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BORON CARBIDE-GRAPHITE NUCLEAR CONTROL-ROD
MATERIAL: PREPARATION, THERMAL STABILITY,
AND IRRADIATION EVALUATION

by

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ABSTRACT

Boron carbide-graphite nuclear control-rod materials containing up to 45 wt-% boron were prepared by hot-pressing techniques. The densities of these compacts decreased with increasing boron content. Compact strengths, however, were noticeably increased with boron content of 20 wt-%, suggesting that the boron acts as a sintering aid.

Thermal stability was evaluated by annealing compacts under a variety of temperature and atmospheric conditions. The upper limit of the loss of boron was 5% to 10% in 200 hr at 2000°C--considerably less than one would predict from the Langmuir evaporation equation. Diffusion of boron atoms through the pores of the graphite, rather than evaporation from the surface, has been found to be the rate-controlling step in the loss of boron.

Compacts containing 20, 30, and 40 wt-% boron were irradiated at 350° and 700°C to 1.0 and 2.4×10^{21} nvt (fast). Physical integrity of the compacts was good after irradiation. Slight increases in tensile strength were noted in several instances.

I. INTRODUCTION

The Peach Bottom High-temperature Gas-cooled Reactor (HTGR) is a solid fuel, semihomogeneous type, employing graphite as the moderator, fuel matrix, and cladding material, and helium as the coolant. The fuel (U^{235}) and fertile material (Th^{232}) are in the form of carbides homogeneously dispersed in annular fuel compacts, which are enclosed in tubes of low-permeability graphite.^{1, 2, 3} Each fuel element for the Peach Bottom HTGR has the appearance of a solid graphite cylinder 3.5 in. in diameter by 144 in. long. A summary of the development and evaluation of graphite-matrix fuel compacts for the Peach Bottom HTGR has been presented in earlier publications.^{4, 5} This paper describes the development and evaluation of the boron carbide-graphite control-rod material for this HTGR.

Approximately 35 of some 840 fuel-element positions in the Peach Bottom reactor are occupied by control rods. The neutron-absorber portion of the control rods consists of five cylindrical sections in a tandem arrangement, separated by spacers, and supported on a gas-cooled metal support tube. The control rod slides inside a graphite tube approximately 12 ft long. The poison sections are composed of graphite loaded with boron carbide. They are approximately 17 in. long, with an outside diameter of 2.25 in. and an inside diameter of 1.25 in. The required boron concentrations are predicted to be in the range of 20 to 40 wt-%. Operating temperatures are calculated to be below 800°C.

The compound B_4C was selected--on the basis of its availability, low cost, and compatibility with graphite to temperatures well in excess of those predicted for the control rods--as the chemical form of boron to be dispersed in graphite. The General Atomic warm-press and sinter process for fuel-compact fabrication^{4, 5} was adapted to the preparation of dispersions of B_4C in graphite. Results of the evaluations of these materials,

including thermal-stability and irradiation testing, are discussed in the following sections.

II. GENERAL ATOMIC B₄C-GRAPHITE CONTROL-ROD MATERIAL

Dispersions of B₄C in graphite were prepared at General Atomic using the warm-press and sinter process developed for Peach Bottom HTGR fuel-compact fabrication.^{4, 5} In this process, B₄C and graphite powders are blended dry and then mixed into a slurry with pitch dissolved in trichlorethylene. Continued mixing produces granules, which are dried of solvent, are cold-pressed at 50,000 psi into either annular or solid cylindrical shape and then warm-pressed at 800°C and 4500 psi, and finally are sintered at 1400°C in vacuo to completely outgas and thermally stabilize the compacts. By using graphite powder for the filler and only a small amount (10 wt-%) of pitch binder, good thermal properties are obtained without taking the bodies to graphitizing temperatures. Also, the warm-press technique produces compacts of high density and good strength, and permits the use of substantially less binder than conventional graphite-fabricating techniques.

Compacts having excellent appearance and containing up to 45 wt-% boron were readily prepared by the warm-press and sinter process. The boron loadings and atom ratios for 1-in. -diameter compacts warm-pressed at 800°C and sintered at 1400°C are given in Table I.

The variation in density as a function of boron content is shown in Fig. 1. Beyond a content of 20 wt-% boron, density decreases with increasing boron content. Annular compacts are less dense than solid ones, as a result of friction on the additional die surface during pressing.

The variation in crushing strength with boron content is shown in Fig. 2. The appreciably higher strength of the 20 wt-% boron material is not fully understood at present, and further studies are in order. It does appear, though, that boron carbide in moderate (10 to 30 wt-%) concentrations may be acting as a sintering aid during hot-pressing and sintering of

Table I

BORON LOADINGS AND ATOM RATIOS OF B₄C-GRAPHITE
COMPACTS PREPARED BY GENERAL ATOMIC
WARM-PRESS AND SINTER PROCESS

Boron Loading (wt-%)	Compact Density (g/cm ³)	Boron Loading (g/cm ³)	Carbon:Boron Atom Ratio
20	1.93	0.39	3.6
30	1.87	0.56	2.1
40	1.79	0.72	1.35

Fig. 1--Density versus boron content for B_4C -graphite control-rod compacts prepared by the General Atomic warm-press and sinter process

Fig. 2--Crushing strength versus boron content for B₄C-graphite control-rod compacts prepared by the General Atomic warm-press and sinter process

the compacts, thereby increasing the strength of the bodies. This apparent contribution by the boron to the strength of the graphite bodies would be an interesting subject to pursue. At this time, one may conclude that the strength of the matrix graphite is not impaired by boron loadings to 30 wt-%. Beyond this concentration, however, there appears to be a definite decrease in strength.

A photomicrograph of a polished section of a B_4C -graphite compact is shown in Fig. 3. The B_4C is seen as angular particles of 20- μ , or smaller, size dispersed throughout the graphite matrix. Many of the particles are too small to be seen clearly at the $\times 250$ magnification.

III. THERMAL STABILITY OF B_4C -GRAPHITE BODIES

A series of experiments was carried out to study the thermal stability of dispersed boron carbide-graphite compacts by determining the rate of loss of boron as a function of time and the nature of the atmosphere. Below the melting point of this system, the only instability of interest was the vaporization of boron from the surface of the compacts and the reactions of B_4C with atmospheric impurities. In these experiments, the compacts, consisting of a dispersion of B_4C in graphite, were annealed at high temperature in vacuum and in the presence of an oxidizing atmosphere. In principle, the evaporation rate of boron carbide can be calculated from a knowledge of its vapor pressure. The evaporation rate, m/tA , is given by the Langmuir equation,

$$\frac{m}{tA} = \frac{44.3 P}{\sqrt{T/M}} \quad g/cm^2\text{-sec}, \quad (1)$$

where m is the mass evaporated in time, t , from free-surface area, A ; P is the vapor pressure in atm; T is the temperature in $^{\circ}K$; and M is the molecular weight of the evaporating species. The vapor pressure⁶ of B_4C is plotted in Fig. 4, and from this figure, assuming that monatomic boron evaporates (i. e., $M = 11$), the calculated evaporation rate at $2000^{\circ}C$ is

(250x)

Fig. 3--Photomicrograph of B_4C particles dispersed
in a graphite compact

Fig. 4--Vapor pressure of boron over B_4C

$1.0 \times 10^{-5} \text{ g/cm}^2\text{-sec.}$ However, this large value is not applicable to the present study, because, since the boron carbide is dispersed in a graphite matrix, the diffusion through this matrix becomes the rate-determining step in the release as soon as the surface of the compact becomes depleted.

Graphite compacts containing approximately 23 wt-% boron as fine particles of B_4C were prepared. These compacts were right-circular cylinders, 1 in. in diameter by 2 in. long. They were prepared by cold-pressing in a steel die followed by warm-pressing in a graphite die at 750°C and 4500 psi for 15 min; they were not sintered at 1400°C .

The compacts were heated in a resistance furnace by passing a large current at low voltage directly through the compacts. The furnace was controlled with the aid of a radiation pyrometer, and temperatures were read with an optical pyrometer. A schematic diagram of the experimental arrangement is shown in Fig. 5. A coaxial graphite sleeve was used to collect the boron which evaporated from the compact. This sleeve was separated from the compact by a 1/4-in. gap, in order to keep the sleeve cool relative to the compact and thus to condense the boron effectively. The temperature difference between the sleeve and the surface of the compact was generally 400° to 500°C .

The experimental procedure consisted in heating a compact under a given set of conditions, cooling it, removing the sleeve, and analyzing the sleeve for deposited boron. The analyses were accomplished by conventional chemical techniques. The compact was then reheated under the same conditions, with a new sleeve, and the process was repeated until two or more time points were obtained.

A brief investigation was made of the effect of oxygen on the rate of transport of boron out of the compacts. Thus, in two cases, the compact was swept with an atmosphere containing CO, since any oxygen entering the furnace would have been immediately reduced to CO at the expense of the graphite. In these experiments, the linear flow rate in the gap between the compact and sleeve was sufficiently low that the boron readily diffused to the sleeve and was not swept away.

Fig. 5--Schematic diagram of experimental apparatus

The boron loss experimental data are summarized in Table II and are plotted on log-log coordinates in Figs. 6 through 8. The dashed lines on these plots are not intended to be best fits but, instead, represent slopes of $\sqrt{t}$ and t .

Following run IV, the compact was reduced to a powder, and a boron analysis was performed. Within experimental error, the boron depletion during the run was negligible, indicating that the enhancement of boron transport as a result of the oxidation of boron carbide by CO is not a serious problem, at least under the conditions studied.⁷

It is desirable to be able to extrapolate the experimental results over a range of temperatures and times. Unless the transport mechanism is known or the experiments are amenable to at least empirical understanding, however, it is difficult to do this with confidence.

Zumwalt⁸ has considered a problem somewhat akin to the one considered here. He has treated the case of vapor-phase transport of boron through an isothermal graphite matrix. In such a case, the boron diffuses from the interior to the surface of the sample, where it evaporates. The peripheral region thus becomes depleted in boron, and a rather well-defined interface is established between the boron-containing region and the boron-depleted region. This "front" moves into the specimen at a rate

$$\frac{dx}{dt} = \frac{\dot{m}}{A\rho}, \quad (2)$$

where x is the radial distance, $\dot{m}$ is the mass of boron evaporating from the surface in time t , A is the evaporating area, and ρ is the concentration of boron in the loaded region. The diffusion in the vapor phase through this depleted region is given by Fick's law, and the solution is

$$\frac{m}{A} = \left(\frac{2\rho D_{\text{eff}} M \Delta P t}{RT} \right)^{1/2}, \quad (3)$$

where D_{eff} is the effective diffusion coefficient, M is the molecular weight of the diffusing species, ΔP is the difference in vapor pressure between

Table II
BORON LOSS

Run No.	Compact No.	Temperature* (°C)	Integrated Time (hr)	Integrated Loss (mg)	Integrated Loss† (%)	Conditions
I-1	1	2000 ±20	18.3	111	1.1	1 atm He
I-2	1	2000 ±20	40.1	230	2.3	1 atm He
I-3	1	2000 ±20	131	373	3.7	1 atm He
II-1	2	1990 ±10	17.4	128	1.3	1 atm He
III-1	3	2000 ±15	25.8	124	1.2	Vacuum
III-2	3	2000 ±15	48.0	241	2.4	Vacuum
IV-1	4	2000 ±20	25.0	134	1.3	3% CO in 1 atm He
IV-2	4	2000 ±20	63.0	218	2.2	3% CO in 1 atm He

*The values shown are for the highest surface temperatures of the compact. The average surface temperature was 30° to 70°C less than the reported value, and the centerline temperature was about 200°C greater than the reported value.

†The data are the per cent of boron in the compact at the start of each series (i. e., I, II, etc.) of runs.

Fig. 6--Boron loss; runs I and II (at 2000°C, in helium atmosphere)

Fig. 7--Boron loss; run III (at 2000°C, in vacuum)

Fig. 8--Boron loss; run IV (at 2000°C , in helium containing 3% CO)

the loaded-depleted interface and the surface of the specimen, and R is the gas constant. Because of the temperature gradients existing in the experiments, Eq. (3) cannot be expected to apply rigorously. Nevertheless, the equation has been found to be useful, as explained below.

The quantity D_{eff} is the diffusion coefficient of gaseous boron through the pores of the graphite. In vacuum, it is related to the Knudsen diffusion coefficient, D_K . With helium in the pores, it is a combination of D_K and an interdiffusion term for boron in helium, D_{12} . These can be combined in the following manner:

$$\frac{C}{D_{\text{eff}}} = \frac{1}{D_K} + \frac{1}{D_{12}}, \quad (4)$$

where C is a geometric factor for porous media. The interdiffusion term, D_{12} , can be estimated as a function of temperature for boron-helium by the method used by Zumwalt,⁸ and the Knudsen term is simply

$$D_K = \frac{1}{3} r \bar{V}, \quad (5)$$

where r is the mean pore radius, and $\bar{V}$ is the molecular velocity.

Equations (3) through (5) have been solved using r equals 3 microns and C equals 9×10^{-3} , with ΔP equal to the vapor pressure of boron at the temperature of the surface of the compact. The values for the pore radius and the geometric factor were taken as typical numbers for a porous graphite.⁹ (The permeability of one of the compacts was measured at the end of the experiments, and the measurement yielded a mean pore radius in qualitative agreement with the assumed value.) Equation (3) gave results which in every case agreed with the experimental values within a factor of five.

Thus, at least to the extent of this agreement, the mechanism of transport appears to be vapor-phase diffusion of boron atoms through the pores of the graphite. The loss rate is considerably less than one would predict from the Langmuir evaporation equation. Diffusion through the

matrix, rather than evaporation from the surface of the compacts, is the rate-determining step. (There is evidence for this statement from other, unpublished results obtained at General Atomic on similar systems.)

Equations (3) through (5) should be useful in extrapolating the present results over time, temperature, and graphite permeability. These equations predict an evaporation rate proportional to $\sqrt{t}$, whereas the experimental data indicate a dependence of the rate that is between $\sqrt{t}$ and t . The experimental evidence for the time dependence is somewhat tenuous, however. The temperature-dependence is predicted to be given approximately by the square root of the temperature-dependence of the vapor pressure.

IV. IRRADIATION STUDIES

When boron was selected as the control-rod poison, a review of the literature indicated that no information was available on boron-loaded graphite exposed to radiation under the conditions of the Peach Bottom HTGR. The properties of particular interest, because of the control rod movement and design, were the integrity, strength, and dimensional stability of the matrix graphite. It was therefore considered necessary to irradiate samples at the temperatures and exposures of the Peach Bottom reactor.

Twelve typical B_4C -graphite compacts prepared by the warm-press and sinter process were irradiated at 350° and $700^\circ C$ in four capsules. Each capsule contained three compacts, one each of 20, 30, and 40 wt-% boron. (The wide range of boron concentration was necessary because the exact concentration required for the reactor was not known at the time these tests were performed.) In order to accelerate boron disintegration in fission by a factor of about six, the samples were exposed in the core of the General Electric Test Reactor (GETR). Concurrently, the samples received a fast-neutron dose in excess of that expected in the Peach Bottom reactor.

Two capsules (one at each temperature) were irradiated for two cycles in GETR (equivalent to about one-third the exposure lifetime of a Peach Bottom HTGR control rod). The other two capsules were irradiated at the same temperatures, but for a total exposure equivalent to that expected for the Peach Bottom HTGR. Sample descriptions, irradiation histories, and postirradiation examination summaries are given in Tables III and IV.

One compact (of 40 wt-% boron, from capsule 2) was broken--possibly when the capsule was accidentally dropped in the hot cell. The other eleven compacts had essentially the same appearance as before irradiation. Photographs of the compacts taken before and after irradiation are shown in Figs. 9 through 13. Small defects on the compacts which were present before irradiation are readily discernible in the postirradiation photographs.

Diameter changes were very small (0.1% to 0.2%) and did not appear to be either temperature- or exposure-dependent. However, length changes were large (1% to 4%), and appeared to be temperature-independent and to increase with exposure.

In another series of tests, similar samples of graphite without B_4C have been irradiated at 650° to $700^\circ C$ for $\sim 1.65 \times 10^{21}$ nvt (>0.10 Mev). Dimensional changes were -0.28% in diameter and +0.09% in length. Therefore, the boron apparently enhances the expansion of the graphite in a direction parallel to the C-axis. The exact mechanism for this expansion has not been determined, although it might be expected that the concentration of the boron will be an influence. Unfortunately, there are insufficient data from which to draw conclusions. However, the samples with 20 wt-% boron and high exposures seemed to show greater expansions than those with 30 wt-% boron and similar exposures.

Strength tests were made by crushing the 1-in. -diameter by 1/2-in. -thick compacts on edge. The values of tensile stress at the center of the disks at failure were calculated by the method of Berenbaum and Brodie,¹⁰

Table III
IRRADIATION HISTORY OF B₄C-GRAPHITE
CONTROL-ROD MATERIAL*

Capsule No.†	No. of GETR Cycles	Sample Temperature from Melt Wire (°C)	Neutron Exposure (nvt)	
			Thermal	Fast
1	2	>330	9.8×10^{20}	9.8×10^{20}
2	5	>330	2.4×10^{21}	2.4×10^{21}
3	2	670-780	9.8×10^{20}	9.8×10^{20}
4	5	670-780	2.4×10^{21}	2.4×10^{21}

* The B₄C-graphite compacts were prepared by warm-pressing, followed by sintering at 1400°C. The size of the samples before irradiation was 1 in. in diameter by 1/2 in. long.

† Each capsule contained one sample each of 20, 30, and 40 wt-% boron in graphite.

Table IV

DIMENSIONAL CHANGES AFTER IRRADIATION OF B_4C -GRAPHITE
CONTROL-ROD MATERIAL*

Irradiation Conditions	Diameter Change (%)			Length Change (%)		
	20 wt-% B	30 wt-% B	40 wt-% B	20 wt-% B	30 wt-% B	40 wt-% B
350°C:						
2 cycles	-0.2	-0.2	0	+0.8	+1.3	+1.2
5 cycles	-0.1	0	(†)	+4.5	+2.5	(†)
700°C:						
2 cycles	-0.2	-0.2	0	+0.6	+1.5	+0.7
5 cycles	-0.2	+0.1	+0.2	+2.6	+2.3	+1.6

*The B_4C -graphite compacts were prepared by warm-pressing, followed by sintering at 1400°C. The size of the sample before irradiation was 1 in. in diameter by 1/2 in. long.

†Sample was broken.

(2.5×)

(a)

(2.5×)

(b)

Fig. 9--Photograph of B_4C -graphite (20 wt-% boron) compact (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $350^\circ C$

(2.5X)

(a)

(2.5x)

(b)

Fig. 10--Photograph of B_4C -graphite (30 wt-% boron) compact (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $350^\circ C$

(2.5×)

(a)

(2.5×)

(b)

Fig. 11--Photograph of B_4C -graphite (20 wt-% boron) compact (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $700^\circ C$

(2.5x)

(a)

(2.5x)

(b)

Fig. 12--Photograph of B_4C -graphite (30 wt-% boron) compact (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $700^\circ C$

(a)

(2.5×)

(b)

(2.5×)

Fig. 13--Photograph of B_4C -graphite (40 wt-% boron) compact (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $700^\circ C$

as illustrated in Fig. 14, from the formula

$$S = \frac{2W}{\pi \tau D} , \quad (6)$$

where S is the tensile stress at failure, W is the applied load at failure, τ is the specimen thickness, and D is the specimen diameter.

A general increase was noted in the strength of the irradiated 20 wt-% boron samples. No other significant changes in compact strengths due to irradiation were observed (see Table V). The increase of strength for the 20 wt-% boron samples was not entirely unexpected, for similar types of graphite without boron have shown increases of compressive strength after irradiation under approximately the same conditions.⁵

Metallographic examination of the compacts revealed no apparent changes in the B_4C particles or in the graphite matrix (see Figs. 15 through 17).

V. SUMMARY AND CONCLUSIONS

Boron carbide-graphite nuclear control-rod materials containing up to 45 wt-% boron have been prepared by hot-pressing techniques. The strength of compacts containing 20 wt-% boron was substantially higher than similar, unboronated graphite. It is suggested that this increased strength is a result of enhanced sintering promoted by the B_4C . Concentrations of boron higher than 30 wt-% result in progressively weaker and less dense bodies.

The thermal stability of these boronated graphites is excellent; at 1100°C (300° above the expected control-rod temperature in the Peach Bottom HTGR) the loss of boron by evaporation is predicted to be less than 0.05% in three years. At 2000°C , the loss has been found to be approximately 5% to 10% in 200 hr either in vacuum, in an atmosphere of helium, or in helium containing a small amount of CO. The diffusion of boron through the graphite matrix, rather than the evaporation from the surface,

Fig. 14--Determination of tensile strength by disk-crushing test:
 $S = 2W/\pi\tau D$

Table V
RESULTS OF DISK-CRUSHING TEST OF B₄C-GRAPHITE CONTROL-ROD MATERIAL^{*}
BEFORE AND AFTER IRRADIATION

Irradiation Conditions	Tensile Strength (psi)					
	20 wt-% B Compacts		30 wt-% B Compacts		40 wt-% B Compacts	
	Preirr.	Postirr.	Preirr.	Postirr.	Preirr.	Postirr.
350°C:						
2 cycles	1450	1900	1350	1800	1225	1400
5 cycles	1450	1400	1350	(600) [†]	1225	(**)
700°C:						
2 cycles	1450	1675	1350	1325	1225	1300
5 cycles	1450	1725	1350	1100	1225	1425

^{*} The B₄C-graphite compacts were prepared by warm-pressing, followed by sintering at 1400°C. The size of the sample before irradiation was 1 in. in diameter by 1/2 in. long.

[†] Sample was cracked.

^{**} Sample was broken.

(500x)

(a)

(500x)

(b)

Fig. 15--Photomicrographs of B₄C particles dispersed in graphite (40 wt-% boron) compacts (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about 350°C

(a)

(1000×)

(b)

(1000×)

Fig. 16--Photomicrographs of B_4C particles dispersed in graphite (40 wt-% boron) compacts (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $700^\circ C$

(a)

(500×)

(b)

(500×)

Fig. 17--Photomicrographs under polarized light of B_4C particles dispersed in graphite (40 wt-% boron) compacts (a) before and (b) after irradiation to 2.4×10^{21} nvt (thermal) at about $350^\circ C$

has been found to be the rate-controlling step in the loss of boron. An equation is given for extrapolating boron loss over time, temperature, and graphite permeability. This equation predicts a time-dependence of $\sqrt{t}$, whereas the experimental data indicate a dependence that is between $\sqrt{t}$ and t .

Irradiation testing was conducted on samples containing 20 to 40 wt-% boron at temperatures of 350° and 700°C and for exposures up to those predicted for the Peach Bottom reactor. In general, the materials were found to be satisfactory. Their physical integrity was good after irradiation, and in several instances their tensile strengths showed slight increases above the preirradiation values. Diametral shrinkages of 0 to 0.2% were equivalent to those obtained on unboronated graphites in similar irradiation tests. However, longitudinal expansions (in the direction of molding) were found to be larger than for unboronated graphites. In the Peach Bottom HTGR, this effect is not critical, but this form of expansion should be considered as possible and would have to be taken into account in other reactor systems.

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FOOTNOTES

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