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AEC RESEARCH AND
DEVELOPMENT REPORT

GA-4557

IRRADIATION OF B₄C-GRAPHITE CAPSULES (GA2-032-1A1 AND -2A2)

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INTRODUCTION

Control rods for the Peach Bottom High-temperature Gas-cooled Reactor (HTGR) will be fabricated of B_4C dispersed in a graphite matrix (30 and 10 wt-% boron). These materials will be exposed to the reactor environment of thermal and fast neutrons in a temperature range of approximately 350° to $800^\circ C$. Experiments were therefore performed to determine the effects of a neutron environment on the dimensional stability of B_4C -graphite fabricated by extrusion. Results obtained with two irradiation capsules (GA2-032-1A1 and -2A2) are described in this report. The samples were obtained from two vendors and contained about 30 wt-% boron dispersed as B_4C powder in a graphite matrix. A hot-pressed sample (fabricated in a similar manner to the Peach Bottom fuel elements) was also included for comparison. In the following discussion the samples of the two vendors are designated the 255 series and 417 series, respectively.

A summary of the fabrication history of the samples is given in Table 1. The B_4C particle size was approximately 20 to 50 microns for the 255 samples as determined by metallographic examination at General Atomic. Irradiation samples were cut from the various experimental pieces as shown in Fig. 1. The samples were disks 1 in. OD by 0.25 in. ID by 0.25 in. long. It should be noted that the parallel direction of the hot-pressed sample is analogous to the perpendicular direction of the extruded samples with respect to preferred crystal orientation.

EXPERIMENTAL PROCEDURE

The capsules were irradiated in the GETR core, capsule 1A1 for two cycles and capsule 2A2 for six cycles. The capsules were designed to operate at $415^\circ C$. A machine thermal calculation using the as-built

Table 1

FABRICATION HISTORY OF B_4C -GRAPHITE AND SAMPLING TECHNIQUE

Sample	Forming Technique	Final Heat Treatment ($^{\circ}C$)	Filler	Boron Content (wt-%)	Size (in.) and Orientation of Sample			
					OD	Orientation	Length	Orientation
255	Extrusion	----	Texas coke	29.84	0.900	$\parallel$ and $\perp^a$	0.2500	$\perp^a$
417	Extrusion	----	Needle coke	31.54	0.9000	$\parallel$ and $\perp^a$	0.2500	$\perp^a$
GA	Hot molding	1450	Petroleum coke ^b	28.22	0.9000	$\parallel$ and $\perp^c$	0.2500	$\parallel^c$

^aOrientation with respect to direction of extrusion.

^bPetroleum coke heated above $2600^{\circ}C$ prior to compact fabrication.

^cOrientation with respect to direction of molding. Anisotropy will be more pronounced than in extruded material.

Fig. 1--Sampling diagram for experimental specimens

dimensions and material of construction was performed by J. M. Davidson of Hanford after the irradiation experiments were concluded to determine the thermal gradients. Since both capsules were identical in construction and were located in about the same neutron flux profile, the thermal gradients were calculated on capsule 1A1 only and were assumed to be similar for capsule 2A2. The data are given in Table 2.

Table 2
TEMPERATURE GRADIENT IN CAPSULES 1A1 AND 2A2 (FROM TWO-DIMENSIONAL, MACHINE CALCULATION USING DESIGN DATA)

Capsule 1A1		Capsule Avg. Temp. ($^{\circ}$ C)	Capsule 2A2	
Sample	Type		Type	Sample
255-1	B ₄ C	329	B ₄ C	255-4
Spacer	Graphite	308	Graphite	Spacer
GA-44-4-1	B ₄ C	316	B ₄ C	GA-44-4-2
Spacer	Graphite	291	Graphite	Spacer
417-1	B ₄ C	297	B ₄ C	417-3
Melt-wire holder	Graphite	241	Graphite	Melt-wire holder
255-2	B ₄ C	298	B ₄ C	255-5
Spacer	Graphite	293	Graphite	Spacer
GA-12-2-1	B ₄ C	319	B ₄ C	GA-12-2-2
Spacer	Graphite	312	Graphite	Spacer
417-2	B ₄ C	340	B ₄ C	417-4

Length

Diam.

Melt wires were included in a midplane position to determine the peak temperature. The results indicated that the maximum temperature was between 480 $^{\circ}$ and 600 $^{\circ}$ C for both capsules. Thermocouples were not

included, so the only temperature records are those derived from the melt wires.

Thermal and fast fluxes were determined from dose wires and from GETR unperturbed flux data. The results are given in Table 3.

The capsules were punctured in the General Atomic hot cell and the accumulated gases were collected and analyzed. The samples were removed and photographed and the dimensions were measured. Metallographic inspection was performed on selected samples, as well as chemical analyses for boron.

EXPERIMENTAL RESULTS

Dimensional Changes

Dimensional changes are given in Table 4. The postirradiation dimensional measurements were quite difficult to obtain because of the rough surfaces. A photograph of a typical sample of the 255 series is shown in Fig. 2. A grey deposit covered the surface of the samples. This deposit was not collected or analyzed. The large variance in dimensional change between samples 255-4 and -5 is unusual because the samples were cut from the same block. The difference cannot be explained satisfactorily.

Metallography

Several samples from capsule 1A1 were polished for metallographic inspection. In all cases the B_4C appeared to remain as discrete particles during the irradiation (see Figs. 3 through 5).

Gas Release

The capsules were punctured in the General Atomic hot cell and the gases were collected. The results are given in Table 5.

Table 3
THERMAL AND FAST FLUXES IN IRRADIATION CAPSULES

Capsule	Flux ($\text{nv} \times 10^{-14}$)						Exposure ($\text{nvt} \times 10^{-21}$)					
	Thermal ^a		Fast ^b				Thermal ^a		Fast ^b			
	GETR Data ^c	Co ⁶⁰	GETR Data	Co ⁵⁸	Mn ⁵⁴	Sc ⁴⁶	GETR Data	Co ⁶⁰	GETR Data	Co ⁵⁸	Mn ⁵⁴	Sc ⁴⁶
1A1	1.5	1.01	2.0	0.54	0.67	0.48	0.75	0.45	1.0	0.24	0.30	0.21
2A2 ^d	1.5	----	2.0	----	----	----	2.60	----	3.47	----	----	----

^aE = 0 to 17 ev.

^bE > 0.18 Mev (GETR data); E > 1 Mev (dose wire data).

^cCorrected for axial flux gradients.

^dDose wires lost.

Table 4

DIMENSIONAL CHANGE DATA ON B₄C-GRAPHITE SAMPLES FROM IRRADIATION CAPSULES
GA2-032-1A1 AND -2A2

Sample	Capsule	Boron Content (wt-%)		Exposure ^a (nvt × 10 ⁻²⁰)		Temp. (°C)	Dimensional change (%)		
		Preirrad.	Postirrad.	0.17 ev	>0.18 Mev		Diameter		Length ^b
							Notch	90° from Notch	
255-1	1A1	29.84	26.48	7.5	10.0	329	-0.04(II)	+0.09(I)	+0.26(I)
-2	1A1	29.84	-----	7.5	10.0	298	-0.14(II)	-0.06(I)	+0.28(I)
-4	2A2	29.84	-----	26.0	34.7	329	+0.58(II)	+0.68(I)	+1.29(I)
-5	2A2	29.84	22.70	26.0	34.7	298	+0.27(II)	+0.03(I)	+0.20(I)
417-1	1A1	31.54	31.46	7.5	10.0	297	+1.01(II)	+1.35(I)	+2.9(I)
-2	1A1	31.54	-----	7.5	10.0	240	+1.10(II)	+1.94(I)	+2.5(I)
-3	2A2	31.54	-----	26.0	34.7	297	broken	-----	-----
-4	2A2	31.54	-----	26.0	34.7	340	broken	-----	-----
GA-44-4-1	1A1	29.05	-----	7.5	10.0	316	+0.32(II)	-0.16(I)	-1.34(I)
GA-12-2-1	1A1	27.40	-----	7.5	10.0	319	+0.91(II)	-0.18(I)	-0.15(I)
GA-44-4-2 ^c	2A2	29.05	-----	26.0	34.7	316	+1.21(II)	+2.91(I)	-0.72(I)
GA-12-2-2	2A2	27.40	-----	26.0	34.7	319	+3.08(II)	+1.60(I)	-0.56(I)

NOTE: The average design temperature of both capsules was 415°C. Melt wires indicated a peak temperature of 480°C to 600°C. Thermal calculations indicated that the melt wire holder operated at an average temperature of 241°C.

^aCalculated from GETR unperturbed flux data.

^bAverage of four measurements.

^cSmall piece was broken off outer diameter of sample during irradiation.

2.5X

Fig. 2--B4C-graphite sample 255-1
from capsule 1A1

200X

200X

Fig. 3--Photomicrographs of irradiated B₄C-graphite sample 255-1
from capsule 1A1

200X

200X

Fig. 4--Photomicrographs of irradiated B_4C -graphite sample 417-1
from capsule 1A1

400X

400X

Fig. 5--Photomicrographs of irradiated B_4C -graphite sample
GA-44-4-1 from capsule 1A1

Table 5
GASES COLLECTED FROM IRRADIATION CAPSULES

Capsule	Gases Collected (cm ³ S.T.P.)			
	Back-filled ^a	Collected	Difference	Species
1A1	15.4	129	113.6	Not analyzed
2A2	15.4	96.0 ^b	80.6	12%-25% He ^b

^a Back-filled with helium.

^b Suspected leak of air into collection bulb (sample remained 2 months in bulb before analysis). Also, pump was not in line during gas collection, so total gas content probably was not collected.

Boron Depletion

Boron analyses were obtained on a few samples before and after irradiation. The results are given in Table 6. The analytical data are only approximate because the preirradiation values were obtained as an average of several samples from the original piece and the postirradiation values were obtained on a single sample. The boron depletion for sample 417-1 appears to be in error, as it shows essentially no burnup. Samples 255-1 and 255-5 indicate an atom burnup of B¹⁰ of about 11% and 23% respectively.

Table 6
BORON CONTENTS OF IRRADIATED SAMPLES

Capsule	Sample	Wt. of Irradiated Sample (g)	Boron Content (wt-%)	
			Preirrad.	Postirrad.
1A1	255-1	4.6066	29.84	26.48
1A1	417-1	4.2905	31.54	31.46
2A2	255-5	4.6091	29.84	22.70

DISCUSSION

If the samples are assumed to be homogeneous with respect to B_4C particle dispersion, the major portion of the thermal neutrons will be captured on the surface. Thus, damage could result from helium recoils and pressure buildup near the surface and from fast neutron effects throughout the structure. The effects of fast neutrons on graphite structures are well known at the irradiation temperature and exposures calculated for these experiments. Data on highly graphitic structures⁽¹⁾ indicate contraction parallel to the layer planes (direction of extrusion). However, the dimensional behavior of the extruded samples (from Table 4) indicates predominantly expansion or swelling in both directions, especially after the six-cycle irradiation. The expansion apparently was more severe in samples 417-3 and -4, and both samples fractured in a radial direction. Similar samples had expanded severely in the two-cycle irradiation. The hot-pressed samples prepared at General Atomic were distorted during both irradiation experiments, and a small piece was broken from one of the samples irradiated for six cycles.

The variance in dimensional behavior indicates that fabrication methods may be quite important. Although complete fabrication histories were not available on the vendor-supplied samples, it appears that high-temperature fabrication ($>2500^{\circ}C$) may be detrimental to dimensional stability. The microstructures of the 417 samples appear to have recrystallized, perhaps as a result of heating to $>2500^{\circ}C$ in the presence of B_4C . Although it is not evident from the metallography, the boron may have permeated the graphite structure.

Valovage, ⁽²⁾ in his work on irradiating hot-pressed and vibration-compacted B_4C particles, indicates that helium release was about 2.5% to 6% for samples irradiated to B^{10} burnups of from 5% to 26% at irradiation temperatures in the range 132° to $540^{\circ}F$. Also, the hot-pressed specimens were broken and distorted. It appears likely that helium may have become trapped within the B_4C particles and perhaps in the graphite

crystals surrounding the B_4C particles. Thus, retention of helium within the samples may account for a portion of the expansion. A quantitative measure of helium release could not be obtained for each sample during the present experiment because all samples were irradiated within a common gas chamber.

These results compare well with some previous experiments at General Atomic⁽³⁾ in which hot-pressed B_4C -graphite samples containing 20 and 30 wt-% boron were observed to have expanded about 2.5% and 4.5%, respectively, across the layer planes and a 40 wt-% sample was observed to have fractured when irradiated at about 370°C for five GETR reactor cycles. Valovage⁽²⁾ also indicated that postirradiation annealing at 815°C released up to 19% of the helium generated in the hot-pressed B_4C samples. This indicates that irradiation at higher temperatures may produce less distortion, assuming again that helium retention is responsible. In the earlier General Atomic work⁽³⁾ samples irradiated for five cycles at approximately 700°C showed less expansion in comparison with samples irradiated at 370°C and no fracture, which is what may be expected if a larger portion of the helium was released during the irradiation at 700°C.

SUMMARY

Extruded B_4C -graphite samples containing 30 wt-% boron dispersed in a graphite matrix expanded when irradiated at approximately 300°C to 340°C for six cycles in the GETR core. Hot-pressed samples exhibited both expansion and contraction.

Several samples fractured, indicating that high-temperature fabrication (>2500°C) may be an important factor resulting in severe radiation damage.

Helium retention within B_4C particles and graphite crystals may cause expansion, although this is only speculative at this time.

The stability of B_4C -graphite of 30 wt-% boron appears to be more dependent on thermal neutron damage (n, α reaction) than on fast neutron damage to the matrix.

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