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POSTIRRADIATION EVALUATION OF
BORON CARBIDE FROM
EBR-II HIGHER WORTH CONTROL ROD

G. L. Copeland
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MASTER

OAK RIDGE NATIONAL LABORATORY

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POSTIRRADIATION EVALUATION OF BORON CARBIDE FROM EBR-II HIGHER WORTH
CONTROL ROD

G. L. Copeland R. G. Donnelly

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ABSTRACT

The EBR-II higher worth control rod, assembly L-4008S, was irradiated to a maximum burnup of about 1.8% of ^{10}B at a maximum temperature of about 500°C. We evaluated the natural ^{10}B boron carbide pellets from two rods of this assembly. Our evaluation included ^{10}B analysis to determine burnup, x-ray analysis to determine lattice swelling, metallography, transmission and scanning electron microscopy to evaluate the structural changes, and vacuum fusion to determine the degree of helium retention. The boron carbide performed well in this application. Only minor microcracking occurred, and pellet swelling was low. Most of the helium generated appears to have been retained in the pellets. Defects observable by transmission electron microscopy were identified as platelike cavities containing helium at a pressure in excess of the surface tension restraint. The behavior of these cavities and the lattice parameters during annealing was similar to previously examined fast-reactor-irradiated boron carbide. Gas bubble coalescence on the grain boundaries was also observed after annealing.

INTRODUCTION

Some of the control rod positions in the Experimental Breeder Reactor II (EBR-II) can be used as instrumented test positions if the control worth of the remaining rods is increased. The EBR-II personnel determined that this could be accomplished by providing movable fuel control rods with boron carbide followers. They fabricated and irradiated a test assembly (L-4008S) to evaluate the performance of the boron carbide follower rods. In view of our interest in the fast-reactor-irradiation performance of boron carbide for the Fast Test Reactor (FTR), the EBR-II project provided rods G and H to Oak Ridge National Laboratory (ORNL), rods A and B to Hanford Engineering Development Laboratory (HEDL), and evaluated rod F themselves. The operating temperature of these rods is lower than the expected operating temperatures in the FTR rods, and the burnup achieved is about 10% of the average expected in the FTR. However, the evaluation of these pellets has provided useful information on the nature of fast-reactor-irradiation damage in boron carbide which will assist in the evaluation of boron carbide for the FTR. This report presents the results of the ORNL evaluation of rods G and H.

PREIRRADIATION CHARACTERIZATION

The boron carbide pellets were produced by Cerac, Inc., Menomonee Falls, Wisconsin. Characterization of the pellets has been reported.¹ The pellets were apparently hot pressed in graphite tooling. However, a number of iron-containing inclusions near the circumferential surface indicate that the pellets were precompacted in steel tooling.

The circumferential surfaces were left in the hot-pressed condition, resulting in variations in the pellet diameter measurements, but the ends of the pellets were ground to meet the requirements for end squareness. Individual pellet weights or densities were not recorded. The density of several test samples ranged from 92.4 to 99.4% of the assumed theoretical density of 2.52 g/cm³.

The pellets were fabricated from boron carbide powders of natural ^{10}B content. The boron and carbon analyses are given in Table 1 for (1) vendor analysis of the boron carbide powder not used, (2) Argonne National Laboratory (ANL) analysis of two pellets, and (3) ORNL analysis of a single pellet. The results are in good agreement and show the material to be nearly stoichiometric B_4C . However, some free carbon exists in the pellets as graphite, which is evident in the microstructures presented here.

1. *Preirradiation Characterization of B_4C Pellets Used in Experiments on High-Worth Control Rods*, coordinated by D. E. Walker, ANL/EBR-051 (January 1972).

Table 1. Boron and carbon content of boron carbide in EBR-II higher worth control rod

Analysis	Chemical composition (wt %)			B/C atomic ratio
	B	C	B + C	
Cerac, Inc. (powder) ^a	78.04	21.28	99.68	4.07
ANL (pellet 525-1) ^a	77.50	21.72	99.22	3.96
ANL (pellet 525-2) ^a	77.62	21.96	99.58	3.93
ORNL (pellet A-6)	76.8	21.6	98.4	3.95

^aPreirradiation Characterization of B₄C Pellets Used in Experiments on High-Worth Control Rods, coordinated by D. E. Walker, ANL/EBR-051 (January 1972).

The boron carbide portion of assembly L-4008S consisted of seven capsules. Each capsule contained 14 pellets nominally 1 in. long by 0.54 in. in diameter. The plenum containing the boron carbide was sealed with an atmosphere of helium at 15.7 psia inside.

IRRADIATION CONDITIONS

Subassembly L-4008S was irradiated in row 5 of EBR-II for a total of 54 1/2 MWd at a maximum power level of 62.5 MW (ref. 2). The subassembly initially occupied grid position 5A3 for 657 hr at power. It was then moved to grid position 5C1 for an additional 1289 hr at power. The boron carbide assembly was positioned above the fuel, resulting in a burnup and operating temperature gradient along the 14-in. length of the column. The maximum ¹⁰B burnup was expected in the first 2 in. of the pellet stack (pellets 14 and 13) and was calculated to be 1.8% ¹⁰B (3.9×10^{20} n,α/cm³). The average burnup was calculated to be 1.2% ¹⁰B (2.6×10^{20} n,α/cm³). The operating temperature calculations were made assuming that the bottom of the pellet stack was 4 in. above the core midplane and indicate a maximum operating temperature of about 500°C at the pellet center line.

GAS COLLECTION AND ANALYSIS

Neutron radiography and dimensional inspection of the capsules were done by ANL prior to shipment of the capsules to ORNL. These measurements showed no detectable change during irradiation.² Our first operation was to collect and measure the gas from the plenum. This was accomplished by placing the capsule in a chamber, evacuating the chamber to about 10⁻⁵ torr, puncturing the plenum with a laser beam, and then pumping the gas into a known volume and measuring the pressure. A sample of the gas was then taken for mass spectrographic analysis. The volume of the gas plenum in the capsule was determined by backfilling with argon.

A valve failure in the gas sampling system during the puncture of capsule G led to incomplete results for this capsule. The only reliable result from this measurement is the total gas volume of 55.58 cm³ at 1 atm and 24°C. After this measurement, mercury from the pump flowed back through the system into the capsule. The system was completely rebuilt and checked out with standards prior to puncturing capsule H

2. Unpublished memorandum, "Information Package on Higher Worth Control Rod, L-4008S," L. C. Walters to C. M. Walter, Argonne National Laboratory, Idaho Facilities, Oct. 27, 1971.

Table 2. Analysis of gas from capsule H

Element	Percent	
	Sample L	Sample R
He	99.1	99.5
O ₂	0.07	0.04
H ₂ O	0.3	0.1
H ₂	0.04	0.02
H ₃	<0.02	<0.02
N ₂ + CO	0.24	0.13
Ar	0.25	0.23
CO ₂	<0.02	<0.02
Kr	<0.02	<0.02
Xe	<0.03	<0.03

so that confidence can be placed in the following results for capsule H:

Plenum volume: 52.18 cm³
 Volume of gas: 64.02 cm³ at 1 atm and 24°C
 Pressure in capsule at 24°C: 17.99 psia

Two samples of the gas were analyzed and contained more than 99% He; complete analyses are shown in Table 2.

Before irradiation, the capsules were filled with helium at 15.7 psia (ref. 3). This indicates that the helium released to the plenum during irradiation was 8.30 cm³ at 1 atm and 24°C. This is 1.3% of the 636 cm³ (1 atm, 24°C) generated in the boron carbide, as calculated from the integrated burnup along the pin. If the helium pressure in the pin before sealing had dropped to 14.7 psia, this would raise the helium release to only 1.86% of that generated. Thus, it appears that most of the helium generated was retained in the boron carbide. This is confirmed by vacuum fusion analysis of the boron carbide as discussed below.

PELLET EVALUATION

The pellets were removed from the cladding after cutting off the top and bottom of the tube. The pellets slid from the tubing easily by tapping the tubing. Mercury from the gas-collection apparatus was observed on the pellets at both ends of capsule G. A couple of small chips were removed from both capsules. The pellets were examined at 10X and appeared to be in excellent condition. No cracks or other irregularities attributable to the irradiation were observed. The cylindrical surfaces of the pellets were very rough and all edges were chipped, but this appears to be the hot-pressed condition.

The activity of the pellets ranged from 30 to 200 mR/hr at 1½ in. However, the pellets smeared at up to 72,000 dis/min beta contamination. It was desirable to clean the pellets before evaluation to lower the beta contamination. For this reason the pellets were rinsed in 50 vol % nitric acid for 30 min, then rinsed in an ultrasonic bath of acetone, and then rinsed in alcohol. The pellets were placed in a 200°F air oven to dry. Visually this cleaning procedure did not appear to affect the pellets. The beta contamination was lowered to less than 20,000 and in most cases to about 3000 dis/min.

3. L. C. Walters, Argonne National Laboratory, Idaho Facilities, unpublished memorandum, Nov. 30, 1971.

Pellet Swelling

The results of length, diameter, and weight measurements are presented in Tables 3 and 4. The spread in diameter measurements on a given pellet due to the irregularity of the surfaces prevented any detection of diameter change. The lengths of the pellets in general increased by 0.001 to 0.002 in. (about 0.1 to 0.2%) during irradiation. Neither weights nor densities were reported for the individual pellets prior to irradiation. The as-irradiated densities, as calculated from average diameter, length, and weight, ranged from 93 to 97% of the theoretical density of 2.52 g/cm³. A total stack weight for the pellets was given so that an average density for the stack can be calculated using the pellet dimensions to calculate their volume. These calculations show an average stack density before irradiation of 95.1% of theoretical for capsule G and 95.3% for capsule H. The average stack densities after irradiation were 94.8 and 95.0% of theoretical for capsules G and H respectively. These density changes indicate an average swelling of 0.28 and 0.38% for the pellets in capsules G and H respectively. Although these calculations are approximate, the swelling is about what would be expected and is consistent with the 0.1 to 0.2% length change observed.

Burnup Analysis

Samples of three pellets from each capsule were analyzed for burnup by mass spectrographic analysis for ¹⁰B/¹¹B ratio. These analyses were made by placing a few particles of the boron carbide and some sodium hydroxide on the filament of the mass spectrograph and fusing the boron carbide on the filament. The results of these analyses are shown in Table 5. Since the burnup of ¹⁰B is quite low, a small difference in ¹⁰B content has a significant effect on the calculated burnup, and, as would be expected, there is

Table 3. Dimensions of pellets from capsule G

Pellet number	Unirradiated ^a			As irradiated						Density ^c	
	Diameter (in.)		Length (in.)	Diameter (in.)			Length (in.)	Weight (g)			
	Minimum	Maximum		Minimum	Maximum	Average ^b				g/cm ³	% of 2.52 g/cm ³
1	0.536	0.541	1.010	0.5381	0.5411	0.5407	1.0111	9.1329	2.401	95.3	
2	0.536	0.542	1.010	0.5373	0.5406	0.5385	1.0111	9.0550	2.400	95.2	
3	0.535	0.540	1.010	0.5366	0.5385	0.5375	1.0108	9.7862	2.338	92.8	
4	0.538	0.542	1.007	d	d	0.5405	1.0080	9.1100	2.404	95.4	
5	0.535	0.537	1.003	0.5355	0.5373	0.5365	1.0042	8.9400	2.403	95.4	
6	0.537	0.540	0.997	0.5385	0.5404	0.5395	0.9990	8.9818	2.400	95.3	
7	0.535	0.540	1.007	0.5365	0.5395	0.5379	1.0085	9.0786	2.404	95.4	
8	0.536	0.539	1.009	0.5368	0.5388	0.5372	1.0107	9.0092	2.400	95.3	
9	0.537	0.540	1.007	d	d	0.5382	1.0102	8.9320	2.372	94.1	
10	0.536	0.544	1.007	0.5322	0.5428	0.5385	1.088	9.0935	2.412	95.7	
11	0.535	0.540	1.016	0.5367	0.5387	0.5420	0.9934 ^e	8.8970	2.369	94.0	
12	0.537	0.540	1.008	0.5382	0.5389	0.5382	1.0099	8.9588	2.380	94.4	
13	0.540	0.544	0.900	d	d	0.5420	0.9015	8.2779	2.429	96.4	
14	0.535	0.538	1.010	0.5352	0.5362	0.5375	1.012	8.7986	2.338	92.8	
Total weight ^f								125.0015	Average	94.8	

^aValues from unpublished memorandum by L. C. Walters, Argonne National Laboratory.

^bAverage of four measurements.

^cCalculated from average diameter, length, and weight.

^dNot available.

^eRechecked.

^fTotal stack weight as loaded = 125.16 g.

Table 4. Dimensions of pellets from capsule H

Pellet number	Unirradiated ^a			As irradiated						
	Diameter (in.)		Length (in.)	Diameter (in.)			Length (in.)	Weight (g)	Density ^c	
	Minimum	Maximum		Minimum	Maximum	Average ^b			g/cm ³	% of 2.52 g/cm ³
1	0.537	0.541	1.003	0.5381	0.5404	0.5395	1.0035	9.0486	2.407	95.5
2	0.535	0.540	1.010	0.5365	0.5375	0.5369	1.0115	9.0421	2.410	95.6
3	0.536	0.540	1.010	0.5382	0.5408	0.5395	1.0115	8.9719	2.368	94.0
4	0.535	0.538	1.002	0.5353	0.5364	0.5357	1.0035	8.6834	2.343	93.0
5	0.536	0.542	1.002	0.5398	0.5417	0.5407	1.0036	9.0658	2.401	95.3
6	0.537	0.541	1.011	0.5386	0.5423	0.5405	1.0117	9.2060	2.420	96.0
7	0.535	0.542	1.003	0.5382	0.5414	0.5400	1.0038	9.0314	2.397	95.1
8	0.536	0.538	1.010	0.5370	0.5379	0.5382	1.0117	8.8695	2.352	93.3
9	0.537	0.539	1.008	0.5372	0.5407	0.5389	1.0094	8.9268	2.366	93.9
10	0.541	0.545	0.908	0.5424	0.5431	0.5428	0.9098	8.4175	2.440	96.8
11	0.534	0.543	1.010	0.5364	0.5384	0.5373	1.0127	9.1886	2.442	96.9
12	0.536	0.539	1.007	0.5366	0.5387	0.5374	1.0088	8.9115	2.377	94.3
13	0.534	0.537	0.996	0.5352	0.5357	0.5354	0.9981	8.8365	2.400	95.2
14	0.536	0.539	1.009	0.5374	0.5387	0.5380	1.0120	8.9734	2.380	94.5
Total weight ^d								125.1730	Average	95.0

^aValues from unpublished memorandum by L. C. Walters, Argonne National Laboratory.^bAverage of four measurements.^cCalculated from average diameter, length, and weight.^dTotal stack weight as loaded = 125.26 g.

Table 5. Burnup analysis of EBR-II higher worth control rod capsules G and H

Sample	At. % of total boron		Calculated burnup	
	¹⁰ B	¹¹ B	% of ¹⁰ B	$n, \alpha/\text{cm}^3$
× 10 ²⁰				
Unirradiated (control A-6)	19.81	80.19		
G-4	19.61	80.39	1.26	2.7
G-9	19.67	80.33	0.88	1.9
G-9 (repeat 6/14/72)	19.42	80.58	2.44	5.2
G-9 (repeat 7/21/72)	19.61	80.39	1.26	2.7
G-13-T-2	19.54	80.46	1.69	3.6
G-13-B-2	19.52	80.48	1.82	3.9
G-13-T-1	19.56	80.44	1.57	3.4
H-2	19.61	80.39	1.26	2.7
H-7	19.57	80.43	1.51	3.2
H-14	19.55	80.45	1.63	3.5
H-14 (repeat 6/14/72)	19.49	80.51	2.01	4.3
H-14 (repeat 7/21/72)	19.61	80.39	1.26	2.7

Fig. 1. ^{10}B burnup analyses on B_4C pellets from EBR-II higher worth control rod L-4008S.

appreciable variation in the burnup calculated for repeated analyses. These values are also shown in Fig. 1, along with the results from ANL and HEDL on the other capsules for comparison.⁴ The burnup calculated from helium content as determined by vacuum fusion (see below) is also included in Fig. 1. The burnup analyses agree well with the predicted burnup for the assembly of 1.8% ^{10}B burnup maximum with 1.2% ^{10}B burnup average.²

Lattice Parameter Measurements

Portions of three pellets from each capsule were annealed at temperatures from 400 to 1900°C in graphite boats. A portion of each piece was crushed to -325 mesh powder and loaded into capillaries for Debye-Scherrer x-ray powder patterns. Chromium $K\alpha$ (V-filtered) x radiation was used. Lattice parameters calculated from these patterns are shown in Figs. 2 and 3 plotted as functions of annealing temperature. The as-irradiated values are plotted at 500°C (estimate of maximum irradiation temperature), and the values from the unirradiated control sample (pellet A-6) are also presented. The volume changes calculated from the as-irradiated lattice parameters show a unit cell volume increase of about 0.3% through an increase in the rhombohedral a parameter and a slight decrease in the angle α . This is in contrast to the unit cell volume decrease of about 0.4% exhibited by boron carbide powders from the EBR-II experiment X-042 (ref. 5). The X-042 powders operated at higher temperatures of 650 to 730°C and exhibited a decrease in a and no change in α . Thermal irradiations of B_4C at temperatures of less than 500°C have generally shown unit cell volume increases. The behavior of the lattice parameters during annealing was similar for these pellets as for the X-042 powders. The a parameter first increased and then decreased with increasing annealing temperature. The angle α first showed a marked decrease and then a rapid increase with annealing

4. L. C. Walters, Argonne National Laboratory, Idaho Facilities, unpublished memorandum, Mar. 2, 1972.

5. G. L. Copeland, H. L. Yakel, and C. K. H. Dubose, *Evaluation of Fast-Reactor-Irradiated Boron Carbide Powders*, ORNL-TM-3729 (May 1972).

Fig. 2. Rhombohedral a parameter of EBR-II higher worth control rod boron carbide pellets.

Fig. 3. Rhombohedral angle α of EBR-II higher worth control rod boron carbide pellets.

temperature. The unit cell volume changed uniformly with annealing temperature as shown in Fig. 4 and returned to about the unirradiated value around 1300°C. This behavior is also similar to that observed for the X-042 powders.

Fig. 4. Unit cell volume change of EBR-II higher worth control rod boron carbide pellets.

Metallography

The microstructure of the unirradiated control sample is shown in Fig. 5. The microstructure is typical of carbon-rich hot-pressed B_4C with some free graphite in the grain boundaries, a typical amount of twinning, some metallic-appearing impurities, some intergranular porosity, and a small amount of intragranular porosity. The samples appeared to be in good condition after irradiation. A very few small cracks such as shown in Fig. 6 were observed which were primarily located in the interface between graphite particles and along adjacent grain boundaries.

We were unable to etch the as-irradiated sample because it would not conduct current for electrolytic etching. However, a sample which had been annealed did etch satisfactorily. We used an ohmmeter to obtain a rough estimate of the electrical resistance of some samples that had been annealed for hardness testing. The sharp points of the ohmmeter contacts were pressed onto the polished sample surface $\frac{1}{4}$ in. apart. The results are shown in Table 6. The as-irradiated and $700^\circ C$ annealed samples did not etch at all. The 1150 and $1600^\circ C$ annealed samples etched satisfactorily but not uniformly. A uniformly good etch was obtained on the unirradiated sample. The etchant used was 10% chromic acid in water at about 0.1 A for 30 sec. We do not have an explanation for this electrical conductivity behavior, nor do we know how typical it is. We note that the return of electrical conductivity during annealing coincides with the return of

Table 6. Electrical resistance of irradiated boron carbide pellets

Sample and condition	Electrical resistance ^a (Ω)
H-13, as irradiated	70,000,000
H-13, irradiated, annealed at $700^\circ C$	20,000,000
H-13, irradiated, annealed at $1150^\circ C$	2,500
H-13, irradiated, annealed at $1600^\circ C$	250
A-6, unirradiated control	150

^aCrude measure of electrical resistance between two sharp probes pressed onto metallographically polished surface $\frac{1}{4}$ in. apart.

Fig. 5. Microstructure of EBR-II higher worth control rod unirradiated control pellet A-6. (a) As polished. (b) Electrolytically etched with 10% chromic acid.

Fig. 6. Microstructure of EBR-II higher worth control rod pellet H-13 as irradiated. As polished.

the lattice parameters to their original values. Boron carbide has previously been electrolytically etched after irradiation in a thermal-spectrum reactor using a solution of 10 g Na_2CO_3 + 10 g NaNO_3 + 80 cm^3 of water.⁶

The purpose of annealing these samples was to conduct microhardness tests on them. Samples which had been annealed cut and broke differently than the as-irradiated samples. However, we were unable to obtain hardness readings because of fracturing of the samples. The anneal did not affect the microstructure in the as-polished condition, as shown in Figs. 7a and 8a. One difference noted in the etched samples is the apparent lack of twinning, as shown in Figs. 7b and 8b. Twins are observed in transmission electron microscopy, both as irradiated and after annealing, as discussed below. The spots in the grain boundaries after the 1600°C anneal (Fig. 8b) may be due to the enlargement of gas bubbles on the grain boundaries during etching. These bubbles are observed by transmission electron microscopy after annealing at 1600°C but not after 1150°C (see below). They are probably too small to be observed metallographically in the as-polished condition (Fig. 8a).

Transmission Electron Microscopy

The irradiation-induced defects in these pellets are similar to those observed previously in fast-reactor-irradiated powders.⁵ The defects in the pellets are smaller than those in the powders due to lower temperature and/or burnup. A representative transmission electron micrograph is shown in Fig. 9. Jostsons and DuBose⁷ have identified the defects as cavities which are ribbonlike with their largest face most commonly parallel to the trace of the {111} plane and less frequently parallel to the {100} and {110} traces of the rhombohedral unit cell. The lattice strain is apparently due to helium pressure in the cavities in excess of the surface tension restraint.

During annealing, the cavities in these pellets behave similarly to those in powders previously examined. No change is evident after 1 hr at 1000°C. After 1 hr at 1150°C, the strain is noticeably reduced and the cavities have begun to grow, indicating that they are indeed gas-filled. After 1 hr at 1450°C, the lattice is essentially strain-free, and the defects appear as slab-shaped bubbles. (In order to be consistent with electron microscopy terminology, the term "bubble" is used only when the gas pressure inside the cavity does not exceed the surface tension restraint.) The large faces of the bubbles are parallel to the traces of the {111}, {100}, and {110} planes, as was the case with the as-irradiated sample. After annealing 1 hr at 1900°C, the bubbles have grown larger and tend to be more equiaxed, as shown in Fig. 10.

During the initial examination of these as-irradiated samples, it was concluded that there were no bubbles on the grain boundaries. However, after observing⁸ extensive bubble precipitation and coalescence on the grain boundaries of pellets irradiated at higher temperatures in test X-099, these pellets were reexamined to look for grain boundary bubbles. A few areas were found where bubbles could be observed on the grain boundaries, as shown in Fig. 11. After annealing 1 hr at 1400°C, these bubbles grow and coalesce, as shown in Fig. 12, and appear very similar to those observed in the X-099 test. The integrity of the grain boundaries in these pellets remains better than those of the higher burnup X-099 pellets, even after the 1400°C anneal.

6. R. G. Gray and L. R. Lynam, *Irradiation Behavior of Bulk B_4C and $\text{B}_4\text{C} + \text{SiC}$ Burnable Poison Plates*, WAPD-261 (October 1963).

7. A. Jostsons and C. K. H. DuBose, "Microstructure of Boron Carbide after Fast Neutron Irradiation," *J. Nucl. Mater.* 44, 91-95 (1972).

8. G. L. Copeland and A. Jostsons, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1972*, ORNL-4820, p. 49.

Fig. 7. Microstructure of EBR-II higher worth control rod pellet H-13, irradiated and annealed at 1150°C for 1 hr. (a) As polished. (b) Electrolytically etched with 10% chromic acid.

Fig. 8. Microstructure of EBR-II higher worth control rod pellet H-13, irradiated and annealed at 1600°C for 1 hr. (a) As polished. (b) Electrolytically etched with 10% chromic acid.

Fig. 9. Transmission electron micrographs of EBR-II higher worth control rod pellet as irradiated. (a) Defect clusters and grain boundary denuded regions at low magnification (20,000X). (b) Contrast to show complex strain fields (100,000X). (c) Contrast to show helium cavities (100,000X). Reduced 38%.

Fig. 10. Transmission electron micrograph of higher worth control rod pellet after annealing 1 hr at 1900°C.

Fig. 11. Transmission electron micrograph of EBR-II higher worth control rod pellet as irradiated, showing network of small bubbles on grain boundary.

Fig. 12. Transmission electron micrograph of EBR-II higher worth control rod pellet annealed 1 hr at 1400°C after irradiation, showing coalescence of gas bubbles on grain boundaries.

Scanning Electron Microscopy

Fractured surfaces of as-irradiated and annealed pellets were examined by scanning electron microscopy. Figure 13 shows that fracture was predominantly transgranular in as-irradiated pellets, but was intergranular after annealing 1 hr at 1150°C. This is probably due to the gas bubble precipitation on grain boundaries during annealing. Gas bubbles on the grain surfaces are not recognizable by scanning electron microscopy at 1000 magnifications, even after annealing at temperatures up to 1600°C for 1 hr.

Vacuum Fusion

Portions of pellets from each rod were vacuum-melted, and the evolved gases were collected, measured, and analyzed to determine the amount of helium retained in the pellets after irradiation. The procedure involved outgassing a graphite crucible at 2500°C in vacuum, cooling, and then loading a previously weighed sample into the crucible without breaking the vacuum. The sample was then melted by heating to 2400°C. The evolved gases were pumped to a calibrated volume, and the pressure was measured. A sample of the gas was then taken and analyzed by mass spectrometry.

The results of the vacuum fusion analyses are shown in Table 7. The burnup for each pellet calculated by assuming that all the helium produced remained in the pellet is within the experimental error for the burnup analyses, although these points fall on the lower edge of the scatter band for rods G and H in Fig. 1. Thus, these results do not contradict the previous observation that most of the helium produced during irradiation remained in the pellets.

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Fig. 13. Scanning electron micrographs of fracture surfaces of EBR-II higher worth control rod pellets. (a) As irradiated. (b) After postirradiation anneal for 1 hr at 1150°C. 1000X. Reduced 40%.

Table 7. Vacuum fusion analysis
of B₄C pellets from rods G and H of EBR-II
higher worth control rod L-4008S

Pellet number	Helium content [cm ³ (STP)/g]	Calculated ¹⁰ B burnup based on helium content ^a (%)
G-4	2.50	0.78
G-8	3.88	1.21
G-14	4.75	1.48
H-2	2.46	0.77
H-7	3.47	1.08
II-14	5.36	1.67

^aAssuming all helium generated was retained in the pellets.

CONCLUSIONS

The boron carbide pellets performed well in this application as a neutron absorber in the EBR-II. Most of the helium generated during irradiation appears to have been retained in the pellets. Swelling was low and within the range which would have been predicted from thermal-reactor performance data. Structural integrity of the pellets was good, with only a few minor microcracks being observed metallographically. The irradiation-induced defect density was low enough so that identification of the defects as cavities containing helium was possible. The behavior of the crystal structure and the helium cavities during annealing was identical to that observed previously in fast-reactor-irradiated boron carbide. During annealing, this material exhibited gas bubble coalescence on the grain boundaries. This phenomenon has been responsible for extensive grain boundary microcracking in boron carbide pellets irradiated in the EBR-II at higher temperature and to higher burnup.

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