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Irradiation Performance of HTGR Fuel Rods in HFIR Experiments HRB-4 and -5

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OAK RIDGE NATIONAL LABORATORY

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IRRADIATION PERFORMANCE OF HTGR FUEL RODS
IN HFIR EXPERIMENTS HRB-4 AND -5

F. J. Homan, E. L. Long, Jr., R. L. Hamner,
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ENCLOSURE

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ABSTRACT

Experiments HBR-4 and -5 were the first tests of weak-acid-resin-derived fissile fuel. This fuel was later selected as the reference recycle fissile kernel. The WAK-derived kernels did not exhibit thermal migration during irradiation, but there was some concentration of rare-earth fission products at the inner surface of the SiC coating. These experiments were also a test of fuel rods carbonized in graphite tubes, to simulate in-block carbonization. The results of these tests showed significant problems associated with in-block carbonization, notably a mechanical interaction between particles and matrix material. A significant development program was launched to solve these problems. HBR-4 and -5 also included three specimens prepared by an extrusion fabrication route. These specimens generally performed well, although some tiny cracks in the SiC layer, apparently caused by the fabrication process but accentuated by the irradiation, were noted.

INTRODUCTION

The fourth and fifth capsules were HBR-4 and -5 in the series of irradiation experiments conducted in the removable beryllium (RB) reflector facility of the High Flux Isotope Reactor. The first three experiments and the sixth experiment have already been documented.¹⁻⁴

¹J. L. Scott et al., *An Irradiation Test of Bonded HTGR Coated Particle Fuels in an Instrumented Capsule in HFIR*, ORNL-TM-3460 (March 1972).

²J. H. Coobs et al., *Irradiation Performance in HFIR Experiment HBR-2 of HTGR Fuel Sticks Bonded with Reference and Advanced Matrix Materials*, ORNL-TM-3988 (January 1973).

³F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HBR-3 and ETR Experiment P13N*, ORNL-TM-4526 (October 1974).

⁴F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HBR-6*, ORNL/TM-5011 (in press).

Some information on the HRB-4 and -5 capsules has been published in annual reports.^{5,6} As has been the case with previous HRB capsules, the space in HRB-4 and -5 was shared with General Atomic Company (GAC). This report will deal mostly with the ORNL fuel rods irradiated in these capsules.

The ORNL fuel rods in the HRB-4 and -5 capsules contained three types of particles. The fissile particles were weak-acid-resin (WAR) loaded kernels, Triso coated. The fissile kernels were fully converted to the carbide (UC_2) form. The fertile particles were Biso-coated ThO_2 prepared by the sol-gel method. The third type of particle was inert and contained a kernel derived from strong-acid resin (SAR). The SAR kernels were heated to 3000°C before coating to eliminate sulfur. Both Biso- and Triso-coated inerts were used in this experiment, which was the first test of WAR-derived fissile fuel.

The objectives of the HRB-4 and -5 experiments were:

1. to test slug-injected bonded fuel rods prepared with different matrix materials and by different carbonization methods,
2. to test experimental fuel rods fabricated by GAC,
3. to test extruded fuel rod specimens,
4. to measure central fuel temperatures and monitor changes in thermal conductivity of extruded rods, and
5. to screen WAR-derived fuel for potential use in fissile particles.

All of these objectives were accomplished except No. 4. HRB-4 contained a tungsten-rhenium central thermocouple which decalibrated badly during the irradiation. In HRB-5 a 2-in.-long rhenium ultrasonic thermometer was used instead of a center thermocouple. This device decalibrated even more than the tungsten-rhenium thermocouple used in HRB-4. The centerline temperature data from the HRB-4 and -5 experiments have been of no value, although efforts are still under way to develop techniques for correction of the readings.

Each capsule contained 13 fuel specimens. Three specimens in each capsule were fabricated by extrusion (ORNL specimens) and the remainder by slug injection. There were six fuel rods in each capsule fabricated by GAC.

DESCRIPTION OF CAPSULES AND FUEL SPECIMENS

Capsules

The HRB-4 and -5 capsules were similar in design to the previous capsules in this series. Thirteen fuel rod specimens were inserted into a tapered, one-piece graphite sleeve made of Poco grade AXF-5Q.

⁵GCR Programs Annu. Progr. Rep. Dec. 31, 1972, ORNL-4911, pp. 107-12.

⁶GCR Programs Annu. Progr. Rep. Dec. 31, 1973, ORNL-4975 (in press).

The sleeve was located inside a primary containment capsule, which was in turn located inside a secondary containment capsule; both capsules were made of stainless steel. A cross section of the HRB-4 and -5 capsules is shown in Fig. 1. The nominal dimensions of the fuel and graphite sleeve are given in Fig. 2. The outside and inside diameters of the secondary containment capsule were 1.292 and 1.140 in., respectively, and 1.137 and 0.9676 in., respectively, for the primary containment. The taper of the graphite sleeve was somewhat different for the HRB-4 and -5 capsules than for other capsules in this series. Figure 2 does not accurately display the taper. There were actually two tapers in the bottom portion of the sleeve, as shown in Fig. 3. Other capsules had only one taper at each end, and capsules from HRB-7 on have had no taper. Figure 3 also shows the postirradiation shape of the capsule, which will be discussed in more detail later in this report. Figure 3 does not show the exact shape of the graphite sleeve; rather it shows the increase in size over the minimum diameter of 0.825 to 0.827 in. (0 and 90° orientations) at the bottom of the capsule.

The tapered graphite sleeve was used to compensate for the axial variation in neutron flux and gamma heating characteristics. Both flux and gamma heating rate (and therefore the fuel specimen heat generation rates) decrease as approximately a cosine function with distance from the reactor horizontal midplane. In order to obtain reasonably uniform specimen temperatures with this axially varying heat generation pattern, the thermal resistance of the radial gas gap between the graphite sleeve and the stainless steel capsule wall was increased through the use of the taper.

Fig. 1. HTGR Instrumented Capsules HRB-4 and -5 Irradiated in HFIR Removable Beryllium Facility.

Temperatures inside the capsules were monitored with eight Chromel-Alumel thermocouples and one centerline thermometer, located as shown in Fig. 4. In HRB-4 the centerline thermometer was a W-3% Re/W-25% Re thermocouple. In HRB-5 it was a rhenium ultrasonic thermometer (see Sect. 7.7 of Ref. 6 for a description of this device). The Chromel-Alumel thermocouples were 0.062 in. in diameter and sheathed with stainless steel. Six of these thermocouples in HRB-4 were coated with a 0.005-in.-thick protective barrier of tungsten applied by vapor deposition; two were uncoated. No difference was noted in the performance of the coated vs the uncoated thermocouples; therefore, in subsequent experiments the coatings were not applied. The bare wire W/Re thermocouple used in HRB-4 was threaded through a two-hole 0.062-in.-diam BeO insulator. The junction and lower 2 in. of the insulator were shielded from the fuel surface by a three-layer wrap of 0.0015-in.-thick rhenium foil. The foil can be seen in the x radiograph (Fig. 5). The upper fuel specimens, 1A, 1B, and 1C, had a 0.092-in. central hole to accommodate the centerline thermometer. High-purity Fe, Ni, Ti, and Co flux monitors in BeO tubes were located in the graphite sleeve in axial holes similar to those for the thermocouples. The graphite sleeve and fuel specimens were continuously swept with a 3600-cm³/hr high-purity helium-neon gas mixture at 1 to 2 atm. Gas samples of the sweep effluent were taken periodically for fission gas release determinations.

Fig. 4. Capsule HRB-4 Fuel Specimen and Thermocouple Locations.

Y-134261

Fig. 5. Preirradiation X Radiograph of Capsules HRB-4 and -5.

The sweep gas is present for temperature control as well as determination of the ratio of fission gas release rate to birth rate. The thermal neutron flux increases drastically at all axial locations during the approximately 23-day irradiation cycle. The percentage increase is greater at the ends of the capsule than at the horizontal midplane. To maintain constant operating temperatures under these conditions, it is necessary to vary the thermal resistance across the gas gaps. This is done with a varying helium-neon gas mixture. A high proportion of low-conductivity neon is used early in the cycle, when the thermal neutron flux is relatively low. More helium is used later in the cycle, when the neutron flux increases. However, because of the difference in the rate of flux increase at different axial locations, it is not possible to maintain exactly constant temperatures for all specimens.

Fuel

Loading Scheme

The types and numbers of specimens were identical for both capsules. A total of 13 specimens were irradiated in each experiment, six of which were fabricated by GAC and seven by ORNL. Of the seven ORNL specimens in each capsule, three were extrusions and the remaining four were fabricated by slug injection. The loading scheme is shown in Fig. 2; specimens 1A, 1B, and 1C are the extrusions; specimens 2A, 2B, 2C, 4A, 4B, and 4C are the GAC rods. The fuel loadings and dimensions for all specimens in both capsules are shown in Tables 1 and 2.

Coated Particles

All ORNL specimens contained the same Triso-coated fissile, Biso-coated fertile, and coated inert particles. The fissile particles were derived from weak-acid resins and were reduced and converted to the carbide form. Fertile particles were ThO_2 microspheres prepared by the sol-gel method. Coatings specified for these particles were conservatively designed. The coated inert particles (used for shim) had kernels that were derived from strong-acid resins, heated to 3000°C before coating to eliminate sulfur. All coated-particle batches were heated to 1800°C before fabrication into fuel rods to relieve residual stresses that might lead to coating breakage during fabrication. The characteristics of these particles are given in Table 3.

Table 1. Capsule HRB-4 Fuel Specimen Location and Loading

	Length (in.)	Volume (cm ³)	Fuel Loading, g/cm ³			Reactor Hmp ^a to Specimen Center (in.)
			²³⁵ U	²³⁶ U	²³² Th	
1 A	1.740	5.1772	0.0049	0.0770	0.1491	5.000
1 B	1.743	5.2355	0.0050	0.0791	0.1533	3.250
1 C	1.758	5.2624	0.0051	0.0795	0.1540	1.500
2A — Hmp	1.008	3.1321	0.0042	0.0660	0.1281	-0.125
2B	1.015	3.1609	0.0042	0.0654	0.1270	-1.125
2C	0.990	3.0743	0.0043	0.0673	0.1306	-2.125
3A	0.953	2.945	0.0043	0.0682	0.1321	-3.125
3B	0.953	2.945	0.0043	0.0682	0.1321	-4.125
3C	0.938	2.922	0.0043	0.0687	0.1331	-5.125
3D	0.956	2.978	0.0043	0.0675	0.1306	-6.125
4A	1.000	3.1113	0.0042	0.0665	0.1290	-7.125
4B	1.017	3.179	0.0041	0.0651	0.1263	-8.125
4C	1.004	3.1228	0.0042	0.0662	0.1285	-9.125

^aHmp = horizontal midplane.

Table 2. Capsule HRB-5 Fuel Specimen Location and Loading

		Length (in.)	Volume (cm ³)	Fuel Loading, g/cm ³			Reactor Hmp ^a to Specimen Center (in.)
				²³⁵ U	²³⁸ U	²³² Th	
	1 A	1.747	5.1980	0.0049	0.0775	7.1502	5.000
	1 B	1.746	5.2243	0.0050	0.0783	0.1518	3.250
	1 C	1.744	5.2318	0.0050	0.0791	0.1533	1.500
	2A	0.996	3.0995	0.0042	0.0667	0.1295	-0.125
	2B	1.005	3.1517	0.0042	0.0656	0.1273	-1.125
	2C	1.005	3.1132	0.0042	0.0665	0.1289	-2.125
	3A	0.949	2.933	0.0044	0.0685	0.1326	-3.125
	3B	0.953	2.945	0.0043	0.0682	0.1321	-4.125
	3C	0.952	2.954	0.0043	0.0680	0.1317	-5.125
	3D	0.956	2.966	0.0043	0.0677	0.1311	-6.125
	4A	0.997	3.1102	0.0042	0.0665	0.1290	-7.125
	4B	1.002	3.1461	0.0042	0.0658	0.1276	-8.125
	4C	0.997	3.1051	0.0042	0.0666	0.1293	-9.125

^aHmp = horizontal midplane.

Table 3. Characterization of Coated Particles for HRB-4 and -5

Batch Number	52A	OR-1856	OR-1833	OR-1815
Kernel				
Type material	UC ₂ ^a	ThC ₂ ^b	Carbon ^c	Carbon ^c
Uranium content, wt %	23.8			
²³⁵ U enrichment, at. %	5.99			
Thorium content, wt %		56.62		
Diameter, μ m	366 ^d $\pm$ 42 ^e	485 $\pm$ 30	370 ^f	370 ^f
Density, g/cm ³	6.2	9.72	1.4	1.4
Buffer				
Thickness, μ m	44.6 $\pm$ 6	88 $\pm$ 8	20 ^f	30 ^f
Density, g/cm ³	0.95 ^f	0.95 ^f	1.0 ^f	1.0 ^f
Inner carbon coating				
Thickness, μ m	30.8 $\pm$ 4		20 ^f	
Density, g/cm ³	1.94 $\pm$ 0.01		1.90 $\pm$ 0.06	
SiC				
Thickness, μ m	31.8 $\pm$ 6.6		15 $\pm$ 2	
Density, g/cm ³	3.21 $\pm$ 0.006		3.21 $\pm$ 0.002	
Outer carbon coating				
Thickness, μ m	28.3 $\pm$ 3.9	79 $\pm$ 7	30 ^f	30 ^f
Density, g/cm ³	1.89 $\pm$ 0.007	1.92 $\pm$ 0.023	1.98 $\pm$ 0.004	1.92 $\pm$ 0.022
Crushing strength, lb	3.44 $\pm$ 0.976	3.96 $\pm$ 0.955	1.88 $\pm$ 0.425	1.33 $\pm$ 0.639

^aWeak-acid resin derived.^bSol-gel derived.^cStrong-acid resin derived.^dMean.^eStandard deviation.^fStandard deviation not determined.

Preparation of Fuel Specimens

A general description of the fuel rod specimens for the two capsules is presented in Table 4. The graphitized Robinson coke was selected for the extrusions because it is isotropic and irradiation resistant. Thermax was added to the filler as an aid for extrusion. Varcum was selected as a binder because of its low viscosity at room temperature. All components were slurry blended with acetone until the solvent had evaporated. The mix was then extruded at room temperature and at a pressure of 1000 psi into graphite trays, cut to length in the green state, cured 16 hr at 90°C, carbonized at 1000°C on a 24-hr cycle, then finally heat treated at 1800°C to dimensionally stabilize the matrix. Due to some variation in the consistencies of different extrusion batches, matrix densities varied between 1.6 and 1.7 g/cm³, but they were considered acceptable for the experiments. Fuel distribution in the specimens appeared to be good, as shown by the radiograph in Fig. 5.

Table 4. General Description of Specimens for HRB-4 and -5

Specimen Number	Supplier	Nominal Dimensions, in.			Filler Material ^a		Particle Volume Loading (%)	Fabrication Technique	Carbonization Mode
		OD	ID	Length	Amount (wt %)	Type ^b			
1A	ORNL	0.490	0.090	1.75	160	Robinson coke	32.5	Extrusion	In covered graphite tray
1B	ORNL	0.490	0.090	1.75	119	Thermax	32.5	Extrusion	
1C	ORNL	0.490	0.090	1.75			32.5	Extrusion	
2A	GAC	0.490		1.0	40	1089 graphite	55-58	Admix	In tube (H-327)
2B	GAC	0.490		1.0	40	1089 graphite	55-58	Admix	In tube (H-327)
2C	GAC	0.490		1.0	110 140	Thermax RC4 graphite	55-58	Admix	In tube (H-327)
3A	ORNL	0.490		1.0	29.6	Asbury 6353	60-62	Slug injection	In packed alumina
3B	ORNL	0.490		1.0	38.7	RC4 graphite	60-62	Slug injection	In packed alumina
3C	ORNL	0.490		1.0	38.7	RC4 graphite	60-62	Slug injection	In block (H-327)
3D	ORNL	0.490		1.0	41.1	1089 graphite	60-62	Slug injection	In block (H-327)
4A	GAC	0.490		1.0	40	1089 graphite	55-58	Admix	In tube (H-327)
4B	GAC	0.490		1.0	40	1089 graphite	55-58	Admix	In tube (H-327)
4C	GAC	0.490		1.0	110 140	Thermax RC4 graphite	55-58	Admix	In tube (H-327)

^aBinders used were Varcum for extruded rods and Ashland A240 pitch for all other rods.

^bFiller materials designation and source are as follows: isotropic Robinson graphitized coke originally produced for Air Force Materials Laboratory by Union Carbide Corporation; Thermax, carbon black, from R. T. Vanderbilt Company; isotropic type 1089 graphite flour from Great Lakes Carbon Corporation; isotropic type RC4 graphite flour from Airco-Speer Corporation; and anisotropic type Asbury natural flake graphite 6353 from Asbury Graphite Mills, Inc.

The fillers selected for the slug-injected specimens consisted of two of the GAC experimental materials and the GAC reference grade NF 6353 natural flake graphite powder. Ashland grade A240 pitch was used in all these specimens as binder.

During fabrication of the slug-injection-bonded fuel rods, special precautions were required to obtain a satisfactory blend of the four types of coated particles. A decade splitter was used to divide the particles required for a single fuel rod into ten equal portions, each portion containing all types of particles. These portions were then transferred to the mold with a probe to minimize free fall of the particles and avoid segregation. Radiographs of fuel rods prepared in this manner showed quite uniform loadings (see Fig. 5). All the matrix materials were injected into the particle beds without difficulty at 180°C and 1000 psi. Two carbonization modes were used: positioned vertically in a packed bed of alumina or positioned horizontally in an H-327 graphite tube. The specimens were given a final heat treatment of 1800°C. Matrix densities and pitch coke yields of slug-injected specimens were highest for those carbonized in graphite tubes.

All specimens for the experiments were characterized by photography, radiography, metallography, weights, measurements, fuel loadings, and, in the case of the slug-injected-bonded specimens, by pitch coke yield. Data summaries of these characterizations are given in Table 5.

Table 5. Physical Characteristics of Specimens for HRB-4 and -5

Specimen Number	Position in Capsule	Average Diameter, in.		Average Length (in.)	Particle Volume Loading (%)	Matrix Density (g/cm ³)	Pitch Coke Yield (%)
		Outside	Inside				
Capsule HRB-4							
JI-11-9	1A	0.4890	0.0910	1.740	30.6	1.60	
JI-11-3	1B	0.4912	0.0905	1.743	31.3	1.69	
JI-11-5	1C	0.4904	0.0910	1.758	31.3	1.70	
M85A002	3A	0.4901		0.953	61.0	0.550	18.7
M86A025	3B	0.4899		0.953	61.0	0.733	25.0
M86A031	3C	0.4916		0.938	61.6	0.846	38.0
M95A002	3D	0.4916		0.956	60.4	0.840	35.8
Capsule HRB-5							
JI-11-10	1A	0.4890	0.0905	1.747	30.7	1.61	
JI-11-16	1B	0.4903	0.0905	1.746	31.1	1.64	
JI-11-4	1C	0.4909	0.0905	1.744	31.3	1.69	
M85A004	3A	0.4899		0.949	61.3	0.557	19.3
M86A026	3B	0.4904		0.953	60.9	0.750	26.0
M86A032	3C	0.4907		0.952	60.9	0.847	39.2
M95A007	3D	0.4911		0.956	60.6	0.854	38.4

It should be noted that in metallographic examination of specimens, matrix-particle interaction occurred in some of the slug-injection-bonded specimens that were carbonized in-tube, resulting in a small tear at the outer edge of the coating. Examples of this are shown later in the report. In some of the fissile particles contained in unirradiated extrusions, fine cracking of the silicon carbide occurred even though the buffer and outer low-temperature isotropic (LTI) coatings were not damaged. These phenomena in both types of specimens could only be seen at high magnifications.

DESCRIPTION OF IRRADIATION

Irradiation Conditions

Both capsules were inserted into the HFIR on October 3, 1972. HRB-4 was placed in the RB-7 facility, and HRB-5 was placed in the RB-5 facility. The locations of these facilities within the HFIR, and the neutron flux and gamma heating characteristics of the facilities have been described previously.⁷

Capsule Operation

Both capsules were inserted at the beginning of HFIR cycle 83. The HRB-4 operating history is given in Table 6. This capsule was irradiated for 11 HFIR cycles and was removed from the reactor on June 26, 1973. HRB-5 began operation at the same time as HRB-4, but was operated for only five cycles, so the influence of burnup and neutron exposure on performance could be determined. It was removed at the end of cycle 87, on February 3, 1973.

The operation of both capsules was stable throughout irradiation. There was an initial drop in the power production from the fuel specimens as the ^{235}U was burned out. This drop was expected. To compensate for the consumption of ^{235}U , the ^{232}Th and ^{238}U contents of each specimen were adjusted so that fissile ^{233}U and ^{239}Pu bred in by neutron capture would provide continuing fission power with a minimum of power variation with time. The fission heat rates vs time for each fuel specimen adjacent to a graphite thermocouple are presented in Fig. 6.

The neutron fluxes and fluences are presented in Tables 7 and 8 for each fuel specimen. Also shown in these tables are the calculated isotropic burnups for each specimen, which are based on the neutron fluxes given. The helium-neon sweep gas effluent was sampled periodically to measure fission gas release. The ratios of release rate to birth rate

⁷F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-3 and ETR Experiment P13N*, ORNL-TM-4526 (October 1974).

Table 6. Capsule HRB-4 Operating History

HFIR Cycle	Cycle		Days Operation	Accumulated Irradiation Time at 100 MW Reactor Power (hr)
	Begin	End		
83	10/8/72	10/23/72	15	364.3
84A	10/24/72	11/11/72	18	
84B	11/13/72	11/19/72	5	925.2
85	11/20/72	12/13/72	23	1479.4
86 ^a	12/13/72	1/7/73	24	2037.6
87A	1/9/73	1/10/73	1	
87B	1/12/73	2/3/73	21	2577.8
88	2/3/73	2/26/73	23	3119.0
89	2/27/73	3/22/73	23	3664.5
90	3/23/73	4/15/73	23	4213.4
91	4/18/73	5/11/73	23	4764.2
92 ^b	5/11/73	6/3/73	23	5310.1
93	6/3/73	6/26/73	23	5854.8

^aIncludes two days at 50 MW.

^bWithin the first three days of this cycle the reactor experienced five momentary power reductions.

Fig. 6. Fission Heat Rate vs Time for HRB-4

Micrographs for HRB-4 Specimens. (a) 1B, (b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

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(b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

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Fig. 6. (Continued) Fission Heat Rate vs Time for HRB-5 Specimens. (g) 1B, (h) 1C, (i)

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Power vs Time for HRB-5 Specimens. (g) 1B, (h) 1C, (i) 2B, (j) 3A, (k) 3B, (l) 4B.

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Table 7. HRB-4 Fuel Specimen Burnups and Neutron Exposures

<div style="display: inline-block; vertical-align: middle; text-align: center;"><div style="border: 1px solid black; padding: 2px;">← 0.49 in →</div><div style="border: 1px solid black; padding: 2px; margin-top: 5px;"><div style="border: 1px solid black; padding: 2px; text-align: center;">1 A</div><div style="border: 1px solid black; padding: 2px; text-align: center;">1 B</div><div style="border: 1px solid black; padding: 2px; text-align: center;">1 C</div><div style="border: 1px solid black; padding: 2px; text-align: center;">2A</div><div style="border: 1px solid black; padding: 2px; text-align: center;">2B</div><div style="border: 1px solid black; padding: 2px; text-align: center;">2C</div><div style="border: 1px solid black; padding: 2px; text-align: center;">3A</div><div style="border: 1px solid black; padding: 2px; text-align: center;">3B</div><div style="border: 1px solid black; padding: 2px; text-align: center;">3C</div><div style="border: 1px solid black; padding: 2px; text-align: center;">3D</div><div style="border: 1px solid black; padding: 2px; text-align: center;">4A</div><div style="border: 1px solid black; padding: 2px; text-align: center;">4B</div><div style="border: 1px solid black; padding: 2px; text-align: center;">4C</div></div></div>		Per Cent Initial Isotope Fissioned			Average Thermal Flux, $E < 0.414$ eV (n cm ⁻² sec ⁻¹)	Thermal Fluence, $E < 0.414$ eV (n/cm ²)	Average Fast Flux, $E > 0.18$ MeV (n cm ⁻² sec ⁻¹)	Fast Fluence, $E > 0.18$ MeV (n/cm ²)
		²³⁵ U	²³⁸ U	²³² Th				
		84.0	20.2	9.8	7.49×10^{14}	1.58×10^{22}	3.70×10^{14}	7.80×10^{21}
		84.0	23.5	12.3	9.38	1.98	4.35	9.17
		84.0	25.9	14.2	10.88	2.29	4.90	10.33
		84.0	26.5	15.0	11.71	2.47	5.00	10.54
		84.0	26.5	15.0	11.80	2.49	4.98	10.50
		84.0	25.0	14.3	11.68	2.46	4.83	10.18
		84.0	23.5	13.5	11.20	2.36	4.56	9.61
		84.0	21.5	12.5	10.47	2.21	4.24	8.94
		84.0	19.0	11.5	9.56	2.01	3.85	8.11
		84.0	16.2	9.6	8.50	1.79	3.43	7.23
		84.0	13.5	8.2	7.32	1.54	2.99	6.30
		84.0	10.5	6.5	6.14	1.29	2.50	5.27
		84.0	7.5	4.8	4.90	1.03	1.97	4.15

Table 8. HRB-5 Fuel Specimen Burnups and Neutron Exposures

←0.49in→		Per Cent Initial Isotope Fissioned			Average Thermal Flux, $E < 0.414$ eV (n cm ⁻² sec ⁻¹)	Thermal Fluence, $E < 0.414$ eV (n/cm ²)	Average Fast Flux, $E > 0.18$ MeV (n cm ⁻² sec ⁻¹)	Fast Fluence, $E > 0.18$ MeV (n/cm ²)
		²³⁵ U	²³⁸ U	²³² Th				
1	A	92.3	8.5	2.8	7.49×10^{14}	7.0×10^{22}	3.70×10^{14}	3.4×10^{21}
1	B	83.5	10.0	3.7	9.38	8.7	4.35	4.0
1	C	84.0	11.5	4.5	10.88	10.0	4.90	4.5
2A		84.0	12.0	4.8	11.71	10.8	5.00	4.6
2B		84.0	11.9	4.8	11.80	10.9	4.98	4.6
2C		84.0	11.0	4.5	11.68	10.8	4.83	4.5
3A		84.0	10.6	4.4	11.20	10.4	4.56	4.2
3B		84.0	9.5	4.0	10.47	9.7	4.24	3.9
3C		83.8	8.0	3.5	9.56	8.9	3.85	3.6
3D		83.5	7.0	3.0	8.50	7.9	3.43	3.2
4A		82.5	5.5	2.5	7.32	6.8	2.99	2.8
4B		81.0	4.0	1.8	6.14	5.7	2.50	2.3
4C		77.5	2.5	1.0	4.90	4.5	1.97	1.8

(Z/B) for selected isotopes vs accumulated irradiation time are shown in Tables 9 and 10 and are plotted in Figs. 7 and 8. The birth rates used in this calculation take into account the depletion of ^{235}U and fissions of bred-in ^{233}U and ^{239}Pu . The daily change in isotopic composition for each fuel rod is calculated, and then the fission product yield from each fissionable isotope.

Operating Temperatures

Capsules HRB-4 and -5 contained eight Chromel-Alumel thermocouples in the graphite sleeves and one centerline thermometer in specimen 1C. As indicated earlier, no useful data were obtained from the centerline thermometers because of the severe decalibration. Efforts are still under way to understand the decalibration kinetics of these devices, and the results of these efforts will be published at a later date.

Attempts have been made to directly calculate the operating temperatures of HRB capsules. The calculated and measured graphite midwall temperatures were used as a check on the accuracy of the calculation. This procedure yielded good comparisons in some cases and poor comparisons in others. There are numerous uncertainties which frustrate such attempts: (1) exact gamma heating rates for the materials involved as a function of time and axial location, (2) exact neutron fluxes and cross-section values, (3) knowledge of the time variation of the neutron flux as a function of axial position. This last item is the most uncertain. The shape of the curve for flux vs time is known only at the reactor horizontal midplane. Flux mapping and gamma heating experiments are under way in the HFIR, but in the meantime we have developed an alternate procedure for calculating specimen operating temperatures. This procedure is outlined in detail in Ref. 8 and will not be described here, except to say that mathematical models^{9,10} are used to determine the specimen fission heat generation rate by forcing the calculated graphite centerline temperature to match the measured temperature. Then, using the calculated fission heat rates in conjunction with sweep gas compositions, the fuel surface and center temperatures are calculated. During irradiation the sweep gas compositions are measured by flow meters. These data are improved through the use of the models and force fitting measured and calculated graphite midwall temperatures. This method has worked very well and has the added advantage of a built-in corrector to compensate for uncertainties. This is explained in Ref. 8.

⁸F. J. Homan et al., *Thermal Analysis of HTGP Fuel Rods Irradiated in the HFIR Removable Beryllium Facility*, ORNL/TM Report (in preparation).

⁹F. J. Homan et al., *HTRANS - A FORTRAN IV Computer Program for Thermal Analysis of Coated Particle Fuels*, ORNL/TM Report (in preparation).

¹⁰K. H. Valentine, *ENCTRK, A Computer Program to Model Heat Transfer Across Asymmetric Gaps in Cylindrical Fuel Rods*, ORNL/TM Report (in preparation).

Table 9. Ratio of Fission Gas Release Rate to Birth Rate (R/B) for HRB-4

Sample Number	Date	Time Irradiated (days)	Release Rate/Birth Rate				
			^{85}Kr	^{87}Kr	^{86}Kr	^{135}Xe	^{133}Xe
1							
2	10-17-72	9.175	1.415×10^{-5}	6.697×10^{-4}	5.414×10^{-5}	1.114×10^{-5}	3.196×10^{-5}
3	10-18-72	10.17	1.498×10^{-5}	5.576×10^{-4}	5.629×10^{-5}	1.339×10^{-5}	3.554×10^{-5}
4	10-23-72	15.14	2.346×10^{-5}	1.246×10^{-3}	7.857×10^{-5}	2.456×10^{-5}	5.198×10^{-5}
5	10-31-72	22.11	7.364×10^{-5}	3.459×10^{-3}	4.179×10^{-5}	7.117×10^{-5}	2.553×10^{-4}
6	11-16-72	35.85	1.725×10^{-4}	7.143×10^{-3}	4.800×10^{-5}	2.076×10^{-4}	5.272×10^{-4}
7	11-27-72	45.53	1.122×10^{-4}	6.383×10^{-3}	7.770×10^{-5}	1.081×10^{-4}	6.604×10^{-4}
8	12-1-72	49.70	1.583×10^{-4}	8.952×10^{-3}	1.237×10^{-4}	1.573×10^{-4}	8.689×10^{-4}
9	12-7-72	55.72	1.623×10^{-4}	8.647×10^{-3}	1.106×10^{-4}	1.590×10^{-4}	8.573×10^{-4}
10	12-19-72	66.94	9.508×10^{-5}	5.210×10^{-3}	7.101×10^{-5}	8.689×10^{-5}	5.270×10^{-4}
11	1-5-73	83.42	7.84×10^{-5}	4.62×10^{-3}	6.21×10^{-5}	7.38×10^{-5}	3.53×10^{-4}
12	1-23-73	96.65	6.04×10^{-5}	4.44×10^{-3}	5.13×10^{-5}	5.54×10^{-5}	3.73×10^{-4}
13	2-2-73	106.78	4.52×10^{-5}	3.27×10^{-3}	3.88×10^{-5}	3.27×10^{-5}	1.89×10^{-4}
13A	2-20-73	124.00	2.87×10^{-5}	2.38×10^{-3}	3.09×10^{-5}	2.85×10^{-5}	1.54×10^{-4}
14	3-12-73	142.35	4.22×10^{-5}	3.37×10^{-3}	3.14×10^{-5}	4.43×10^{-5}	2.71×10^{-4}
15	3-21-73	151.38	4.43×10^{-5}	2.89×10^{-3}	3.17×10^{-5}	3.47×10^{-5}	1.77×10^{-4}
16	3-29-73	158.35	4.65×10^{-5}	2.63×10^{-3}	3.56×10^{-5}	3.93×10^{-5}	2.35×10^{-4}
17	4-2-73	162.35	5.65×10^{-5}	3.33×10^{-3}	4.37×10^{-5}	5.19×10^{-5}	2.67×10^{-4}
18	4-10-73	170.42	5.64×10^{-5}	3.55×10^{-3}	4.26×10^{-5}	3.85×10^{-5}	4.60×10^{-4}
19	4-23-73	180.79	6.21×10^{-5}	3.77×10^{-3}	4.46×10^{-5}	4.58×10^{-5}	5.57×10^{-4}
20	4-26-73	183.80	6.32×10^{-5}	3.93×10^{-3}	4.66×10^{-5}	4.77×10^{-5}	5.23×10^{-4}
21	5-4-73	191.81	7.58×10^{-5}	4.04×10^{-3}	5.42×10^{-5}	4.51×10^{-5}	1.17×10^{-3}
22	5-9-73	196.74	7.19×10^{-5}	4.08×10^{-3}	5.08×10^{-5}	5.07×10^{-5}	6.81×10^{-3}
23	5-18-73	205.12	1.47×10^{-4}	8.32×10^{-3}	1.10×10^{-4}	8.00×10^{-5}	1.99×10^{-3}
24	5-24-73	211.29	1.42×10^{-4}	8.90×10^{-3}	1.06×10^{-4}	7.75×10^{-5}	1.47×10^{-3}
25	6-1-73	223.46	1.50×10^{-4}	8.11×10^{-3}	1.12×10^{-4}	7.80×10^{-5}	1.36×10^{-3}
26	6-11-73	228.62	1.64×10^{-4}	8.44×10^{-3}	1.06×10^{-4}	8.31×10^{-5}	2.35×10^{-3}
27	6-18-73	235.69	2.71×10^{-4}	1.28×10^{-2}	1.89×10^{-4}	1.07×10^{-4}	2.17×10^{-3}
28	6-25-73	242.63	3.16×10^{-4}	1.56×10^{-2}	2.63×10^{-4}	1.03×10^{-4}	2.46×10^{-3}

Table 10. Ratio of Fission Gas Release Rate to Birch Rate (R/B) for HRB-5

Sample Number	Date	Time Irradiated (days)	Release Rate/Birch Rate				
			^{85}Kr	^{87}Kr	^{88}Kr	^{133}Xe	^{135}Xe
2	10-17-72	9.2	6.36×10^{-6}	2.32×10^{-6}	2.28×10^{-6}	1.49×10^{-5}	5.35×10^{-6}
3	10-18-72	10.2	6.93×10^{-6}	3.13×10^{-6}	2.59×10^{-6}	1.50×10^{-5}	5.81×10^{-6}
4	10-23-72	15.1	1.44×10^{-5}	3.85×10^{-6}	4.65×10^{-6}	2.81×10^{-5}	1.25×10^{-5}
5	10-23-72	15.2	5.21×10^{-5}	1.09×10^{-5}	2.34×10^{-5}	2.71×10^{-5}	3.04×10^{-5}
6	10-31-72	22.1	1.93×10^{-4}	8.02×10^{-5}	1.14×10^{-4}	7.51×10^{-4}	1.86×10^{-4}
7	11-16-72	35.8	3.54×10^{-4}	1.83×10^{-4}	2.77×10^{-4}	9.00×10^{-4}	4.06×10^{-4}
8	11-27-72	45.8	8.93×10^{-5}	5.20×10^{-5}	6.08×10^{-5}	5.27×10^{-4}	5.67×10^{-5}
9	12-1-72	49.7	2.11×10^{-4}	1.07×10^{-4}	1.76×10^{-4}	1.42×10^{-3}	1.82×10^{-4}
10	12-7-72	55.7	1.43×10^{-4}	6.47×10^{-5}	1.02×10^{-4}	8.85×10^{-4}	1.11×10^{-4}
11	12-19-72	67.2	7.20×10^{-5}	3.45×10^{-5}	4.80×10^{-5}	4.17×10^{-4}	1.24×10^{-4}
12	1-5-73	84.1	7.46×10^{-5}	4.47×10^{-5}	5.62×10^{-5}	4.98×10^{-4}	6.95×10^{-5}
13	1-23-73	97.5	4.90×10^{-5}	3.51×10^{-5}	3.97×10^{-5}	2.18×10^{-4}	3.81×10^{-5}
14	2-2-73	106.8	4.66×10^{-5}	3.70×10^{-5}	3.85×10^{-5}	2.84×10^{-4}	4.12×10^{-5}

Fig. 7. HFIR HRB-4 Experiment Fission Gas Sample Data.

Fig. 8. HFIR HRB-5 Capsule Fission Gas Sample Data.

The calculated graphite midwall, fuel surface, and fuel center temperatures for the HRB-4 and -5 capsules are given in Fig. 9. Although not labeled explicitly, the uppermost curves of Fig. 9 are always fuel centerline temperatures, and the lowermost are always graphite midwall temperatures. As was the case with the fission heat rates shown in Fig. 6, only fuel temperatures adjacent to graphite midwall thermocouples are presented.

POSTIRRADIATION EXAMINATION AND ANALYSIS

Disassembly and Physical Examination

Visual examination of the exterior surfaces of capsules HRB-4 and -5 revealed no unusual or unexpected features. Gamma scans (0.55-0.75 MeV) made along the lengths of the capsules showed the fuel rods to be intact, with no gross segregation of fuel within the fuel rods or accumulation of fission products at the ends of the fuel columns. The graphite sleeves which contained the fuel rods were removed from the primary containment after cutting off the bottom and top of the capsules. The fuel rods were removed from the graphite sleeves. Fuel rods 2A, 2B, 2C, 4A, 4B, 4C, and 4D were visually examined and dimensionally inspected by a GAC representative; arrangements were made to return the GAC fuel rods to their hot cells for detailed examinations.

The extruded fuel rods (1A, 1B, and 1C) from HRB-4 and -5 were in excellent condition, as shown in Figs. 10-12. The appearance of the slug-injected rods 3A, 3B, 3C, and 3D from HRB-4 and -5 is shown in Figs. 13-16. The slug-injected rods from HRB-5 were in excellent condition. The following comments pertain to the slug-injected fuel rods from HRB-4, the higher fluence capsule. Fuel rod 3A was in good condition, with only slight debonding noted on the top and bottom edges. Although no failures were seen on the outer surface of rod 3A, a sooty region was noted on the bottom end of the rod that indicated a failure had occurred. Rod 3B was badly debonded in two regions at the top of the rod; stereo examination revealed only one failed particle on the surface of the rod. A rather large sooty region was noted on one side of fuel rod 3C in a partially debonded region; stereo examination revealed that numerous outer LTI layers had failed, exposing SiC layers on the side, top, and bottom surfaces of the fuel rod. Fuel rod 3D was in excellent condition; stereo examination revealed only one sooty region and that occurred on the top of the rod.

Dimensional Changes

The correlation of fuel rod dimensional change with fast fluence (>0.18 MeV) is shown in Fig. 17. As stated earlier, these fuel rods contained Biso- and Triso-coated particles; the particle volume loading is 60-62% for the slug-injected rods and about 30% for the extruded rods.

Fig. 9. Graphite Midwall, Fuel Surface, and Center Temperature

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Temperatures for HRB-4 Specimens. (a) 1B, (b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

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(a) 1A, (b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

3

TEMPERATURE (DEG. C)

(i)

TEMPERATURE (DEG. C)

(l)

Fig. 9. (Continued) Graphite Midwall, Fuel Surface, and Center Temperatures for HRB-5 Specimens. (g) 1B, (h) 1C, (i) 3A, (k) 3B, (l) 4B.

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Fig. 9. (Continued) Graphite Midwall, Fuel Surface, and Center Temperatures for HRB-5 Specimens. (g) 1B, (h) 1C, (i) 2B, (j) 3A, (k) 3B, (l) 4B.

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Fig. 10. Appearance of Extruded Fuel Rod 1A. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x. (c) Higher magnification of rod 1A from HRB-4, 5x.

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Y-138251

(b)

Appearance of Extruded Fuel Rod 1A. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x.
Magnification of rod 1A from HRB-4, 5x.

Fig. 11. Appearance of Extruded Rod 1B. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x. (c) Higher magnification of rod 1B from HRB-4, 5x.

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(a)

(b)

(c)

Fig. 12. Appearance of Extruded Fuel Rod 1C. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x. (c) Higher magnification of rod 1C from HRB-4, 5x.

Fig. 13. Appearance of Slug-Injected Fuel Rod 3C. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x. Higher magnification views of rod 3C, 5x; (c) side, (d) top, and (e) bottom.

Fig. 14. Appearance of Slug-Injected Fuel Rod 3B. (a) From HRB-5, 1.8x. (b) From HRB-4, 1.8x. Higher magnification views of rod 3B, 5x; (c) side, (d) top, and (e) bottom.

Fig. 15. Appearance of Slug-Injected Fuel Rod 3A. (a) From HRB-5, 1.8 $\times$. (b) From HRB-4, 1.8 $\times$. Higher magnification views of rod 3A, 5 $\times$; (c) side, (d) top, and (e) bottom.

Fig. 16. Appearance of Slug-Injected Fuel Rod 3D.
(a) From HRB-5, 1.8 $\times$. (b) From HRB-4, 1.8 $\times$. Higher magnification views of rod 3D, 5 $\times$; (c) side, (d) top, and (e) bottom.

Fig. 17. Dimensional Changes of Fuel Rods from HRB-4 and -5 Experiments.

The matrix densities were about 1.7 g/cm³ for the extruded rods and ranged from about 0.6 to 0.8 g/cm³ for the slug-injected rods. The onset of anisotropic shrinkage expected in the extruded rods was observed at about 6.5×10^{21} neutrons/cm². The surprising result was that the close-packed (slug-injected) rods showed the same anisotropic behavior. A check of the dimensions of the slug-injected green rods vs carburized vs annealed rods did not show any trend toward anisotropic shrinkage. One way to explain this effect would be to assume that the matrices of not only the extruded rods, but the slug-injected rods as well, have a preferred orientation; and at fluences above 6.5×10^{21} neutrons/cm², Biso coatings have densified sufficiently so that the matrix becomes the controlling factor, resulting in anisotropic shrinkage of the slug-injected fuel rods. The continuous matrix of an extruded rod is always the controlling shrinkage factor, since the volume of the rod occupied by coated particles is only about 50%.

Metallographic Examination

Metallographic sections of the four slug-injected rods were examined to determine the effect of carbonization mode on the irradiation performance of the three matrix compositions. The appearance of radial sections through the slug-injected rods from HRB-5 is shown in Figs. 18-21; no gross differences between the rods were apparent in these low-magnification views. Examination of the matrices at higher magnification revealed significant microstructural differences, as shown in Figs. 22-25. These differences reflected the matrix composition, carbonization mode, and densification from fast-neutron damage. The relationship between matrix type and microporosity is

Fig. 18. Radial Section Through Slug-Injected Fuel Rod 3A Irradiated in HRB-5. This fuel was bonded with FSV reference matrix and carbonized in packed alumina.

Fig. 19. Radial Section Through Slug-Injected Fuel Rod 3B Irradiated in HRB-5. This fuel was bonded with 40% RC4 in A240 pitch and carbonized packed alumina.

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rough Slug-Injected Fuel Rod 3A Irradiated in HRB-5. This fuel rod
matrix and carbonized in packed alumina.

R-43254

rough Slug-Injected Fuel Rod 3B Irradiated in HRB-5. This fuel
pitch and carbonized packed alumina.

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**Fig. 20. Radial Section Through Slug-Injected Rod 3C Irradiated in HRB-5, Bonded w
in A240 Pitch and Carbonized in H-327 Graphite Block.**

**Fig. 21. Radial Section Through Slug-Injected Fuel Rod 3D Irradiated in HRB-5, Bond
40Z GCL 1089 in A240 Pitch and Carbonized in H-327 Graphite Block.**

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Section Through Slug-Injected Rod 3C Irradiated in HRB-5, Bonded with 40Z RC4
Carbonized in H-327 Graphite Block.

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Section Through Slug-Injected Fuel Rod 3D Irradiated in HRB-5, Bonded with
Carbon and Carbonized in H-327 Graphite Block.

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Fig. 22. Appearance of Matrix in Fuel Rod 3A That Resulted From 29.6 wt % Asbury 6353 and Ashland A240 Pitch Carbonized in Packed Al_2O_3 (PSV Reference Matrix). (a) As-fabricated. (b) After 4.2×10^{21} n/cm². (c) After 9.6×10^{21} n/cm² ($E > 0.18$ MeV).

Fig. 24. Appearance of Matrix in Fuel Rod 3C That Resulted from 38.7 wt % RC4 Graphite and Ashland A240 Pitch Carbonized in H-327 Graphite Block. (a) As-fabricated. (b) After 3.6×10^{21} n/cm². (c) After 8.1×10^{21} n/cm² ($E > 0.18$ MeV).

Fig. 25. Appearance of Matrix in Fuel Rod 3D That Resulted from 41.1 wt % 1089 Graphite Carbonized in H-327 Graphite Block. (a) As-fabricated. (b) After $3.2 \times 10^{21} \text{ n/cm}^2$. (c) After $7.2 \times 10^{21} \text{ n/cm}^2$ ($E > 0.18 \text{ MeV}$).

given in Table 11 for the as-fabricated controls and the irradiated rods. As can be seen in this table, there was considerable variation in microporosity between the as-fabricated rods, with a low of 21% occurring in rod 3C, which also had the highest matrix density and pitch-coke yield. The individual graphite filler particles were discernable in the as-fabricated rods that contained the Fort St. Vrain (FSV)-type (3A) and experimental matrices that were carbonized in packed Al_2O_3 . The matrices of the rods carbonized in graphite tubes were more dense, and the individual filler particles were not discernable. The denser matrices resulted from the higher yield of coke from the pitch in rods carbonized in the graphite tubes. With the exception of rod 3D, additional densification (decrease in microporosity) of the matrices occurred during irradiation.

Table 11. Variation in Microporosity of Matrices Used in Fabricating Fuel Rods for HRB-4 and -5 With Matrix Density, Pitch-Coke Yield, and Fast-Neutron Damage

Fuel Rod Identification ^a	Micro-porosity (%)	Fast Fluence ($E > 0.18 \text{ MeV}$)	Matrix Density (g/cm^3)	Pitch-Coke Yield (%)
3A, as fabricated	63		0.56	~20
3A, HRB-5	4	4.2×10^{-21}		
3A, HRB-4	23	9.6		
3B, as fabricated	27		0.73	~25
3B, HRB-5	28	3.9		
3B, HRB-4	15	8.9		
3C, as fabricated	21		0.83	~40
3C, HRB-5	16	3.6		
3C, HRB-4	9	8.1		
3D, as fabricated	25		0.84	~38
3D, HRB-5	30	3.2		
3D, HRB-4	25	7.2		

^aSee Table 4 for matrix composition and carbonization mode.

Close examination of the fuel rods revealed no failed particles in the rods fabricated with the FSV matrix (3A), except for an occasional inert Biso particle that had been crushed during fabrication. Nor were any failed fuel particles observed in the fuel rods with an experimental matrix carbonized in packed Al_2O_3 (3B). However, a significant number of pyrolytic carbon coatings in the rods with experimental matrices carbonized in graphite tubes (3C and 3D) had failed as a result of matrix-particle interaction. Two typical areas from rods 3C and 3D irradiated in HRB-4 are shown in Fig. 26; as can be seen, the failure frequency of the pyrocarbon coatings was greatest in 3C. Higher-magnification views of failed Biso and Triso coatings on inert kernels are shown in Fig. 27. Although no failed Biso-coated ThO_2 particles were observed, tears in the LTI coating were present that extended about halfway through the coating thickness (Fig. 28). Examination of the as-fabricated rods in carbonized graphite (3C and 3D) revealed that small tears were present in the outer coatings of the Biso- and Triso-coated particles (Fig. 29). These tears formed during fuel rod fabrication and were the initiation sites of coating failure as the matrix and coatings densified with fast fluence.

The appearance of a radial section through an extruded rod, 1C, from HRB-4 is shown in Fig. 30; except for a slight exaggeration of the cracks, no obvious changes were apparent in the continuous matrix. Examination of the coated particles at a higher magnification revealed that a significant number (~25%) of the SiC coatings on the Triso-coated fissile kernels contained cracks [Fig. 31(a)]. None of the SiC layers on the Triso-coated inert kernels exhibited cracks. (The thickness of the SiC layer was about 15 μm on the inert particles and 32 μm on the fissile particles.) Close examination of the archive sample revealed that fine cracks were present in the SiC layers on the fissile particles of the as-extruded rods and were exaggerated by irradiation. None of the LTI outer layers on the fissile particles had failed, although a few (~2%) of the inner LTI coatings had failed in particles that contained cracked SiC layers [Fig. 31(b)]. With the exception of one cracked inert Biso particle, none of the Biso-coated particles showed evidence of potential failure, nor was there any evidence of matrix-particle interaction.

Thus, it appears that in-block carbonization has presented some unexpected problems. Also, there are apparent problems with the extruded fuel rods, since an unacceptable fraction of cracked SiC layers were present in the Triso fissile particles in the as-fabricated condition. Irradiation testing either increased the number of cracked SiC layers or merely enhanced them, so that they were much more readily detected during metallographic examination.

Metallographic examination of the Triso-coated WAR fissile particles contained in the slug-injected rods revealed several potential problems for the SiC layers. In the relatively small number of fissile particles exposed in the planes of polish, about one-third showed evidence of attack in inner surface regions of the SiC layers. This attack appeared as localized regions and was not temperature-gradient dependent. The attack varied in degree of severity from only slight

Fig. 26. Typical Areas from (a) Rod 3C and (b) Rod 3D Irradiated in HRB-4 Showing Failed Outer LTI Layers of Triiso Coatings on Inert and Fissile Kernels. The kernels missing from the Triiso coatings were lost during sample preparation.

Fig. 27. Failed (a) Triso and (b) Biso Coatings on Inert Kernels That Failed Because of Matrix-Particle Interaction in Rods Carbonized in Graphite Tubes.

Fig. 2C. Examples of the More Severely Damaged Biso Coating of Fertile Particles Contained in Rods Carbonized in Graphite Tubes. (a) From HRB-5, rod 3D. (b) From HRB-4, rod 3C.

Fig. 29. Unirradiated Triso-Coated Particle in As-Fabricated Control Fuel Rod 3D. Note small tears in the peripheral region of the outer coating.

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Fig. 30. Radial Section Through Extruded Fuel Rod Specimen 1C Irradiated in HRB-4. This fuel rod contained Triso-coated weak-acid resin fissile particles, Biso-coated ThO_2 fertile particles, and Triso and Biso inert particles bonded with Robinson graphitized coke and Varcum and carbonized in a covered graphite tray.

Fig. 31. Triso Coatings With Cracked SiC Layers in Extruded Fuel Rod 1C, HRB-4. (a) Cracked SiC. (b) Cracked SiC and inner LTI layer.

to complete penetration of the thickness of the SiC. A general degradation of the inner surface of the SiC was also apparent in many of the coated particles. Analysis of a region of attack with the aid of an electron microprobe revealed the fission product palladium to be the culprit. The results of this analysis are shown in Fig. 32. The attacked regions appeared to have been molten. A palladium-silicon phase diagram shows three eutectics that have melting points below the surface temperature of the fuel rods (15.5% Pd, mp 800°C; approximately 45% Pd, mp 720°C; 58% Pd, mp 850°C).¹¹ This result would be atypical of HTGR conditions, since the bulk of the palladium present was a fission product from ^{239}Pu and not ^{235}U (the UC_2 was enriched to about 6% ^{235}U). However, this result could become significant should a decision be made to incorporate plutonium in the HTGR fuel cycle.

In addition to the above problem, about one-fourth of the UC_2 kernels appear to have phase segregation and were in intimate contact with the inner LTI coating, and a metallic-like ingot had attacked the SiC coating. Although no SiC coatings had failed as a result, up to one-half the thickness of the SiC appeared to have been dissolved by the metallic-like material; one such particle is shown in Fig. 33; the results from an electron microprobe analysis are shown in Fig. 34. The rare earths La, Ce, Pr, and Nd were the fission products associated with the localized dissolution of the SiC layer. Note also that uranium was not associated with this reaction zone and that there is no obvious concentration of silicon in the kernel.

The rare-earth fission products were also found concentrated on the inner surface of the cooler side of the SiC coating, as has been reported in other coated-particle systems that have been exposed to a thermal gradient.¹² The presence of the rare earths led to penetration of about half the thickness of the SiC coating; both rare-earth and palladium attacks are shown in Fig. 35. The fissile kernels were varied in appearance, and it is therefore not possible to show a "typical" particle; some of the types of particles examined from HRB-4 are shown in Fig. 36. Generally, except for kernels that reacted with the SiC, the kernel and buffer layer densified, creating a gap at the inner surface of the inner LTI coating.

The kernels of the Biso fertile particles in the slug-injected fuel rod 3A from HRB-4 exhibited a slight "amoeba" effect. The ThO_2 kernels had migrated about 20 μm [Fig. 37(a)]. Although there was no measurable movement of the ThO_2 kernels in the other three fuel rods, the densified-buffer recoil zone was noticeably darker on the hot side of the kernel in rod 3B [Fig. 37(b)]. An electron microprobe analysis was unable to detect any chemical difference between the hot and cold side of the buffer layer.

¹¹Max Hansen, *Constitution of Binary Alloys*, 2nd ed., McGraw-Hill, New York, 1958, p. 1126.

¹²HTGR Base Program Quart. Progr. Rep. May 31, 1973, GULF-GA-A12599, pp. 87-93.

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Fig. 32. Palladium-SiC Reaction in a Triso-Coated WAR Fissile Particle Contained in Fuel Rod 3A, HRB-4, as Displayed by Electron Microprobe. (a) Optical micrograph. (b) Backscattered electrons. (c) U M_β. (d) Pd L_α. (e) Si K_α.

Fig. 33. Triso-Coated WAR Fissile Particle in Slug-Injected Fuel Rod 3B, HRB-4, Showing Chemical Interactions Between Kernel and Coatings. (a) Bright field. (b) Polarized light.

Fig. 34. X-Ray Displays From an Electron Microprobe Analysis of a Triso-Coated WAR Fissile Particle Contained in Fuel Rod 3B, HRB-4. Note chemical attack of SiC layer by rare-earth fission products Ce, Nd, La, and Pr. (a) Optical photograph. (b) Backscattered electrons. (c) U M_{α} . (d) La L_{α} . (e) Pr L_{α} . (f) Si L_{α} . (g) Ce L_{α} . (h) Nd L_{α} .

Fig. 35. Triso Coatings on WAR Fissile Particles Irradiated in HRE-4. The SiC coating on the left shows the result of attack by rare-earth fission products on the cold side of the particle. The SiC coating on the right shows attack by palladium.

Fig. 36. Appearance of Triso-Coated WAR Fissile Particles Contained in Slug-Injected Rods Irradiated in HRB-4.

Fig. 37. Bisco-Coated ThO₂ Fertile Particles in Slug-Injected Fuel Rods 3A and 3B Irradiated in HRB-4. (a) Rod 3A. Note migration of ThO₂ kernel - hot side of particle is on the right. (b) Rod 3B. Note dark region in buffer on hot side (bottom) of the kernel.

The extruded rods operated at a higher heat rating and in a higher flux than did the slug-injected rods. However, the overall temperature of the extruded rod was lower than that of the slug-injection rods because of the higher matrix density. Metallography showed that the WAR kernels had densified, as expected (Fig. 38); but in contrast to the fissile particles in the slug-injected rods, there was *no* evidence of fission product attack on the SiC layers. Examination of the fertile particles showed no evidence of the "amoeba" effect or indication of potential failure.

SUMMARY AND CONCLUSIONS

HRB-4 and -5 were the first tests of weak-acid-resin-derived fissile fuel. Approximately nine months after the completion of this test, the WAR fissile fuel was selected as the reference recycle fissile kernel, replacing the dense mixed oxide kernel. The decision was made on the basis of the performance of the WAR kernels in HRB-4 and -5. The good behavior of WAR fuel in GAC irradiations tests (P13P)¹³ was also a factor in the decision to make this fuel the reference recycle kernel.

The primary deficiency in the dense mixed oxide kernel was the amoeba effect, or thermal migration of the kernels up the temperature gradient. This effect was not present in the WAR kernels irradiated in HRB-4 and -5. There was some concentration of fission products at the inner SiC boundary, primarily Pd, Ce, Pr, La, and Nd. Yields of Pd from Pu fissions are higher by a factor of 8 than from ²³⁵U or ²³³U fissions. The HRB-4 and -5 fuel was only partially enriched (about 6-7% ²³⁵U) compared with the fully enriched material which will be used in HTGRs. Therefore, Pd will not be a problem. The yields of the rare-earth fission products (Ce, Pr, La, and Nd) are about the same for Pu and U fissions, but subsequent irradiations have shown that these fission products can be retained in the kernels in oxide form by adjustment of the initial kernel stoichiometry.

A significant observation from the HRB-4 and -5 tests was the interaction between particle and matrix in the specimens carbonized in graphite tubes. Apparently the outer coatings of some particles were torn away due to a strong bond between particle and matrix and irradiation-induced shrinkage of both coatings and matrix. This problem, which was first identified by the HRB-4 and -5 experience, has received considerable attention in the Thorium Utilization Program development work subsequent to this experiment. The particle-matrix interaction is thought to be dependent upon pitch-coke yields during carbonization, on the amount and distribution of macro- and microporosity, and on the strength of the coatings themselves. In HRB-4 and -5 the worst cases of particle-matrix interaction were noted in specimens which had the lowest fraction of microporosity after fabrication. These slug-injected specimens also had the highest matrix density and the highest pitch-coke yield. The crush strength values reported in

¹³HTGR Base Program Quart. Progr. Rep. May 31, 1974, GULF-CA-A13030, pp. 9-8 through 9-20.

(a)

(b)

Fig. 38. Appearance of Typical Triso-Coated WAR Fissile Particles in Fuel Rod 1C Irradiated in HRE-4. Note kernel and buffer have densified but there is no evidence of migration or fission product attack.

Table 3 are typical for fissile and fertile particles. The crush strengths of the inert particles may be a little low, but this does not necessarily reflect "weak" coatings, because the coating thicknesses are low compared with the thicknesses on the fissile and fertile particles. Much progress has been made in understanding the matrix-particle interaction problem, and potential solutions are being checked in additional irradiation tests.

The performance of the extruded fuel rods in HRB-4 and -5 was generally good. Only eight extruded fuel rods have been tested in the ORNL HTGR fuel development program, six of them in HRB-4 and -5 and the remaining two in HRB-6. The appearance of the extruded rods in HRB-4 and -5 after irradiation was very good - better than the appearance of the slug-injected rods. The presence of small cracks in the SiC layer after fabrication suggests that the fabrication techniques for extruded fuel require additional development. Unfortunately, the scope and funding level of the ORNL program have not permitted additional development of extrusion technology beyond what has been demonstrated in HRB-4, -5, and -6.

The dimensional behavior of the fuel rod specimens in HRB-4 and -5 was about what was expected from the results obtained in previous experiments. The anisotropic behavior of the slug-injected fuel rods above a fast fluence of 6.5×10^{21} neutrons/cm² was somewhat surprising, since it had been thought that the dimensional changes in these high-particle-volume rods would be controlled by the dimensional changes in the particles themselves, and would therefore be isotropic. However, it is now thought that the particles shrink away from the matrix, and at high exposures the dimensional changes are caused by the matrix itself.

All of the experimental objectives of HRB-4 and -5 were met except one - the measurement of fuel centerline changes and monitoring the change in thermal conductivity with irradiation for the extruded fuel rods. This was not accomplished due to the severe decalibration of the centerline thermometers. Analytical methods are being investigated for correction of the thermometer readings, and if these methods are successful, the last objective may still be met.

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APPENDIX

Thermal Analysis

APPENDIX

Thermal Analysis

The calculated specimen fission heat rates and operating temperatures have already been presented in this report. This Appendix will present some of the backup information used in making these calculations. As indicated previously, HRB capsules contain a number of Chromel-Alumel thermocouples in the graphite sleeve. One of these thermocouples is selected to control the temperatures of the capsules. In HRB-4 and -5 the thermocouple selected was No. 7, adjacent to specimen 4B. In operating HRB-4 and -5 it was desired to keep the temperature of the graphite midwall between 900 and 940°C in order for the fuel specimens to operate at their design temperatures. Due to the increase in the fission heat rate during each cycle, control was maintained by reducing the thermal resistance across the fuel-graphite gap and the graphite-stainless steel gap, with increasing heat rate. This was done by varying the composition of the helium-neon sweep gas. Early in the cycle, when the heat rates were low, a high concentration of low-conductivity neon was used in the sweep gas. As the heat rates increased, the concentration of high-conductivity helium was increased. Knowledge of the exact composition of the sweep gas is necessary for accurate thermal analysis. Flowmeter data give indication of the gas composition, but detailed analysis of these data have shown them to be unreliable. Also, the variation of thermal flux as a function of time at different axial positions is not constant. Therefore, constant operating temperatures cannot be achieved for all specimens using this control procedure. This is evident from examination of the curves in Figs. 6 and 9.

Data for sweep gas composition vs time for HRB-5 are presented in Fig. A-1. This information is derived from flowmeter readings. No sweep gas data were taken during operation of HRB-4. There are gaps in the HRB-5 data where no readings were taken, and in some regions the data are inconsistent with the thermocouple readings. For example, during cycle 86 the flowmeter data suggest a big change (10% increase) in the helium content at day 9. The thermocouple data suggest a much more modest change. During cycle 87 the flowmeter data suggest a 14% increase in helium content at day 20. The thermocouple data reflect a much more modest change.

The deficiencies noted with the HRB-4 and -5 sweep gas data caused some modifications to be made in the thermal analysis procedure. Three parameters are of concern in the analysis - the graphite midwall temperatures, the fuel specimen fission heat generation rate, and the sweep gas composition. With any two of these parameters, the third can be calculated. Beginning with cycle 88 a modification was made in the way the capsules were operated. Time periods were interspersed throughout each cycle where the sweep gas was 100% helium. This fixed one of the above-mentioned parameters. Knowing the graphite midwall temperature during these periods permitted a calculation of the specimen heat generation rate. Assuming that the change in heat rate was a smooth

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function of time in the cycle, a curve was drawn. From this curve, a back calculation of the sweep gas compositions throughout the cycle was made. Unfortunately, the interspersed periods of 100% helium operation were not available for cycles 83 through 87. To permit use of the analytical techniques perfected for other capsules, the graphite midwall temperature profiles expected for 100% helium operation have been "drawn" in as shown in the example given in Fig. A-2. The sweep gas compositions inferred by using this procedure are shown in Figs. A-3 and -4. Figure A-4 can be compared with Fig. A-1 for HRB-5. The compositions shown in Figs A-3 and -4 were used to make the fuel surface and center temperature calculations presented earlier in this report. Without accurate measurements of the fuel centerline temperatures, it is not possible to determine the precision of the calculated temperatures. However, since each fuel specimen located adjacent to a graphite midwall thermocouple is treated in the same manner, it is possible to use the agreement between sweep gas compositions calculated for these specimens as a rough indicator of the precision of the calculations. Examination of the plots shown in Figs. A-3 and -4 reveal that the gas compositions are usually within $\pm 2\%$ of each other, and always within $\pm 5\%$. This agreement is considered to be "fair." Much better precision has been achieved in other experiments, where periods of operation with 100% helium were interspersed among periods of operation with mixed sweep gas. The errors associated with uncertainty in the sweep gas composition have been described.¹

Additional information needed for the fuel temperature calculations includes the dimensional changes of the fuel rods and the graphite sleeve during irradiation. The dimensional behavior of the fuel rods was shown in Fig. 17. The pre- and postirradiation graphite sleeve dimensions are shown in Figs. A-5-A-8. As before, these figures show the increase in graphite diameter over the minimum diameter of 0.825 to 0.827 in. (for 0° and 90° orientation respectively). The fuel dimensional change data were fit to a parabola, and the graphite data were fit to a straight line (linear swelling with fast fluence).

¹F. J. Homan, *Thermal Analysis of HTGR Fuel Rods Irradiated in the HFIR Removable Beryllium Facility*, ORNL/TM report (in preparation).

Fig. A-1. Helium-Neon Sweep Gas Composition vs Time in HRB-5.

Fig. A-2. Temperature Readings for Thermocouple 7, HRB-4, During HPFR Cycle 85.

Fig. A-3. Calculated Sweep Gas Composition vs Time for H

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for HKB-4 Specimens. (a) 1B, (b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

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1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

Fig. A-4. Calculated Sweep Gas Composition vs Time for HRB-5 Specimens. (a) 11

ORNL-DWG 75-18049

ORNL-DWG 75-18050

ORNL-DWG 75-18052

ORNL-DWG 75-18053

vs Time for HRB-5 Specimens. (a) 1B, (b) 1C, (c) 2B, (d) 3A, (e) 3B, (f) 4B.

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Fig. A-5. Graphite Sleeve Dimensions for Capsule HRB-4 0° Orientation.

Fig. A-6. Graphite Sleeve Dimensions for Capsule HRB-4 90° Orientation.

Fig. A-7. Graphite Sleeve Dimensions for Capsule HRB-5 0° Orientation.

Fig. A-8. Graphite Sleeve Dimensions for Capsule HRB-5 90° Orientation.