGR service conditions.

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