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IRRADIATION PERFORMANCE IN HFIR EXPERIMENT HRB-2 OF HTGR FUEL STICKS BONDED WITH REFERENCE AND ADVANCED MATRIX MATERIALS

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OAK RIDGE NATIONAL LABORATORY
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METALS AND CERAMICS DIVISION
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J. H. Coobs, J. L. Scott, B. H. Montgomery,[1]
J M Robbins, C. B. Pollock, and J. A. Conlin[1]

ABSTRACT

Fuel elements for HTGR's may contain carbide or oxide fuel particles that are coated with various combinations of pyrolytic carbon and SiC. The blended particles can be fabricated into fuel element segments, or "fuel sticks," by several techniques such as intrusion bonding or compacting. In the second long-term irradiation experiment in the HFIR reflector facilities, we tested fuel sticks prepared by intrusion bonding with the reference Fort St. Vrain matrix and with various other matrix filler materials. This instrumented and swept experiment, HRB-2, was irradiated for 257 days (11 cycles) at temperatures of 1100 to 1300°C to test the integrity and behavior of the coated particles and fuel sticks at high burnup (12% FIMA in fertile ThC_2 and ThO_2 particles and 24% FIMA in mixed carbide particles) and the design maximum HTGR fast-neutron fluence of about 8×10^{21} neutrons/cm^2 (>0.18 MeV). The fission-gas release rates were steady at acceptable levels (R/B approx 10^{-5}). After irradiation, the 14 short (0.7-in.-long) fuel sticks were intact although six of them showed slight debonding at the edges. Both 2-in.-long fuel sticks broke into three segments, probably due to bowing of the graphite sleeve, but they were otherwise in good condition. Polished sections confirmed the good performance of the matrix structure and of the TRISO- and BISO-coated particles. Only two failed particles were detected, and these were BISO-coated resin-derived particles observed in a specimen that received maximum exposure. These failures were attributable to interactions with the shrinking matrix. Linear shrinkage of the six fuel sticks that contained TRISO-coated particles were 1.0 to 1.6%, or well within design limits, while shrinkage of the specimens that contained BISO-coated particles was much greater (3.0 to 4.5%), as expected. Results from this experiment demonstrate that HTGR fuel sticks bonded with reference and alternate matrix materials will survive to the maximum design exposure.

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[1]Reactor Division.

INTRODUCTION

The basic fuel materials for the HTGR are coated particles of uranium carbide or oxide, thorium carbide or oxide, or a mixture of uranium and thorium as carbide or oxide. The coatings consist of two or more layers of pyrolytic carbon and sometimes an additional layer of silicon carbide that is used to improve the retention of fission product nuclides within the fuel. The coated particles plus binder and carbonaceous filler are formed into cylindrical fuel sticks carbonized and inserted into holes in large prismatic graphite fuel elements. The fuel elements, which also contain coolant passages, are stacked in a close-packed array to make up the reactor core.[2]

During service in the HTGR environment, the components of the fuel element — the graphite block, the pyrolytic carbon coatings on the particles, and the carbonaceous bonding matrix — undergo dimensional changes induced by fast-neutron damage. These changes are complex functions of density, preferred orientation, crystallite size of the carbon structure, restraint, fast-neutron fluence and flux, and temperature.[3] Interactions between the components could produce stresses that cause rupture of particle coatings or debonding of the fuel stick. Irradiation testing of composite structures representative of the fuel element is guiding the development of bonding materials and techniques just as such testing guided and assisted the recent development of coated particle technology.[4,5]

Several recent irradiation experiments have demonstrated that fuel sticks bonded with selected carbonaceous matrix materials will survive and perform well at exposures representing about 70% of the maximum

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[2]R. C. Dahlberg, R. F. Turner, and W. V. Goeddel, "Fort St. Vrain Nuclear Power Station; Core Design Characteristics," *Nucl. Eng. Int.* 14(n163): 1073–77 (1969).

[3]J. C. Bokros, G. L. Guthrie, R. W. Dunlap, and A. S. Schwartz, "Radiation-Induced Dimensional Changes and Creep in Carbonaceous Materials," *J. Nucl. Mater.* 31: 25 (1969).

[4]T. G. Godfrey et al., *Coated Particle Fuels*, USAEC Report ORNL-4324, Oak Ridge National Laboratory, November 1968.

[5]C. S. Luby and D. P. Harmon, "HTGR Coated Particle Fuel Performance Under High Irradiation Exposures," *Trans. Amer. Nucl. Soc.* 13: 135 (1970).

fast-neutron fluence of the HTGR (maximum approx 8×10^{21} neutrons/cm^2). For instance, in the HRB-1 experiment,[6] fuel sticks bonded with prepolymerized furfuryl alcohol containing large amounts of Poco graphite flour and carbon black performed well at fluences up to 5.6×10^{21} neutrons/cm^2 (>0.18 MeV). In other experiments,[7] fuel stick specimens bonded with coal tar pitch containing 30 wt % natural flake graphite as filler behaved well at similar exposures, but gave marginal performance (i.e., moderate cracking, swelling, and debonding) at greater exposures ($\geq 7 \times 10^{21}$ neutrons/cm^2) that approximated the full HTGR fluence.[8] These results were complicated, however, by failure of a significant number of particle coatings in the high-fluence specimens. On the other hand, excellent performance at high fluences ($>7 \times 10^{21}$ neutrons/cm^2) was demonstrated on specimens consisting of coated inert particles bonded with pitch containing at least 40 wt % Poco graphite flour as filler.[9]

Because the filler materials used in the more successful specimens were carefully selected specialty items, we conducted fabrication experiments on bonding of fuel sticks with matrices consisting of a variety of readily available and inexpensive filler materials in 15V pitch.[10] The second experiment, HRB-2, in the removable beryllium facility of HFIR was designed and constructed with the objective of testing a series of fuel stick specimens bonded with these matrices. Another objective of this experiment was to determine the integrity, at 1100°C and a fluence of 8×10^{21} neutrons/cm^2, of preproduction Fort St. Vrain fuel rod specimens

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[6]J. L. Scott et al., *An Irradiation Test of Bonded HTGR Coated Particle Fuels in an Instrumented Capsule in HFIR*, USAEC Report ORNL-TM-3640, Oak Ridge National Laboratory, March 1972.

[7]C. S. Luby, D. P. Harmon, and W. V. Goeddel, *HTGR Fuel Design and Irradiation Performance*, USAEC Report GA-10468, Gulf General Atomic, January 1971.

[8]*PSC High-Temperature Gas-Cooled Reactor Quart. Progr. Rep. Sept. 30, 1970*, USAEC Report GA-10313, pp. 160–91, Gulf General Atomic.

[9]J. L. Scott et al., "Development of Bonded Coated-Particle Beds for HTGR Fuel Elements, pp. 456–73 in *Proceedings of Gas-Cooled Reactor Information Meeting at Oak Ridge National Laboratory, April 27–30, 1970*, USAEC Report CONF-700401, USAEC Division of Technical Information.

[10]Grade 15V coal tar pitch from Allied Chemical Company.

and the performance of early-production TRISO-coated[11] fissile and fertile particles in these specimens. The specimens of Fort St. Vrain fuel were prepared for this experiment by Gulf General Atomic (GGA).

DESCRIPTION OF CAPSULE AND SPECIMENS

The design of instrumented capsule HRB-2 was the same as that of the first capsule, HRB-1,[6] with the exception of the test fuel and the graphite fuel support sleeve. The capsule, shown in Fig. 1, was a double-walled water-cooled stainless steel vessel having a 1.292-in. OD and a 0.967-in. ID. The test fuel specimens consisted of a series of 0.412-in.-diam bonded fuel sticks having a total stack length of 15 in. The specimens were supported in a one-piece sleeve made of Poco graphite, grade AXF-5Q. Design test conditions (1180°C peak fuel temperature and 940°C in the graphite sleeve at 3.3 kW/ft fission heat rate) were intended to match conditions typical of an HTGR.

Fuel temperatures were monitored by nine sheathed 0.062-in.-diam Chromel-P—Alumel thermocouples. The stainless steel thermocouple sheaths were coated with a 0.005-in.-thick protective barrier of chemically vapor-deposited tungsten. The thermocouples were located in axial holes in the graphite sleeve adjacent to fuel specimens as shown in Fig. 1. Three titanium flux monitor wires, each enclosed in a platinum tube, were located in the graphite sleeve in similar axial holes. The graphite sleeve and the fuel specimens were continuously swept with a 3600 cm^3/hr high-purity helium-neon gas mixture at 1- to 2-atm pressure. Gas samples of the sweep effluent were taken periodically for fission-gas release determination.

The specifications for the fueled specimens are given in Table 1. Included with the table is a drawing showing the relative length and position of the various specimens. A set of ten fuel stick specimens made at ORNL all contained three types of coated particles and were bonded with various filler materials in 15V pitch. The three types of particles

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[11]Four-layer coating consisting of an inner buffer layer and a SiC layer sandwiched between two outer layers of pyrolytic carbon.

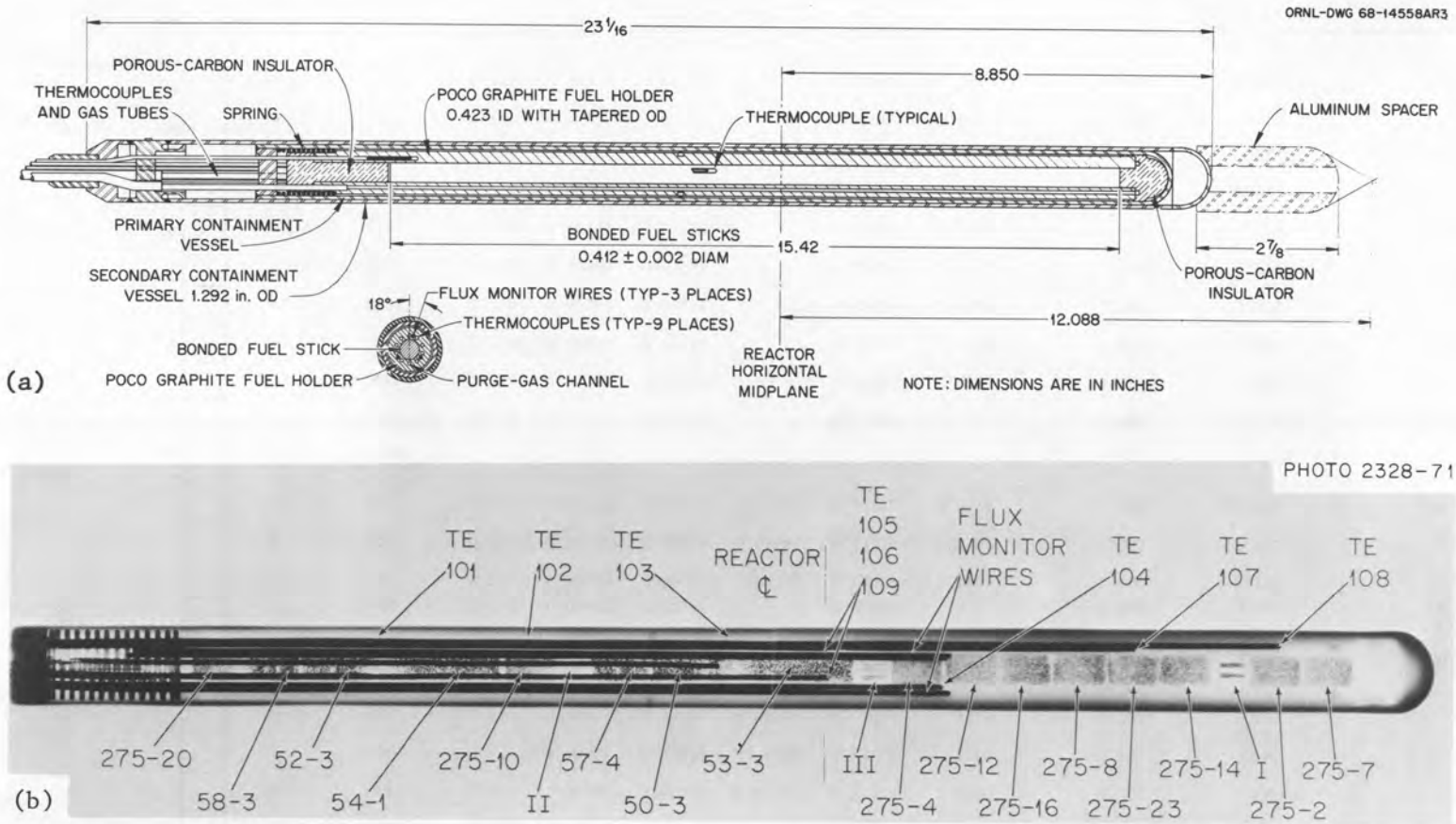
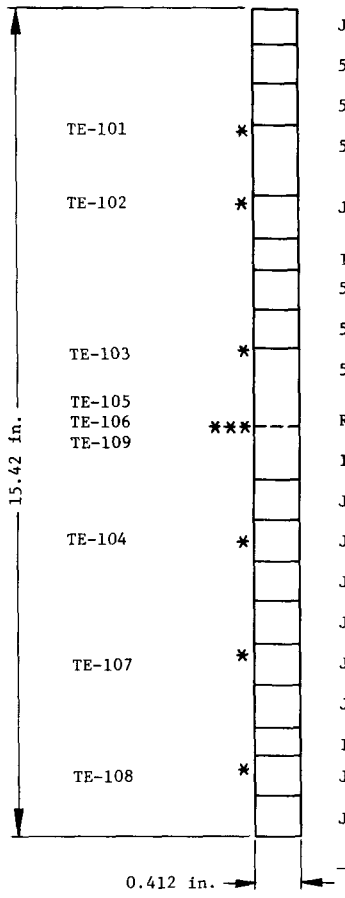


Fig. 1. HTGR Instrumented Capsule HRB-2 for Irradiation in HFIR Removable Beryllium Facility. (a) Schematic drawing of capsule; (b) radiograph of capsule after assembly.

Table 1. Fuel Loading Data for Capsule HRB-2



Thermocouple Number	Axial Position	Specimen Number	Fabricator	Length (in.)	Bonded Bed Loading: Particles, g/cm³: Fissile ^{235}U	Fissile ^{233}U	Fissile ^{232}Th	Fertile ^{233}Th	Inert Material (g)	Average[a] Flux, neutrons cm⁻² sec⁻¹: Thermal (<0.414 eV)	Fast (>0.183 MeV)	Average Gamma[b] Plus Fission Heat (W/g)
										× 10^{15}	× 10^{14}	
		JF275-20	ORNL	0.705	0.00370	0.03292		0.10992	1.9792	0.385	1.850	9.00
		58-3	GGA	0.77	0.00380	0.00029	0.01755	0.13029	2.1720	0.465	2.050	9.75
		52-3	GGA	0.77	0.00368	0.00029	0.01680	0.13116	2.2592	0.560	2.250	10.70
TE-101	*	54-1	GGA	1.95	0.00378	0.00028	0.01737	0.12976	5.9578	0.720	2.710	12.10
TE-102	*	JF275-10	ORNL	0.7145	0.00365	0.03250		0.10853	1.9355	0.866	3.100	13.30
		II[c]	ORNL	0.450	0.00023	0.00741		0.00810		0.946	3.240	13.85
		57-4	GGA	0.77	0.00380	0.00029	0.01755	0.13029	2.1720	1.000	3.333	14.20
		50-3	GGA	0.77	0.00368	0.00029	0.01680	0.13116	2.2592	1.063	3.440	14.50
TE-103	*	53-3	GGA	1.95	0.00378	0.00028	0.01737	0.12976	5.9578	1.155	3.575	14.70
TE-105 TE-106 TE-109	***	Reactor Midplane[d]										
		III[c]	ORNL-GGA	0.450		0.00440		0.05393		1.185	3.575	14.55
		JF275-4	ORNL	0.710	0.00366	0.03258		0.10880	2.0765	1.185	3.530	14.35
TE-104	*	JF275-12	ORNL	0.7101	0.00369	0.03283		0.10963	1.8816	1.170	3.440	14.00
		JF275-16	ORNL	0.7107	0.00368	0.03271		0.10922	2.0310	1.145	3.333	13.65
		JF275-8	ORNL	0.7105	0.00366	0.03261		0.10888	1.9761	1.105	3.210	13.20
TE-107	*	JF275-23	ORNL	0.7090	0.00367	0.03263		0.10895	1.9395	1.055	3.050	12.80
		JF275-14	ORNL	0.7109	0.00368	0.03278		0.10945	2.0078	0.998	2.875	12.30
		I[c]	ORNL-GGA	0.450	0.00011	0.00023		0.07222		0.930	2.650	11.75
TE-108	*	JF275-2	ORNL	0.7025	0.00372	0.03312		0.11060	2.0640	0.872	2.470	11.30
		JF275-7	ORNL	0.7058	0.00370	0.03293		0.10998	1.9664	0.780	2.225	10.50

[a]Average flux over reactor cycle based on latest evaluation of flux monitors from previous experiments.

[b]Gamma heat as measured in graphite.

[c]Graphite holder with fissile and fertile loose particles.

[d]Three thermocouples at ≈ 120-deg radial orientation at the midplane.

have kernels that are: (1) 10.16% enriched uranium as UOS in a carbonized strong-acid resin, (2) sol-gel-derived ThO_2 particles, and (3) inert, carbonized resin particles. These particles are described in Table 2.

Table 2. Characteristics of BISO-Coated Particles Used in ORNL Bonded Beds for Experiment HRB-2

Particle Batch Number	OR-1295	OR-1290	OR-1331
Kernel Material	UOS	ThO_2	Carbon
Uranium Content, wt %	9.81		
^{235}U Enrichment, at. %	10.16		
Thorium Content, wt %		34.1	
Average Particle Dimensions, μm			
Total diameter	462	435	675
Kernel diameter	252	201	437
Outer-coating thickness	74	61	76
Total-coating thickness	105	117	119
Density[a] of Coated Particle, g/cm^3	1.89	2.48	1.50
Density[b] of Outer Coating, g/cm^3	1.925	1.910	1.87
Density[c] of Buffer Coating, g/cm^3	0.9	0.9	0.9

[a]Measured by mercury pycnometer.

[b]Measured by density gradient column.

[c]Estimated from deposition conditions.

In addition to the ten fuel stick specimens, six fuel rods prepared at GGA and three magazines containing samples of loose coated particles were included. The six GGA fuel rods include two 2-in.-long rods that contain preproduction Fort St. Vrain fissile and fertile particles that were bonded with the reference matrix material, 27 wt % N.F. 6353 graphite in 15V pitch. The bonded specimens are described in more detail in Table 3.

Table 3. Characteristics of Bonded Fuel Sticks for Experiment HRB-2

Specimen Designation	Filler Material[a]		Particle Packing Fraction (%)	Matrix Density (g/cm^3)
	Type	Amount (wt % in binder)		
JF275-20	Asbury	35	63.9	0.76
GGA58-3	N.F. 6353	27	59.3	0.60
GGA52-3	N.F. 6353	27	58.6	0.63
GGA54-1	N.F. 6353	27	60.8	0.59
JF275-10	Robinson	40	63.1	0.65
GGA57-4	N.F. 6353	27	58.9	0.57
GGA50-3	N.F. 6353	27	58.8	0.63
GGA53-3	N.F. 6353	27	60.6	0.58
JF275-4	Thermax	50	63.2	0.90
JF275-12	Robinson	40	59.7	0.67
JF275-16	JOZ	40	63.5	0.83
JF275-8	Santa Maria	40	63.3	0.73
JF275-23	Poco AXZ	35	63.3	0.67
JF275-14	JOZ	40	63.6	0.80
JF275-2	Thermax	50	64.3	0.92
JF275-7	Santa Maria	40	63.9	0.73

[a]Mixed with 15V pitch as binder material.

The fuel mixtures were designed to give a uniform axial ^{235}U loading of 0.0037 g/cm^3, which would provide an initial fission power generation of 3.3 kW/ft at the reactor midplane. The heat rate is reduced at either end of the fuel stack in direct proportion to the local neutron flux. Because of the high thermal flux in the HFIR-RB facility (approximately 1.5×10^{15} neutrons/cm^2) the ^{235}U is consumed rapidly (50% in about 16 days at the reactor midplane). To provide for continuing fission power, the fuel specimens were also loaded with ^{232}Th and ^{238}U such that the fission power from the ^{233}U and ^{239}Pu produced would match that of the initial ^{235}U loading. However, this is not quite possible in practice. There is an initial drop in power, as the ^{235}U is consumed, followed by a gradual buildup that varies with time, axial position, and fertile material (^{232}Th or ^{238}U) to a power approximating that produced by the

initial fissile loading. This is shown graphically in Fig. 2, which presents the calculated fission power generated by two specimens (JF275-4 and GGA specimen 53-3) located near the reactor midplane. The linear fission-heat rate for the GGA specimen drops significantly more than the ORNL specimen during the first cycle. This is a consequence of the rapid burnout of the initial loading of ^{235}U and the fact that additional fission power in the GGA specimen depended primarily on the $^{232}Th \rightarrow {}^{233}U$ chain with its longer characteristic conversion time than the $^{238}U \rightarrow {}^{239}Pu$ chain of the ORNL specimen.

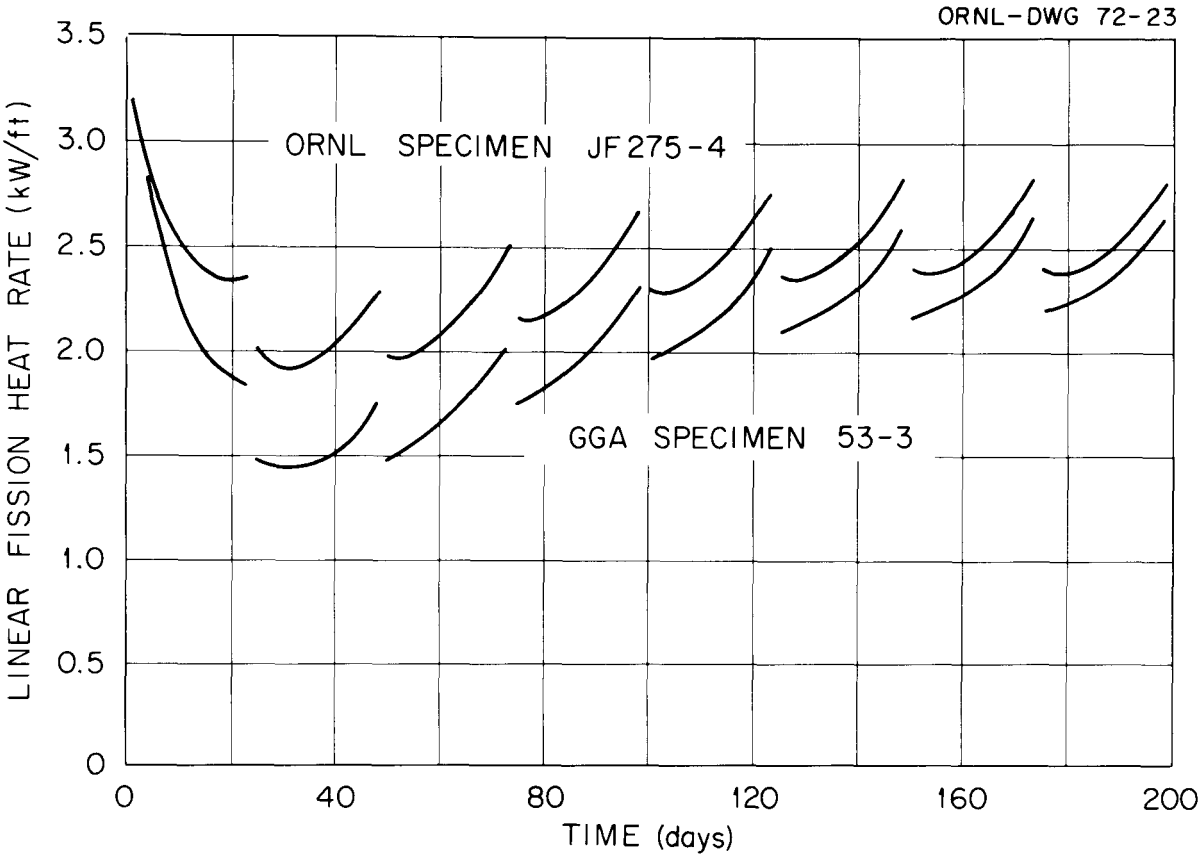


Fig. 2. Calculated Fission-Heat Rate for Two Specimens in Capsule HRB-2.

The capsule was designed to obtain reasonably uniform axial temperatures by tapering the graphite sleeve so as to increase the gas gap at either end, thus varying the thermal resistance of the gas gap between

the graphite sleeve and the water-cooled capsule inversely with the calculated initial axial power. To compensate for overall power variations with time, the composition of the helium-neon sweep gas mixture, which occupies the gap, was varied to maintain the peak fuel temperature at the design level.

OPERATING DATA

Capsule HRB-2 was inserted in the removable beryllium facility of HFIR on December 17, 1970, and was irradiated for eleven fuel cycles (256.5 days at 100 MW reactor power). The irradiation was completed and the capsule removed as scheduled on September 14, 1971. Capsule operation was stable throughout the irradiation, and all test parameters were within design limits. The calculated fuel burnup and fluence for each specimen are shown in Table 4. The maximum heavy metal fissile particle burnup in the ORNL specimens (at the reactor midplane) was 31% and in the GGA specimen 23.6%. The maximum heavy metal burnup in the fertile particles for both the ORNL and GGA specimens was 12%.

Table 4. Calculated Fuel Burnup and Fast-Neutron Fluence for Bonded Specimens in Capsule HRB-2

Specimen Designation	Fuel Burnup (% FIMA)		Exposure (neutrons/cm^2)	
	Fissile	Fertile	Thermal (<0.414 eV)	Fast (>0.18 MeV)
			$\times 10^{21}$	$\times 10^{21}$
JF275-20	18.6	3.9	8.5	4.1
GGA58-3	13.2	5.0	10.3	4.5
GGA52-3	15.2	6.0	12.4	5.0
GGA54-1	18.2	7.9	16.0	6.0
JF275-10	28.0	9.3	19.2	6.9
GGA57-4	22.0	10.4	22.2	7.4
GGA50-3	22.7	11.0	23.6	7.6
GGA53-3	23.6	12.0	25.6	7.9
JF275-4	31.0	12.0	26.3	7.8
JF275-12	30.5	11.9	25.9	7.6
JF275-16	29.9	11.6	25.4	7.4
JF275-8	29.2	11.2	24.5	7.1
JF275-23	28.1	10.6	23.4	6.8
JF275-14	27.0	9.9	22.1	6.4
JF275-2	24.6	8.5	19.3	5.5
JF275-7	23.0	7.4	17.3	4.9

The helium-neon sweep gas effluent was sampled periodically to measure fission-gas release. The ratios of release rate to birth rate (R/B) of selected isotopes versus accumulated irradiation time are shown in Fig. 3. The birth rates used in this calculation take into account the depletion of ^{235}U and the fissions of bred-in ^{233}U and ^{239}Pu. We first calculated the daily change in isotopic composition of the fuel for each specimen and then the fission product yield from each fissionable isotope. As may be seen in Fig. 3, the fractional release rate remained moderate throughout the irradiation.

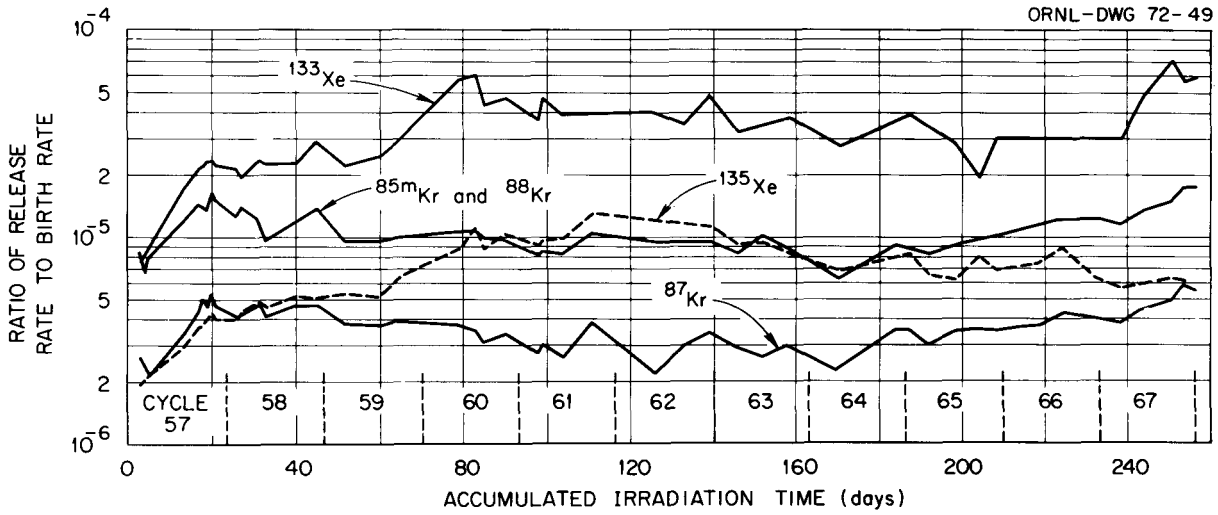


Fig. 3. Ratio of Fission-Gas Release Rate to Birth Rate Versus Irradiation Time in Capsule HRB-2.

Typical plots of the time-temperature history of the nine thermocouples for one cycle of operation (cycle 62) are given in Figs. 4 through 7. Operating temperature was controlled by maintaining TE-109 at 940°C through March 13, 1971. At that time the control temperature was reduced to 915°C in order to avoid temperatures higher than the design conditions. The data of Figs. 4 through 7 show an additional reduction in the control setting on May 3 as the temperature of TE-107 rose somewhat above that of the control thermocouple. The maximum graphite temperature of 966°C occurred during cycle 60 on TE-107, adjacent to fuel specimen JF275-23.

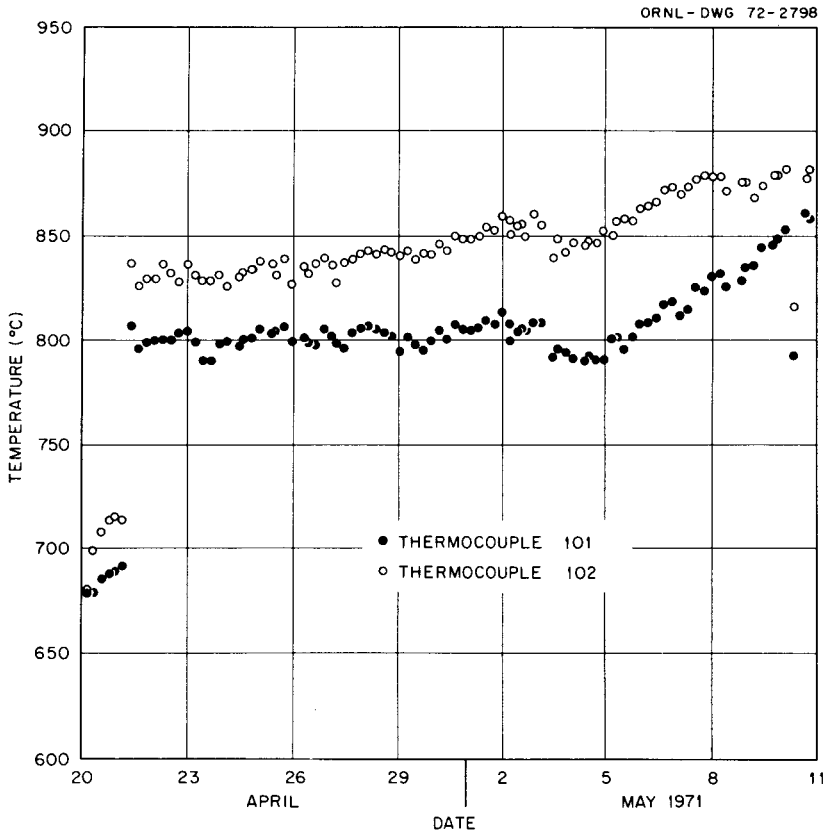


Fig. 4. Temperatures Recorded by Thermocouples 101 and 102 of Capsule HRB-2 During HFIR Cycle 62. This was about midway in the irradiation period (see Fig. 3).

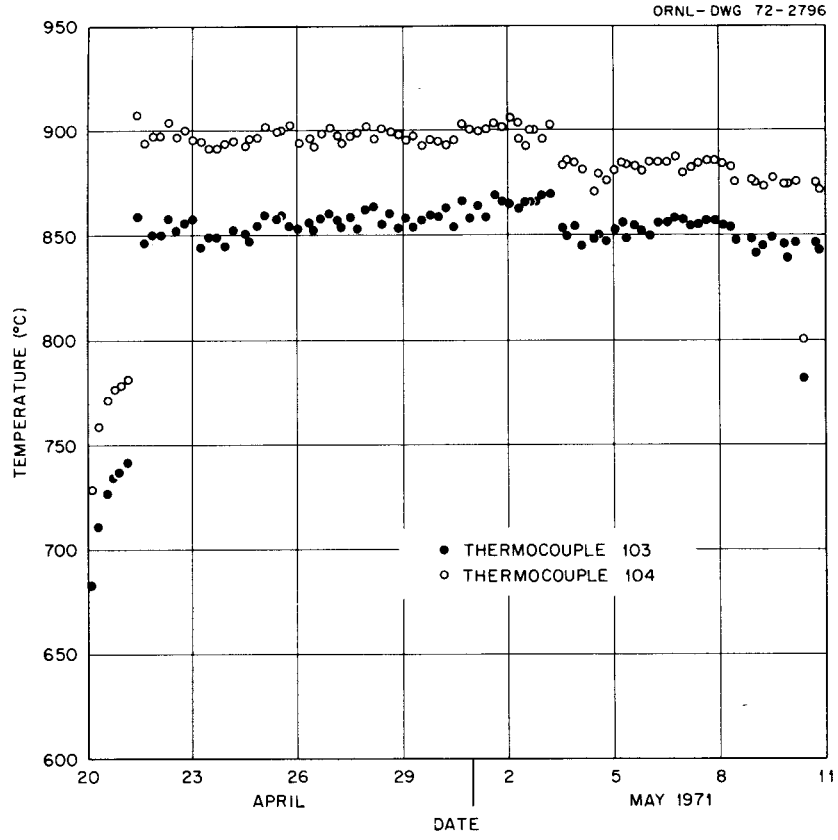


Fig. 5. Temperatures Recorded by Thermocouples 103 and 104 of HRB-2 During HFIR Cycle 62.

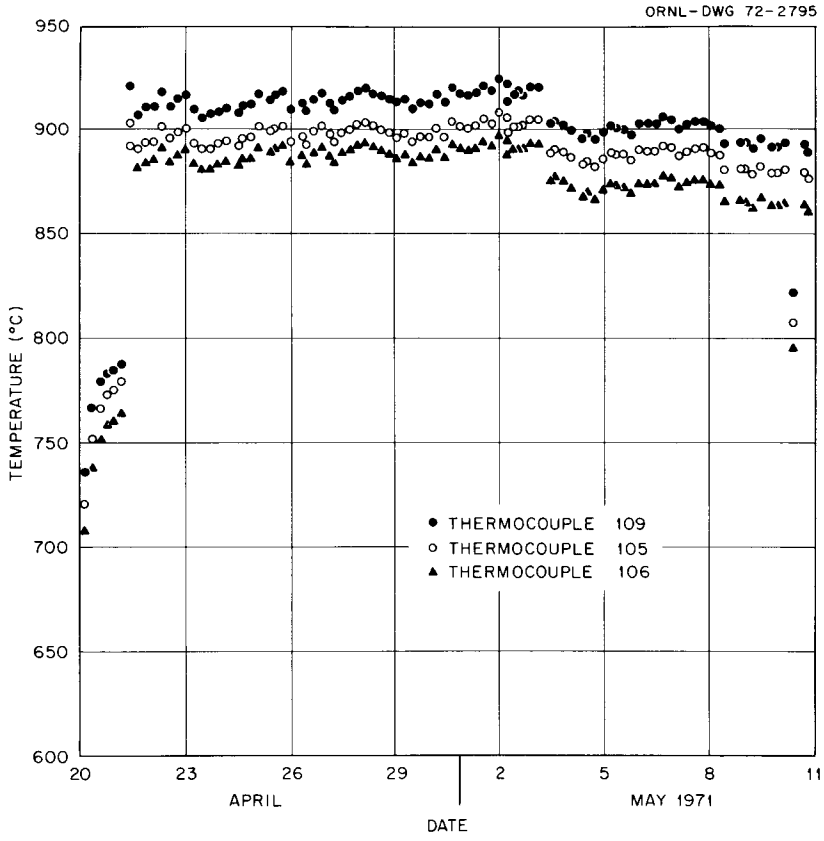


Fig. 6. Temperatures Recorded by Thermocouples 105, 106, and 109 of HRB-2 During HFIR Cycle 62.

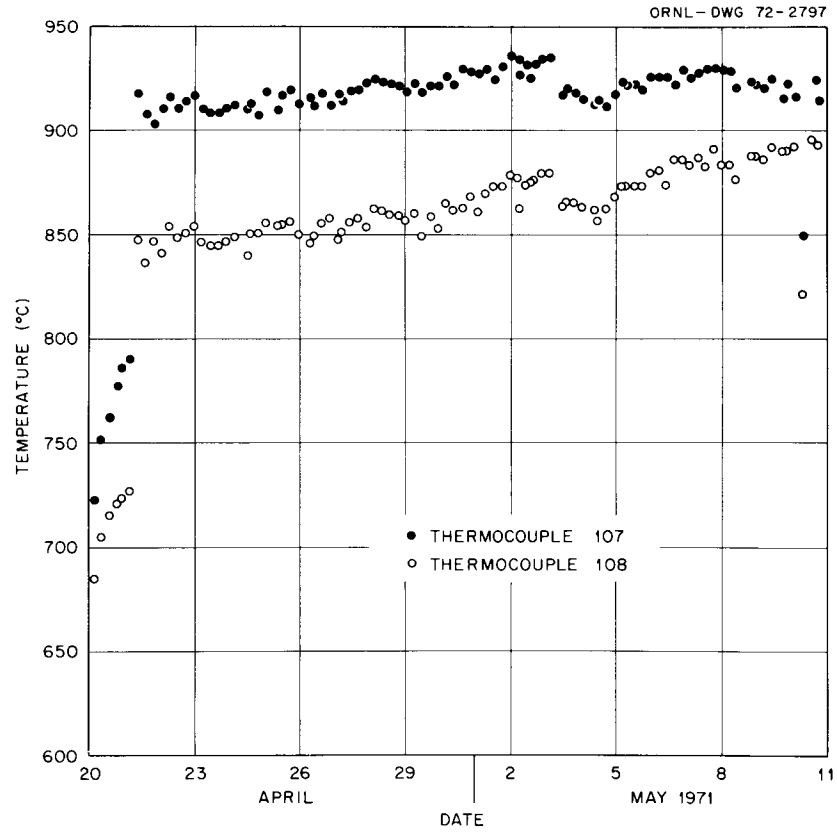


Fig. 7. Temperatures Recorded by Thermocouples 107 and 108 of HRB-2 During HFIR Cycle 62.

In Figs. 4 through 7 one observes that the temperatures near the midplane of the capsule (TE-103, -104, -105, -106, -109, and -107) remain reasonably constant whereas the temperatures at either end show significant changes (low temperatures at the start of a cycle which increase at the end) as illustrated by TE-101, -102, and -108. This is a consequence of several factors. The greatest part of the change is caused by the large increase in neutron flux at either end of the reactor core towards the end of each cycle. The capsule is located immediately outside the cylindrical control plates which surround the core. As the core burns out, the plates are pulled out — one set upward, the other downward — thus increasing the flux at either end of the core. Since the midplane temperatures are in general the highest, we adjust our sweep gas mixture to control midplane temperatures and accept the resulting temperature variation at the ends. Other factors also contribute to the changes, in particular the continuing variation with time and reactor cycle life of the local fissile atom content of a given fuel specimen. This fissile content is at a peak at the start of a core cycle due to the buildup of ^{238}Pu or ^{233}U which occurs during reactor shutdowns. This buildup is greatest at the midplane.

The time-temperature plots also show a short low-temperature period for all thermocouples at the beginning of each cycle. This was caused by capsule operation with a 100% He sweep, which reduces the thermal resistance of the gap between the graphite sleeve and the capsule wall and results in the lower temperatures. The capsule was operated with 100% He periodically to provide a relative measure of the power generation as a function of temperature without the complication of an uncertain gas conductivity.

RESULTS FROM POSTIRRADIATION EXAMINATION

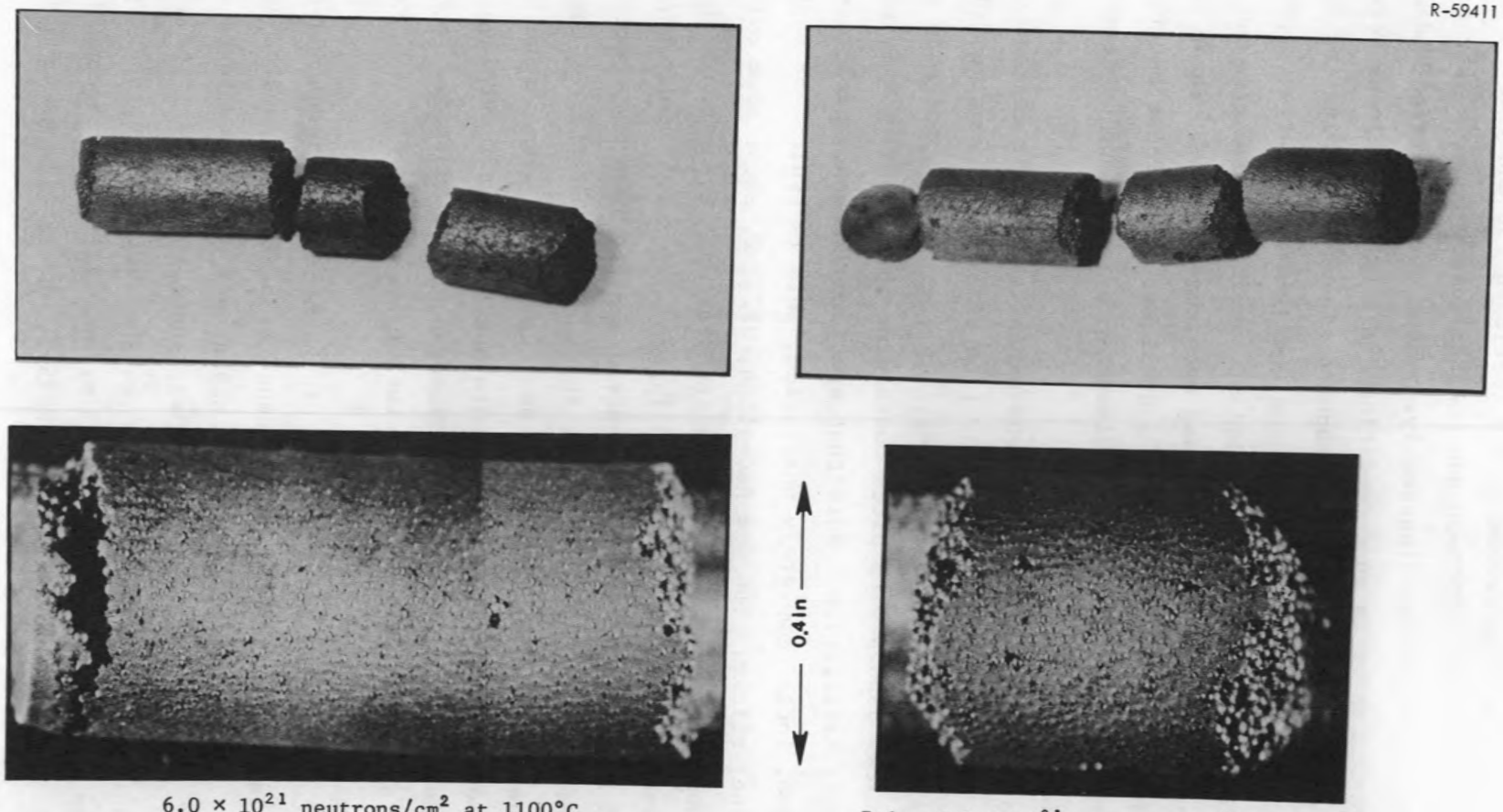
Visual and Dimensional Examination

The graphite sleeve holding the column of fuel stick specimens was removed intact from the capsule by cutting off the ends of the capsule and slitting the inner and outer containment. The three titanium

dosimeter wires were recovered and sectioned for analysis. Two of the bonded fuel sticks (JF275-20 and GGA58-3) dropped out of the top of the graphite sleeve during disassembly. These were recovered, identified, and observed to be in good condition. All other specimens were forced out through the bottom of the graphite sleeve by tapping the sleeve against a metal pan and by bouncing the sleeve on blotter paper on the cell floor. A considerable period of tapping and bouncing was required in order to get the two 2-in.-long specimens to move. These specimens came out in three segments plus a small amount of loose material, as shown in Fig. 8. All other specimens were intact and in reasonably good condition.

Some of the shorter specimens exhibited varying degrees of debonding at the edges; this was especially true of one ORNL specimen (JF275-4) and two GGA specimens (50-3 and 57-4). In Fig. 9 the appearance of two ORNL fuel sticks bonded with pitch containing 50 wt % Thermax is shown before and after irradiation. Note that specimen JF275-4, which received full exposure, looks completely unaffected; this is typical of most of the ORNL fuel sticks. The one exception, JF275-2, shows some evidence of poor bonding at the edges before irradiation. Even though this specimen received somewhat lower exposure [approx 5.5×10^{21} neutrons/cm^2 (>0.18 MeV)], some debonding occurred and the loss of material was significant. On the other hand, the surface condition of the GGA fuel rod specimens after irradiation was quite good, even though the long specimens broke into three segments and the shorter specimens exhibited some debonding. Their surface appearance is illustrated in Fig. 8, which shows portions of the two fuel rods, and in Fig. 10, which shows a short fuel stick (GGA50-3).

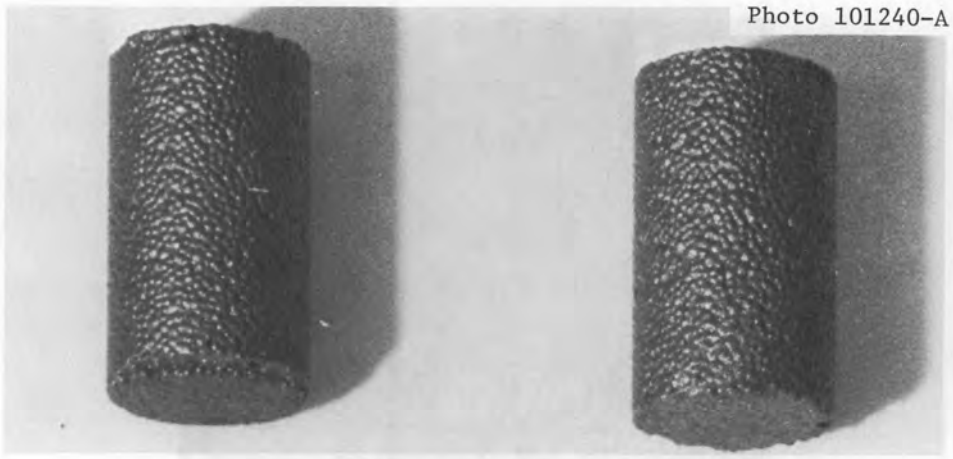
Careful visual examination of the bonded specimens and the debonded material did not reveal any evidence of broken coatings on particles. This is best illustrated by the appearance of the fractured ends of the middle segment of specimen GGA53-3, shown in Fig. 11. The 14 short specimens (approx 0.7 in. long) were weighed before being measured. Weight losses ranged from negligible (3 mg or 0.1%) for specimens JF275-8 and JF275-14, to 27 mg (1.2%) for JF275-2, and as high as 48 mg (1.7%) for GGA57-4.



6.0×10^{21} neutrons/cm^2 at 1100°C

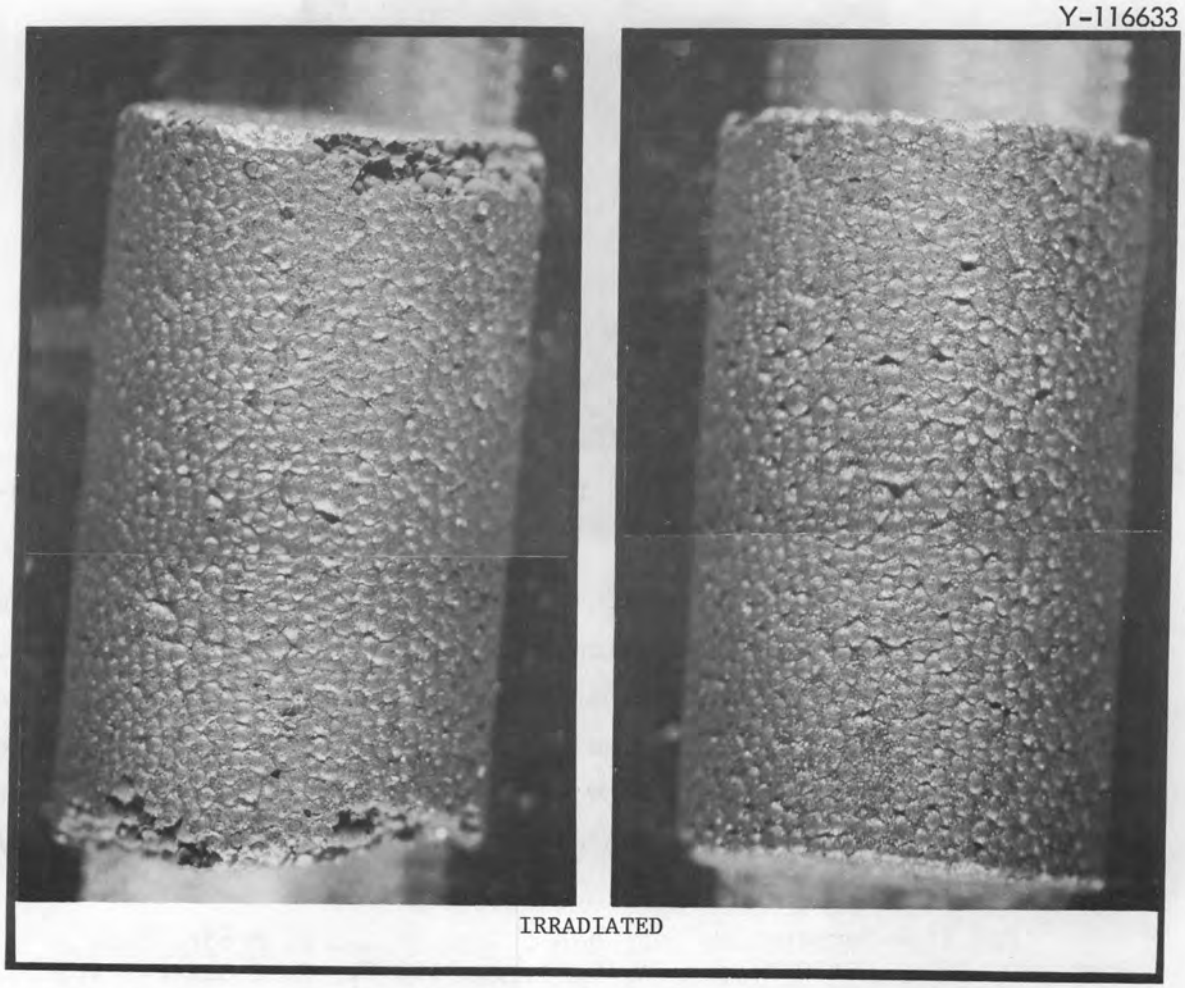
7.9 7.9×10^{21} neutrons/cm^2 at 1200°C

Fig. 8. GGA Fuel Rods Bonded with 27 wt % N.F. 6353 Graphite in Pitch After Irradiation in HRB-2.



JF275-2 JF275-4

BEFORE IRRADIATION



IRRADIATED

Fig. 9. Fuel Sticks Bonded with 50 wt % Thermax in Pitch and Irradiated in Capsule HRB-2.

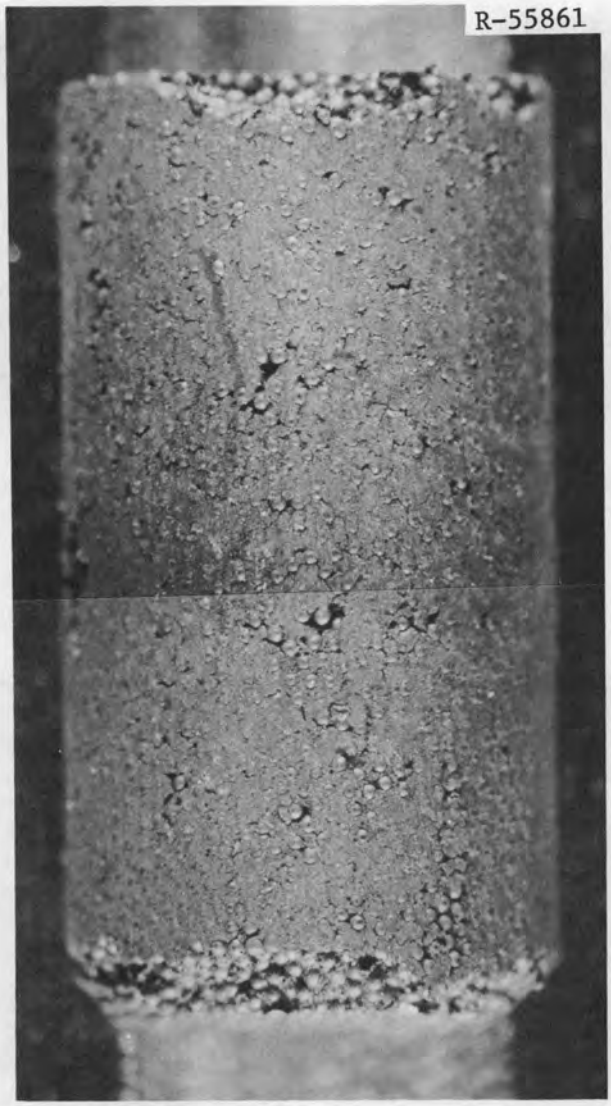


Fig. 10. Appearance of Fuel Stick Specimen GGA50-3 After Irradiation. Note good condition of outer surface.

The postirradiation dimensions of all specimens were recorded before further examination. The dimensional changes are presented in Table 5, and the diametral changes are plotted in Fig. 12 as functions of the calculated fast-neutron fluence. The two smooth curves in Fig. 12 represent calculated[12] shrinkages for the two types of fuel sticks, assuming that the outer carbon coatings in both cases have a density of about 1.85 g/cm^3.

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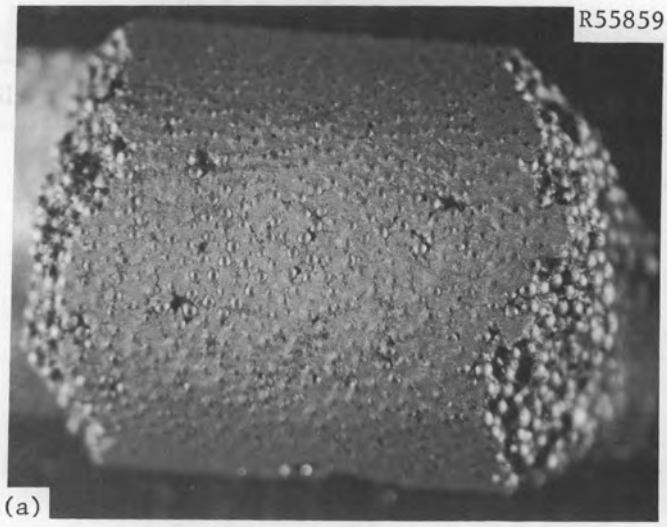
[12] *PSC High-Temperature Gas-Cooled Reactor Quart. Progr. Rep. Sept. 30, 1970,* USAEC Report GA-10313, pp. 160–91, Gulf General Atomic.



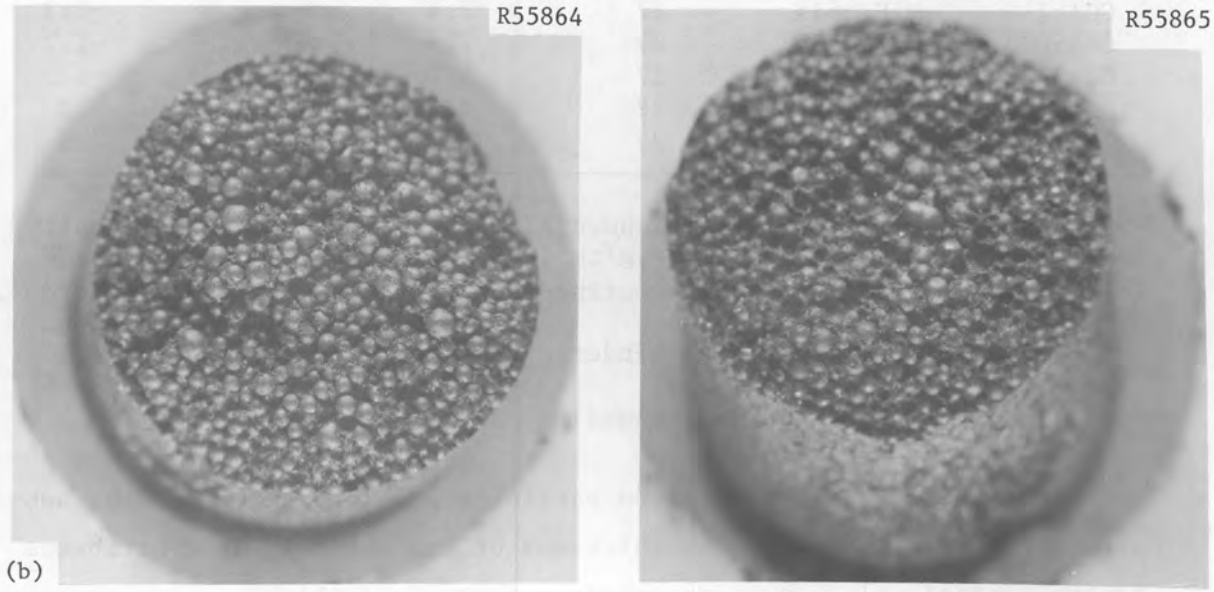


Fig. 11. View of Fuel Stick Specimen Containing TRISO-Coated Particles. (a) Middle segment of GGA specimen 53-3. (b) End Surfaces of segment shown in (a) above. Note good appearance of outer surface and good condition of particles exposed at debonded endges. 6×. (Reduced 10%).

Table 5. Fuel Stick Performance in HRB-2 Experiment

Specimen[a] Designation	Filler Material[b]		Fast Neutron Fluence (neutrons/cm^2)	Net Dimensional Change, %	
	Type	Amount (wt %)		Diameter	Length
			$\times 10^{21}$		
JF275-2	Thermax	50	5.5	−3.7	−3.0
JF275-4	Thermax	50	7.8	−3.95	−4.1
JF275-7	Santa Maria	40	4.9	−3.6	−3.5
JF275-8	Santa Maria	40	7.1	−4.0	−4.4
JF275-10	Robinson	40	6.9	−3.4	−2.7
JF275-12	Robinson	40	7.6	−4.0	−4.2
JF275-14	JOZ	40	6.4	−4.3	−4.4
JF275-16	JOZ	40	7.4	−4.5	−4.4
JF275-20	Asbury	35	4.1	−3.1	−3.1
JF275-23	Poco AXZ	35	6.8	−4.4	−4.3
GGA58-3	N.F. 6353	27	4.5	−1.6	−1.0
GGA57-4	N.F. 6353	27	7.4	−1.6	−1.1
GGA50-3	N.F. 6353	27	7.6	−1.55	−1.2
GGA52-3	N.F. 6353	27	5.0	−1.35	−1.3
GGA53-3	N.F. 6353	27	7.9	−1.25	c
GGA54-1	N.F. 6353	27	6.0	−1.0	c

[a]ORNL specimens contain blended BISO-coated batches; outer-coating densities; 1.92, 1.91, and 1.87 g/cm^3. GGA specimens contain blended TRISO-coated particles; outer-coating-density range: 1.71 to 1.86 g/cm^3.

[b]Mixed with 15V pitch as binder.

[c]Specimens broke during unloading.

The SiC layer of the TRISO-coated particles provides an unyielding substrate, so only the change in thickness of the outer layer contributes to the dimensional changes observed in these fuel sticks.

Dimensional changes of the graphite sleeve were recorded after unloading. The graphite bowed slightly (0.030 in.) from center line and increased in diameter and length. The diametral swelling, which was observed on both inside and outside dimensions, was only slight at the top end (<0.2%) but was greater than 1% at the reactor midplane and at the bottom end. The net length increase amounted to 0.32%.

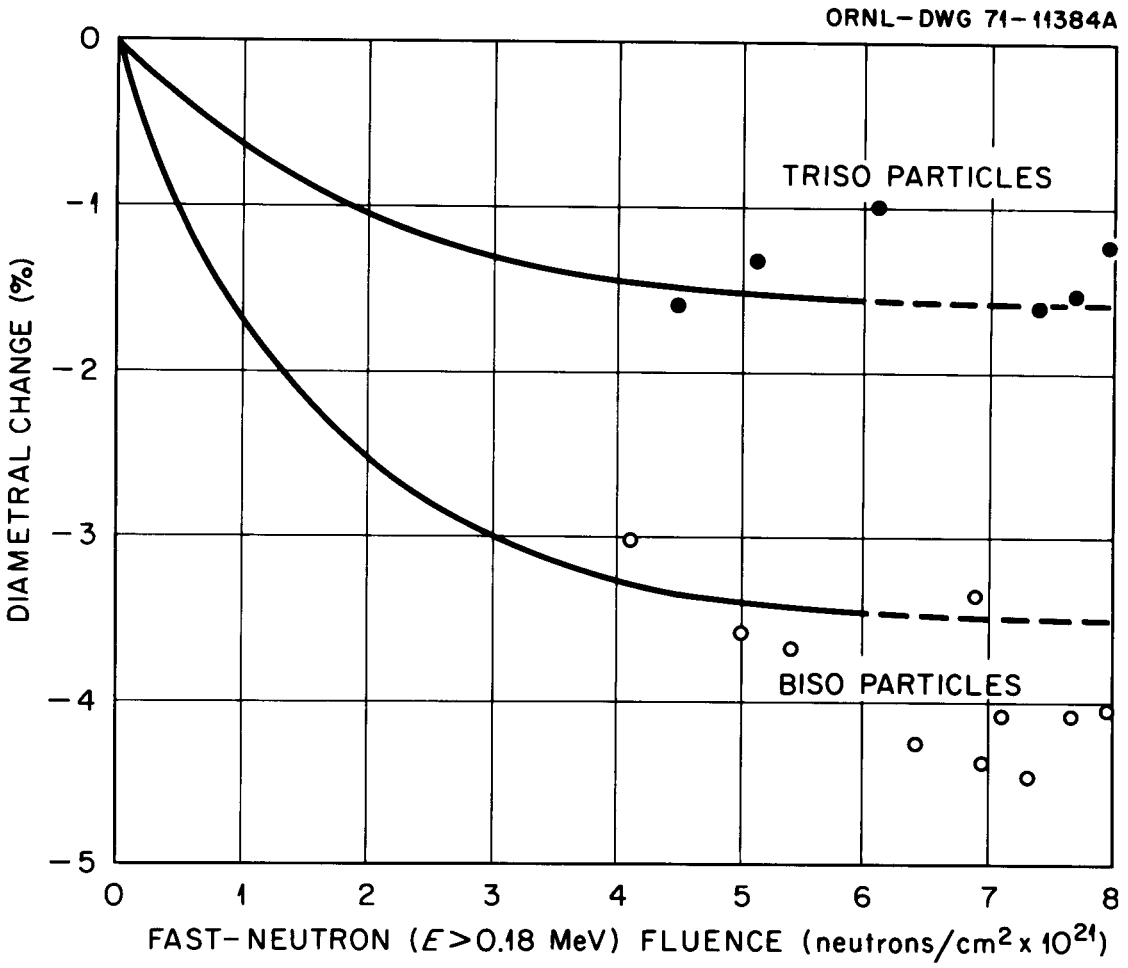


Fig. 12. Shrinkage of Fuel Sticks in HRB-2 Experiment.

Analysis of Flux Dosimeters

The three titanium dosimeter wires were located in holes 120 deg apart in the graphite sleeve. The wires extended from 1 1/2 in. below to 9 1/8 in. above core midplane, and were encapsulated in platinum tubes. After capsule disassembly, the tubes were removed to a "clean" hot cell and segments were cut from both ends and the middle of each wire. Initially seven segments were cut from each wire. Nine large segments (three from each wire) were weighed and dissolved and the solutions analyzed by gamma ray spectrometry. It was intended to analyze the small samples directly, but only three of them were cool enough to permit this initially.

The counting results for samples from the cold ends of the wires gave fair agreement with flux levels obtained from the HRB-γ2 capsule dosimeters.[13] In addition, the solid and solution sample results agreed, and the results from the three wires were consistent. However, the solution sample results from the middle and hot ends of the wires were completely inconsistent, disagreeing both with each other and with expected values by factors of 10 to 20.

The remaining small samples were allowed to decay to low enough levels for counting. In addition, four new samples were cut from the wires for analysis. These samples, counted approximately 2 months later than the original set, gave data that essentially reproduced the earlier results, which indicated that the original results were not entirely spurious. Thus, the disagreement of the results from the expected values must stem from some other, as yet unknown, source.

Even though the radiochemical analyses were reproducible, the results from individual samples were highly inconsistent with any reasonable flux profile. The difficulty stems, in part, from the fact that in a long-term irradiation significant secondary reactions occur, for which the capture cross sections are not well known. As a consequence, no conclusions on fast flux dose to HRB-2 can be drawn from the dosimeter data. Fortunately, a consistent set of dosimeter data is available from previous short-exposure capsules.[14] Since the HFIR characteristically produces a stable flux distribution,[15] we believe it is reasonable to apply the short-exposure distribution to longer term tests. Therefore, fast and thermal flux doses for the HRB-2 experiment were obtained from the earlier data.

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[13] R. L. Senn and W. R. Mixon, "Experimental Measurement of Gamma Heat in the High Flux Isotope Reactor," *Nucl. Technol.* 12: 235–40 (1971).

[14] J. L. Scott et al., *An Irradiation Test of Bonded HTGR Coated Particle Fuels in an Instrumented Capsule in HFIR,* USAEC Report ORNL-TM-3640, Oak Ridge National Laboratory, March 1972.

[15] R. D. Cheverton and T. M. Sims, *HFIR Core Nuclear Design,* USAEC Report ORNL-4621, Oak Ridge National Laboratory, July 1971.

Metallographic Examination

Two of the GGA specimens and two ORNL fuel sticks were sectioned and polished for metallographic examination. The two GGA specimens included a short fuel stick (50-3) and the middle portion of a long fuel rod (53-3). No broken coatings on fissile or fertile particles were observed in either specimen. A composite radial section of specimen 53-3 in Fig. 13 shows no evidence of detrimental matrix-coating interactions, and no large voids were observed in the matrix. The preponderance of inert particles (coated SiC kernels) is obvious in this section. A marked degree of kernel swelling was observed in some coated ThC_2 fertile particles as a result of the high burnup (calculated as 12% FIMA, maximum). This effect is illustrated in Fig. 14, which shows in a polished section of a specimen with 11% FIMA burnup, that the ThC_2 consumed or penetrated the buffer coating, but did not damage the outer coating layers. The smaller fissile particles generally showed some shrinkage of the buffer layer, leaving a gap next to the inner dense coating as shown in Fig. 14.

The two ORNL fuel sticks examined by metallography included one bonded with 50 wt % Thermax in pitch (JF275-4) and another bonded with 40 wt % JOZ graphite in pitch (JF275-16). Both specimens received high exposure [$>7 \times 10^{21}$ neutrons/cm^2 (>0.18 MeV)]. The matrix of both specimens was in good condition although the specimen with JOZ filler showed less cracking, as shown in Fig. 15. The large voids in specimen JF275-4 are typical of as-fabricated specimens bonded with 50 wt % Thermax in pitch. Dimensional changes of these two specimens were quite similar, ranging from 4.0 to 4.5% in diameter and from 4.1 to 4.4% in length, as would be expected due to their similar loading and environment.

The performance of the BISO-coated ThO_2 fertile particles in the ORNL specimens was excellent. The appearance of typical coated ThO_2 particles before and after irradiation is shown in Fig. 16. Note that the ThO_2 kernels show some porosity and agglomerated fission products (small, bright particles) and the swelling associated with the 12% burnup has partially densified the buffer coating layer. The outer coatings were unaffected, and examination of the two specimens did not disclose any broken fertile particles or any distinguishable increase in anisotropy of the outer coating layers. On the other hand, some of the fissile particles, which had

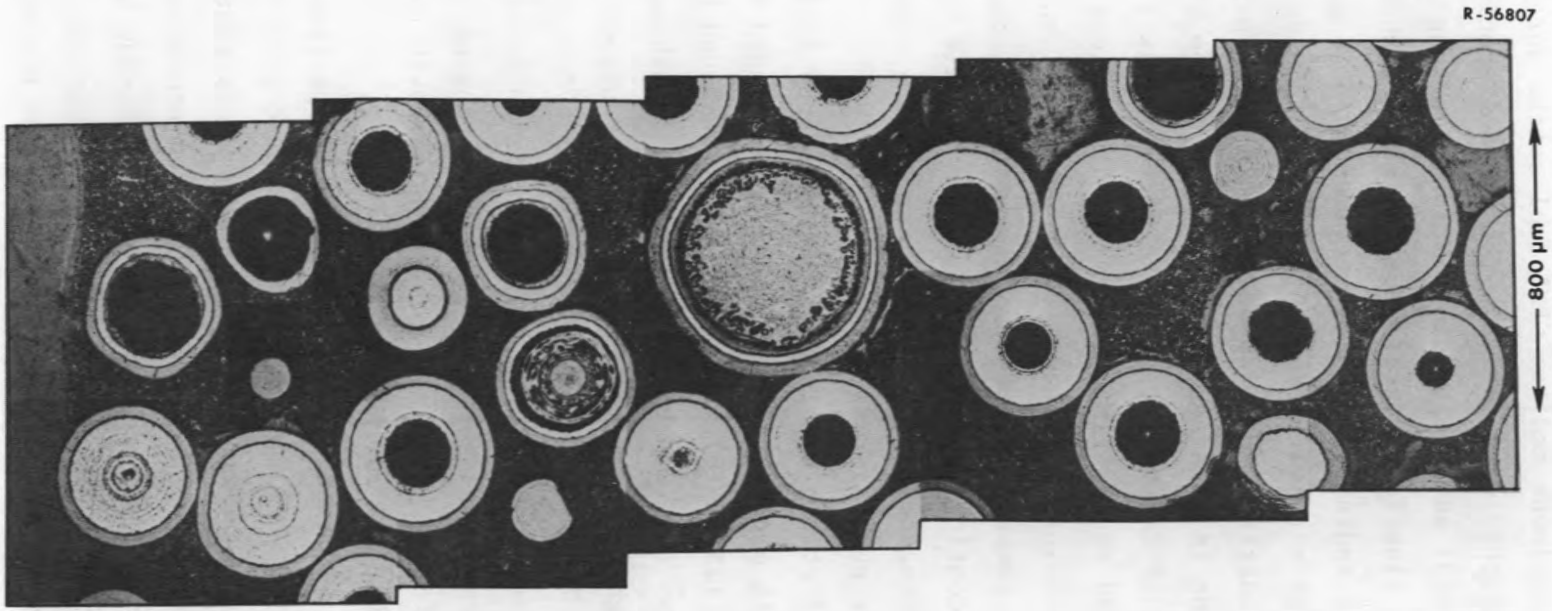

Fig. 13. Polished Section of GGA Specimen 53-3 Irradiated to a Fast Fluence of 7.9×10^{21} neutrons/cm^2 in HRB-2 Capsule.

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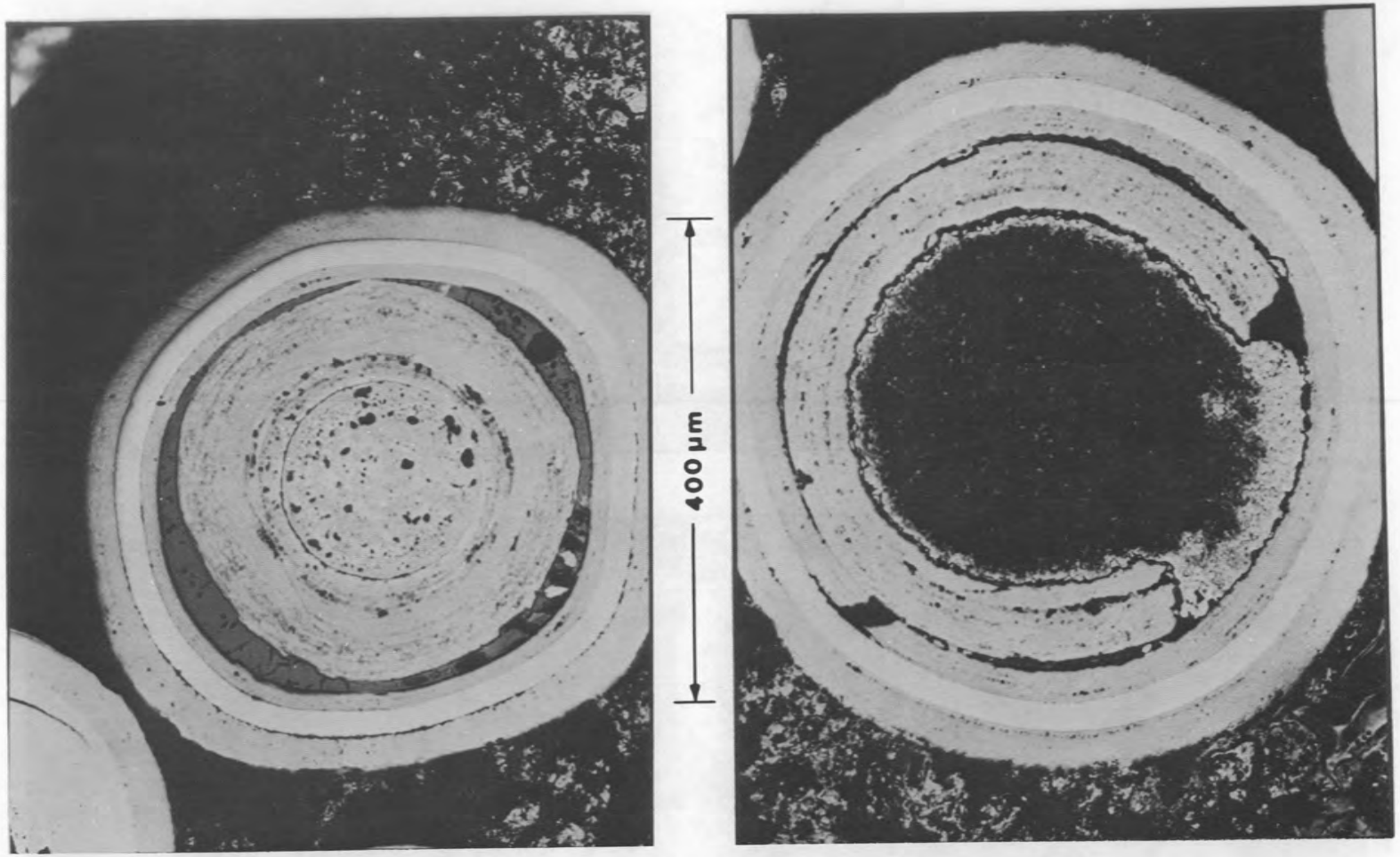

$(Th,U)C_2$ Fissile, 23% FIMA

ThC_2 Fertile, 11% FIMA

Fig. 14. TRISO-Coated Particles Irradiated to 7.6×10^{21} neutrons/cm^2 at 1200°C.

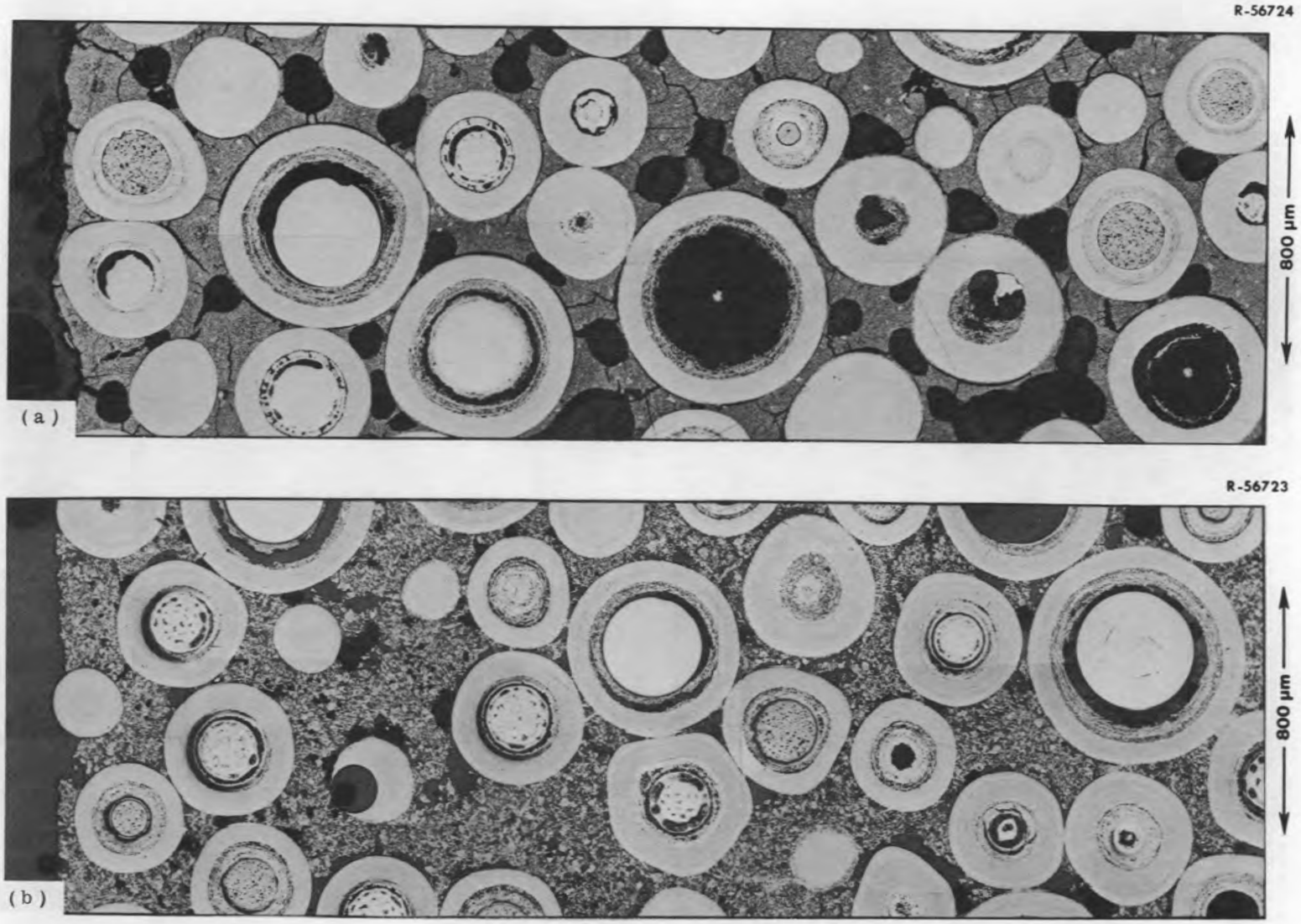

Fig. 15. Polished Sections of ORNL Specimens Irradiated to a Fast Fluence of $>7 \times 10^{21}$ neutrons/cm^2 in Experiment HRB-2. (a) Section of specimen JF275-4, bonded with 50 wt % Thermax in pitch. (b) Section of specimen JF275-16, bonded with 40 wt % JOZ pitch. Note the large pores and numerous cracks in matrix JF275-4 as compared with specimen bonded with JOZ.

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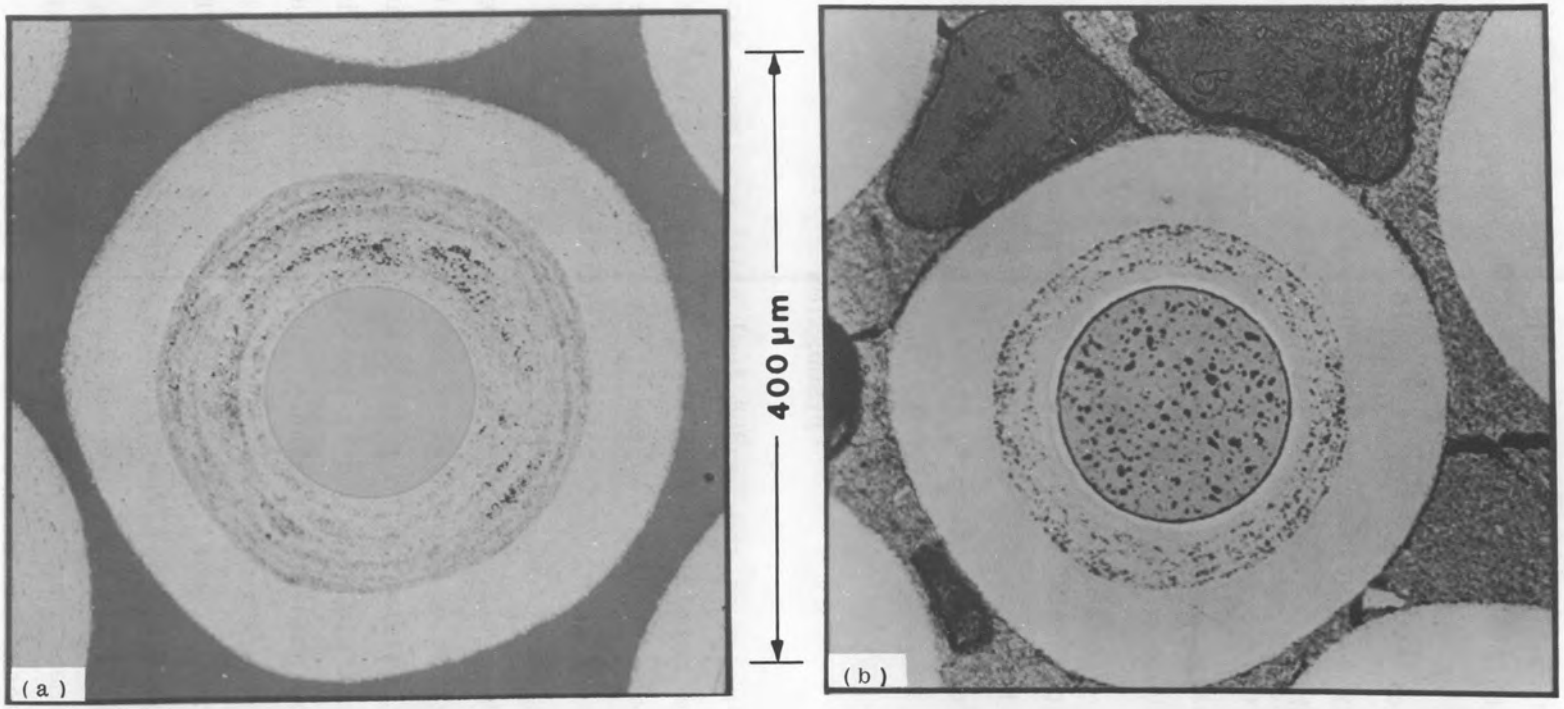

Fig. 16. Polished Sections of BISO-Coated ThO_2 Particles from HRB-2. (a) Unirradiated. (b) Particle irradiated in specimen JF275-4 to a burnup of 12% FIMA and a fast fluence of 7.9×10^{21} neutrons/cm^2 at 1280°C. Note porosity in ThO_2 kernel and densification of buffer coating layer.

carbonized strong-acid resin fuel kernels, showed coating damage and fuel redistribution during the irradiation. The polished section of the specimen bonded with Thermax in pitch (JF275-4) showed two fissile particles with broken coatings and one that was severely cracked but not completely broken. One of the broken particles is shown in Fig. 17; it appears that the cracks may have propagated from the matrix into and through the coatings. The other specimen, JF275-16, showed no failed coatings. In other particles of this type, some radial redistribution of fuel and kernel components was observed, as illustrated by the center photograph in Fig. 17. This redistribution did not seem to affect the performance of the coated particles.

The inert particles that made up an appreciable volume of each ORNL specimen consisted of BISO-coated carbon kernels. These particles performed well in the experiment; the carbon kernels densified and shrank considerably, as shown in Fig. 15, but the coatings survived.

DISCUSSION

The shrinkage of the bonded fuel stick specimens was evidently independent of the matrix material used to bond the coated particles and was strongly affected by the neutron fluence, especially in the case of the fuel sticks containing BISO-coated particles. Because of the nature of the bonded sticks (i.e., closely packed particles bonded together with a carbonaceous matrix), the shrinkage is expected to be strongly affected by the properties of the particle coatings. The observed dimensional changes are consistent with this model. The six specimens that contained TRISO-coated particles would be expected to shrink less because the SiC layer does not change appreciably during irradiation, and the observed shrinkage is due primarily to densification and changes in thickness of the outer carbon coating. On the other hand, two-layer BISO coatings can shrink and densify almost without restraint during early stages of the irradiation; the much greater dimensional changes (up to 4.5%) of the ten specimens that contained BISO-coated particles are consistent with the expected behavior of the actual coatings (average density of outer layers approx 1.85 g/cm^3). The fact that the observed dimensional

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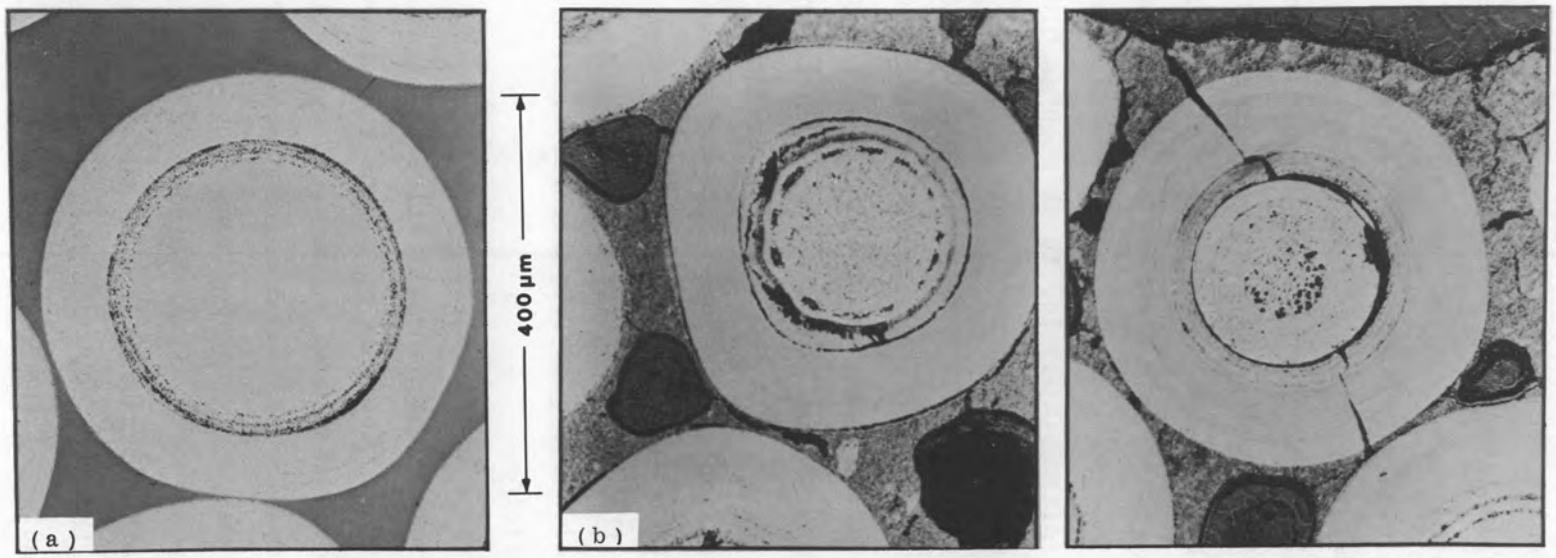

Fig. 17. Polished Section of BISO-Coated Strong-Acid Resin Fissile Particles Irradiated in Experiment HRB-2. (a) Unirradiated. (b) Particle irradiated to a burnup of 31% FIMA and a fast fluence of 7.9×10^{21} neutrons/cm^2 at 1280°C.

changes at highest fluence exceed somewhat the calculated changes (see Fig. 12) probably results from uncertainties in the extrapolation of the curves, which were calculated from density measurements on coatings irradiated at fluences up to about 6×10^{21} neutrons/cm^2.

The dimensional changes of the fuel stick specimens and the graphite sleeve caused the fuel temperatures to increase during the irradiation. This increased fuel temperature may be an impediment to the use of fuel sticks containing BISO-coated particles. In experiment HRB-2 the effect was more pronounced because the Poco graphite sleeve actually swelled slightly instead of shrinking in diameter as would be expected of needle-coke graphites at this exposure. These changes permitted the radial gap to increase from an initial 0.005 in. to as much as 0.017 in. The effects of these changes on specimen temperature are reflected in the comparison of calculated temperatures for the midplane specimens (GGA53-3 and JF275-4) given in Table 6 for the initial and final test conditions. Condition 1 is for the initial startup using the initial fuel loadings, reactor fluxes, and as-built dimensions. Condition 2 corresponds to the conditions at the end of the irradiation. In the latter calculation we used the calculated isotopic compositions, the end-of-core-life reactor fluxes, and the postirradiation measured dimensions of the bonded fuel sticks and graphite sleeve. Even though the linear heat ratings are lower at the end of the irradiation, the final calculated temperatures are significantly higher than the initial condition.

We observed during examination that the debonding and weight loss of the short fuel sticks could be qualitatively correlated with their preirradiation condition. The only specimens containing BISO-coated particles that lost more than 6 mg of material during irradiation were two that did not have well-bonded and sharp edges before irradiation. The same generalizations apply to specimens containing TRISO-coated particles as well, even though all of these lost more material and were in poorer condition after irradiation than the sticks containing BISO particles. The fact that the long specimens each broke into three segments during unloading of the graphite sleeve is not considered serious, since considerable effort was required to dislodge these specimens and very little loose material was observed.

Table 6. HFIR-HTGR HRB-2 Capsule Calculated Midplane Fuel Temperature for Startup and End of Irradiation

Experimental Parameters	Condition 1[a]		Condition 2[b]	
	GGA53-3	JF275-4	GGA53-3	JF275-4
Linear Heat Ratings, kW/ft				
Fission heat rate	3.33	3.33	2.65	2.80
Total heat generation rate	4.03	3.92	3.32	3.33
Bonded Bed Physical Properties				
Density, g/cm^3	1.78	1.485	~ 1.8	~ 1.5
Thermal conductivity, Btu hr^{-1} ft^{-1} $°F^{-1}$	3.47	3.47	3.47	3.47
Dimensions, in.				
Bonded bed diameter	0.4115	0.4125	0.406	0.396
Graphite sleeve inner diameter	0.423	0.423	0.4273	0.4273
Graphite sleeve outer diameter	0.904	0.904	0.9115	0.9115
Temperatures, °C				
Bonded bed center	1228	1213	1294	1356
Bonded bed surface	1052	1041	1147	1209
Mean graphite sleeve (measured)	905	905	905	905

[a]Initial startup using initial fuel loadings, fluxes, and as-built dimensions.

[b]End of irradiation using calculated isotopic compositions, fluxes, and postirradiation dimensions of the beds and graphite sleeve. We assumed that thermal conductivity did not change.

The performance of the coated fuel particles in this experiment was excellent as far as could be determined by fission-gas release and metallographic examination. Reference to Fig. 3 shows that fission-gas release rate was moderate all during the experiment. The significant increase during the first cycle was expected as the experiment approached equilibrium, while the increase in ^{133}Xe and Kr activity during the last cycle was not accompanied by bursts of activity and may have been a result only of the higher operating temperature of some specimens, as discussed above. A few (2 or 3) bursts of activity were observed just prior to shutdown at the end of the eighth cycle, but no bursts occurred thereafter. Metallography confirmed the good performance of all TRISO-coated particles and of the BISO-coated ThO_2 fertile particles, although the coatings on the latter were intentionally overdesigned to avoid the effects of coating failure on fuel stick integrity. The only failed BISO-coated resin-derived particles were observed in ORNL specimen JF275-4, which was bonded with 50 wt % Thermax in pitch and received maximum exposure at high temperature. Failure of these few particles seemed to be due to a combination of stresses in the coating deriving from interactions with the shrinking matrix and from property gradients within the outer coating (see Fig. 17). The gradation of properties, which manifests itself as banding in the coating, was much more evident after irradiation and was somewhat variable; other particles that received the same exposure but showed less of the property gradient did not fail.

CONCLUSIONS

The results from irradiation testing of bonded fuel stick specimens in this experiment show that:

1. Fuel sticks bonded with the Fort St. Vrain reference matrix material and with various other filler materials in pitch binder survive irradiation to a maximum exposure of 8×10^{21} neutrons/cm^2 at about 1250°C.

2. The moderate shrinkage of fuel sticks containing TRISO-coated particles does not produce significant changes in operating temperature during irradiation. On the other hand, the more drastic shrinkage of

fuel sticks containing BISO-coated particles can lead to temperatures significantly higher than initial conditions.

3. Fertile particles with conservatively designed BISO and TRISO coatings perform well to the maximum HTGR fast-neutron exposure and at burnups 50% greater than the design conditions for a large HTGR.

4. Fissile particles consisting of fueled strong-acid resin kernels with BISO coatings generally perform well at the maximum fast-neutron exposure and a burnup of 31 at. % heavy metal.

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