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Irradiation Performance of HTGR Fuel Rods in HFIR Experiments HRB-7 and -8

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HTGR BASE TECHNOLOGY PROGRAM

Fueled Graphite Development (189a 01330)

**IRRADIATION PERFORMANCE OF HTGR FUEL
RODS IN HFIR EXPERIMENTS HRB-7 AND -8**

K. H. Valentine, F. J. Homan, E. L. Long, Jr., T. N. Tiers,
B. H. Montgomery, R. L. Hamner, and R. L. Beatty

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ABSTRACT

The HRB-7 and -8 experiments were designed as a comprehensive test of mixed thorium-uranium oxide fissile particles with Th:U ratios from 0 to 8 for HTGR recycle application. In addition, fissile particles derived from Weak-Acid Resin (WAR) were tested as a potential backup type of fissile particle for HTGR recycle. Several other minor objectives were also accomplished in these experiments, which were conducted at two temperatures (1250 and 1500°C) to determine the influence of operating temperature on the performance parameters studied. The minor objectives were comparison of advanced coating designs where ZrC replaced SiC in the Triso design, testing of fuel coated in laboratory-scale equipment with fuel coated in production-scale coaters, comparison of the performance of ^{233}U -bearing particles with that of ^{235}U -bearing particles, comparison of the performance of Biso coatings with Triso coatings for particles containing the same type of kernel, and testing of multijunction tungsten-rhenium thermocouples. All objectives were accomplished, although the performance of the tungsten-rhenium thermocouples was disappointing. As a result of these experiments the mixed thorium-uranium oxide fissile kernel was replaced by a WAR-derived particle in the reference recycle design. A tentative decision to make this change had been reached before the HRB-7 and -8 capsules were examined, and the results of the examination confirmed the accuracy of the previous decision. Even maximum dilution (Th/U $\approx$ 8) of the mixed thorium-uranium oxide kernel was insufficient to prevent annealing of the kernels at rates that are unacceptable in a large HTGR. Other results showed the performance of ^{233}U -bearing particles to be identical to that of ^{235}U -bearing particles, the performance of fuel coated in production-scale equipment to be at least as good as that of fuel coated in laboratory-scale coaters, the performance of ZrC coatings to be very promising, and Biso coatings to be inferior to Triso coatings relative to fission product retention.

INTRODUCTION

Capsules HRB-7 and -8 were seventh and eighth in the series of irradiation experiments conducted in the removable beryllium (RB) reflector facility of the High Flux Isotope Reactor (HFIR).¹⁻⁵ Most of the coated particles were fabricated at ORNL. Four fuel rods contained particles fabricated by General Atomic Company (GAC), but all the fuel rods were made by ORNL.

During the planning stages of the HRB-7 and -8 experiment, it was apparent that the former reference recycle fissile particle Biso coated $(\text{Th}_{0.9}\text{U}_{0.2})\text{O}_2$ was a marginal performer, due to the thermal migration of the kernel up the temperature gradient through the coating layers. WAR-derived fissile particles showed promise in the HRB-4 and -5 tests. The HRB-7 and -8 capsules were designed to compare the performance of a number of candidate fissile particles, both for fresh and recycle fuel application, under different conditions of operating temperature and exposure. A complete listing of the objectives of HRB-7 and -8 is given below:

To test a theoretical model for amoeba migration. The kernels of primary interest were (1) sol-gel-derived mixed oxide-carbide, because the two-phase system is thought to be more stable than the one-phase oxide system, and (2) sol-gel-derived uranium oxynitride, because thermochemical calculations suggest improved performance under HTGR conditions of fluence and temperature. A sol-gel-derived UO_2 particle was used as a reference for comparison with the other two particles.

To test potential recycle fissile kernels. These included sol-gel-derived UO_2 , WAR-derived UC_2 , and sol-gel-derived, $(\text{U}_{0.2}\text{Th}_{0.8})\text{O}_2$. Comparisons were also made between ^{233}U - and ^{235}U -bearing particles, and between Biso-coated and Triso-coated particles.

To establish the relative stability of mixed oxide sol-gel-derived kernels having Th/U ratios of 0, 1, 2, 4, and 8. The latter was also of interest to GAC for possible use in Initial and Makeup (IM) fuel elements.

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 HRB-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 HRB-3 and ETR Experiment P13N*, ORNL/TM-4526 (October 1974).

⁴F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiments HRB-4 and -5*, ORNL-5115 (June 1976).

⁵F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-6*, ORNL/TM-5011 (December 1975).

To test advanced coating designs from GAC, in which ZrC was substituted for SiC in the reference IM fissile particle design.

To compare the performance of a HAR-derived $\text{UO}_2\text{-UC}_2$ kernel to that of the reference IM fissile kernel at very high temperatures.

To test mixed oxide fissile particles prepared in a production-size coater (5 in.).

To test a multijunction tungsten-rhenium thermocouple for measuring centerline temperatures at different locations in the capsule.

All of the above objectives were accomplished. The test showed that all dense mixed-oxide kernels had unacceptably high rates of amoeba, that ^{233}U and ^{235}U behaved identically, that the uranium oxynitride had unacceptably high amoeba migration, and that the ZrC coatings performed very well.

DESCRIPTION OF CAPSULES AND FUEL SPECIMENS

Capsules

The HRB-7 and -8 capsules were similar in design to earlier HRB capsules, with two major differences. In previous capsules the graphite sleeve was tapered at the ends to provide a higher thermal resistance to compensate for the lower neutron flux (and hence lower heat generation rate for constant fissile loadings) at the ends of the capsules. The same objective was achieved in HRB-7 and -8 with an untapered sleeve by adjusting the heavy metal loadings so as to flatten the axial power density profile, resulting in axially constant fuel operating temperatures. The second major difference was the central thermocouple concept. Previous capsules have used a central thermocouple at one location, usually just above the horizontal midplane position. HRB-7 and -8 employed a multijunction tungsten-rhenium thermocouple, so that central temperatures could be measured at several locations. As usual, Chromel vs Alumel thermocouples were positioned at various axial positions.

The design configuration for HRB-7 and -8 is shown in Fig. 1. This design is a significant improvement over the design used in previous HRB capsules (HRB-1 through HRB-6). Improvements have continued in later HRB capsules, especially in thermometry.

The decalibration of the tungsten-rhenium thermocouples due to transmutation of the rhenium to osmium presented problems in interpreting the temperature data taken from these thermocouples, and efforts are still under way to analytically correct the data.⁶ Inspection of Fig. 1 reveals

⁶M. W. Waddell et al., *HTGR Base Program Progress Report for January 1, 1974, through June 30, 1975*, ORNL-5108, pp. 428-35.

Fig. 1. Capsule Design for HRB-7, -8, -9, and -10.

that all the fuel specimens contained a central hole to accommodate the multijunction centerline thermocouple used in HRB-7 and -8. The locations of the five junctions are not shown in Fig. 1, but they were located in about the same axial positions as the Chromel-Alumel thermocouples adjacent to specimens 3, 6, 10, 14, and 17.

The central hole was used in HRB-9 and -10 also, even though a Johnson Noise Thermometer was employed on those capsules, at only one axial location. In future experiments, where Johnson Noise Thermometers are used, only the top half of the capsule will contain annular fuel specimens.

Some additional details of the HRB-7 and -8 capsule design are shown in Fig. 2. As before, the sleeve was Poco grade AXF-5Q graphite. 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 $3600\text{-cm}^3/\text{hr}$ 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. 2. Cutaway View of HRB-7 and -8 Capsule Configuration.

Fuel

Loading Scheme

The two capsules were essentially identical. Each capsule contained 19 fuel specimens fabricated by using the slug injection process. The heavy metal loadings were varied with axial location in the capsules to achieve equal power production rates and to minimize the axial variation in operating temperatures. The heavy metal loadings are given in Table 1.

Table 1. Heavy Metal Loadings for HRB-7 and HRB-8

Position in Capsule	Contents, g/cm ³					
	HRB-7			HRB-8		
	²³⁵ U	²³⁸ U	²³² Th	²³⁵ U	²³⁸ U	²³² Th
1	0.0185	0.1593	0.1782	0.0186	0.1602	0.1791
2	0.0186	0.1599	0.1787	0.0186	0.1605	0.1793
3	0.0166	0.1161	0.1731	0.0166	0.1159	0.1727
4	0.0167	0.1170	0.1744	0.0167	0.1169	0.1743
5	0.0149 ^a	0.0833	0.1639	0.0149 ^a	0.0834	0.1640
6	0.0150	0.0851	0.1657	0.0151	0.0853	0.1661
7	0.0150	0.0847	0.1649	0.0149	0.0845	0.1646
8	0.0143	0.0735	0.1634	0.0143	0.0734	0.1632
9	0.0143	0.0734	0.1632	0.0142	0.0728	0.1618
10	0.0140	0.0722	0.1604	0.0143	0.0734	0.1630
11	0.0143	0.0732	0.1628	0.0141	0.0727	0.1615
12	0.0144	0.0739	0.1641	0.0144	0.0739	0.1641
13	0.0148	0.0837	0.1631	0.0148	0.0836	0.1630
14	0.0149	0.0840	0.1638	0.0148	0.0839	0.1635
15	0.0148	0.0837	0.1632	0.0148	0.0837	0.1632
16	0.0165	0.1151	0.1715	0.0165	0.1155	0.1722
17	0.0163	0.1142	0.1701	0.0164	0.1144	0.1705
18	0.0183	0.1580	0.1767	0.0182	0.1564	0.1748
19	0.0184	0.1585	0.1772	0.0186	0.1601	0.1789

^a96.11% ²³³U.

Coated Particles

A total of 17 types of fissile particles were tested in the HRB-7 and -8 capsules. These types and their purposes in the experiment are listed in Table 2. Each fuel specimen contained one type of fissile particle and various amounts of fertile ThO₂ particle, inert Strong-Acid-Resin (SAR) particles, and WAR ²³⁸UO₂ + 15% UC₂ driver particles to make up the required loadings. The characterization of all types of coated particles included in the experiment is tabulated in Table 3.

Table 2. Types of Coated Particles and their Positions in HRB-7 and -8 Capsules^a

Position in Capsule	Kernel Type	Coating Type	Purpose in Experiments
1	$^{235}\text{UC}_2 + 1\% \text{ThC}_2$	Triso	GAC reference
2	$(4\text{Th}, ^{235}\text{U})\text{O}_2$ SG ^b	Biso	Test particle prepared in large coater
3	$^{235}\text{UO}_2 + 7\% \text{U}_3\text{N}_4$ SG	Biso	To study amoeba effect
4	$^{235}\text{UC}_2 + 1\% \text{UC}_2$ SG	Biso	To study amoeba effect
5	$^{235}\text{UO}_2$ SG	Triso	Potential candidate for recycle; compare with $^{235}\text{UO}_2$
6	$^{235}\text{UO}_2$ SG	Biso	Reference for amoeba effect; compare Biso with Triso
7	$^{235}\text{UO}_2$ SG	Triso	Reference for amoeba effect; compare Triso with Biso
8	$(2\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Biso	Relative stability; Th:U ratio
9	$(4\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Triso	Recycle candidate
HRB-10	$(8\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Triso	Relative stability; Th:U ratio; interest for first-core loadings
11	$(4\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Biso	Relative stability; Th:U ratio
12	$(\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Biso	Relative stability; Th:U ratio
13	WAR $^{235}\text{UO}_2 + 14\% \text{UC}_2$	Triso	Compare with GAC reference makeup particle
14	WAR $^{235}\text{UO}_2 + 92\% \text{UC}_2$	Triso	Recycle candidate; compare Triso with Biso
15	WAR $^{235}\text{UO}_2 + 14\% \text{UC}_2$	Biso	Compare Biso with Triso
16	WAR $^{235}\text{UO}_2 + 92\% \text{UC}_2$	Biso	Recycle candidate; compare Biso with Triso
17	$^{235}\text{UC}_2$ with ZrC in coating ^c	Triso	Test GAC advanced coating design
18	$^{235}\text{UC}_2 + 1\% \text{ThC}_2$	Triso	GAC reference
19	$(4\text{Th}, ^{235}\text{U})\text{O}_2$ SG	Biso	Test particle prepared in large coater

^aSG = derived by sol-gel process.

WAR = derived from weak-acid resin.

HRB = reactor horizontal midplane

^bSupplied by GAC.

Table 3. Characterization of Coated Particles for HRB-7 and -8

Coated Particle	GAC6151-00-035 ^a	J263	OR2098H	OR2097H	Pu303H
Batch Number	VSM-UC ₂ + 1% ThC ₂	SG (4Th,U)O ₂	SG UO ₂ + 7% U ₂ N ₃	SG UO ₂ + 1% UC ₂	SG UO ₂
Type of Kernel Material	18.86	7.25	23.65	24.91	25.5
Uranium Content, wt %	93.15	93.17	93.17	93.17	
²³⁵ U Enrichment, at. %					98.05
²³³ U Enrichment, at. %	~1.0	29.74			
Thorium Content, wt %	Mean ^b	Mean ^b	Mean ^b	Mean ^b	Mean ^b
Kernel					
Diameter, μ m	196(16)	353.8(5.5)	192.8(4.7)	193.8(5.8)	226.8(11.0)
Density, g/cm ³	10.99	9.80	9.7	10.5	10.08
Buffer					
Thickness, μ m	99(10.6)	101.2(13.8)	70.7(9.6)	67.6(8.2)	84.7(10.9)
Density, g/cm ³	1.07	1.30 ^c	1.2	1.19	0.90
Inner Carbon Coating					
Thickness, μ m	33(3.5)				35.1(5.7)
Density, g/cm ³	1.92				2.0(ND)
SiC					
Thickness, μ m	32(5.3)				27.1(1.5)
Density, g/cm ³	3.20				3.19(0.001)
Outer Carbon Coating					
Thickness, μ m	38(5.0)	91.8(9.6)	75.6(7.2)	78.4(4.9)	40.7(2.9)
Density, g/cm ³	1.85	2.01(0.007) ^c	2.006(0.004)	1.998(0.004)	2.02(0.005)
Deposition Rate (μ m/min)		7.6			
Particle Crushing Strength (lb)		6.2(1.2)			
Boron Content, ppm	ND ^d	12	5	20	1
Iron Content, ppm	ND	40	10	3	30
Fraction of Actinide Exposures ^e		8×10^{-5}			

∞

Table 3 (Continued)

Coated Particle	OR2089H	OR2114H	OR2094H	OR2116H	GAC6155-01-020 ^a
Batch Number	SG UO ₂	SG UO ₂	SG (2Th,U)O ₂	SG (4Th,U)O ₂	SG (8Th,U)O ₂
Type Kernel Material	22.41	16.99	10.44	6.50	5.23
Uranium Content, wt %	93.17	93.17	93.17	93.13	93.15
²³⁵ U Enrichment, at. %					
²³³ U Enrichment, at. %			20.79	26.42	43.59
Thorium Content, wt %	Mean ^c	Mean ^b	Mean ^b	Mean ^b	Mean ^b
Kernel					
Diameter, μm	195.6(5.9)	195.9(5.2)	282.0(13.2)	360.0(4.7)	502(23.1)
Density, g/cm ³	9.97	9.83	10.17	9.8	9.77
Buffer					
Thickness, μm	74.8(8.6)	85.0(9.8)	84.5(7.9)	97.7(8.7)	106(17.6)
Density, g/cm ³	1.25	1.28	1.26	1.36	1.24
Inner Carbon Coating					
Thickness, μm		37.0(3.6)		38.8(2.9)	32.0(4.3)
Density, g/cm ³		2.0(ND)		2.0(ND)	1.93(ND)
SiC					
Thickness, μm		27.3(1.8)		33.6(1.9)	34.0(6.7)
Density, g/cm ³		3.17(0.009)		3.21(0.003)	3.22(ND)
Outer Carbon Coating					
Thickness, μm	77.1(4.7)	46.4(2.3)	85.0(6.2)	42.9(3.0)	42.0(6.1)
Density, g/cm ³	2.01(0.003)	1.98(0.001)	2.01(0.003)	2.04(0.002)	1.81(ND)
Boron content, ppm	5	20	20	3	ND
Iron Content, ppm	7	20	20	20	ND

Table 3 (Continued)

Coated Particle	OR2120H	GAC6171-63E ^a	J-262 ^h	OR2104H ⁱ	OR2079H
Batch Number	WAR UC ₂	USH UC ₂	SG ThO ₂	WAR UO ₂ + 14% UC ₂	SAR Carbon
Type Kernel Material					
Uranium Content, wt %	23.66	20.70		29.11	
²³⁵ U Enrichment, at. %	93.32	93.15		0.70	
²³³ U Enrichment, at. %					
Thorium Content, wt %			54.05		
	Mean ^b	Mean ^b	Mean ^b	Mean ^b	Mean ^b
Kernel					
Diameter, μm	315.3(29.1)	197.0(24)	496.5(3.2)	426.2(31.4)	471.5(24.6)
Density, g/cm ³	5.28	ND	10.0	3.34	1.37
Buffer					
Thickness, μm	74.6(11.3)	102(9.5)	84.0(13.4)	42.3(7.7)	77.3(ND)
Density, g/cm ³	1.10	1.00	1.07 ^j	1.11	
Inner Carbon Coating					
Thickness, μm		8.4(1.2)			
Density, g/cm ³		1.80(ND)			
SiC					
Thickness, μm		21.0(0.0) ^k			
Density, g/cm ³		6.5(ND)			
Outer Carbon Coating					
Thickness, μm	88.6(6.6)	65.0(3.8)	86.1(10.9)	83.1(9.0)	95.3(6.6)
Density, g/cm ³	1.99(0.003)	1.73(ND)	2.01(0.006) ^l	2.02(0.006)	1.96(0.013)
Deposition Rate ($\mu\text{m}/\text{min}$)			8.3		
Crushing Strength (lb)			6.5(0.8)		
Boron Content, ppm	2	ND	ND	20	3
Iron Content, ppm	10	ND	ND	3	10
Fraction Actinide Exposed ^g			8×10^{-6}		

Table 3 (Continued)

Coated Particle	OR2111H	OR2090H	OR2115H	OR2121H	OR2106H
Batch Number	SG (4Th,U)O ₂	SG (Th,U)O ₂	WAR UO ₂ + 14% UC ₂	WAR UC ₂	WAR UO ₂ + 14% UC ₂
Type Kernel Material	7.24	13.5	19.13	20.03	21.75
Uranium Content, wt %	93.17	93.17	93.18	93.32	93.18
²³⁵ U Enrichment, at. %					
²³³ U Enrichment, at. %					
Thorium Content, wt %	29.98	13.36			
	Mean ^b	Mean ^b	Mean ^b	Mean ^b	Mean ^b
Kernel					
Diameter, μ m	360.0(4.7)	245.5(19.0)	385.8(29.0)	315.5(29.1)	385.8(29)
Density, g/cm ³	9.8	6.58	3.18	5.28	3.18
Buffer					
Thickness, μ m	95.1(7.2)	78.5(9.0)	40.9(6.8)	74.6(11.3)	40.9(6.8)
Density, g/cm ³	1.36	1.28	1.37	1.10	1.37
Inner Carbon Coating					
Thickness, μ m			42.9(3.6)	36.1(3.8)	
Density, g/cm ³			2.0(ND)	2.0(ND)	
SiC					
Thickness, μ m			32.1(1.9)	28.4(2.7)	
Density, g/cm ³			3.19(0.003)	3.19(0.006)	
Outer Carbon Coating					
Thickness, μ m	94.6(5.5)	84.4(7.0)	48.8(4.0)	49.2(3.3)	99.0(7.1)
Density, g/cm ³	2.04(0.002)	2.01(0.004)	1.99(0.005)	1.98(0.004)	2.00(0.005)
Boron Content, ppm	5	20	5	5	20
Iron Content, ppm	3	80	20	20	3

^aSupplied by General Atomic Company; VSM is GAC's proprietary process; SG means derived by the sol-gel process.

^bNumbers in parentheses are standard deviations.

^cAfter application of the LTI, the buffer density was 1.39.

^dGradient column density.

^eCorrected value; that is, the bulk density was 1.93.

^fND means not determined.

^gMeasured by aqueous leach.

^hFertile particles included in all rods.

ⁱU-235 containing driver particles included in all rods.

^jAfter application of the sealer coat, buffer density was 1.19 g/cc.

^kZrC substituted for SiC.

^lCorrected value; that is, the bulk density was 1.92.

Kernels

The primary fissile kernel variables investigated in the ORNL fuels were thorium-uranium ratios in sol-gel oxides and oxygen potential in both sol-gel and WAR-derived materials. The fissile isotope in all particles for this experiment was ^{235}U except for one batch of $^{233}\text{UO}_2$ for direct comparison with a similar batch of $^{235}\text{UO}_2$. Since the oxygen-uranium ratio of as-received sol-gel UO_2 and $(\text{Th},\text{U})\text{O}_2$ kernels varied over a wide range and since analyses⁷ had shown that oxygen potential was likely to be a critical factor influencing the stability of irradiated coated-oxide particles, a standardizing treatment was employed with each batch of sol-gel oxide. This treatment consisted of applying the buffer coating in the usual way to provide a large excess of carbon and then heating at 1500°C for 30 min in a CO atmosphere before applying the seal coating. It was determined that this treatment should fix the oxygen-uranium ratio at 2.000 ± 0.0002 , stoichiometric beyond the resolution of available analytical techniques.⁸ This standardizing treatment was considered necessary to eliminate initial oxygen potential as a variable in those samples in which comparisons were made between different thorium-uranium ratios or between $^{235}\text{UO}_2$ and $^{233}\text{UO}_2$.

Two batches of sol-gel $^{235}\text{UO}_2$ were altered especially to reduce their anticipated increase in oxygen potential. One batch was prepared by taking as-dried microspheres containing residual carbon from the organic compounds used in sphere forming and heating these in a tungsten crucible at 1400°C for 22 hr. The atmosphere was initially argon, but was changed to a 2.5 to 5.0×10^{-2} torr vacuum during the final 21 hr. Chemical analysis of this product showed 0.08 wt % combined carbon, which corresponds to a nominal composition of 98.23 mole % UO_2 -1.77 mole % UC. The other batch was prepared by mixing as-dried sol-gel UO_2 with powdered carbon and heating for 21 hr at 1500°C in an atmosphere of 600 torr N_2 , 52 torr CO , 108 torr (Ar and 4% H_2). Chemical analysis of this product showed a nominal composition of $\text{UO}_{1.7}\text{N}_{0.1}$; this is a solid solution of 93 mole % UO_2 -7 mole % $\text{UN}_{1.5}$. However, the preparation conditions for this material indicate that the composition should be $\text{UO}_{1.68}\text{N}_{0.24}$ (ref. 9).

Three batches of uranium-loaded amberlite IRC-72 WAR particles were fabricated for these experiments. Two were candidate fissile particles, and the third was a normal U-loaded fertile particle to be included in

⁷T. B. Lindemer and H. J. de Nordwall, *An Analysis of Chemical Failure of Coated UO_2 and Other Oxide Fuels in the High-Temperature Gas-Cooled Reactor*, ORNL-4926 (January 1974).

⁸Ref. 7, Figs. 1 and 2.

⁹J. M. Leitnaker and T. B. Lindemer et al., *Oxynitride Fuel Kernel for Gas-Cooled Reactor Fuel Particles* (U.S. Atomic Energy Commission). U.S. Patent 3,878,041. Apr. 15, 1975.

all specimens for maintaining the desired test temperature while the ^{235}U burned out and before sufficient ^{233}U was generated in the fertile ThO_2 particles. One of the fissile WAR types was partially reduced; approximately 15% of the UO_2 was converted to UC_2 to produce an O/U of about 1.7. This was considered to be the minimum reduction level necessary to avoid buildup of CO in the irradiated fuel.¹⁰ Processing of these kernels included carbonizing at 5.5°C/min and then heating at 1600°C for 19 min in an argon-fluidized bed to achieve the desired reduction. Kernel density was 3.17 g/cm³. Kernel analysis showed 74.60% U, 8.72% O, and 16.72% C. The second fissile WAR type was fully reduced to produce UC_2 for comparison with dense UC_2 . This batch was carbonized at 6°C/min and then heated at 1750°C for 45 min to effect full reduction. Final kernel analysis, however, showed 87.11% U, 11.62% C, and 0.93% O, indicating that the reduction had not been completed as predicted. This was attributed to the densification which occurred so that the final stage of reduction was diffusion limited; final kernel density was 5.28 g/cm³. Apparently, reduction rate is constant for a given temperature, depending on the specific gas sweep rate, only as long as the material does not sinter significantly. The importance of using slower carbonization rates near 2°C/min to avoid densification on subsequent reduction was learned later.

The fertile WAR kernels were carbonized at 3.7°C/min and then were heated at 1600°C for 20 min to effect approximately 15% reduction. Final kernel analysis showed 74.50% U, 17.09% C, and 8.55% O for an O/U of about 1.7. Kernel density was 3.35 g/cm³.

Coatings.

All particle coatings for this experiment were deposited under similar, established conditions, except for the use of MAPP gas for certain LTI coatings, since coating structure was not an experimental variable. Details of all coating conditions are listed in Table 4. All fissile particle LTI coatings were deposited from undiluted propylene; MAPP gas was used only for the inert and normal-U fertile particles. Process steps not evident in Table 4 included the post-buffer 30-min, 1500°C treatment in CO for sol-gel oxides mentioned earlier, a surface impregnation of outer LTI coatings to reduce coating-matrix interactions, and a 10-min, 1800°C heat treatment after final coating. Because of different batch sizes, Coaters ranging in size from 1 to 2.5 in. were used for the OR-batches. The Pu-batch, containing ^{233}U , was coated in a 1.75-in. furnace contained in a plutonium-handling box. The J-batches were coated in a 5-in. coater.

¹⁰F. J. Homan, et al., "Stoichiometric Effects on Performance of HTGR Fuels from the U-C-O System" (Accepted for publication in *Nuclear Technology*).

Table 4. Conditions for Coating Particles for HRB-7 and -8^a

Batch Number	Buffer ^b			Inner LTI			SIC ^c			Outer LTI		
	Temperature (°C)	Deposition Rate (μm/min)	Coating Gas	Temperature (°C)	Deposition Rate (μm/min)	Coating Gas	Deposition Rate (μm/min)	Coating Gas	Temperature (°C)	Deposition Rate (μm/min)	Coating Gas	
OR-2079-N	1300	5.52						MAPP ^d	1275	6.81		
OR-2089-N	1100	13.60						C ₃ H ₆	1275	7.71		
OR-2090-N	1100	14.27						C ₃ H ₆	1275	8.44		
OR-2094-N	1100	16.90						C ₃ H ₆	1275	11.13		
OR-2097-N	1100	16.90						C ₃ H ₆	1275	8.71		
OR-2098-N	1100	20.20						C ₃ H ₆	1275	9.45		
OR-2104-N	1300	10.57						MAPP ^d	1275	9.23		
OR-2106-N	1300	6.82						C ₃ H ₆	1275	7.62		
OR-2111-N	1100	13.59						C ₃ H ₆	1250	9.46		
OR-2114-N	1100	15.45		1275	9.25	C ₃ H ₆	0.33	C ₃ H ₆	1250	6.63		
OR-2115-N	1300	6.82		1275	7.80	C ₃ H ₆	0.34	C ₃ H ₆	1275	6.97		
OR-2116-N	1700	13.96		1250	11.94	C ₃ H ₆	0.37	C ₃ H ₆	1250	10.75		
OR-2120-N	1100	6.78						C ₃ H ₆	1275	5.37		
OR-2121-N	1100	6.78		1275	6.02	C ₃ H ₆	0.32	C ₃ H ₆	1275	4.92		
PR-303-N	1075	15.40		1300	14.04	C ₃ H ₆	0.32	C ₃ H ₆	1300	10.18		
J-262	1450	24.71						C ₃ H ₆	1350	8.52		
J-263	1160	13.66						C ₃ H ₆	1350	4.85		

^aAll coating temperatures are as read with an optical pyrometer on outside wall of coating tube.^bCoating gas - C₃H₆^cCoating gas - CH₃SiCl₃ + N₂ at 1550°C^dMAPP gas - Mixture of methylacetylene-propadiene, propane, butane, iso-butane, propylene, butadiene.

Preparation of Fuel Specimens

The coated particles were fabricated into fuel rods, nominally 0.49 in. in diameter and 0.8 in. long, with a 0.132 in. central hole for placement of a multijunction tungsten-rhenium thermocouple to measure fuel centerline temperatures. An orientation-identification mark was formed on one end of each specimen so diameter measurements could be made at the same location both before and after irradiation.

All coated particles, except those supplied by GAC, were heat treated at 1800°C before incorporation into the fuel rods. Coatings generally shrink during heat treatment of the fuel rods unless given the 1800°C anneal. The matrix used for bonding the coated particles was composed of 28.5 wt % Ashbury Mills Graphite flour (grade NF6353) in Ashland Oil Company grade 270 petroleum pitch. The matrix was injected through the particle bed at 175°C and 900 psi pressure. A metal mold was used, and a metal rod was used to form the thermocouple hole. The mold, mandrel, and punches used to form the fuel rods are shown in Fig. 3. Note that the bottom punch has a small ridge at the surface contacting the specimen to make an indentation, which is the orientation-identification mark mentioned earlier. If a flat-surface punch is used, the specimen can be easily separated from the mandrel and punches by twisting, but with the ridged punch, it was necessary to design a device that pushes the green rod and mandrel away from the bottom punch so that the specimen can be twisted off without damage.

Fig. 3. Hardware Used for Preparation of Slug-Injected Specimens for HRB-7 and -8.

After forming, all specimens except those containing ^{233}U were carbonized horizontally in a bed of graphite powder by heating in argon to 1000°C at a rate of 40°C/hr . Because of glove-box considerations (carbon contamination), the ^{233}U -bearing specimens were carbonized vertically in alumina powder at a heating rate as follows: 300°C/hr to 300°C , 60°C/hr to 650°C , and then 300°C/hr to 800°C . All were heat treated at 1800°C for 30 min to dimensionally stabilize the matrix. A total of 152 specimens were prepared for the experiment, archives, and characterization.

The specimens were characterized by dimensional measurements, weights, radiography, metallography, and calculated fuel and particle volume loadings and matrix densities. All specimens were not characterized for pitch coke yield. However, the average coke yield for 23 ^{235}U -bearing specimens was 26.1% (± 1.2). The yield for the ^{233}U -type specimens was not determined. End and side views, respectively, of typical specimens are shown in Figs. 4 and 5. A fuel rod characterization summary for each specimen for HRB-7 and -8 is given in Table 5. Exposed actinide in the specimens was not determined, because a satisfactory technique had not been developed.

Fig. 4. End View of Typical Specimens for HRB-7 and -8.

Fig. 5. Side View of Typical Specimens for HRB-7 and -8.

Table 5. Characterization of Specimens for Capsules HRB-7 and -8

Position in Capsule	Average Dimensions (in.) ^a		Volume (cm ³)	Weight (g)	Density (g/cm ³)		Particle Volume Loading (%)
	Outside Diameter	Length			Matrix	Total	
1	0.4889	0.7802	2.2272	3.4449	0.61	1.55	60.2
2	0.4895	0.7758	2.2205	3.4511	0.63	1.55	60.5
3	0.4896	0.7695	2.2034	3.3046	0.61	1.50	60.9
4	0.4895	0.7635	2.1859	3.2804	0.61	1.50	60.8
5	0.4921	0.7753	2.2445	3.1480	0.50	1.40	59.3
6	0.4897	0.7748	2.2193	3.2226	0.61	1.45	60.0
7	0.4891	0.7807	2.2303	3.2605	0.60	1.46	60.5
8	0.4893	0.7723	2.2083	3.1989	0.61	1.45	60.3
9	0.4891	0.7741	2.2115	3.2365	0.62	1.46	59.7
10	0.4900	0.7844	2.2498	3.2177	0.61	1.43	59.3
11	0.4890	0.7662	2.2165	3.1864	0.58	1.44	58.0
12	0.4899	0.7669	2.1986	3.1838	0.62	1.45	60.3
13	0.4895	0.7878	2.2546	3.2705	0.61	1.45	60.0
14	0.4893	0.7852	2.2452	3.2772	0.62	1.46	60.3
15	0.4891	0.7888	2.2535	3.2562	0.61	1.44	60.0
16	0.4891	0.7782	2.2232	3.3105	0.61	1.49	60.4
17	0.4896	0.7827	2.2410	3.3656	0.61	1.49	60.0
18	0.4888	0.7872	2.2459	3.4843	0.62	1.55	60.8
19	0.4897	0.7819	2.2397	3.4687	0.63	1.55	60.0
HRB-8							
1	0.4893	0.7748	2.2157	3.4450	0.63	1.55	60.5
2	0.4897	0.7723	2.2124	3.4535	0.64	1.56	60.6
3	0.4891	0.7729	2.2083	3.2955	0.60	1.49	60.6
4	0.4890	0.7660	2.1876	3.2772	0.61	1.50	60.7
5	0.4914	0.7770	2.2426	3.1420	0.50	1.40	59.3
6	0.4892	0.7745	2.2136	3.2292	0.62	1.46	60.2
7	0.4890	0.7826	2.2348	3.2678	0.60	1.46	60.3
8	0.4893	0.7734	2.2114	3.1950	0.61	1.44	60.2
9	0.4897	0.7785	2.2300	3.2433	0.62	1.45	59.2
10	0.4883	0.7776	2.2136	3.2185	0.63	1.45	60.2
11	0.4898	0.7800	2.2352	3.1874	0.57	1.43	57.5
12	0.4892	0.7693	2.1987	3.1812	0.62	1.45	60.3
13	0.4897	0.7878	2.2566	3.2783	0.62	1.45	59.7
14	0.4893	0.7868	2.2497	3.2764	0.61	1.46	60.2
15	0.4895	0.7864	2.2534	3.2518	0.61	1.44	60.0
16	0.4888	0.7761	2.2143	3.3139	0.62	1.50	60.7
17	0.4873	0.7820	2.2360	3.3469	0.62	1.55	60.0
18	0.4896	0.7928	2.2699	3.4846	0.60	1.56	60.2
19	0.4892	0.7762	2.2184	3.4650	0.64	1.56	60.5

^a Inside diameter = 0.132 in.

DESCRIPTION OF IRRADIATION

Irradiation Conditions

The design changes incorporated into the HRB-7 and -8 capsules have been described. There were also some changes made in the manner of operating the capsules. Previous capsules received all their irradiation in RB positions in HFIR. The locations of these facilities within HFIR, the neutron flux, and gamma heating characteristics have been described previously.¹¹ Previous capsules contained either a modest enrichment (about 7% ^{235}U) of the fissile isotope or a fully enriched (about 93% ^{235}U) fissile isotope contained in a mixed-oxide ($\text{Th}_x\text{U}_{1-x}\text{O}_2$) kernel. Because of the very high (relative to the HTGR spectrum) thermal flux characteristic of the RB facilities, it was not possible to irradiate a pure fissile particle containing 93% enriched fuel. The power production rates would be so high at the beginning of the irradiation that the fissile particles would likely fail. Beginning with HRB-7 and -8 this problem was solved by employing a "preirradiation" in a HFIR position (VXF-13) further from the target region. The location of this position is shown in Fig. 6. The irradiation conditions present in the permanent beryllium (PB) facilities (VXF-13) are not as well characterized as those in the RB facilities, but it is estimated that the thermal flux in a PB position is about half of that in an RB position.

¹¹F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-3 and ETR Experiment P13N*, ORNL/TM-4526 (October 1974).

Fig. 6. Location of Irradiation Facilities in the HFIR Core.

Capsule Operation

HRB-7 and -8 were both irradiated for two HFIR cycles in the VXF-13 facility where 46 to 60% of the ^{235}U was fissioned (at the ends and horizontal midplane positions respectively). The capsules were then moved to RB positions for the remainder of their exposure. With most of the ^{235}U burned out, the watt/particle values in the RB positions were acceptable. HRB-7 was irradiated for six HFIR cycles in the RB-7 facility at a design fuel centerline temperature of 1500°C . HRB-8 was irradiated for eight cycles in the RB-5 facility at a design centerline temperature of 1250°C . Detailed operating histories are given in Table 6.

The operation of both capsules was stable throughout irradiation. There was an initial drop in the power production rates for all fuel specimens, as expected, due to the burnout of the ^{235}U . The calculated fission heat rate profiles for those specimens adjacent to Chromel vs Alumel thermocouples are shown in Fig. 7 (a-1). The blank space between cycles 2 and 3 (about 24 days) represents reactor downtime due to maintenance problems. As shown in Fig. 7, the heat generation rates at the beginning of the irradiation were quite high in some specimens, even in the lower neutron fluxes of the VSF-13 facility. Because of these high power-production rates, fissile particles began failing immediately, and the level of activity in the capsule sweep gas became so high that it was not possible to sample the sweep gas for release-to-birth rate (R/B) determinations. The reactor off-gas system could not handle the high level of activity in the sweep gas, so the capsules had to be operated in stagnant gas during most of the irradiation. The capsules were periodically swept by clearing the control room of all personnel while the gas in the capsules was replaced.

The high gas release from the particles in this experiment should not be interpreted as unsatisfactory fuel performance. As shown in the section on metallography there were some failures, particularly in the UC_2 fissile fuels, due to fission product interaction with the SiC layer. However, those failures are probably not sufficient to explain the high gas release throughout the operation of both HRB-7 and -8. As mentioned earlier, this was the first HRB experiment in which fully enriched uranium was irradiated in undiluted fissile particles. Because this process produces very high power production rates at the beginning of the experiment, the preirradiation in the VXF-13 facility at lower neutron flux was instituted. However, examination of fission heat rate vs time plots in Fig. 7 reveals very high power-production rates at the beginning of the first cycle (first VXF-13 cycle) and again at the beginning of cycle 3 (first RB cycle), sometimes reaching as high as 6.5 kW/ft (213 W/cm). These high heat rates produced power production rates of nearly a factor of 10 higher than those envisioned for the LHTGR. Using the following data from Tables 1 and 3:

kernel diameter = $386 \mu\text{m} = 386 \times 10^{-4} \text{ cm}$,
 ^{235}U loading = 0.0148 g/cm^3 ,
 fuel rod diameter = $0.49 \text{ in.} = 1.245 \text{ cm}$,
 kernel density = 3.18 g/cm^3 ,
 uranium in kernels = $74.6 \text{ wt } \%$,
 ^{235}U enrichment = 93.2% ,

Table 6. Capsule HRB-7 and -8 Reactor Power History

Facility	Cycle	Begin		End		Irradiation time (hr)	
		Time	Date	Time	Date	During Cycle	Accumulated
<u>HRB-7^a</u>							
PB-13	101	1540	12/18/73	2400	1/10/74	559.44	559.44
PB-13	102	1237	1/11/74	2400	2/3/74	562.56	1122.00
RB-7	103	1440	2/28/74	1915	3/23/74	556.80	1678.80
RB-7	104	0907	3/24/74	1006	4/16/74	552.72	2231.52
RB-7	105	1750	4/17/74	1135	5/9/74	521.28	2752.80
RB-7	106	0120	5/10/74	2200	6/1/74	547.92	3300.72
RB-7	107	2326	6/4/74	0100	6/28/74	552.72	3853.44
RB-7	108	1406	6/28/74	1340	7/21/74	551.52	4404.96
<u>HRB-8</u>							
PB-13	103	1440	2/28/74	1915	3/23/74	556.80	556.80
PB-13	104	0907	3/24/74	1006	4/16/74	552.72	1109.52
RB-5	105	1750	4/17/74	1135	5/9/74	521.28	1630.80
RB-5	106	0120	5/10/74	2200	6/1/74	547.92	2178.72
RB-5	107	2326	6/4/74	0100	6/28/74	552.72	2731.44
RB-5	108	1406	6/28/74	1340	7/21/74	551.52	3282.96
RB-5	109	1842	7/22/74	0400	8/13/74	546.88	3829.84
RB-5	110	1707	8/15/74	2012	9/9/74	549.60	4379.44
RB-5	111	1030	9/12/74	0736	10/5/74	548.88	4928.32
RB-5	112	0525	10/6/74	0.218	10/31/74	547.20	5475.52

^aReactor down 24.61 days between cycles 102 and 103.

Fig. 7. Fission Power Density vs Irradiation Time for a given specimen: (a) rod

ORNL-ENG 75-14615

ORNL-ENG 75-14618

rod 1 in HRB-7, (b) rod 3 in HRB-7, (c) rod 6 in HRB-7, (d) rod 10 in HRB-7, (e) rod 14 in HRB-7, (f) rod 17

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ORNL-DWG 75-14615

ORNL-DWG 75-14616

ORNL-DWG 75-14618

ORNL-DWG 75-14619

rod 6 in HRB-7, (d) rod 10 in HRB-7, (e) rod 14 in HRB-7, (f) rod 17 in HRB-7.

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Fig. 7. (Continued) Fission Power Density vs Irradiation Time for: (g) rod 1 in HRB-8

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ORNL-86-75-14621

ORNL-86-75-14629

in HRB-8, (h) rod 3 in HRB-8, (i) rod 6 in HRB-8, (j) rod 10 in HRB-8, (k) rod 14 in HRB-8, (l) rod 17 in HRB-8

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6 in HRB-8, (j) rod 10 in HRB-8, (k) rod 14 in HRB-8, (l) rod 17 in HRB-8.

the power production rate for rod 13Z HRB-7 was calculated:

$$\begin{aligned} & \left[\frac{\pi}{6} (0.0386 \text{ cm})^3 \text{ of kernel material} \right] (0.746 \text{ g of uranium per gram of kernel material}) \\ & \times (0.932 \text{ g of } ^{235}\text{U per gram of uranium}) (3.18 \text{ g/cm}^3) \\ & = 6.65 \times 10^{-5} \text{ g of } ^{235}\text{U per particle;} \end{aligned}$$

$$\begin{aligned} & (0.0148 \text{ g of } ^{235}\text{U per cm}^3 \text{ of rod}) \left[\frac{\pi}{6} (1.245)^2 \text{ cm}^3 \text{ per cm of rod} \right] \\ & = 0.0180 \text{ g of } ^{235}\text{U per cm of rod;} \end{aligned}$$

$$\begin{aligned} & \frac{(6.5 \text{ kW/ft})(1000 \text{ W/kW})}{(12 \text{ in./ft})(2.54 \text{ cm/in.})} = 213.3 \text{ W per cm of rod;} \end{aligned}$$

$$\begin{aligned} & \frac{(213.3 \text{ W per cm of rod})(6.65 \times 10^{-5} \text{ g of } ^{235}\text{U per particle})}{(0.0180 \text{ g of } ^{235}\text{U per cm of rod})} = 0.788 \text{ W per particle} \end{aligned}$$

This power production rate of about 0.8 W/particle early in life is compared with the maximum rate of about 0.1 W/particle calculated for a LHTR. This overpowering of the particles is thought to be a contributing factor to the high gas release rate.

The estimated neutron fluxes and fluences for the two capsules are given in Tables 7 and 8, and the calculated burnups, in both the PB and RB facilities are given in Tables 9 and 10.

Operating Temperatures

The operating temperatures for HRB-7 and -8 were calculated using the same general procedures used for other HRB capsules.¹²⁻¹⁷ HRB-7 and -8 had some unique features which required that more sophisticated calculational procedures be developed. Previous HRB capsules were rotated

¹²J. L. Scott et al., *An Irradiation Test of Bonded HTGR Coated Particle Fuels in an Instrumented Capsule in HFIR*, ORNL/TM-3460 (March 1972)

¹³J. H. Coobs et al., *Irradiation Performance in HFIR Experiment HRB-2 of HTGR Fuel Sticks Bonded with Reference and Advanced Matrix Materials*, ORNL/TM-3988 (January 1973).

¹⁴F. J. Noman et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-3 and ETR Experiment P13N*, ORNL/TM-4526 (October 1974)

¹⁵F. J. Noman et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiments HRB-4, -5, ORNL-5115* (June 1976).

¹⁶F. J. Noman et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-6*, ORNL-5011 (December 1975).

¹⁷F. J. Noman, *Thermal Analysis of HTGR Fuel Rods Irradiated in the HFIR Removable Beryllium Facility* (in preparation).

Table 7. Capsule HRB-7 Specimen-Calculated Thermal and Fast Fluence

Specimen Number	Reactor Wg ^a to specimen center (in.)	Flux in PB Facility, neutrons cm ⁻² sec ⁻¹		Flux in RB-7 Facility, neutrons cm ⁻² sec ⁻¹		Total Fluence, neutrons/cm ²	
		Average Thermal, E < 0.414 eV	Fast, E > 0.18 MeV	Average Thermal, E < 0.414 eV	Fast, E > 0.18 MeV	Thermal E < 0.414 eV	Fast, E > 0.18 MeV
1	7.2	4.44 × 10 ¹⁵	3.42 × 10 ¹¹	6.18 × 10 ¹⁵	2.95 × 10 ¹⁵	0.91 × 10 ²²	3.62 × 10 ²¹
2	6.4	4.86	3.80	7.07	3.31	1.03	4.07
3	5.6	5.22	4.31	7.95	3.66	1.15	4.50
4	4.8	5.56	4.65	8.80	3.97	1.26	4.88
5	4.0	5.89	4.98	9.57	4.28	1.37	5.26
6	3.2	6.23	5.17	10.27	4.55	1.47	5.59
7	2.4	6.57	5.40	10.90	4.78	1.55	5.87
8	1.6	6.85	5.58	11.40	4.92	1.62	6.04
9	0.8	6.98	5.67	11.71	5.00	1.67	6.14
10	0	7.03	5.70	11.80	5.00	1.68	6.14
11	0.8	6.98	5.67	11.71	5.00	1.67	6.14
12	1.6	6.85	5.58	11.40	4.92	1.62	6.04
13	2.4	6.57	5.40	10.90	4.78	1.55	5.87
14	3.2	6.23	5.17	10.27	4.55	1.47	5.59
15	4.0	5.89	4.98	9.57	4.28	1.37	5.26
16	4.8	5.56	4.65	8.80	3.97	1.26	4.88
17	5.6	5.22	4.31	7.95	3.66	1.15	4.50
18	6.4	4.86	3.80	7.07	3.31	1.03	4.07
19	7.2	4.44	3.42	6.18	2.95	0.91	3.62

^aWg = horizontal midplane.

Table 8. Capsule HRB-8 Specimen-Calculated Thermal and Fast Fluence

Specimen Number	Reactor 10W ^a to specimen center (in.)	Flux in PB Facility, neutrons cm ⁻² sec ⁻¹		Flux in RB-5 Facility, neutrons cm ⁻² sec ⁻¹		Total Fluence, neutrons/cm ²	
		Average Thermal, E < 0.414 eV	Fast, E > 0.18 MeV	Average Thermal, E < 0.414 eV	Fast, E > 0.18 MeV	Thermal E < 0.414 eV	Fast, E > 0.18 MeV
1	7.2	4.44 × 10 ¹⁴	3.42 × 10 ¹¹	4.18 × 10 ¹⁴	2.95 × 10 ¹⁴	1.15 × 10 ¹⁷	4.78 × 10 ²¹
2	6.4	4.86	3.80	7.07	3.31	1.30	5.35
3	5.6	5.22	4.31	7.95	3.66	1.46	5.92
4	4.8	5.56	4.65	8.80	3.97	1.60	6.42
5	4.0	5.89	4.90	9.57	4.28	1.73	6.92
6	3.2	6.23	5.17	10.27	4.55	1.86	7.36
7	2.4	6.57	5.40	10.90	4.78	1.97	7.73
8	1.6	6.85	5.58	11.40	4.92	2.07	7.95
9	0.8	6.98	5.67	11.71	5.00	2.12	8.09
10	0	7.03	5.70	11.80	5.00	2.14	8.09
11	0.8	6.98	5.67	11.71	5.00	2.12	8.09
12	1.6	6.85	5.58	11.40	4.92	2.07	7.95
13	2.4	6.57	5.40	10.90	4.78	1.97	7.73
14	3.2	6.23	5.17	10.27	4.55	1.86	7.36
15	4.0	5.89	4.90	9.57	4.28	1.73	6.92
16	4.8	5.56	4.65	8.80	3.97	1.60	6.42
17	5.6	5.22	4.31	7.95	3.66	1.46	5.92
18	6.4	4.86	3.80	7.07	3.31	1.30	5.35
19	7.2	4.44	3.42	6.18	2.95	1.15	4.78

^a10W = horizontal midplane.

Table 9. Capaule HRB-7 Specimen-Calculated Burnup

Specimen Number	Distance from NRP ^a to Specimen Center (cm)	Calculated Burnup, FIWA									
		235U			238U			239Pu			232Th
		PB-13	RB-7	Total	PB-13	RB-7	Total	PB-13	RB-7	Total	Total
1	18.3	46.19	37.34	83.53	0.42	10.21	10.63	0.11	3.94	4.05	4.05
2	16.3	48.97	54.95	83.92	0.50	11.69	12.19	0.12	4.57	4.69	4.69
3	14.2	51.56	32.64	84.20	0.58	13.25	13.83	0.15	5.27	5.42	5.42
4	12.2	54.02	30.66	84.68	0.65	14.45	15.10	0.16	5.93	6.09	6.09
5	10.2	55.68	28.90	84.58	0.69	15.62	16.31	0.18	6.47	6.65	6.65
6	8.1	57.14	27.40	84.54	0.77	16.57	17.34	0.20	6.99	7.15	7.15
7	6.1	58.28	26.28	84.57	0.82	17.41	18.23	0.21	7.37	7.58	7.58
8	4.1	59.14	25.57	84.71	0.86	18.04	18.90	0.22	7.71	7.93	7.93
9	2.0	59.60	25.09	84.69	0.89	18.38	19.27	0.23	7.85	8.08	8.08
10	0	59.84	24.84	84.68	0.90	18.58	19.48	0.23	8.05	8.28	8.28
11	2.0	59.60	25.09	84.69	0.89	18.38	19.27	0.23	7.85	8.08	8.08
12	4.1	59.14	25.57	84.71	0.86	18.84	18.90	0.22	7.71	7.93	7.93
13	6.1	58.28	26.28	84.57	0.82	17.41	18.23	0.21	7.37	7.58	7.58
14	8.1	57.14	27.40	84.54	0.77	16.57	17.34	0.20	6.99	7.15	7.15
15	10.2	55.68	28.90	84.58	0.69	15.62	16.31	0.18	6.47	6.65	6.65
16	12.2	54.02	30.66	84.68	0.65	14.45	15.10	0.16	5.93	6.09	6.09
17	14.2	51.56	32.64	84.20	0.58	13.25	13.83	0.15	5.27	5.42	5.42
18	16.3	48.97	34.95	83.92	0.50	11.69	12.19	0.12	4.57	4.69	4.69
19	18.3	46.19	37.34	83.53	0.42	10.21	10.63	0.11	3.94	4.05	4.05

^a NRP = horizontal midplane.

Table 10. Capsule HRB-8 Specimen-Calculated Burnup.

Specimen Number	Distance from HMP ^a to Specimen Center (cm)	Calculated Burnup, FIMA									
		²³⁵ U			²³⁸ U			²³² Th			
		PB-13	RB-5	Total	PB-13	RB-5	Total	PB-13	RB-5	Total	
1	18.3	46.19	38.03	84.22	0.42	13.46	14.38	0.11	5.89	6.00	
2	16.3	48.97	35.37	84.34	0.50	15.84	16.34	0.12	6.79	6.91	
3	14.2	51.56	32.87	84.43	0.58	17.83	18.41	0.15	7.77	7.92	
4	12.2	54.02	30.79	84.81	0.65	19.31	19.96	0.16	8.70	8.86	
5	10.2	55.68	28.95	84.63	0.69	20.75	21.44	0.18	9.45	9.63	
6	8.1	57.14	27.42	84.56	0.77	21.92	22.69	0.20	10.18	10.38	
7	6.1	58.28	26.29	84.57	0.82	22.96	23.78	0.21	10.70	10.91	
8	4.1	59.14	25.57	84.71	0.86	23.75	24.61	0.22	11.17	11.39	
9	2.0	59.60	25.09	84.69	0.89	24.17	25.06	0.23	11.36	11.59	
10	0	59.84	24.84	84.68	0.90	24.41	25.31	0.23	11.64	11.87	
11	2.0	59.60	25.09	84.69	0.89	24.17	25.06	0.23	11.36	11.59	
12	4.1	59.14	25.57	84.71	0.86	23.75	24.61	0.22	11.17	11.39	
13	6.1	58.28	26.29	84.57	0.82	22.96	23.78	0.21	10.70	10.91	
14	8.1	57.14	27.42	84.56	0.77	21.92	22.69	0.20	10.18	10.38	
15	10.2	55.68	28.95	84.63	0.69	20.75	21.44	0.18	9.45	9.63	
16	12.2	54.02	30.79	84.81	0.65	19.31	19.96	0.16	8.70	8.86	
17	14.2	51.56	32.87	84.43	0.58	17.83	18.41	0.15	7.77	7.92	
18	16.3	48.97	35.37	84.34	0.50	15.84	16.34	0.12	6.79	6.91	
19	18.3	46.19	38.03	84.22	0.42	13.46	14.38	0.11	5.89	6.00	

^aHMP = horizontal midplane.

with respect to the reactor centerline during the shutdown period between cycles. This rotation was necessary to prevent graphite-sleeve bowing caused by the steep fast flux gradient across the RB facilities. However, rotation of the capsules makes the interpretation of flux-monitor data difficult. To get accurate data from the flux monitors, the capsules must be kept in the same position relative to the reactor centerline during the entire irradiation. This was done in HRB-7 and -8 in an attempt to get high quality flux-monitor data. During PIE of the capsules, the graphite sleeves were found to be bowed so badly that it was difficult to remove them from their stainless steel containment. Since the fuel specimens could not be removed from the graphite sleeves in the usual manner, the sleeves had to be split apart. When the flux monitors were removed from the sleeve, they became mixed and, thus, impossible to identify. Therefore, no useful flux monitor information was obtained.

The bowing of the graphite sleeves presented some unusual problems for thermal analysis. The procedures used in previous capsules reflected an assumption of concentric, cylindrical geometry, where the fuel, graphite, and both containment vessels had the same center. This assumption was not valid for HRB-7 and -8 because of the bowing of the graphite sleeve. A computer code was developed¹⁸ to deal with this situation. This code can handle any eccentricity pattern among the four cylinders (outer containment, inner containment, graphite sleeve, and fuel specimen) involved in the HRB capsules. In the worst case (shown in Fig. 8), the graphite sleeve is touching the inner containment on one side, and the fuel is touching the graphite sleeve on the same side. Using this computer model, the temperatures in HRB-7 and -8 were calculated. Maximum fuel temperatures calculated for the geometry shown in Fig. 8 were always within

¹⁸K. H. Valentine, *ECNTRK, A Computer Program to Model Heat Transfer Across Asymmetric Gaps in Cylindrical Fuel Rods* (in preparation).

Fig. 8. Isotherms for Gap Eccentricities of 0, 0.9, and 0.9 in three gaps in HRB-7 and -8 Geometry.

50°C of those calculated using concentric geometry. The concentric geometry case always yields the highest calculated centerline temperature. While the temperatures are close, the thermal center for the geometry shown in Fig. 8 is considerably different from the geometric center.

HRB-7 and -8 contained Chromel vs Alumel thermocouples at six axial locations. In addition, there were junctions for a centerline tungsten-rhenium thermocouple at five axial locations in each capsule. As stated earlier, no useful information was obtained from the centerline thermocouples due to severe decalibration; however, efforts are still under way to analytically correct these data.¹⁹ The fuel operating temperatures are calculated as described elsewhere.²⁰ These calculations are based on the temperatures measured with the Chromel vs Alumel thermocouples. The measured graphite midwall temperatures and the calculated fuel surface and center temperatures are shown in Fig. 9(a-m). Although not labeled explicitly, the uppermost curves always correspond to fuel centerline temperatures, and the lowermost to graphite midwall temperature. Additional discussion on the thermal analysis is contained in Appendix A.

Examination of Fig. 9(a-l) reveals several trends. The temperature swings during an HFIR cycle are more severe at the ends of the capsule (rods 1 and 17) than at the center of the capsule (rod 10). This type of behavior could be easily predicted from the shape of the fission heating curves shown in Fig. 7. The specimens at the center of the capsule experience somewhat higher operating temperatures, and significantly more constant operating temperatures. Performance trends related to temperature and temperature gradient are, therefore, much easier to establish in the specimens at the center of the capsule. Figure 9(m) is a plot of the axial temperature distribution at two points in time for HRB-7 and -8. With the exception of the HRB-8 plot at the end of the first RB cycle, the shapes of the curves follow similar trends. The deviation of the HRB-8 plot can be explained using the sweep gas data shown in Appendix A. The HRB-8 capsule was run with 100% He throughout this cycle, while the sweep gas in HRB-7 was only 40% He at the end of this cycle. The fission heat rates drop off rapidly toward the end of the first RB cycle as ^{235}U is burned out and ^{233}U has not yet been bred in sufficient quantities to keep the heat rates high. With constant sweep gas composition throughout this cycle the temperatures would be lower at the end of the cycle in the center of the capsule. This trend can be controlled by changing the sweep gas composition during the cycle, as shown in the HRB-7 plots.

¹⁹M. W. Waddell et al., "Correction of W-Re Thermocouple Data," *HTGR Base Program Prog. Rep. Jan. 1, 1974-June 30, 1975*, ORNL-5108, Sect. 6.10.5.

²⁰F. J. Homan, "Performance Assessment of HTGR Fuels," *HTGR Base Program Prog. Rep. Jan. 1, 1974-June 30, 1975*, ORNL-5108, Sect. 6.11.

Fig. 9. Graphite Midwall, Fuel Surface, and Center Temperature vs. Time from Beginning of Irradiation (DAYS) for (d) rod 10 in HRB-7, (e) rod 14 in HRB-7, (f) rod 17 in HRB-7 (g) :

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SECTION 10

SECTION 10

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Temperatures for a given specimen: (a) rod 1 in HRB-7, (b) rod 3 in HRB-7, (c) rod 6 in HRB-7, (d) rod 1 in HRB-8, (e) rod 3 in HRB-8, (f) rod 6 in HRB-8, (g) rod 1 in HRB-8, (h) rod 3 in HRB-8.

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(a) rod 1 in HRB-7, (b) rod 3 in HRB-7, (c) rod 6 in HRB-7,
-8.

Fig. 9. (Continued) (i) rod 6 in HRB-8, (j) rod 10 in HRB-8, HRB-8, (m) axial temperature distributions for capsules HRB-7 and

and 10 in HRB-8, (k) rod 14 in HRB-8 (l) rod 17 in
 HRB-7 and -8.

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POSTIRRADIATION EXAMINATION AND ANALYSIS

Disassembly and Visual Examination

The graphite sleeve containing the fuel rods in HRB-7 was removed by cutting off the top and bottom of the capsule and slitting the containment vessel longitudinally. An attempt was made to remove the fuel rods by removing the graphite plug from one end of the graphite sleeve and tapping the sleeve against the hot-cell floor. This technique has been used on previous HRB capsules, but was unsuccessful for HRB-7 because of the bowing of the graphite sleeve. Fuel rod removal was accomplished by slitting the graphite sleeve longitudinally at 0 and 90° orientation. The appearance of the major portion of the fuel stack, without a portion of the graphite sleeve, is shown in Fig. 10. A detailed summary of the condition of individual rods is given below.

Disassembly of HRB-8 was accomplished by cutting off the top and bottom of the capsule and using a new fixture (see Fig. 11). The graphite sleeve containing the fuel rods was pushed out of the containment vessel with this device. The graphite sleeve was in good condition with no evidence of soot. After end-milling out the bottom porous graphite plugs, the fuel rods were removed by tapping the graphite sleeve on the cell floor. A detailed summary of the condition of the individual rods is given below.

Visual examination of the HRB-7 and -8 capsules showed them to be normal in appearance (i.e., no unusual marks or evidence of localized hot spots). A white substance, thought to be an oxide, had formed on the surface of the stainless steel capsule along the fueled sections. All rods from each capsule were visually inspected with the aid of a shielded stereomicroscope. Each end and a typical side view of each rod is shown in Figs. 12 through 30. A count of those surface particles with visibly broken coatings was made and is reported in Table 11, along with the position in the capsule, type of coating and kernel, and purpose in the experiment. Although the type of failed particle could not be positively identified in most cases (i.e., fissile, fertile, driver, or inert), these results give an indication of relative performance. As can be seen in the above figures, the general condition of the majority of rods from HRB-7 ranged from excellent to good except for rod 10, which exhibited considerable debonding (particles broken loose from the matrix) along its length. The condition of the rods from HRB-8 (higher fluence and lower operating temperatures than HRB-7) was excellent to good, except for rods 1, 5, 10, 19. One end of rods 1 and 19 had debonded, while rods 5 and 10 had debonded along their lengths.

An arbitrary failure grouping of rod vs number of failed particles observed on the surfaces of the rods, shown in Table 12, gives an indication of relative performance. Although about 2000 particles were visible on the rod surfaces, only about 5 to 10% of the particles were fissile particles. If we assume that the only particles that failed were the fissile particles (over half of the fuel rods showed only 0 to 1 failure), then the poorer performers were rods 1, 4, 5, 7, 10, and 18. Only rod 4 of the poor performers contained Biso-coated fissile particles. However, when these results are compared to the results of gamma scans of the empty

Fig. 10. Fuel Rod Specimens Contained in Graphite Sleeve of HRB-7. Seventeen of the 19 rods are shown; the top of the fuel stack is on the right ($\sim 1/2$ scale).

Fig. 11. Disassembly Fixture Used to Remove the Fueled Graphite Sleeve from HRB-8. After cutting off the top and bottom ends, the capsule was placed in the universal chuck and the steel push rods were inserted in the Jacob's Chuck. The rack and pinion was then used to push the graphite sleeve out of the capsule.

Fig. 12. Fuel Rods from Position 1 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 was irradiated to a fast neutron fluence of 3.6×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 4.8×10^{21} neutrons/cm²; ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 13. Fuel Rods from Position 2 of Capsules HRR-7 (top row) and HRR-8 (bottom row). The HRR-7 rod was irradiated to a fast neutron fluence of 4.1×10^{21} neutrons/cm²; the HRR-8 rod was irradiated to a fast neutron fluence of 5.1×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top (b) Side, (c) Bottom, 5x.

Fig. 14. Fuel Rods from Position 3 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 4.5×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 5.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 15. Fuel Rods from Position 4 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 4.9×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 6.4×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 16. Fuel Rods from Position 5 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.3×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 6.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 17. Fuel Rods from Position 6 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.6×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.4×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 18. Fuel Rods from Position 7 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.9×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.7×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 19. Fuel Rods from Position 8 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 6.0×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 20. Fuel Rods from position 9 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 6.1×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 8.1×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 21. Fuel Rods from Position 10 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 6.1×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 8.1×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom 5 $\times$.

Fig. 22. Fuel Rods from Position 11 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 6.1×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 8.1×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 23. Fuel rods from Position 12 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 6.0×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 24. Fuel Rods from Position 13 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.9×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.7×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

"Top view not photographed because of presence of segment of Re foil, which was left in for radiographic examination."

Fig. 25. Fuel Rods from Position 14 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.6×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 7.4×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top (b) Side (c) Bottom, 5 $\times$.

Fig. 26. Fuel Rods from Position 15 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 5.3×10^{21} neutrons/cm²; the HRB-8 was irradiated to a fast neutron fluence of 6.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 27. Fuel Rods from Position 16 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 4.9×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 6.4×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5x.

Fig. 28. Fuel Rods from Position 17 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 4.5×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 5.9×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Fig. 29. Fuel Rods from Postion 18 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 4.1×10^{21} neutron/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 5.3×10^{21} neutron/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom 5%.

Fig. 30. Fuel Rods from Position 19 of Capsules HRB-7 (top row) and HRB-8 (bottom row). The HRB-7 rod was irradiated to a fast neutron fluence of 3.6×10^{21} neutrons/cm²; the HRB-8 rod was irradiated to a fast neutron fluence of 4.8×10^{21} neutrons/cm² ($E > 0.18$ MeV). (a) Top, (b) Side, (c) Bottom, 5 $\times$.

Table 11. Results of Visual Examination for Failed Particles on the Surfaces of Rods from HRB-7 and -8^a

Position in Capsule	Failed Particles on Rod Surface ^b		Kernel Type	Kernel Diameter (μm)	Coating Type	Purpose in Experiment
	HRB-7	HRB-8				
1	13	16	²³⁵ UC ₂ -1% Th ^c	200	Triso	GAC reference
5	52	14	²³⁵ UO ₂ SG	215	Triso	Potential candidate for recycle; compare with ²³⁵ UO ₂
7	11	1	²³⁵ UO ₂ SG	200	Triso	Reference for amoeba effect; compare Triso with Biso
9	2	0	(4 Th, ²³⁵ U)O ₂ SG	150	Triso	Recycle Candidate
RHMP-10	12	>30	(8 Th, ²³⁵ U)O ₂ ^c SG	500	Triso	Relative stability; Th/U ratio; interest for first-core loadings
13	1	0	WAR ²³⁵ UO ₂ + UC ₂	400	Triso	Compare with GAC reference makeup particle
14	3	0	WAR ²³⁵ UC ₂	100	Triso	Recycle candidate; compare Triso with Biso
17	0	2	²³⁵ UC ₂ with ZrC in coating ^b	200	Triso	Test GAC advance coating design
18	28	23	²³⁵ UC ₂ -1% Th ^c	200	Triso	GAC reference
2	0	5	(4Th, ²³⁵ U)O ₂ SG	350	Biso	Test particle prepared in large coater
3	1	0	²³⁵ UO ₂ + U ₂ N ₃ SG	200	Biso	Study amoeba effect
4	4	11	²³⁵ UO ₂ + UC ₂ SG	200	Biso	Study amoeba effect
6	5	6	²³⁵ UO ₂ SG	200	Biso	Reference for amoeba effect; compare Biso with Triso
8	0	0	(2 Th, ²³⁵ U)O ₂ SG	100	Biso	Relative stability; Th/U ratio
11	4	0	(4Th, ²³⁵ U)O ₂ SG	150	Biso	Relative stability; Th/U ratio
12	4	1	(Th, ²³⁵ U)O ₂ SG	250	Biso	Relative stability; Th/U ratio
15	0	0	WAR ²³⁵ UO ₂ + UC ₂	400	Biso	Compare Biso with Triso
16	0	0	WAR ²³⁵ UC ₂	100	Biso	Recycle candidate; compare Biso with Triso
19	0	5	(4 Th, ²³⁵ U)O ₂ SG	350	Biso	Test particle prepared in large coater

^aSG = derived by sol-gel process.

RHMP = reactor horizontal midplane.

WAR = derived from weak-acid resin.

^bAll failed particles not positively identified as being the fissile particle.

^cCoated by GAC.

Table 12. Summary of the Number of Failed Particles Observed the Surface of HRB-7 and -8 Fuel Rods

Failure Range	Rod Number	
	HRB-8	HRB-7
0	2, 8, 15, 16, 17, ^a 19	3, 8, 9, ^a 11, 13, ^a 14, ^a 15, 16
1-5	3, 4, 6, 11, 12, 13, ^a 14, ^a	2, 7, ^a 12, 17, ^a 19
6-10		6
11-15	1, ^a 7, ^a 10 ^a	4, 5 ^a
>15	5, ^a 18, ^a	1, ^a 10, ^a 18 ^a

^aTriso-Biso system.

graphite sleeves it becomes apparent that visual examination alone is not adequate for performance evaluations. For example, the visual examination indicated that fuel rod 15 showed no failures, whereas the gamma scans indicated that it was the poorest performer with respect to fission-product retention.

Gamma Scan of Capsule and Graphite Sleeve

After removal of the thermocouple and sweep gas leads, gamma scans were obtained along the lengths of the capsules. The gamma scans (see Fig. 31) showed the fuel rods to be intact with no gross inhomogeneities of fuel within the individual rods and no accumulation of fission products at the ends of the fuel stack. The activity peaks shown above the fuel stacks are from the stainless steel bulkheads. Note also that the average count rate reflects the axial flux profile of the HFIR. The integrated gamma scans were made using a 0.01- by 1.0- in. collimated slit 17 in. long and NaI detector over an energy range of 0.55-0.75 MeV. No attempt should be made to compare the relative magnitudes of the gamma scans between capsules, as the numbers on the scan charts in Fig. 31 have no meaning.

Gross gamma scans along the lengths of the empty graphite sleeves from HRB-7 and -8 aided in determining the relative performance of the fuel rods. In the gross scan of the HRB-7 graphite sleeve, shown in Fig. 32, the relative retention of the volatile fission products in the individual fuel rods is apparent. A more detailed analysis was performed on the graphite sleeve from HRB-8. In addition to the gross gamma scan, gamma spectra for several elements were acquired at select locations using a germanium-lithium detector. A composite of the gross gamma scans made of the fueled and empty graphite sleeve is shown in Fig. 33. The results of this composite show that rods 4 and 15 released more fission products than rods 8 and 9. Other interesting comparisons can be made with Fig. 33.

Fig. 31. Gamma Scans Made Along the Lengths of Capsules HRB-7 (top row) and HRB-8 (bottom row). The activity peaks beyond the fuel rod stack are from the stainless steel bulkheads of the capsule and not from an accumulation of fission products.

Fig. 32. Gamma Scan Made Along the Length of the Empty Graphite Sleeve from HRB-7. The scan was made to include energy peaks in the range of 550 to 750 keV.

Fig. 33. Gamma Scans of HRB-8 Capsule. Detector was set to respond to any photon in the energy range of 550 to 750 keV. This range includes ^{134}Cs , ^{114}Ce , ^{154}Eu , and ^{91}Zr .

For example, rods 6 and 7 both contain $^{235}\text{UO}_2$ kernels made via the sol-gel process. The fissile kernels in rod 6 were Biso coated; in rod 7 they were Triso coated. Figure 33 suggests better fission product retention for the Triso-coated particles. Rods 13 and 15 both contain $\text{UC}_{0.44}\text{O}_{1.72}$ (15% converted WAR) kernels in the fissile particles. The fissile particles in rod 13 are Triso coated; in rod 15 they are Biso coated. Again, Fig. 33 suggests better fission product retention for the Triso-coated particles. Rods 8, 9, 10, 11, and 12 all contain mixed-oxide fissile particles with varying Th/U ratios, and either Biso or Triso coatings. There is very little difference in fission product retention capability for these particles, according to the gamma scan.

Capsules HRB-7 and -8 were the first capsules to be gamma scanned under this program. The information is very useful, but still qualitative in nature. Quantitative information will require the application of geometry corrections and the use of standards. One useful conclusion is that there appears to be little general smearing of fission products. The Grafoil spacers between fuel rods help prevent smearing.

After completing the gross gamma scans, a high resolution germanium-lithium detector was used to acquire gamma spectra at eight different locations along the length of the sleeve. The locations were chosen to correspond to the center positions of the eight fuel rods listed in Table 13.

Table 13. Fissile Particle Design Data for HRB-8 Rods
Examined by Gamma Spectroscopy

Fuel Rod	Kernel Diameter (um)	Coating Thickness (um)				Kernel Type	Coating Type
		Buffer	Inner LTI	SIC	Outer LTI		
1	196	99	33	32	38	$^{235}\text{UC}_2 + 12 \text{ ThC}_2$	Triso
2	354	101			99	(4 Th, $^{235}\text{U})\text{O}_2$ SC	Biso
9	360	98	39	34	43	(4 Th, $^{235}\text{U})\text{O}_2$ SC	Triso
10	502	106	32	34	42	(8 Th, $^{235}\text{U})\text{O}_2$ SC	Triso
13	386	41	43	32	49	$^{235}\text{UO}_2 + 142 \text{ UC}_2$ WAR	Triso
14	315	75	36	28	49	$^{235}\text{UO}_2 + 922 \text{ UC}_2$ WAR	Triso
15	386	41			99	$^{235}\text{UCO}_2 + 142 \text{ UC}_2$ WAR	Biso
17	197	102	8	21 ²	65	$^{235}\text{UC}_2$	Triso ⁴

²ZrC is substituted for SIC.

The concentrations of fission products were measured using gamma spectroscopy techniques for these eight positions, and the concentrations were corrected to October 31, 1974, the last day of the irradiation. These data are presented in Table 14. No attempt was made to measure the actual diametral distribution of fission products on the sleeve. But due to a fortuitous counting geometry, the numbers in Table 14 also represent (within a factor of 2) a surface concentration by changing the units to atoms/cm².

Table 14. Fission Product Concentrations Measured by Gamma Spectroscopy in Graphite Sleeve Adjacent to Selected Fuel Rods from HRE-8

Fuel Rod	Fission Product Concentration, 10^{16} atoms/cm ³						
	¹³⁴ Cs	¹³⁷ Cs	^{110m} Ag	⁹⁵ Zr	¹⁴⁴ Ce	¹⁵⁴ Eu	²³³ Pa
1	10.3	57.8	1.08				
2	12.7	69.6	1.00	0.139	0.316		
9	6.7	44.8	1.07	0.130	0.244		0.369
10	8.6	48.7	1.22				
13	7.0	39.8	1.29				
14	12.1	67.6	1.32				
15	14.0	83.5	1.22	0.138	0.241	4.49	
17	6.0	34.5	1.15				

Dimensional Changes

The length and diameter of each fuel rod from HRE-7 and -8 were determined and compared to as-fabricated dimensions. The diameters were averaged from six measurements taken at the top, midlength, and bottom of each rod at 0 and 90°. The bottom and 0° orientation of the rods were identified from a groove that was molded into the rods during fabrication. The maximum length was determined for the fuel rods. The resultant dimensional changes are given in Table 15. Correlation of fuel system, dimensional changes, and fast fluence are shown in Fig. 34. The presence of the more dimensionally stable Triso-coated particles did not significantly reduce the shrinkage of the fuel rods relative to the fuel rods that contained Biso-coated particles only. This is not a surprising result since the volume occupied by the fissile particles was only about 10% of the fuel rod volume. The maximum shrinkage of the fuel rods in HRE-7 and -8 was about 5%. The minimum in the $\Delta D/D$ vs fluence plots occurred at fast fluence levels of 5.0 and 6.5×10^{21} neutrons/cm², respectively, for the two capsules. The data shown also that the shrinkage was slightly anisotropic in nature, with more shrinkage occurring along the length of the rods.

The graphite sleeves (POCO AXFSQ)²¹ were measured along the lengths and compared to as-machined dimensions. Maximum swelling occurred in the graphite at the region of maximum fluence (reactor horizontal midplane). The graphite swelled a maximum of 1.2% in HRE-7 and 0.95% in HRE-8. Additional information on the swelling behavior of these graphite sleeves can be found in Appendix A.

²¹POCO Graphite, Inc., a subsidiary of Union Oil Company of California, Decatur, Texas.

Table 15. Dimensional Changes of Fuel Rods Irradiated
in Capsules HRB-7 and -8 vs Fast Fluence

Fuel Rod	Coating Combination ²	Change, %		$\frac{\Delta D/\Delta L}{D/L}$	Fast Fluence, E > 0.18 MeV (neutrons/cm ²)
		Diameter	Length		
<u>HRB-7</u>					
1	T-B	-3.72	-3.54	1.05	3.62 × 10 ²¹
2	B-B	-4.69	-4.99	0.94	4.07
3	B-B	-5.02	-4.95	1.01	4.50
4	B-B	-5.01	-5.33	0.94	4.88
5	T-B	-4.16	-4.82	0.86	5.26
6	B-B	-4.57	-4.75	0.96	5.59
7	T-B	-4.55	-5.03	0.90	5.87
8	B-B	-4.67	-5.06	0.92	6.04
9	T-B	-3.71	-4.33	0.86	6.14
10	T-B	-3.70	-3.74	0.99	6.14
11	B-B	-4.50	-5.09	0.88	6.14
12	B-B	-4.91	-5.09	0.96	6.04
13	T-B	-4.51	-5.13	0.88	5.87
14	T-B	-4.60			5.59
15	B-B	-4.82	-5.31	0.91	5.26
16	B-B	-5.35	-5.77	0.93	4.88
17	T-B	-4.92	-5.11	0.96	4.50
18	T-B	-4.54	-5.07	0.90	4.07
19	B-B	-4.21	-4.31	0.98	3.62
<u>HRB-8</u>					
1	T-B	-3.93	-3.92	1.00	4.78 × 10 ²¹
2	B-B	-4.33	-4.59	0.94	5.25
3	B-B	-4.77	-4.61	1.03	5.92
4	B-B	-4.61	-6.07	0.75	6.42
5	T-B	-4.03	-6.68	0.60	6.92
6	B-B	-4.80	-4.67	1.03	7.36
7	T-B	-4.67	-5.03	0.93	7.73
8	B-B	-4.77	-5.52	0.86	7.95
9	T-B	-3.92	-3.52	1.11	8.09
10	T-B	-3.52	-3.87	0.91	8.09
11	B-B	-4.58	-4.81	0.95	8.09
12	B-B	-4.65	-4.90	0.95	7.95
13	T-B	-4.49	-4.93	0.91	7.73
14	T-B	-4.55	-5.25	0.87	7.36
15	B-B	-4.95	-5.17	0.96	6.92
16	B-B	-4.99	-5.42	0.92	6.42
17	T-B	-5.00	-5.14	0.97	5.92
18	T-B	-4.32	-4.39	0.98	5.35
19	B-B	-4.07	-4.53	0.90	4.78

^aT-B means Triso-coated fissile particles and Biso-coated particles; B-B means both fissile and fertile particles are Biso coated.

Fig. 34. Dimensional Change vs Fast Fluence for Fuel Rods Irradiated in HRB-7 and -8.

Metallographic Examination

All 38 fuel rods from the two irradiation capsules were examined metallographically. Transverse sections were made at about the midlength of each rod for metallographic preparation. A summary of the metallographic results for the various fissile particles is given in Tables 16 and 17. The appearance of the fuel particles in HRB-7 and -8 were essentially identical, and subtle differences could be attributed to the different operating temperatures. Representative particles shown in Figs. 35 through 57 are in the same order as listed in Table 16 and 17. The following general observations were noted during the metallographic examinations:

1. There was no evidence of migration of the ThO_2 kernels in any of the fuel rods, whereas a measureable amount of migration was observed with all of the fissile-oxide kernels. The kernel migration coefficients have been calculated by Lindemer and Pearson.²² A comparison of the performance of the various mixed-oxide kernels is shown in Fig. 58.

2. Examination of over 350 Biso-coated fertile particles (from Batch J-262 see Table 3) from HRB-7 and -8 fuel rods disclosed that about 3% of the buffer coatings had failed (Fig. 59), and about 1% of the buffer and inner LTI coatings had failed (Fig. 60). Examination of the coatings under polarized light revealed a noticeable degree of anisotropy, indicating that the coating failures resulted from fast neutron damage.

3. The performance of the WAR-derived kernels showed that the 100% converted kernels behaved like the dense seeded UC_2 kernels. When compared to the fully converted kernel, the 15% converted kernel showed a marked improvement in performance. A comparison of the three is shown in Fig. 61.

4. During the planning for HRB-7 and -8 there was some concern that the fission product palladium, which is formed in high concentrations in the ^{238}U -bearing kernels (used in the Biso driver particles), would migrate out of the driver particles and attack adjacent particles. No evidence of this was observed in metallographic examination.

5. During the planning of HRB-7 and -8, there was also concern that chemical reactions might occur between the rhenium foil used to protect the central thermocouple and the coated particles on the inner surfaces of the annular fuel rods. The rhenium foil was included in the metallographic section taken through rod 14 from HRB-7. No reaction was observed in the metallographic examination.

²²T. B. Lindemer and R. L. Pearson, "Mass Transport Mechanisms in Temperature Gradients," *Gas-Cooled Reactor Program HTGCR Base-Technology Program Progress Report Jan. 1, 1974-June 30, 1975*, ORNL-5108, pp. 205-8.

Fig. 35. Triso-Coated UC_2 Particle Irradiated in Rod 1 of HRB-7. Top row: the average temperature was $810^\circ C$, and the average temperature gradient was $940^\circ C/cm$. Bottom row: the average temperature was $1120^\circ C$, and the average temperature gradient was $0^\circ C/cm$. (a) and (b) Bright-field illumination; (c) and (d) polarized light. The hot side of the particle in (a) and (c) is on the bottom.

Fig. 36. Triso-Coated UC_2 Fissile Particle Irradiated in Rod 1 of HRB-8. The average temperature was $1180^\circ C$ and the average temperature gradient was $580^\circ C/cm$. The hot side of the particle is on the right; (a) bright-field illumination; (b) polarized light.

Fig. 37. Triso-Coated UC_2 Particle Irradiated in Rod 18 of HRB-8. (a) the average temperature 1280°C and the average temperature gradient was $340^\circ\text{C}/\text{cm}$; the cold side of the particle is on the top. (b) A higher magnification of the cold side of the particle, showing crack in the SiC coating and the fission product attack of the SiC .

Fig. 38. Failed Triso-Coated UC_2 Particle in Rod 18 of HRB-8. The average temperature was $1250^{\circ}C$ and the average temperature gradient was $490^{\circ}C/cm$. The hot side is on the bottom; (a) bright-field illumination (b) polarized light.

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Fig. 39. Triso-Coated UC₂ Particle. (a) Rod 17 of HRB-7 was operated at an average temperature of 1360°C and an average temperature gradient of 420°C/cm with the hot side of the particle on the top. (b) Rod 17 of HRB-8 was operated at an average temperature of 1225°C and an average temperature gradient of 590°C/cm with the hot side of the particle on the top. This coating design employed ZrC instead of SiC and a thin seal coat rather than the reference inner LTI.

P-65348

Fig. 40. High Magnification View of ZrC Coating from Fig. 39 Showing (a) Cold Side of the particle and (b) Hot Side. The "peppery" appearance of ZrC is thought to be a chemically induced porosity due to residual chlorine from the coating run.

50 100 200X 300 350
MICRONS
0.005 INCHES 0.010 0.015

50 100 200X 300 350
MICRONS
0.005 INCHES 0.010 0.015

Fig. 41. Biso-Coated Sol-Gel UO_2 Located on the Outer Surface of Rod 6 of HRB-7 Where the Average Temperature was 1180°C and the Average Temperature Gradient was $990^\circ\text{C}/\text{cm}$. The hot side of the particle is on the bottom; (a) bright-field illumination; (b) polarized light. Particles from the companion rod in HRB-8 had a similar appearance.

Fig. 42. Triso-Coated Sol-Gel UO_2 Particles from Rod 7 of HRB-7. (a) The average temperature was 1180°C and the average temperature gradient was $990^\circ\text{C}/\text{cm}$; the hot side is on the bottom; (b) attack of SiC layer on the cold side of a particle that was operated at an average temperature of 1295°C and an average temperature gradient of $690^\circ\text{C}/\text{cm}$. Particles from the companion rod in HRB-8 had a similar appearance, except the attack of SiC was not seen there. The attack is oxidation and is not fission-product related, as a microprobe analysis shows that all fission products are retained in the kernel.

Fig. 43. Triso-Coated Sol-Gel $^{233}\text{UO}_2$ Particle. (a) For rod 5 of HRB-7, the average temperature was 1295°C and the average temperature gradient was 690°C/cm ; the hot side is on the top. A portion of the kernel was lost during metallographic preparation. (b) For rod 5 of HRB-8 the average temperature was 1180°C and the average temperature gradient was 560°C/cm . This is an example of a pressure vessel failure. The oxide kernel has been converted by carbide as evidenced by the high optical reflectivity of the kernel.

Fig. 44. Triso-Coated WAR Particle (85% UO_2 -15% UC_2 kernel) from Rod 13 of HRM-7. The particle's average temperature was 1350°C , and the average temperature gradient was $720^\circ\text{C}/\text{cm}$. (a) Bright-field illumination; (b) polarized light. The hot side of the particle is on the bottom.

50 100 MICRONS 200X 300 350
0.005 INCHES 0.010 0.015

50 100 MICRONS 200X 300 350
0.005 INCHES 0.010 0.015

Fig. 45. Triso-Coated WAR Particle (85% UO_2 -15% UC_2 kernel) from Rod 13 of HRB-8 Irradiated at an Average Temperature of 1330°C and an Average Temperature Gradient of $260^\circ\text{C}/\text{cm}$. (a) Bright-field illumination; (b) polarized light. Note the two-phase appearance of the kernel; the optically active phase under polarized light is nonmetallic and contains carbon from the buffer coating. The hot side is on the bottom.

Fig. 46. Biso-Coated WAR Particle (85% UO_2 -15% UC_2) from Rod 15 of HRB-7 Irradiated at an Average Temperature of 1460°C and an Average Temperature Gradient of 0°C/cm .
(a) Bright-field illumination; (b) polarized light.

Fig. 47. Triso-Coated WAR Particle (100% UC_2) in Rod 14 of HRB-7 Irradiated at an Average Temperature of $1442^\circ C$ and an Average Temperature Gradient of $260^\circ C/cm$. The hot side of the particle is on the top. (a) Bright-field illumination; (b) polarized light. Note the accumulation of fission products on the cold side of the particle (optically active in polarized light), which resulted in failure of the SiC layer.

50 100 MICRONS 300 350
0.005 INCHES 0.010 0.015

50 100 MICRONS 300 350
0.005 INCHES 0.010 0.015

Fig. 49. Triso-Coated WAR particle (100% UC_2) from Rod 14 of HRB-8 Irradiated at an Average Temperature of $1130^\circ C$ and an Average Temperature Gradient of $990^\circ C/cm$. The hot side of the particle is on the bottom. (a) Bright-field illumination; (b) polarized light.

50 100 MICRONS 200X 300 350
0.005 INCHES 0.010 0.015

50 100 MICRONS 200X 300 350
0.005 INCHES 0.010 0.015

Fig. 49. Biso-Coated WAR Particle (100% converted) from Rod 16 of HRB-7 Irradiated at an Average Temperature of 1380°C and an Average Temperature Gradient of $240^{\circ}\text{C}/\text{cm}$. The hot side of the particle is on the right. (a) Bright-field illumination; (b) polarized light. There was no evidence of amoeba although the kernel did move unilaterally through the buffer layer. There was no correlation between this movement and the apparent temperature gradient.

Fig. 50 Biso-Coated Sol-Gel $\text{UO}_2 + 7\% \text{U}_2\text{N}_3$ Particle from Rod 3 of HRB-7. (a) Unfailed particle was irradiated at an average temperature of 1105°C and an average temperature gradient of $890^\circ\text{C}/\text{cm}$ with the hot side of the particle on the bottom. (b) Failed particle was irradiated at an average temperature of 1170°C and an average temperature gradient of $730^\circ\text{C}/\text{cm}$ with the hot side of the particle on the right. Movement of the kernel is not amoeba, because it is independent of the temperature gradient. Unfailed particles have an oxide appearance, but kernels of failed particles appear to have been converted to carbide.

Fig. 51. Biso-Coated Sol-Gel ($\text{Th}_{0.5}\text{U}_{0.5}\text{O}_2$) Particle from Rod 12 of HRB-8. The average irradiation temperature was 1240°C and the average temperature gradient was $640^\circ\text{C}/\text{cm}$. The hot side of the particle is on the upper left. (a) Bright-field illumination; (b) polarized light. Note severe amoeba, gas bubbles, and metallic fission products in the kernel; also note the deposition of a carbon phase on the cold side of the kernel.

Fig. 52. Biso-Coated Sol-Gel ($\text{Th}_{0.67}\text{U}_{0.33}\text{O}_2$) Particle from Rod 8 of HRB-7 Irradiated at an Average Temperature of 1220°C and an Average Temperature Gradient of $1060^\circ\text{C}/\text{cm}$. The hot side of the particle is on the top. (a) Bright-field illumination; (b) polarized light. The particles in this rod were similar in appearance to the ($\text{Th}_{0.5}\text{U}_{0.5}\text{O}_2$) particles irradiated in rod 12 (Fig. 51). Particles from rod 8 of HRB-8 had a similar appearance also.

Fig. 53. Biso-Coated Sol-Gel $(\text{Th}_{0.8}\text{U}_{0.2})\text{O}_2$ Particle from Rod 11 of HRB-7 Irradiated at an Average Temperature of 1350°C and an Average Temperature Gradient of 715°C/cm . The hot side of the particle is on the top. (a) Bright-field illumination; (b) polarized light.

Fig. 54. Biso-Coated Sol-Gel ($\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$) Particle from Rod 2 of HRB-8 Irradiated at an Average Temperature of 1225°C and an Average Temperature Gradient of $540^\circ\text{C}/\text{cm}$. The hot side of the particle is on the top. (a) Bright-field illumination; (b) polarized light.

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R-65406

Fig. 55. Biso-Coated Sol-Gel $(\text{Th}_{0.6}\text{U}_{0.4})\text{O}_2$ Particle from Rod 19 of MKB-7 Irradiated at an Average Temperature of 1180°C and an Average Temperature gradient of 970°C/cm . The hot side of the particle is on the bottom. (a) Bright-field illumination; (b) polarized light.

125 MICRONS 375
180X
0.005 INCHES 0.015

125 MICRONS 375
180X
0.005 INCHES 0.015

Fig. 56. Triso-Coated Sol-Gel $(Th_{0.6}U_{0.3})O_2$ Particle from Rod 9 of HKB-7 Irradiated at an Average Temperature of $1300^{\circ}C$ and an Average Temperature Gradient of $860^{\circ}C/cm$. The hot side of the particle is on the bottom. (a) Bright-field illumination; (b) polarized light. Note the slight gap that developed between the buffer and the inner I.TI on the hot side.

Fig. 57. Triso-Coated Sol-Gel $(\text{Th}_{0.66}\text{U}_{0.34})\text{O}_2$ Particle from Rod 10 of HKB-8 Irradiated at an Average Temperature of 1320°C and an Average Temperature Gradient of $340^\circ\text{C}/\text{cm}$. The hot side of the particle is on the top. Failed outer LTI coatings were typical for this batch of particles and were due to poor coating properties, as evidenced by the high optical activity for the outer LTI shown in the polarized-light view. (a) Bright-field illumination; (b) polarized light.

Table 16. Summary of Metallographic Observations for HRB-7 and -8 Rods

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NOT RECORDED AND INDEXED

Table 17. Summary of Particle Failure and Amoeba Characterization for HKB-7 and -8

Fuel Rod	HKB-7					HKB-8				
	Number of Fissiles Examined	Number of Failed Fissiles	Amoeba		Maximum Migration (μ m)	Number of Fissiles Examined	Number of Failed Fissiles	Amoeba		Maximum Migration (μ m)
			Yes	No				Yes	No	
1	9	3		✓		24	23		✓	
18	8	4		✓		13	13		✓	
17	13	0		✓		7	0		✓	
6	7	0	✓		65	8	0	✓		28
7	11	1	✓		100	22	0	✓		95
5	4	2	✓		65	7	4	✓		100
13	5	0		✓		7	0		✓	
15	10	0		✓		7	0		✓	
14	11	5		✓		2	2		✓	
16	8	0		✓	~100	6	0		✓	
3	16	4		✓	>150	7	1		✓	>150
4	12	2 ^a		✓	>150	5	1		✓	
12	14	1 ^a	✓		55	15	2	✓		55
8	10	0	✓		80	12	0		✓	
11	12	0	✓		75	7	0	✓		20
2	10	0	✓		40	6	0	✓		13
19	10	0	✓		30	6	0	✓		20
9	23	0	✓		25	11	0	✓		20
10	8	1	✓		25	14	0	✓		85

^aFailed during rod fabrication.

Fig. 58. Relative Thermal Stabilities of Candidate HTGR Recycle Fuel Particles Irradiated in HRR-7. Average irradiation temperatures ranged from 1200 to 1220°C; average thermal gradients ranged from 1000 to 1080°C/cm. The hot side of the particle is on the right in all cases.

R-65478

R-65479

Fig. 59. Failed Buffer Coating in Biso-Coated Sol-Gel ThO_2 Particle. This batch of fertile particles was used in all fuel rods in HRB-7 and -8. Failures were due to fast neutron damage in coating. (a) Bright-field illumination; (b) polarized light.

Fig. 60. Failed Biso-Coated Sol-Gel ThO_2 Fertile Particle. This particle was irradiated in rod 10 of HRB-7 at an average temperature of 1410°C and an average temperature gradient of $420^\circ\text{C}/\text{cm}$. The hot side is on the bottom of the lower left.

Fig. 61. Relative Performance of Candidate and reference HTGR Fissile Particles Irradiated in HRB-7 in Rods 18 (left), 14 (center), and 13 (right). The average irradiation temperatures were, respectively, 1390, 1440, and 1350°C; average temperature gradients were, respectively, 210, 260, and 720°C/cm. The hot side of the particle was always on the right. Note the graphitization of the inner LTI in the dense UC₂ and WAR UC₂ particles. The 15% UC₂ particle shows partial consumption of the buffer layer by the kernel, but there is no evidence of graphitization of the inner LTI.

Electron Microprobe Analysis

Electron microprobe analyses were performed on a select number of metallographic specimens from HRB-7 and -8. These included 100% converted WAR, 15% converted WAR, sol-gel UO_2 , and dense melted UC_2 -1% Th (with ZrC coating instead of SiC) fissile particles with Triso coatings. The irradiation conditions of the particles used in the microprobe analyses are given in Table 18.

Table 18. Irradiation Conditions of Particles from HRB-7 and -8 Used in Electron Microprobe Analysis

Figure Number	Kernel Composition	Experiment and Rod Numbers	Distance of Particle from Rod to Outside Diameter (cm)	Time-Averaged Temperature ($^{\circ}\text{C}$)	Time-Averaged Thermal Gradient ($^{\circ}\text{C}/\text{cm}$)	Burnup (at. %)
62, 63	$\text{UC}_{2.01}\text{O}_{0.10}$ WAR (100% reduced)	HRB-8, rod 14	On outside diameter	1130	990	84.5
64	Sol-Gel UO_2	HRB-8, rod 7	On outside diameter	1200	910	84.5
65	$\text{UC}_{2.00}\text{O}_{0.15}$ WAR (15% reduced)	HRB-8, rod 13	1300	1240	690	84.5
66	99% UC_2 -1% Th (ZrC coating)	HRB-8, rod 17	1700	1225	600	84.5
67	99% UC_2 -1% Th (ZrC coating)	HRB-7, rod 17	1700	1225	600	84.5

² Rods are 0.24 in. or 6100 μm .

One of the major factors limiting the performance of recycle fissile particles is the migration of the high-yield rare-earth fission products lanthanum, praseodymium, cerium, and neodymium down the temperature gradient. If the fissile kernel does not contain sufficient oxygen to form stable oxides with the rare earths, they will move out of the kernel and accumulate on the cold side of the particles, as shown in Fig. 62. The presence of the rare earths can result in a corrosive attack of the SiC coating whose strength could seriously be affected. In addition, the buildup of the rare earths (presumably in the form of carbides with high volume expansion) can cause a mechanical interaction with the coatings and can lead to particle failure. The 100% reduced WAR particle shown in Fig. 62 is a good example of the results of massive rare-earth accumulation.

The behavior of other important elements (from the same particle as in Fig. 62) in a 100% reduced WAR particle is presented in Fig. 63. As shown, both the uranium and zirconium remain within the kernel. The cesium, however, was found in the surrounding carbon coatings, except where the SiC coating had failed. Being extremely mobile at elevated temperatures, the cesium evidently distilled out of the kernel into the surrounding carbon coatings. The absence of cesium where the SiC has broken is evidence that the particle indeed failed during operation, and not during metallographic preparation.

Y-127988

Optical Micrograph

La L α Pr L α

Backscattered Electron Image

Ce L α Nd L α

Fig. 62. Distribution of Rare-Earth Fission Products in a Triso-Coated WAR UC₂ Particle Irradiated in Rod 14 of HRB-8 at an Average Irradiation Temperature of 1130°C and an Average Temperature Gradient of 990°C/cm. The hot side of the particle is at the top. Note the heavy concentration of fission products on the cold side of the particle at the inner LFI SiC interface.

Y-127907

Optical Micrograph

Umap

CsL α ZrL α

Fig. 63. Distribution of Uranium, Zirconium, and Cesium in the Triso-Coated WAR UC_2 Particle Irradiated in Rod 14 of HRB-8 at an Average Irradiation Temperature of $1130^\circ C$ and an Average Temperature Gradient of $990^\circ C/cm$. The hot side of the particle is at the top. Note the heavy concentration of fission products on the cold side of the particle at the inner LTI-SiC interface.

The retention of the rare earths within the kernel of sol-gel UO_2 is shown in Fig. 64. Apparently the rare earths are tied up as stable oxides and no migration takes place. Also contained in the kernels, although not present in Fig. 64, are uranium, zirconium, and barium. The cesium (also not shown) was observed in the surrounding carbon coatings and not in the kernel, but always within the SiC coating. This indicates the SiC coating did not fail during irradiation.

With a 15% converted MAR particle, the majority of the rare earths are retained in the kernel with the uranium, as shown in Fig. 65. Concentrations of the rare earths also appear in separate inclusions dispersed in the densified buffer (associated with uranium). The cesium distribution, although not shown, was observed in the densified buffer and the inner LTI. All cesium was found within the SiC coating, indicating the SiC remained intact throughout irradiation. Results of the microprobe analysis on a dense melted UC_2 -1% Th particle with a ZrC coating are shown in Fig. 66. Uranium and fission product zirconium are contained within the kernel only. The cerium display, which is representative of the other high-yield rare earths, shows a high concentration on the periphery of the kernel and is present in the kernel and the carbon coatings. In addition, cerium appears to be in the ZrC coating and on the outside of the ZrC as well. This result, coupled with the fact that no evidence of graphitization of the inner LTI was found during metallographic examination of ZrC coated particles, suggests that the cerium and other rare earths were able to diffuse through the ZrC coating. Cesium, on the other hand, was contained within the ZrC in the carbon coatings, proving that the ZrC coating remained intact during irradiation.

Metallographic examination of dense melted UC_2 -1% Th particles with ZrC coatings from HRB-7 showed corrosive attack of the ZrC. Microprobe analysis, presented in Fig. 67, shows the attack attributable to uranium. It was discovered that the ZrC contained an appreciable amount of chlorine (less than 1%)²³, which is associated with the deposition of the coating. Evidently, during irradiation, highly volatile uranium chlorides were formed, leading to the uranium migration and eventual attack of the ZrC. Although a great deal of corrosion occurred, the ZrC remained intact, as evidenced by the cesium display, which shows all cesium within the ZrC.

²³Personal communication between T. N. Tiegs (ORNL) and G. A. Reynolds (GAC). Reynolds is no longer with GAC but is now employed by Tapco International, Houston, Texas.

Fig. 64. Distribution of Rare-Earth Fission Products in a Triso-Coated Sol-Gel UO_2 Particle Irradiated in Rod 7 of H₂B-8 at an Average Irradiation Temperature of 1280°C and an Average Temperature Gradient of 910°C/cm. The hot side of the particle is at the bottom. Note the retention of rare-earth fission products within the kernel.

Fig. 65. Distribution of Rare-Earth Fission Products in a Triso-Coated WAR 85% UO_2 -15% UC_2 Particle Irradiated in Rod 13 of HRB-8 at an Average Temperature of 1240°C and an Average Temperature Gradient of 680°C/cm . The hot side of the particle is at the top. Note the relatively good retention of rare-earth fission products within the kernel, as compared to the WAR UC_2 particle shown in Fig. 62.

Optical Micrograph

U M

Zr M

Cs M

Cd M

Fig. 66. Distribution of Zr, U, Cd, and Cs in a Triso-Coated Dense UC_2 -1% ThC_2 Particle Irradiated in Rod 17 of HRB-8 at an Average Temperature of $1225^\circ C$ and an Average Temperature Gradient of $600^\circ C/cm$. The hot side of the particle is on the top. ZrC was substituted for SiC in this particle. Note the good appearance of the ZrC coating and the high concentration of fission products outside the kernel.

Fig. 67. Distribution of U, Cr, Cs, and Ce in a Triso-Coated Dense UC_2 -1% ThC_2 Particle Irradiated in Rod 17 of HRB-7 at an Average Temperature of $1225^\circ C$ and an Average Temperature Gradient of $600^\circ C/cm$. The hot side of the particle is on the top. ZrC substituted for SiC. Note the good appearance of ZrC and the high concentration of rare-earth fission products outside the kernel.

SUMMARY AND CONCLUSIONS

The HRB-7 and -8 capsules were the first HRB capsules to undergo a preirradiation in the PB-13 facility, and they were the first capsules to employ certain capsule design changes that have been used in all subsequent HRB capsules. The experiment was generally successful, and all objectives were accomplished. There were, however, some areas in the experiment which were less than satisfactory:

1. Because there was, from the beginning of the experiment, a very high fission gas release from the fuel specimens, it was not possible to make fission gas release rate to birth rate (R/B) measurements. These measurements are of marginal value in this type of experiment, where there are so many different types of particles being irradiated that it is not possible to tell how any one type of particle is performing from R/B data. Inability to measure R/B is only one consequence of the high fission gas release rate. Another consequence is the difficulty imposed on temperature control of the capsule. Because the capsule cannot be swept with a helium-neon mixture, there is greater uncertainty about the composition of the gas in the various gaps within the capsule. The capsules can not be swept when the off-gas system cannot handle the large amount of activity coming from the sweep gas. Changes in gas composition within the capsule are made with great difficulty. The entire control area at HFIR must be cleared of personnel so the capsule can be swept. Even the reduced thermal flux characteristic of the PB-13 facility results in a severely overpowered fissile particle (compared to LHTR conditions) at the beginning of the experiment. Weak particles cannot withstand this power level, and they fail. For future tests of fully enriched, undiluted fissile particles, only very high-quality fuel, with a high probability of successful operation, can be considered. This precludes HRB tests for screening candidate fuels.²⁴
2. The performance of the tungsten-rhenium centerline thermocouples was disappointing.²⁵ In HRB-7 the thermocouple decalibrated over 300°C and failed after 2200 hr of operation, about half way through the test. In HRB-8 the thermocouple decalibrated over 500°C but survived the entire irradiation. The decalibration of each thermocouple was measured by removing the capsule from the RB facility periodically,

²⁴Screening tests with low-enriched fissile fuel can still be done in HRB capsules, but the fission product yields will be characteristic of plutonium fuels. Hopefully, the proven success to date of the WAR fissile fuels will preclude the need for future screening tests.

²⁵R. L. Shepard and B. J. Montgomery, "Thermometry in HRB Capsules," *GCR Program HTGR Base-Technology Program Progress Report January 1, 1974-June 30, 1975*, ORNL-5108, pp. 418-25.

during a reactor shutdown for refueling. An in-pool thermocouple calibration furnace, designed specifically for use with HRB capsules, was used for the measurement.²⁶

3. The decision not to rotate the capsule with respect to the HFIR axial centerline turned out to be unwise. Because of the radial fast flux gradient across the RB positions it is not possible to get good flux monitor data when the capsules are rotated. However, if the capsules are not rotated, this same flux gradient causes bowing of the graphite sleeve. In the case of HRB-7 and -8 this bowing made removal of the graphite sleeves from the stainless steel containment difficult, and removal of the fuel rods from the graphite sleeve was even more difficult. The sleeve had to be split open, which spilled the flux monitors out on the hot cell floor; therefore, no flux monitor data were obtained from these capsules.

Several conclusions related to the performance of the fuel in these capsules were made:

1. There was extensive thermal migration of the kernels in the UO_2 and mixed-oxide particles. There was some reduction in the amount of migration by diluting the fissile kernel with thorium, but even at a Th/U ratio of 8:1, the migration rate was higher than desirable. Further dilution is not possible, because 8:1 is the overall Th/U ratio on recycle fuel elements. This observation supported the decision made in early 1974 to switch from the mixed-oxide kernel to the WAR-derived oxide-carbide kernel for the reference recycle fissile particle.
2. There was extensive graphitization of the inner LTI coating and some attack of the SiC layer in the Triso particles containing VSM UC_2 kernels. This same behavior was observed in the Triso particles containing WAR derived UC_2 . In addition, failures of the SiC layer were observed on the cold side of the particle for both of these fuel types.
3. The appearance of the Biso particles was considerably better than that of the Triso particles containing the same type of WAR-derived kernels. This difference is attributed to the accumulation of fission products at the inner LTI-SiC interface on the cold side of the Triso particles, causing graphitization of the inner LTI, and sometimes attack of the SiC. In the Biso particles, the fission products migrated through the coatings without attacking them. The higher fission product release from the Biso fuel is evident in the gamma scan of the graphite sleeve from HRB-8, shown in Fig. 33.
4. The sol-gel-derived $UO_2 + 7\% U_2N_3$ and $UO_2 + 1\% UC_2$ performed very badly, and will not be considered further. While these kernels did not exhibit amoeba *per se*, there was a multidirectional movement of the kernels through the coatings;

²⁶B. H. Montgomery, J M Robbins, and R. L. Hamner, "Irradiation Tests in HFIR Removable Beryllium Facility," *GCR Programs Annu. Prog. Rep.* Dec. 31, 1973, ORNL-4975, pp. 89-127.

in some severe cases, these kernels attacked the outer coatings of adjacent particles. The failed particles appeared to have converted to carbide. The kernels of unfailed particles had the appearance of UO_2 .

5. The VAR-derived $^{235}\text{U}_{0.39}\text{O}_{1.72}$ (15% converted from oxide to carbide) generally performed well. There was some slight attack of the SiC layer in the Triso particle, but no graphitization of the inner LTI. There was apparent phase segregation within the kernel for both the Biso and Triso particles. The performance of this fuel, high in oxygen content relative to the 1974 reference fissile particle (75% converted WAR, was very encouraging, since, from a processing standpoint, minimum conversion of UO_2 to UC_2 is preferred.
6. The performance of the GAC-prepared Triso particles, containing ZrC coatings instead of SiC, was very favorable. The results of this experiment led to a decision to continue the development of ZrC coatings as an advanced HTGR fuel concept. There is some evidence that ZrC may not be as effective in retaining fission products as SiC. Microprobe analysis of the ZrC-coated particles showed rare-earth fission products on the outside of the ZrC coating, whereas all cesium was retained within the coating. Further, there was no graphitization of the inner LTI coating by fission products in the particles with ZrC coatings. When SiC coatings are used in the Triso design there is usually severe graphitization of the inner LTI in the regions associated with rare-earth fission product accumulation at the inner LTI-SiC interface.
7. The performance of $^{235}\text{UO}_2$ and $^{233}\text{UO}_2$ was compared. The appearance of the two kernels, both in Triso particles, was identical. There was somewhat more migration in the ^{235}U -containing particle, but this was probably because of differences both in temperature gradient during operation, rather than a difference in the performance of the two isotopes. Previous experiments indicate that ^{235}U and ^{233}U behave identically under irradiation,²⁷ and this conclusion is not challenged by the results of this experiment.
8. The performances of $(\text{Th}_{0.8}\text{U}_{0.2})\text{O}_2$ fabricated in laboratory-scale and production-scale equipment were compared. The rods fabricated in production-scale equipment and irradiated at the ends of the capsules performed well and showed less migration than the rod fabricated in laboratory-scale equipment and irradiated in the center of the capsules. The amount of migration is probably not due to the fabrication equipment used, but more likely is due to the temperature and temperature gradient characteristics used during the test. These characteristics differ between the ends of the capsule and the center. The gamma scan of the graphite sleeve after irradiation of HRB-8 showed more fission product activity adjacent to fuel rod 2 (fabricated in production-scale equipment) than in fuel rod 11 (fabricated in laboratory-scale equipment). This

²⁷F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-8*, ORNL/TM-5011 (December 1975).

indicated better fission product retention capability for the Biso coating deposited in the laboratory coater. This observation is reinforced by the fact that the burnup was higher in the center of the capsule than on the ends.

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APPENDIX A - Thermal Analysis

The calculated specimen fission heat rates and operating temperatures have already been presented in the body of this report. This Appendix will present some of the backup information that was used in making these calculations. As indicated previously, HRB capsules contain a number of Chromel vs Alumel thermocouples in the graphite sleeve. The thermocouple at one selected axial location is used to control the temperature of the capsule. The sweep gas mixture (helium and neon) is adjusted to give the desired graphite temperature at that location. For HRB-7 and -8 the location adjacent to specimen 10 was selected. The graphite midwall temperature was maintained at about 850°C for HRB-7 and at about 780°C for HRB-8.

The mathematical models used to make the temperature calculations have been referenced in the body of the report. The materials property data and calculational techniques are described in those reports. Some of the geometric data needed for the calculations, and other data peculiar to capsules HRB-7 and -8, are described in this appendix.

The temperature calculation begins with the coolant water. The axial coolant water temperature profiles in both R3 and PB facilities is given in Fig. A1. Another vital piece of data needed in the calculation is the dimensional behavior of the graphite sleeve under irradiation. Poco graphite swells slightly during irradiation at these temperatures, and the final dimensions of the sleeve are represented in Figs. A2 through A4. For the temperature calculations it was assumed that the swelling was a linear function of fast fluence at each axial location. The assumed dimensional behavior of the fuel rods is shown in Fig. A5. From this and other experiments,¹⁻⁷ it is known that the dimensional change relationship

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 HRB-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 HRB-3 and ETR Experiment P13N*, ORNL/TM-4526 (October 1974).

⁴F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiments HRB-4 and -5*, ORNL-5115 (June 1976).

⁵F. J. Homan et al., *Irradiation Performance of HTGR Fuel Rods in HFIR Experiment HRB-6*, ORNL/TM-5011 (December 1975).

⁶E. L. Long et al., *Fabrication of ORNL Fuel Irradiated in the Peach Bottom Reactor and Postirradiation Examination of Recycle Test Elements 7 and 4*, ORNL/TM-4471 (September 1974).

⁷R. B. Fitts et al., "Summary of Fuel Rod Dimensional Changes Under Irradiation," *Gas-Cooled Reactor Programs Ann. Prog. Rep. Dec. 31, 1972*, ORNL-4911, p. 142.

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Fig. A1. Coolant Temperature vs Axial Location for RB and PB Facilities in HFIR.

Fig. A2. Diametral Change vs Fast Fluence for Graphite Sleeves in HRB-7 and -8.

Fig. A3. Outer Sleeve Diameters vs Fast Fluence for HRB-7 and -8.

Fig. A4. Inside Sleeve Diameters vs Fast Fluence for HRB-7 and -8.

Fig. A5. Fuel Rod Dimensional Change vs Fast Fluence for HRB-7 and -8.

of fuel rods with fast fluence is parabolic, with a relatively rapid shrinkage initially, followed by a leveling off of the shrinkage rate, and then swelling of the rods beginning at a fast fluence level of $5-7 \times 10^{21}$ neutrons/cm² ($E > 0.18$ MeV). The HRB-7 and -8 data were fit to the parabolas shown in Fig. A5.

The calculational techniques employed in the thermal analysis of HRB capsules are described in detail in refs. 8 and 9. Briefly, there are three pieces of vital information, and by knowing any two of these, the third can be computed. The three pieces are: (1) graphite midwall temperature, (2) fuel fission heat rate, and (3) the composition of the sweep gas. The graphite midwall temperature is measured at selected axial locations throughout the experiment. Periodically in each cycle the sweep gas composition is switched to pure helium from mixed He-Ne. The composition of the sweep gas is measured with flow meters, but our experience has been that the flow meter data are unreliable; therefore, these data are used only as a guide in the calculation. Knowing the graphite midwall temperatures during the periods of operation with 100% helium sweep gas permits a determination of the fission heat rate vs time curves. Now it is possible to calculate the sweep gas compositions for all times during the irradiation, and also calculate the fuel surface and center temperatures.

Without accurate measurements of the centerline temperatures, it is not possible to establish the precision of these calculations. However, since each fuel specimen located adjacent to a graphite midwall thermocouple is treated using this procedure, it is possible to compare the calculated gas composition profiles and to use this as a rough indicator of the precision of the calculation. Presumably, the gas composition should be constant for all axial positions, although some minor local variations are probably possible, especially in a sealed capsule (as was necessary for HRB-7 and -8 due to the high concentration of fission gas). The calculated sweep gas composition profiles are shown in Fig. A6, and examination of these profiles shows that they are in very good agreement.

⁸F. J. Homan, *Thermal Analysis of HTGR Fuel Rods Irradiated in the HFIR Removable Beryllium Facility* (in preparation).

⁹K. H. Valentine, *ECNTRK, A Computer Program to Model Heat Transfer Across Asymmetric Gaps in Cylindrical Fuel Rods* (in preparation).

Fig. A6. Sweep gas Composition Profile for a Specific Specimen: (a) Rod 1 in HRB

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HRB-7, (b) Rod 3 in HRB-7, (c) Rod 6 in HRB-7, (d) Rod 10 in HRB-7, (e) Rod 14 in HRB-7, (f) Rod 17 in HRB-7

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ORNL-DWG 75-14628

(c)

ORNL-DWG 75-14630

ORNL-DWG 75-14631

(f)

6 in HRB-7, (d) Rod 10 in HRB-7, (e) Rod 14 in HRB-7, (f) Rod 17 in HRB-7.

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Fig. A6. (Continued) (g) Rod 1 in HRB-8, (h) Rod 3 in HRB-8,

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200 220
RTSI

200 220
RTSI

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HRB-8, (i) Rod 6 in HRB-8, (j) Rod 10 in HRB-8, (k) Rod 14 in HRB-8, (l) Rod 17 in HRB-8.

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in HRB-8, (k) Rod 14 in HRB-8, (l) Rod 17 in HRB-8.

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