ORNL-TM-1142

Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION

IRRADIATION BEHAVIOR OF THORIUM-URANIUM ALLOYS AND COMPOUNDS*

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R. E. Adams, and D. A. Douglas

JUNE 1965

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*Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

For presentation at the IAEA Panel on "Utilization of Thorium in Power Reactors," Vienna, Austria, June 14-18, 1965.

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ABSTRACT

Irradiation programs at Oak Ridge National Laboratory have been evaluating thorium-base compounds as part of the overall Thorium Utilization Fuel Cycle Program and in support of the Advanced Gas-Cooled Reactor Program. Thorium-base metal fuels are being considered for lower temperature reactor systems.

Emphasis in the clad bulk oxide fuel program has been on the evaluation of the sol-gel derived ThO_2-UO_2 fuels, and vibratory compaction for fuel rod fabrication. The objectives of this program and the reported results are reviewed. Vibratorily compacted, sol-gel ThO_2-UO_2 has the characteristics of a useful power-reactor fuel. Experimental rods up to 36 in. in length fabricated in this manner have compared favorably with vibratorily compacted rods containing arc-fused ThO_2-UO_2 fuels and with rods containing pressed and sintered ThO_2-UO_2 pellet fuels. Tests at linear heat ratings between 300 and 400 w/cm have been examined with burnups in excess of 80,000 Mwd/tonne (Th+U) with less than 20% fission-gas release. Although the maximum performance capabilities have not been defined, rods have been operated at linear heat ratings of 1,000 w/cm to 22,000 Mwd/tonne (Th+U) burnup, with no evidence of swelling, or central melting and with fission-gas release rates less than 30%. Preliminary comparisons with similar UO_2 fuel rods indicate that higher power ratings are required to produce similar microstructural changes in the ThO_2–5% UO_2 fuels.

Static and sweep capsules have been used in the irradiation tests of thorium-bearing fuels, both carbides and oxides, for application to high-temperature gas-cooled reactor designs. Fuel in the form of small particles, coated with pyrolytic carbon has been irradiated in loose beds and in graphite matrix elements. Reported data are reviewed, showing that spherical particles, with multilayer coatings are superior

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and that fission-gas release rates (R/B) for ^{85}Kr of 10^{-5} to 10^{-7} have been achieved in loose bed tests. Coated particles of $(Th,U)C_2$ and $(Th,U)O_2$ compare favorably with similar UC_2 and UO_2 particles. The oxide particles are compatible with the intact pyrolytic carbon coatings.

Metallic fuels, particularly thorium-base metal fuels, are being considered for reactors to supply heat for desalting water. A review of the limited experiments to date on thorium irradiations indicated that such isotropic fuels are not subject to the "growth" phenomena of anisotropic uranium and that they do not exhibit the "cavitation swelling" reported for uranium metal fuels in the 400 to 600°C temperature range. Additional research and development is required to define the usefulness of thorium-base metallic fuels.

INTRODUCTION

Thorium metal has an isotropic body-centered cubic crystal structure, higher strength, and a 600°C higher melting point than uranium which has an anisotropic orthorhombic crystal structure. Thorium dioxide crystallizes in the same stable cubic fluorite lattice as uranium dioxide but has a melting point some 500°C higher and forms a complete series of solid solutions with uranium dioxide with the same lattice configuration. The carbides of thorium and uranium are similar and also form a series of solid solutions. All of these physical facts indicate the promise of improved performance for thorium-base fuels even if one ignores its breeding potential.

Historically, thorium and thorium alloys or compounds have been studied much less intensively than uranium and its alloys. Only recently have a number of investigators begun to seriously study the thorium-base compounds, both oxides and carbides for potential application to power reactors.

The program at Oak Ridge National Laboratory has emphasized the sol-gel[1] process for the preparation of thorium-base fuel compounds as potentially more economical with a denser, more uniform product. Calcined sol-gel thoria-urania fuel has been semiremotely processed and fabricated into rods by vibratory compaction techniques.[2] The sol-gel material can be processed into truly round, smooth microspheres of theoretical density for coated particle applications.[1]

The data from the irradiation tests reported have shown that thorium-base fuels, both metallic and ceramic, have the basic characteristics required for power reactors. Interest has shifted to determining the maximum performance characteristics to take advantage of the indicated potentials for higher temperature operation and increased power ratings with improved stability. This paper will review the data obtained from different irradiation-test programs on sol-gel and conventionally processed thorium-base fuels, as metal-clad bulk oxides and as-coated particles. Finally, there is a section which reviews the available data on the irradiation characteristics of thorium-base metallic fuels.

BULK OXIDE FUELS OF ThO_2-UO_2

In support of the Thorium Utilization Fuel Cycle Program a series of irradiation tests on metal-clad bulk oxide fuels containing intimately mixed ThO_2 and UO_2 was started at the Oak Ridge National Laboratory in 1961. This program has concentrated on ceramic fuels produced by the sol-gel[1] process and fabricated into rods by the use of vibratory compaction. The initial objectives were to compare the basic nuclear fuel performance characteristics of this chemically produced ceramic with those of arc-fused material and the performance of vibratorily compacted fuel with pressed and sintered pellets. The early tests indicated a favorable comparison and the objectives have been expanded to include:

1. the effects of sol-gel processing variables,
2. the effects of fuel-rod geometry (diameters and lengths),
3. the determination of the maximum performance characteristics of vibratorily compacted sol-gel fuels,
4. the potential of mixed progeny sol-gel fuels (i.e., ThO_2-PuO_2), and
5. the effects of semiremote fabrication of vibratorily compacted ThO_2-$^{233}UO_2$ sol-gel fuels.[2]

Table 1 lists the groups of fuel rods, their principal objectives, and their current status.

Most of the tests have been made with noninstrumented rods in the Materials Test Reactor (MTR), Engineering Test Reactor (ETR), and the Chalk River National Research Experimental Reactor (NRX). Three tests

Table 1. Summary of Thor'um Fuel Cycle Program Installations of Powder-Packed Capsules (Rods) of Type 304 Stainless Steel*

Designation	No. of Rods	Type of Oxide	Density, % theor.	Fuel Rod Dimensions, in. Length	OD	Wall	Linear Heat Rating, Btu/h-ft	Peak Burnup, Mwd/tonne metal	Status	Objective
MTR-I	7	Arc-fused Sol-Gel E	86 to 87	11	0.312	0.025	40,000	15,000 to 100,000	2 examined; 4 being examined; 1 in pile	Provide base-line data to use in comparing sol-gel and arc-fused oxide
MTR-II	2	Sol-Gel S	88 to 89	22	0.312	0.025	62,000	100,000	In Pile	Obtain higher heat rating by increasing enrichment
MTR-III	6	Sol-Gel 25	86 to 89	12	0.438	0.025	85,000	100,000	In Pile	Compare oxide containing atmosphere and higher heat ratings obtained by increasing diameter
ETR-I	4	Sol-Gel 25	86 to 89	12	0.438	0.025	> 100,000	22,000	Being Examined	Same as for MTR-III
NRX-I	8	Sol-Gel A and B	86 to 87	11	0.312	0.025	17,000	16,000	Examined	Provide base-line data
NRX-II	4	Sol-Gel C Arc-fused	83 to 86	22	0.312	0.025	22,000	5,000	Examined	Study effect of increased length
NRX-III	6	Sol-Gel S	88 to 89	30	0.312	0.025	28,000	22,000	Being Examined	Study effect of increased length
NRX-III	3	Sol-Gel ThO_2-PuO_2	74 to 76**	11	0.312	0.025	27,000	22,000	Being Examined	Study ThO_2-PuO_2 oxide and lower packed density
OMR Loop	3	Sol-Gel 36	84 to 85	22	0.440	0.015	32,000	2,100	Examined	Study in pressurized water at 260°C and 1750 psi
OMR Pendulum	2	Sol-Gel D	85	7	0.625	0.030	35,000	5,000	Examined	Measure effective thermal conductivity using a central thermocouple in Na-K at 315 psi, 540 and 705°C
ETR-II	6	BNL-Sol-Gel	90	19	0.499	0.035	~45,000	30,000 to 100,000	In Pile	Study effects of remote fabrication and oxide reconsolidation
ETR-III	6	Sol-Gel ThO_2	88	19	0.499	0.035	~80,000	10,000 to 70,000	In Pile	Study ThO_2 blanket material with gradually increasing heat rating and provide high Pa low-fission-gas-product material for chemical processing

*Except ETR-II, III and two of the three OMR loop specimens were Zircaloy clad

** lump packed

were conducted in the Oak Ridge Research Reactor (ORR),[3] one a trefoil cluster in a pressurized-water loop operating at 250°C and 1750 psi and the other two were instrumented rods in the Poolside Facility with 538 and 704°C cladding temperatures.

Postirradiation examinations include dimensional analysis for swelling or bowing, fission-gas release measurements, gamma scanning, burnup determinations, x-ray diffraction analysis, and metallographic examinations.

As with any irradiation program, and particularly with one as broad in scope as this, there are time overlaps in obtaining data to evaluate the various objectives. The initial scoping studies are complete and all other portions are being pursued. Many of the earlier tests have been reported in detail[4-8] and have shown that sol-gel ThO_2-UO_2 has the basic performance characteristics for a power reactor fuel.

Effects of High Burnup

The effects of higher burnup on three types of ThO_2-UO_2 fuels have recently become available. One of the advantages of the ThO_2 base fuels is the relatively high breeding ratio. It can be calculated that in an appropriate thermal flux a thoria fuel containing approximately 1 wt % ^{235}U will produce essentially constant power to a burnup of approximately 50,000 Mwd/tonne (Th+U). However, in most of our tests a 4 to 5 wt % ^{235}U has been used to shorten the time required for significant burnup. Total depletion of this initial uranium will produce approximately 30,000 Mwd/tonne but it is necessary to move the test rods periodically to higher flux positions in order to maintain essentially constant heat ratings. Test rods from the MTR Group I containing either sol-gel or arc-fused vibratorily compacted ThO_2-4.5% UO_2 have been examined after burnups in excess of 70,000 Mwd/tonne (Th+U). A drawing of these fuel rods is shown in Fig. 1. In addition, four rods, containing pressed and sintered pellets of ThO_2-4.5% UO_2 at burnups between 37,000 and 119,000 Mwd/tonne (Th+U), have been examined.

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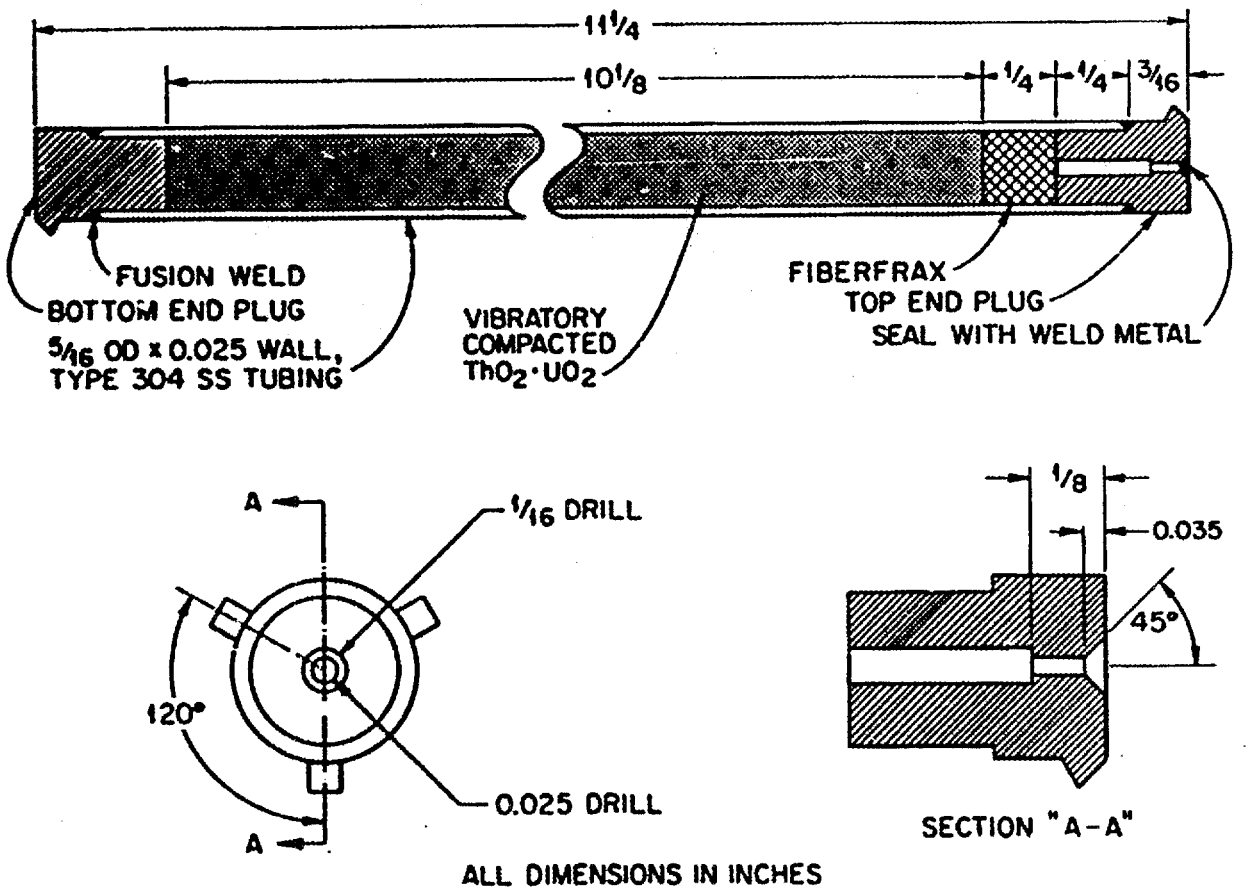

Fig. 1. ThO_2-UO_2 Fuel Cycle Capsule Design for MTR Group I Irradiations.

The fabrication details, irradiation histories, and the significant postirradiation findings and calculations are presented in Tables 2a, b, and c. The time average values for the various heat ratings are for the peak burnup region in each rod calculated from the maximum burnups at the same location, while the maximum instantaneous values are based on fuel depletion calculations and flux steps during irradiation and do not include any changes in flux profile during the various test-reactor cycles. All of the values for

$$\int_{T_o}^{T_c} k\,d\theta$$

assume that the surface temperature of the fuel (T_s) is the same as the temperature calculated for the inside surface of the cladding and that

Table 2a. Effects of High Burnup on ThO_2-UO_2 Fuels: Description of Irradiation Test Rods[a]

Group	Number	Fuel Rod Identification	Type of Oxide	Total Uranium[b] (wt %)	Oxide Particle Size	Vibrated Density[c] (% TD)	Fuel Rod Dimension Length (cm)	Outside Diameter (cm)	Wall (cm)
MTR-I	43-59	U-1	Arc fused	3.96	d	85.7	28.70	0.797	0.0635
	43-60	U-2	Arc fused	3.96	d	85.6	28.70	0.797	0.0635
	43-61	U-3	Arc fused	3.96	d	85.5	28.70	0.797	0.0635
	43-62	U-5	Sol-gel E	3.96	d	87.0	28.70	0.797	0.0635
	43-63	Z-5	Sol-gel E	3.96	d	86.2	28.70	0.797	0.0635
	43-64	Z-7	Sol-gel E	3.96	d	86.2	28.70	0.797	0.0635
	43-65	Z-8	Sol-gel E	3.96	d	85.5	28.70	0.797	0.0635
MTR	43-39	712	Arc fused	3.92	Pellets	93	11.43	0.795	0.0635
	43-40	729	Arc fused	3.92	Pellets	93	11.43	0.795	0.0635
	43-41	730	Arc fused	3.92	Pellets	93	11.43	0.795	0.0635
ETR	43-44	645	Arc fused	3.92	Pellets	93	11.43	0.795	0.0635

[a]Clad with type 304 stainless steel with operating cladding temperature at 100°C.

[b]Enriched in ^{235}U to 93%.

[c]Based upon a calculated density depending on composition (approx 10.04 g/cm^3).

[d]Distribution A: 60 wt % −10 +16 mesh; 15 wt % −70 +140 mesh; 25 wt % −200 mesh.

Table 2b. Effects of High Burnup on ThO_2-UO_2 Fuels: Irradiation History

Experiment	Fuel Rod Identification	Average External Pressure (psia)	Exposure Time Reactor Full Power Days	Peak Linear Heat Rating		Peak Cladding Heat Flux		Maximum Burnup[a]	
				Time Average (w/cm)	Maximum Instantaneous (w/cm)	Time Average (w/cm^2)	Maximum Instantaneous (w/cm^2)	(Mwd/tonne Th+U)	(fissions/cm^3)
									$\times 10^{20}$
MTR-I	U-1	40	110.2	299	362	120	145	12,300	2.50
	U-2	40	376.2	297	388	119	155	42,100	8.56
	U-3	40	707.5	267	419	107	168	71,000	14.43
	Z-5	40	110.2	341	426	137	172	14,000	2.87
	Z-7	40	375.2	286	347	114	138	40,200	8.23
	Z-8	40	691.1	311	507	124	202	81,000	16.46
MTR	712	40	905	420	652	168	261	119,400	26.41
	729	40	660	461		185		96,000	24.6
	730	40	497	270		108		37,000	10.8
ETR	645	180	406	394		158		50,000	12.8

[a]Based on 200 Mev/fission.

Table 2c. Effects of High Burnup on ThO_2-UO_2 Fuels: Postirradiation Observations and Calculations

Experiment	Fuel Rod Identification	$\int_{T_o}^{T_c} k d\theta$ Time Average (w/cm)	$\int_{T_o}^{T_c} k d\theta$ Maximum Instantaneous (w/cm)	^{85}Kr Release (%)	Dimensional Changes Average (mm)	Dimensional Changes Maximum (mm)	Radial Extent of Microstructural Changes Equiaxed Grains (cm)
MTR-I	U-1	33.8	39.8	2.4	+0.025	+0.025	
	U-2	33.7	41.9	7.2	0.000	−0.051	a
	U-3	31.6	44.7	6.4	+0.005	+0.013	a
	Z-5	37.8	44.4	0.5	+0.025	+0.075	
	Z-7	32.8	38.4	13.2	−0.025	−0.037	a
	Z-8	34.6	52.7	17.0	+0.015	+0.058	0.1[b]
MTR	712	44.8	65.4	22.8			
	729	48.2		12.4			
	730	31.5	c				
ETR	645	42.6	c				

[a]Sintering boundary not delineated.

[b]Calculated heat flux for equiaxed grain growth $\int_{T_o}^{T_{eg}} k d\theta = 31.5$ w/cm.

[c]Gas samples diluted with air in sampling and partially lost.

the thermal conductivity data of Kingery[9] apply for

$$\int_{T_o}^{T_s} k d\theta$$

at temperatures below this surface temperature. Although the reliability of these assumptions is questionable, as will be seen later in this paper, they do provide a consistent method of comparison.

The dimensional changes on the MTR-I group indicate no significant swelling in these highly restraining clads. Preirradiation measurements were not made on the pellet rods, since they were originally intended for chemical processing studies only; however, the postirradiation diameters were within the tolerance specifications for the tubing used as cladding. The fission-gas data indicate a slightly higher release for the sol-gel material, but there is no evidence of a breakaway phenomenon. The principal factor in gas release at these heat ratings seems to be time at temperature, which is consistent with a diffusion mechanism for gas release.

The macroscopic appearance of the various sections shown in Figs. 2, 3, and 4 shows a remarkable similarity in the appearance of the fuels at these high burnup levels. Sections of the rods with less burnup are similar, although the radial position of the more or less circumferential cracks appear to move outward with increased exposure. The radial location of these cracks appears to be associated with the extent of in-pile sintering or equiaxed grain growth. Since this phenomenon is dependent on time as well as temperature, the radial extent of such changes would be expected to increase with higher burnup (i.e., time) at a constant heat flux. The microstructures on these rods clearly show the time dependence of such diffusional processes as sintering and grain growth. Figures 5 and 6 are composite radial micrographs of the arc-fused vibratorily compacted rods at various levels of exposure. Figures 7 and 8 are similar composites of the sol-gel vibratorily compacted rods, while Figs. 9, 10, 11, and 12 show selected micrographs at various radial positions for the pressed and sintered pellets.

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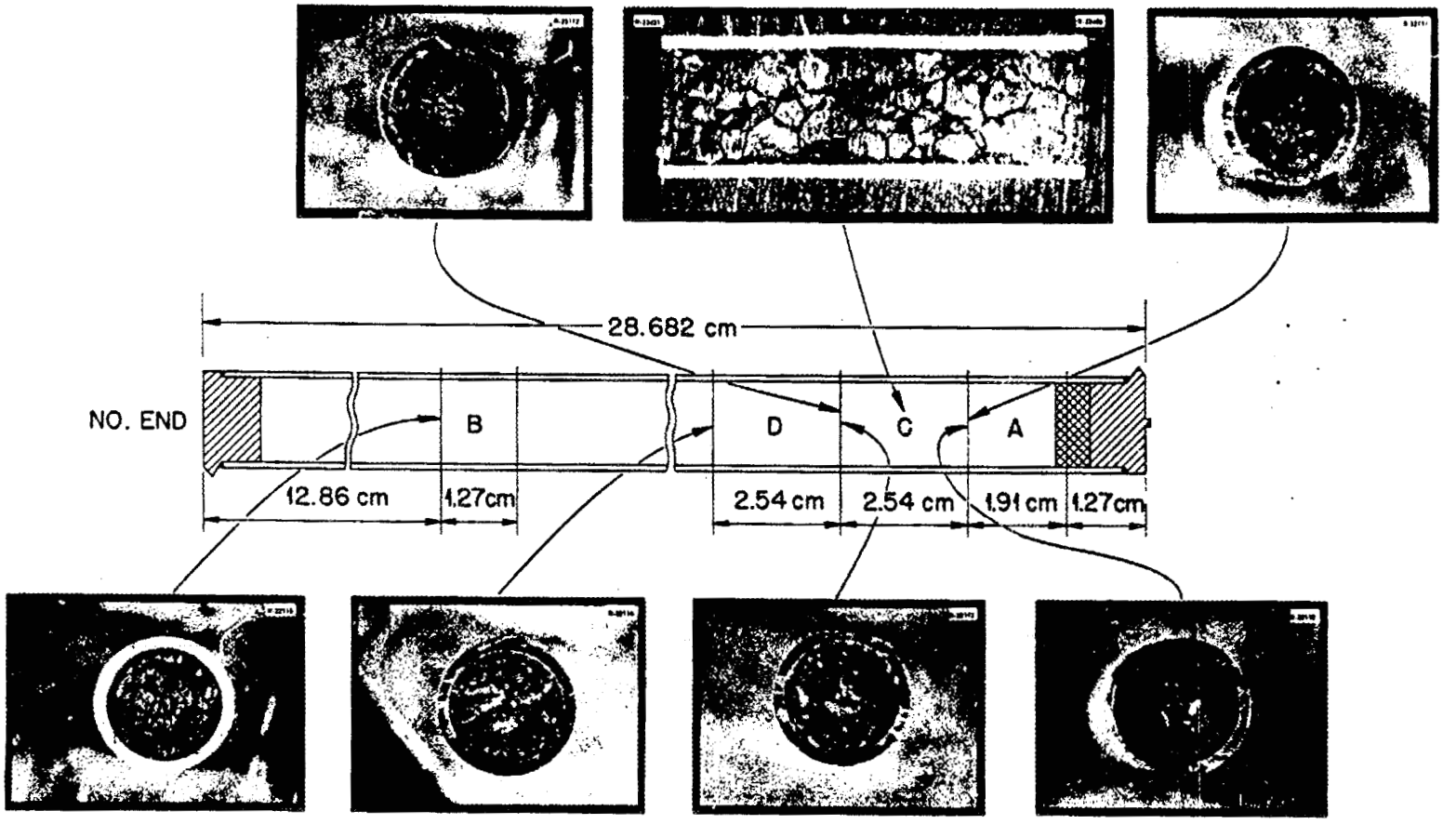
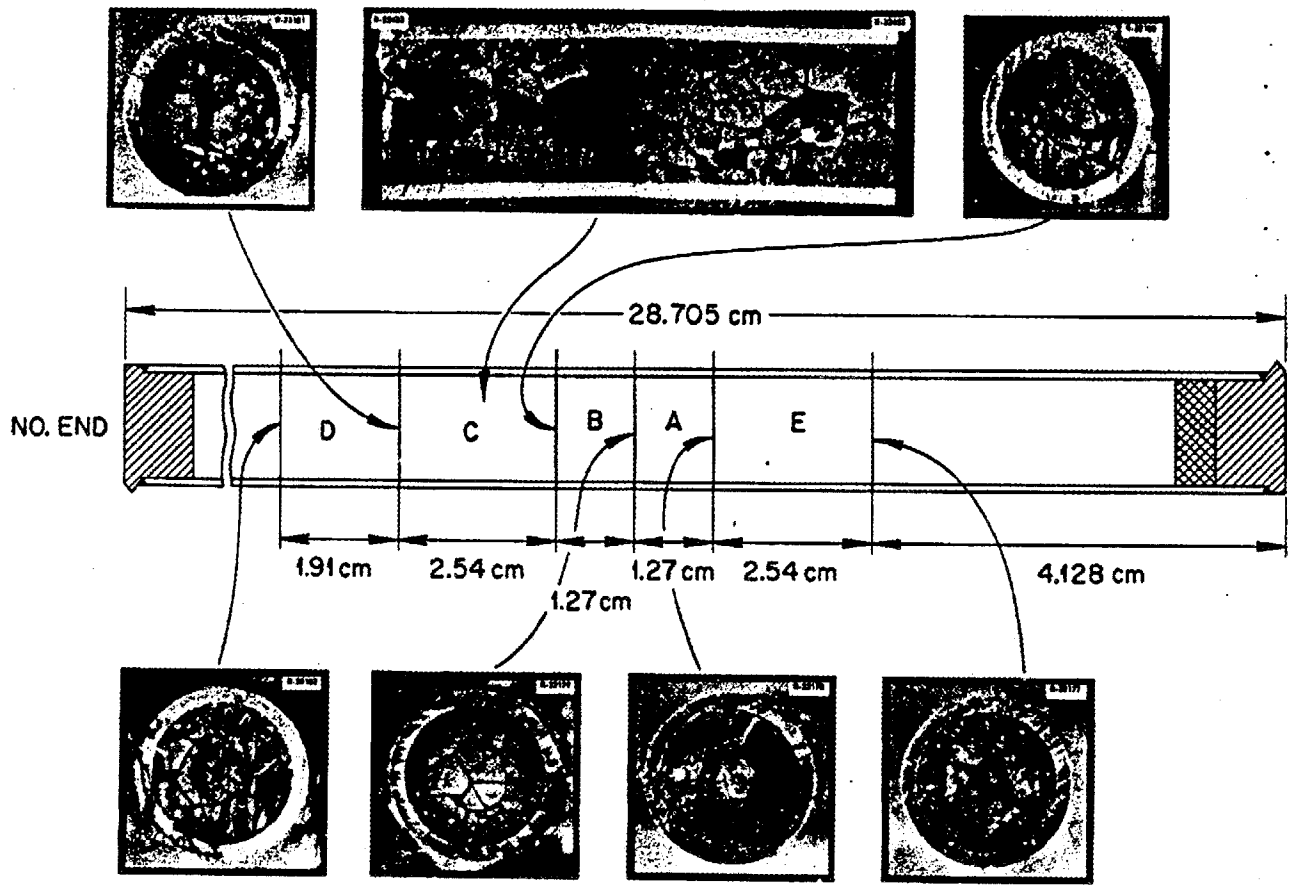


A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION

Fig. 2. MTR Group I, Capsule U-3 – Macroscopic Views of Selected Section Locations, As Cut.

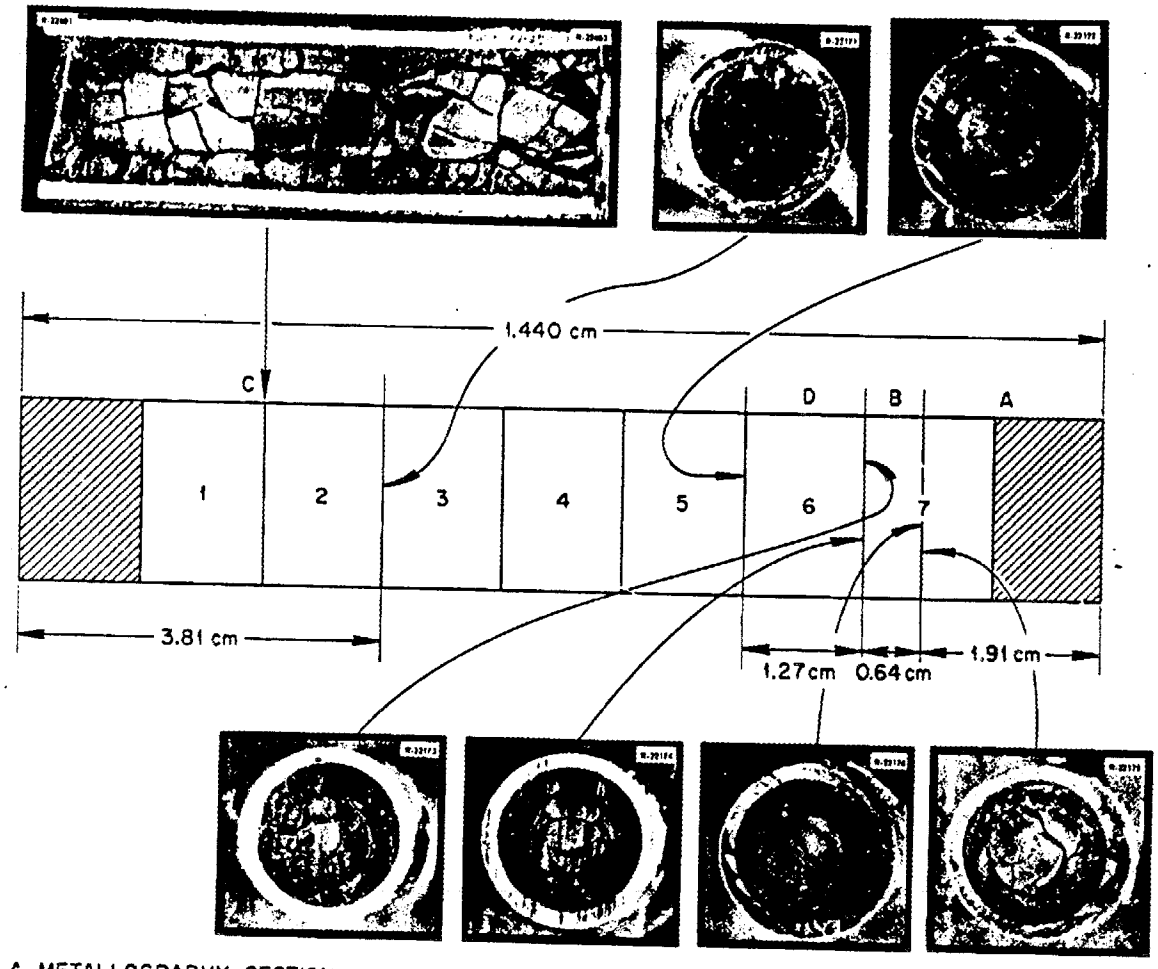
ORNL-DWG 65-2480



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION
E. NEUTRON ACTIVATION SECTION

Fig. 3. MTR Group I, Capsule Z-8 — Macroscopic Views of Selected Section Locations, As Cut.

ORNL-DWG 65-2476



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. NEUTRON ACTIVATION SPECIMEN

Fig. 4. Pellet Rod No. 712 – Macroscopic Views of Selected Section Locations, as Cut.

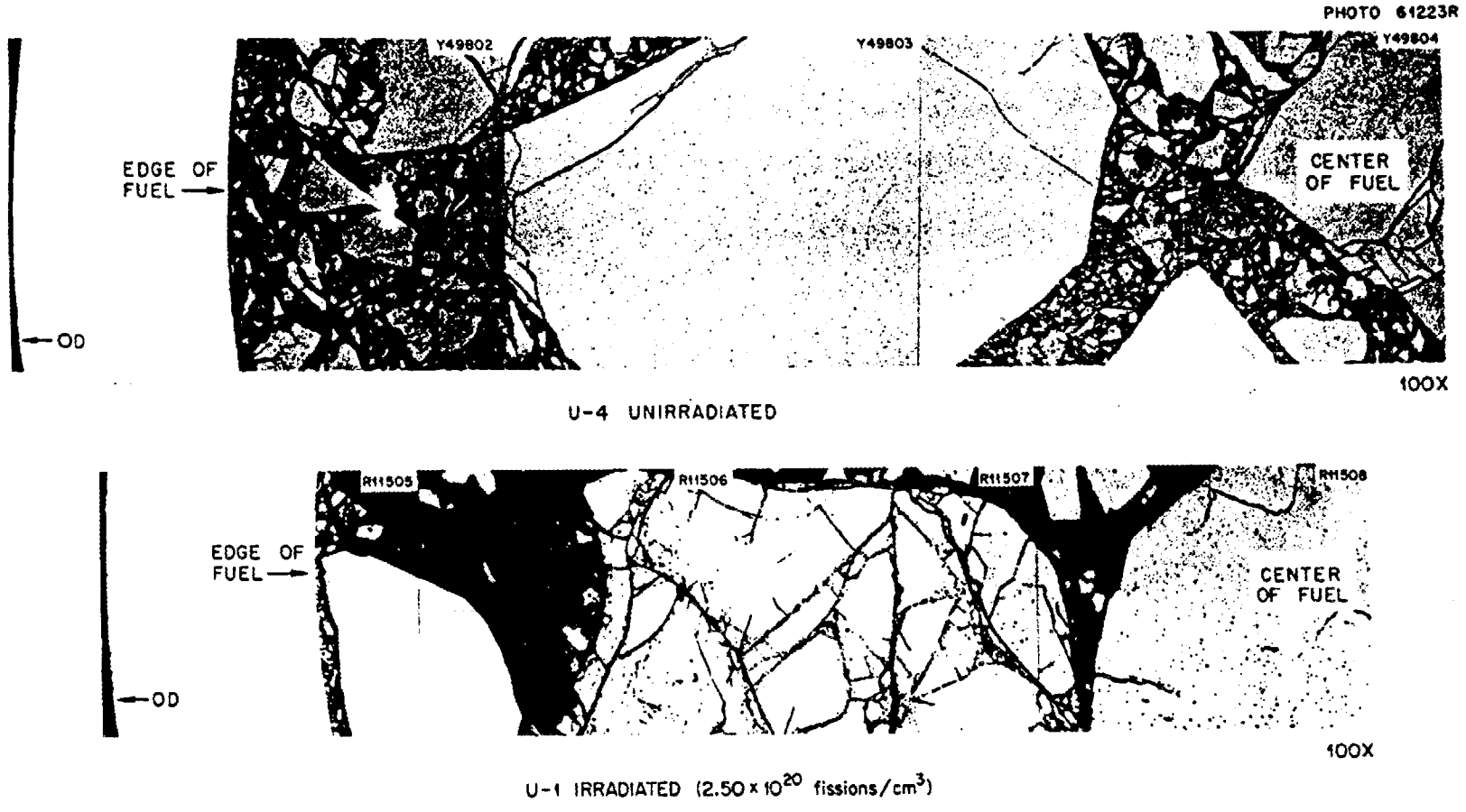
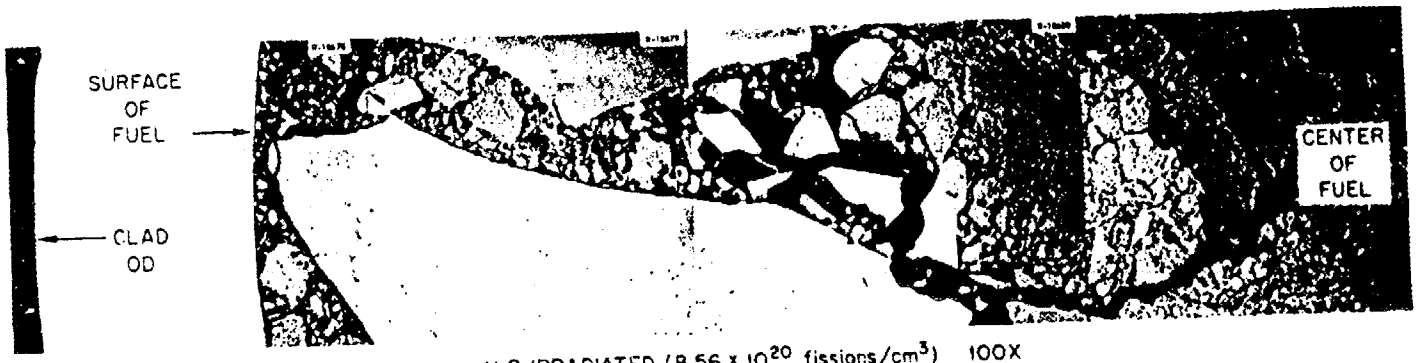
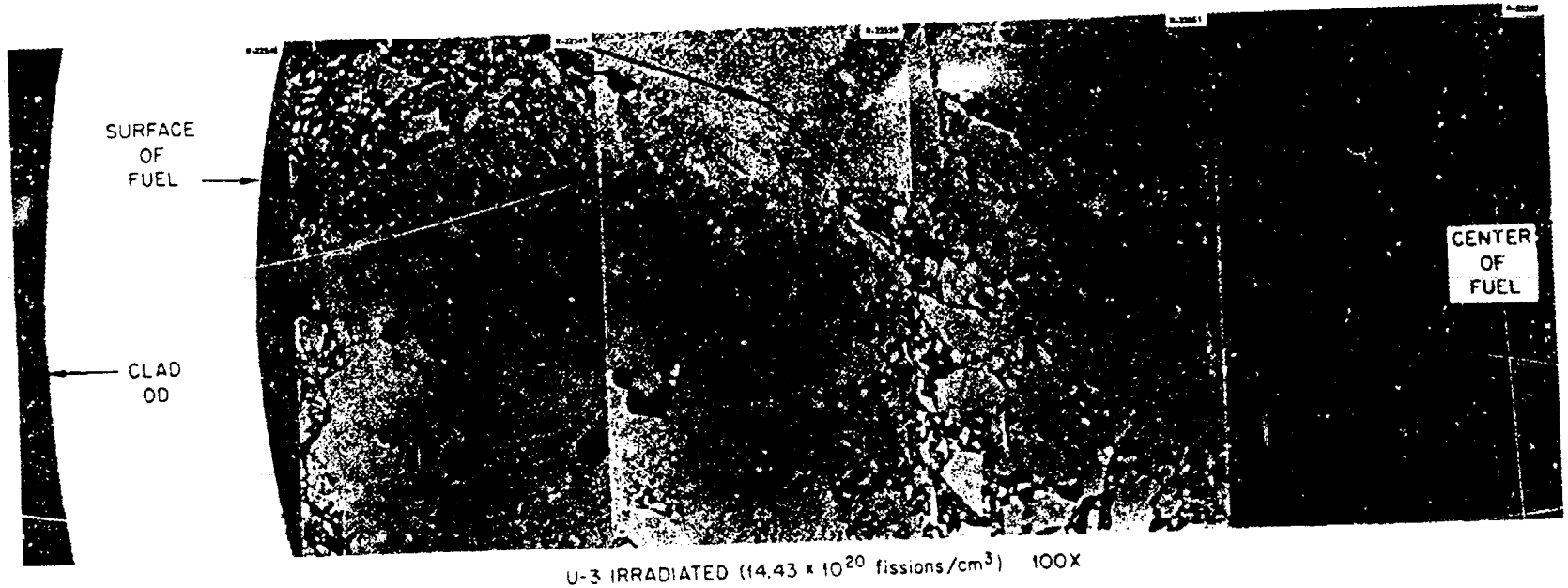


Fig. 5. Composite Photomicrographs Showing Typical Transverse Sections of Unirradiated and Irradiated Vibratorily Compacted Arc-Fused ThO_2–4.5% UO_2. Radial area from the outside diameter to the center of the fuel. As polished.

PHOTO 69435



U-2 IRRADIATED (8.56×10^{20} fissions/cm^3) 100X



U-3 IRRADIATED (14.43×10^{20} fissions/cm^3) 100X

Fig. 6. Composite Photomicrographs Showing Typical Transverse Sections of Irradiated Vibratorily Compacted Arc-Fused ThO_2–4.5% UO_2. Radial area from the outside diameter to the center of the fuel. As polished.

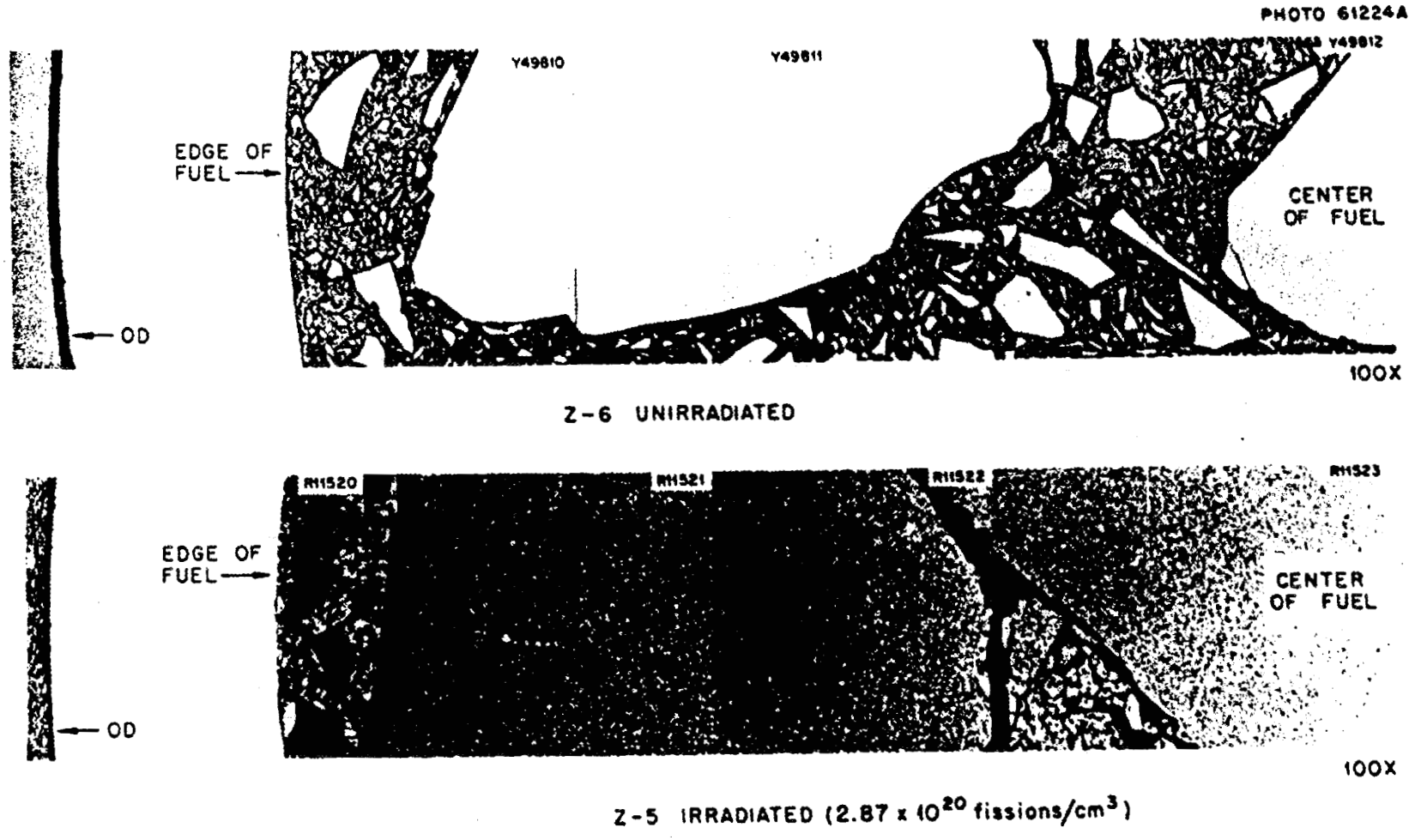
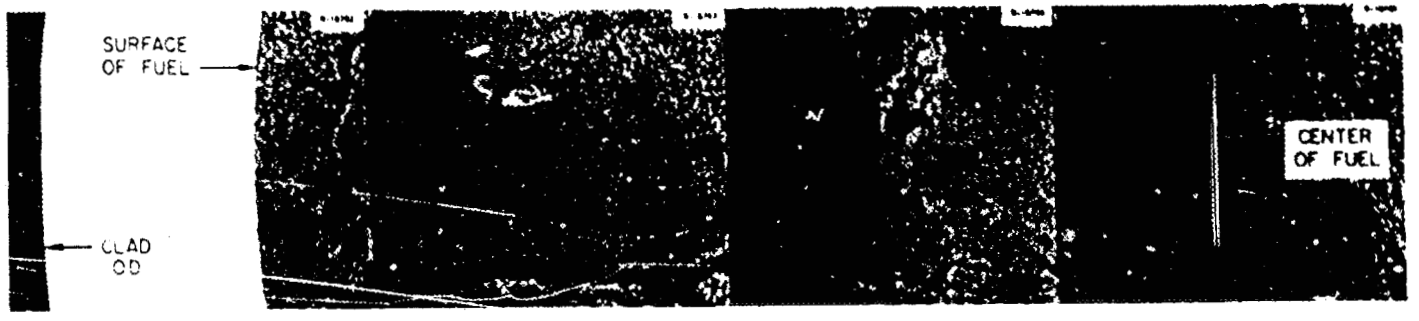
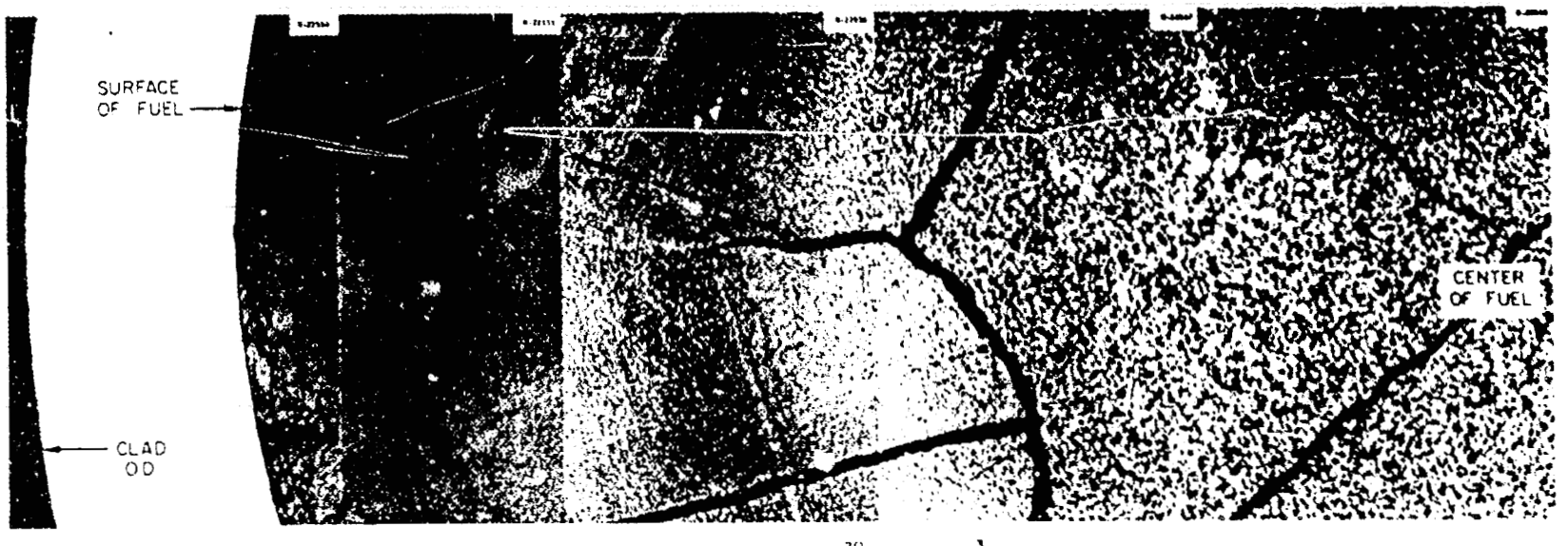


Fig. 7. Composite Photomicrographs Showing Typical Transverse Sections of Unirradiated and Irradiated Vibratorily Compacted Sol-Gel E ThO_2-4.5% UO_2. Radial area from the outside diameter to the center of the fuel. As polished.

PHOTO 69433



Z-7 IRRADIATED (8.23×10^{20} fissions/cm^3) 100X



Z-8 IRRADIATED (16.46×10^{20} fissions/cm^3) 100X

Fig. 9. Composite Photomicrographs Showing Typical Transverse Sections of Irradiated Vibratorily Compacted Sol-Gel ThO_2-4.5% UO_2. Radial area from the outside diameter to the center of the fuel. As polished.

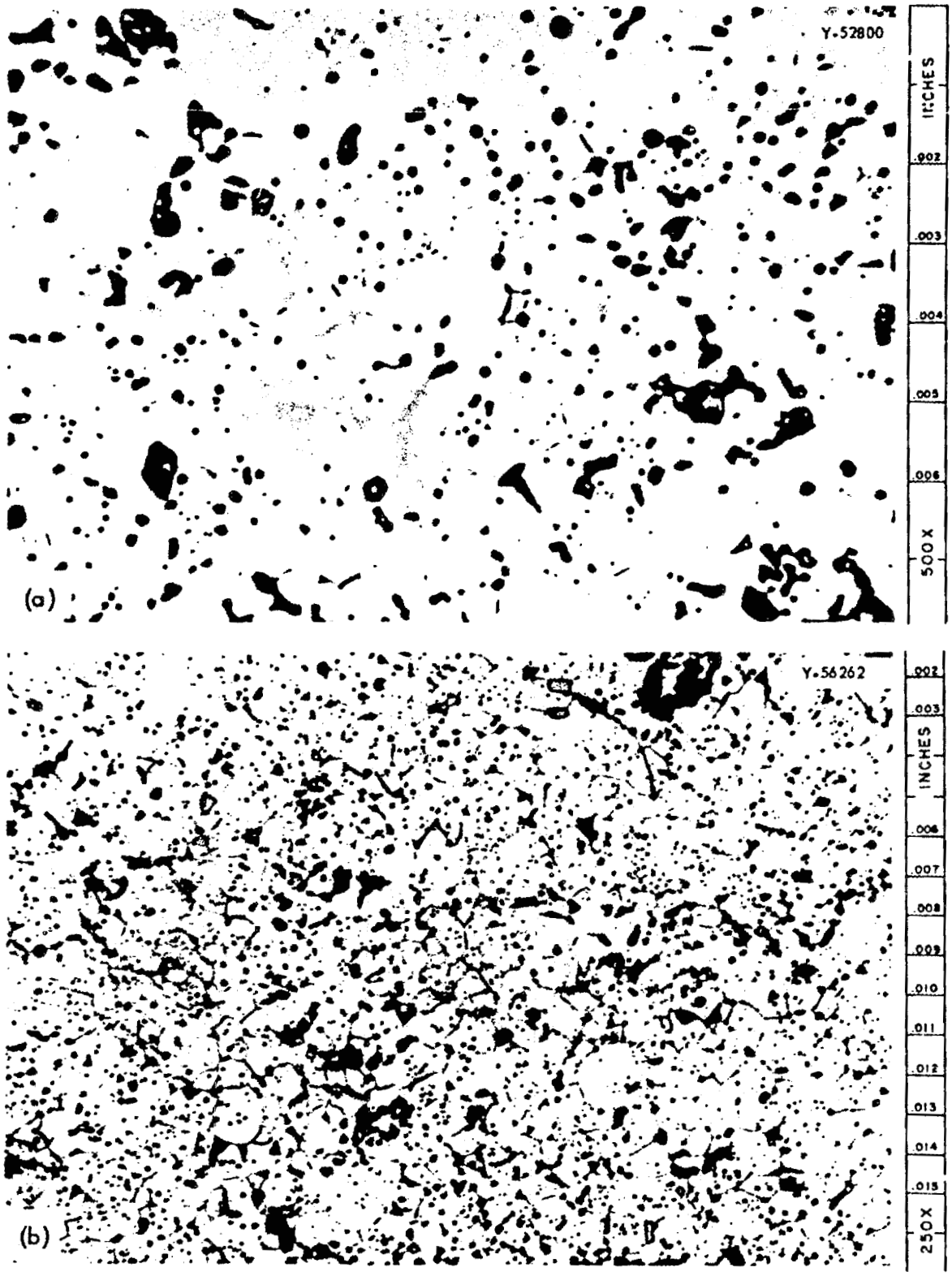


Fig. 9. Unirradiated Microstructure of ThO_2-UO_2 Pellet. Note copious quantity of gray impurity phase. (a) As polished. (b) Etched.

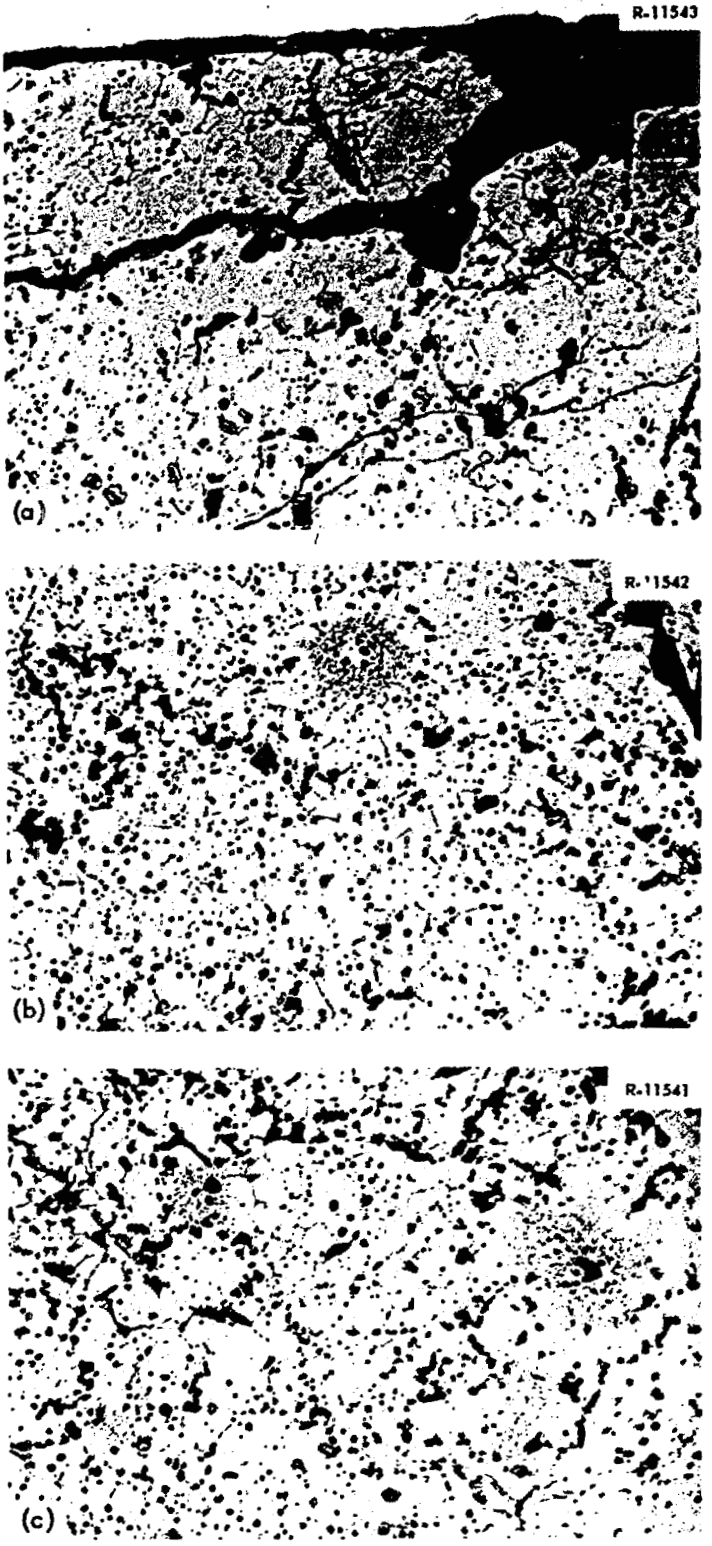


Fig. 10. Representative Regions of Fuel from Fuel Rod 43-41. (10.8×10^{20} fissions/cm^3). (a) Peripheral region. (b) Midradial region. (c) Central region. As polished. 250×. Reduced 16.5%.

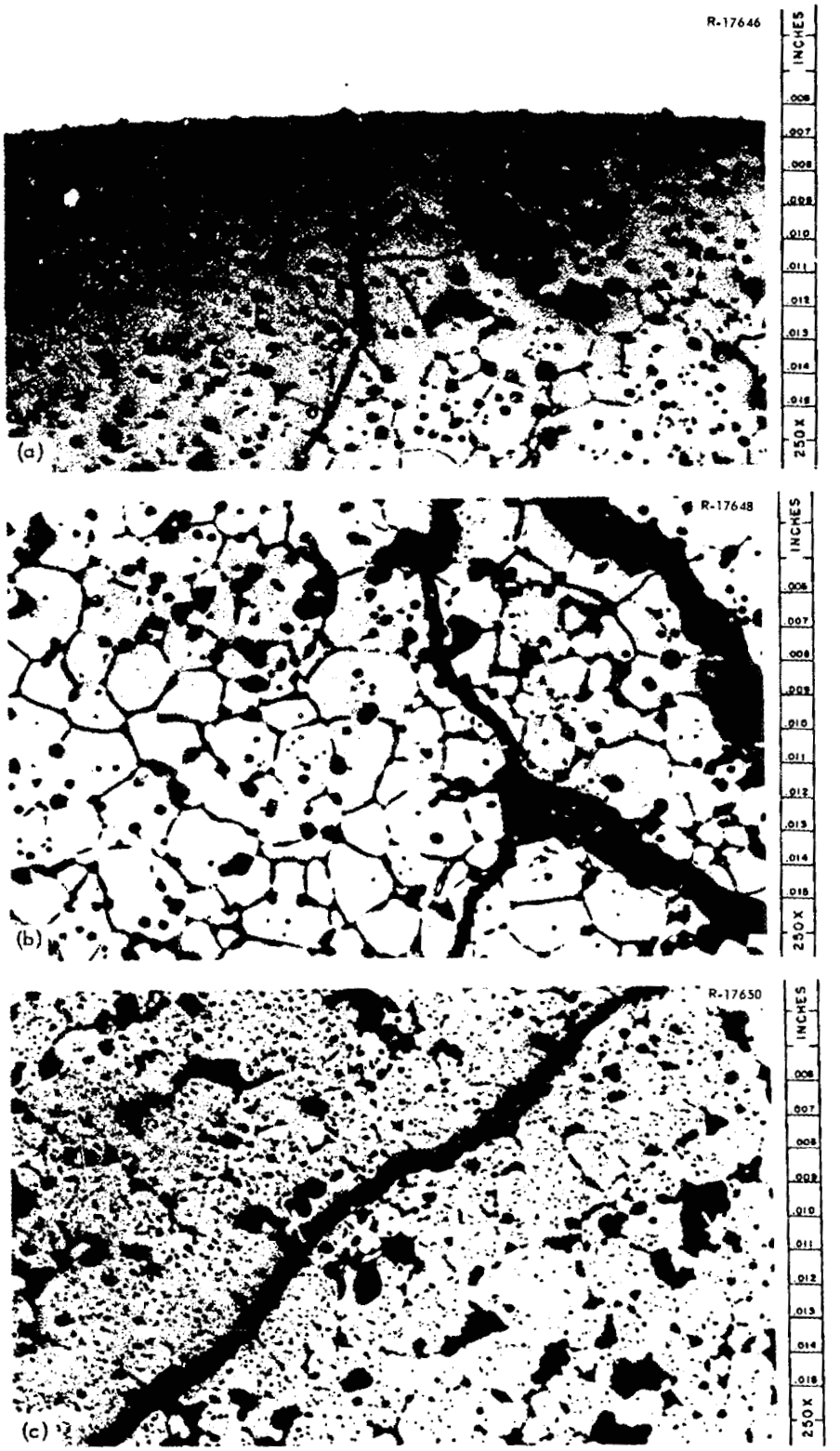


Fig. 11. Representative Regions of Fuel From Fuel Rod 43-44. (12.8×10^{20} fissions/cm^3). (a) Cladding-fuel interface. (b) Midradial region. (c) Central region. As-polished. Reduced 25%.

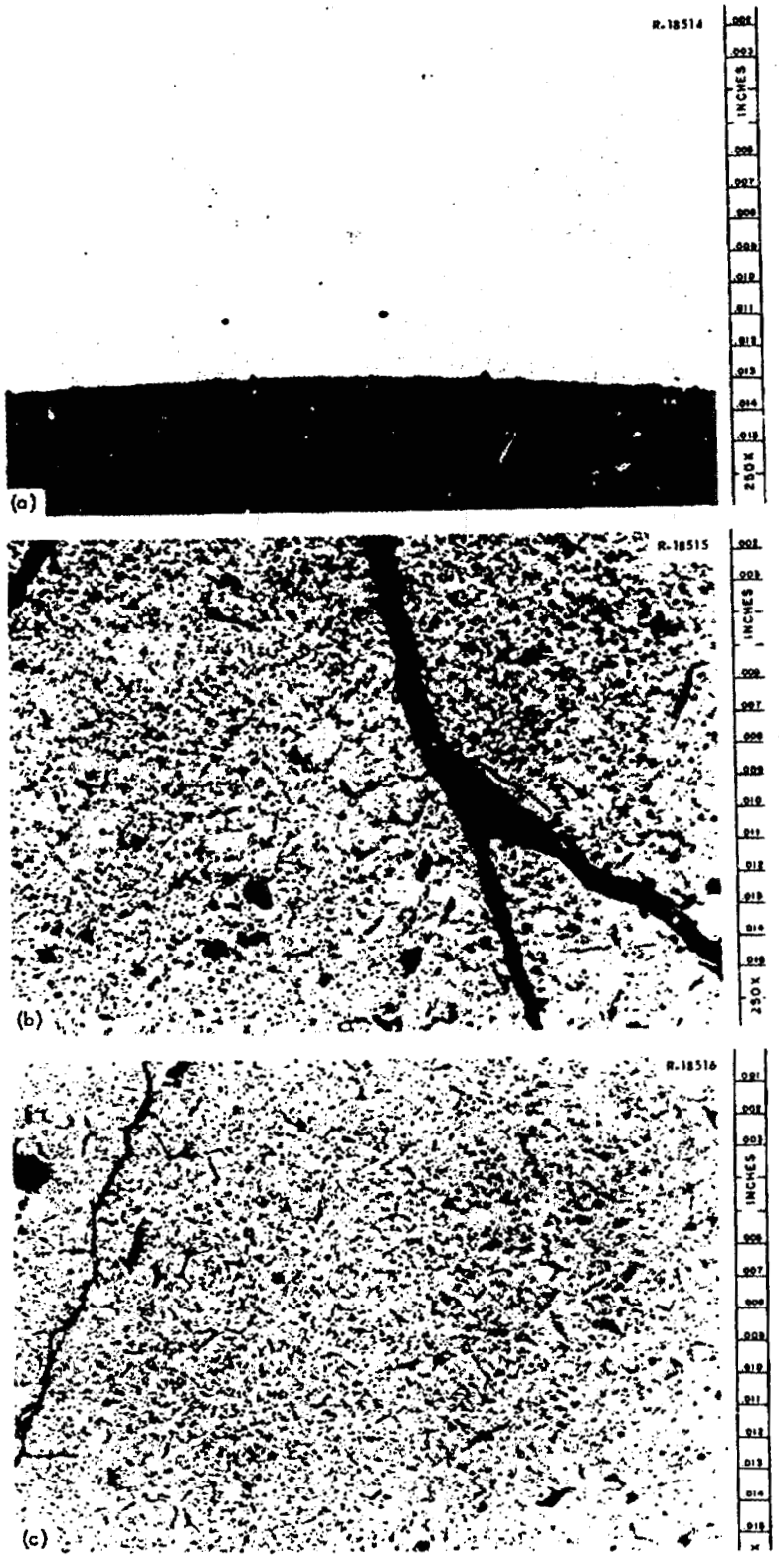


Fig. 12. Representative Areas From Fuel Rod 43-40. (24.6×10^{20} fissions/cm^3). (a) Cladding fuel interface. (b) Central region. As-polished. Reduced 34%.

In the case of the arc-fused material, the cracks in the original fuel fragments were first accentuated in the midradius region while the porosity in the center agglomerated. With continued irradiation, the cracks apparently healed and a grain structure developed within the fuel fragments while the pore sizes in the center increased. At 70,000 Mwd/tonne, 14×10^{20} fissions/cm^3, the central region appears to have developed a dense laminar-like structure with some interparticle sintering and the outer two-thirds shows a uniform dispersion of fine porosity with a band of larger pores at the dividing line between the two structures.

The sol-gel fuel fragments in the unirradiated condition are crack free but do contain uniformly distributed submicroscopic pores. On irradiation, this porosity first appears to agglomerate and accentuate the fine-particle size. Continued irradiation develops a distinct grain structure in the center of the fuel with equiaxed grains 6 to 8 μ in size separated from the balance of the fuel by a circumferentially oriented crack. At 81,000 Mwd/tonne, 16×10^{20} fissions/cm^3, the development of equiaxed grains is very distinct with a maximum size of 60 μ. The circumferential crack still separates this area from the remainder of the fuel, but there appears to be some interfragment sintering in the outer two-thirds and, as with the arc-fused material, a band of moderately large porosity is found in this area.

With both fuels, after irradiation there is a second white metallic-appearing phase developed in the central region. Initially, this appears to be closely associated with porosity but at exposures of 8×10^{20} fissions/cm^3, or 375 reactor full-power days, this phase appears to be randomly distributed throughout the matrix. A third light-grey phase was also found located in discrete limited areas of some specimens. Although this phase has not been definitely identified, the appearance and the association with fine fuel fragments indicate that it is probably aluminum silicate contamination from the ball-milling operations.

The evolution of structural changes in the pressed and sintered pellets is not as clearly defined since an equiaxed grain structure exists before irradiation. As with the other fuels, there is an accumulation of porosity. There is a gradual degradation of the distinct grain structure

starting at the center and spreading out towards the cladding. The one exception to this is the one rod exposed in the ETR where the maximum instantaneous heat flux may have been considerably higher; in this case the central degradiation is apparent but distinct grain growth is noted between the midradius and the cladding. All structures show a second white metallic phase particularly in the central regions. The grey impurity phase seen in the unirradiated, as well as the irradiated, structures has been identified by x-ray diffraction as Al_2O_3 and 6 $Al_2O_3 \cdot CaO$. The effect of these high levels of impurities on the microstructures is not known.

One rod of vibratorily compacted sol-gel fuel is still under irradiation and will be exposed to a burnup in excess of 100,000 Mwd/tonne (Th+U). However, from the findings to date it is apparent that there is no sifnificant difference between the performance of vibratorily compacted sol-gel and arc-fused ThO_2-UO_2 fuels. Both compare favorably with pressed and sintered pellet fuels at linear heat ratings between 300 and 400 w/cm ($\int k d\theta$ 34-45 w/cm). In addition, the ThO_2-5% UO_2 fuels show no evidence of breakaway swelling or fission-gas release at burnups as high as 2.6×10^{21} fissions/cm^3, 120,000 Mwd/tonne, where three-fourths of the fission energy has been derived from the ^{233}U bred into the fuel during irradiation. According to swelling estimates[10] on the pellet rods, the ThO_2-4.5% UO_2 change in volume was limited to 0.46% $\Delta V/10^{20}$ fissions/cm^3. This is roughly one-half the rate proposed by Anderson[11] and others for UO_2 (0.8 $\Delta V/10^{20}$ fissions/cm^3). X-ray diffraction data on the various irradiated ThO_2-UO_2 fuels have also tended to confirm the stability of the cubic fluorite crystal structure lattice and indicate a considerable capacity for fission products, as can be seen from the negligible changes in lattice parameters in Table 3.

Effects of High Heat Ratings

Although the high-burnup test rods have shown the usefulness of the thoria-base fuels at moderate heat ratings comparable to current pressurized water-power reactors, the full utilization of the sol-gel ThO_2-UO_2 fuels requires an investigation of the maximum performance characteristics and

Table 3. Effect of Irradiation on Lattice Parameters of ThO_2-UO_2 Fuels

Group	Rod	Fuel Material[a]	Burnup (fissions/cm^3)	Unit Cell[b] Preirradiation	Unit Cell[b] Postirradiation
			$\times 10^{20}$		
MTR-I	U-2	Arc-fused	8.6	5.594 ± 0.001	5.599 ± 0.009
	U-3	Arc-fused	14.4	5.594 ± 0.001	5.592 ± 0.002
	Z-7	Sol-gel	8.2	5.593 ± 0.001	5.594 ± 0.003
	Z-8	Sol-gel	16.5	5.593 ± 0.001	5.586 ± 0.004
Pellet	645	Pressed and sintered	12.8	5.590 ± 0.001	5.590 ± 0.002
	729	Pressed and sintered	24.6	5.590 ± 0.001	5.584 ± 0.002
	712	Pressed and sintered	26.4	5.590 ± 0.001	5.587 ± 0.002

[a] ThO_2–4.5% UO_2, ^{235}U enriched 93%.

[b] Nelson-Riley Function.

an understanding of the effect of processing variables on these characteristics. The ORR loop and ORR Poolside Experiments were the first steps in this phase of the investigation. More recently, experiment groups MTR-II, MTR-III, and ETR-I were prepared and inserted in the reactor for this purpose. Tables 4a, b and c present the fabrication characteristics, irradiation histories, and principal postirradiation findings on the rods examined to date.

The findings on the ORR loop and Poolside experiments have been reported[5-8], and the ETR-I rods are currently being evaluated. This latter group of three rods was subjected to the highest flux with the expectation of producing central melting. A common sol-gel ThO_2–5.7% UO_2 preparation was used as fuel in these rods, however, it was calcined in three different atmospheres: air, A–4% H_2 (the standard atmosphere), and nitrogen to produce sol-gel batches 35-1, 35-2, and 35-3, respectively. The postirradiation dimensional analyses have shown no indications of swelling or bowing. Fission-gas samples and gamma scans were obtained. Based on the gamma scans, which showed no anomalies, and the structures found, these rods were sectioned and are currently being evaluated metallographically. Figures 13, 14, and 15 show the resulting as-cut structures at the various section locations.

From these sections, it is apparent that the linear heat ratings were sufficient to develop a central void in all three of the rods. The levels of the gamma scans and the results of the burnup analysis indicate that there was a real difference in fission rate and consequently in linear heat rating as shown in Tables 4b and c. The extent of the central voids appears to match the indicated heat ratings, but the lack of any anomalies in the gamma scans and the appearance of the sections indicate that the anticipated condition of central melting was not achieved although columnar grain growth is significant. Columnar grain growth has been shown to occur without melting in UO_2 (ref. 12) and in ThO_2-UO_2 in the ORR loop experiment L-1-B (ref. 7), but the probability of the effective thermal conductivity of the as-packed oxides being high enough to permit this in these experiments at full power was unlikely. Examination of the reactor startup on the first cycle of exposure of these rods shows that for the first 9 1/2 hr of exposure the reactor was

Table 4a. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO_2-UO_2 Fuel Rods: Description of Irradiation-Test Fuel Rods

Experiment	Fuel Rod Identification	Type of Oxide	Total Uranium[a] (wt %)	Cladding	Oxide Particle Size	Vibrated Density (% TD)[b]	Fuel Rod Dimensions (cm) Length	Outside Diameter	Wall
ORR Loop	L-1-A	Sol-gel 26	5.35	304 Stainless Steel	c	85.2	54.61	1.168	0.0381
	L-1-B	Sol-gel 26	5.35	Zircaloy 2	c	84.1	54.61	1.168	0.0381
	L-1-C	Sol-gel 26	5.35	Zircaloy 2	c	84.1	54.61	1.168	0.0381
ORR Poolside	03-5	Sol-gel D	2.50	304H Stainless Steel	d	84.8	17.80	1.587	0.0508
	03-6	Sol-gel D	2.50	304H Stainless Steel	d	85.5	17.78	1.587	0.0508
ETR-I 43-80	18	Sol-gel 35-1	5.02	304 Stainless Steel	e	88.1	30.48	1.111	0.0635
43-80	10	Sol-gel 35-2	5.02	304 Stainless Steel	e	89.4	30.48	1.111	0.0635
43-81	8	Sol-gel 35-3	5.02	304 Stainless Steel	e	85.1	30.48	1.111	0.0635

[a]Enriched in ^{235}U to 93%.

[b]Based upon a calculated density depending on composition (approx 10.04 g/cm^3).

[c]Distribution H: 60 wt % −6 +16 mesh; 25 wt % −50 +140 mesh; 15 wt % −200 mesh.

[d]Distribution A: 60 wt % −10 +16 mesh; 15 wt % −70 +140 mesh; 25 wt % −200 mesh.

[e]Distribution C: 60 wt % −8 +16 mesh; 15 wt % −70 +100 mesh; 25 wt % −200 mesh.

Table 4b. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO_2-UO_2 Fuel Rods: Irradiation History

Experiment	Fuel Rod Identification	Cladding Temperature (°C)	Average External Pressure (psia)	Exposure Time Reactor Full-Power Days	Peak Linear Heat Rating		Peak Cladding Heat Flux		Maximum Burnup[a]	
					Time Average (w/cm)	Maximum Instantaneous (w/cm)	Time Average (w/cm²)	Maximum Instantaneous (w/cm²)	(Mwd/tonne Th+U)	(fissions/cm³) × 10^{20}
ORR Loop	L-1-A	260	1750	29.6	381	389	104	106	1,600	0.34
	L-1-B	260	1750	29.6	499	509	136	139	2,100	0.45
	L-1-C	260	1750	29.6	410	418	112	114	1,730	0.38
ORR Poolside	03-5	705	315	215.2	304	337	61	68	5,220	1.13
	03-6	540	315	215.2	275	297	55	60	4,040	0.88
ETR-I	18	100	180	140.5	870	1050	304	367	20,400	4.28
	10	100	180	140.5	865	1044	302	365	20,000	4.26
	8	100	180	140.5	914	1103	319	385	22,000	4.46

[a]Based on 200 Mev/fission.

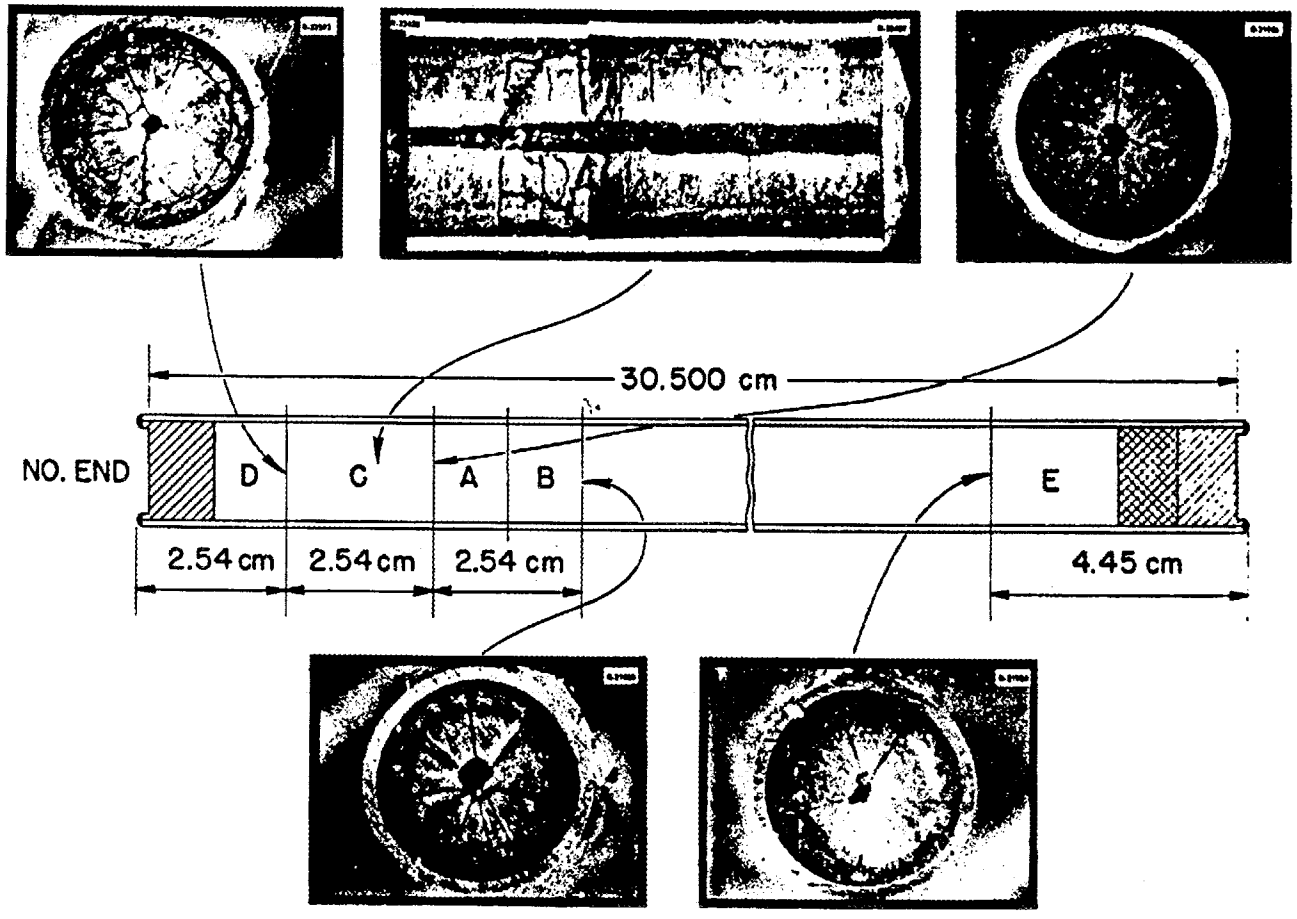
Table 4c. Effects of High Heat Ratings on Powder Compacted Sol-Gel ThO_2-UO_2 Fuel Rods: Postirradiation Observations and Calculations

Experiment	Fuel Rod Identification	$\int_{T_o}^{T_c} k d\theta$ Time Average (w/cm)	$\int_{T_o}^{T_c} k d\theta$ Maximum Instantaneous (w/cm)	Release (%)	Dimensional Changes Average (mm)	Dimensional Changes Maximum (mm)	Radial Extent of Microstructural Changes: Void (cm)	Radial Extent of Microstructural Changes: Columnar Grain Growth (cm)	Equiaxed Grains (cm)	Various Changes[a] (w/cm): $\int_{T_o}^{T_v} k d\theta$	Various Changes[a] (w/cm): $\int_{T_o}^{T_{cg}} k d\theta$	Various Changes[a] (w/cm): $\int_{T_o}^{T_{eg}} k d\theta$
ORR Loop	L-1-A	46.5	47.4	2.3	+0.025	+0.102			0.311			31.2
	L-1-B	57.0	58.1	Failure	0.000	+0.229		0.152	0.406		54.2	35.4
	L-1-C	49.6	50.6	3.9	0.000	+0.102			0.330			38.2
ORR Poolside	03-5	54.5	60.5	18.3	−0.025	−0.229			0.660			35.5
	06-5	46.7	50.4	Gas lost	0.000	+0.051			b			b
ETR-I	18	82.3	99.4	38.6	+0.025	+0.051	0.076	0.340	0.346	76.5	49.3	48.2
	10	82.2	99.2	27.8	+0.037	+0.056	0.04	0.394	0.415	79.9	38.1	33.3
	8	86.2	104.0	21.4	+0.027	+0.041	0.109	0.383	0.396	75.3	42.2	39.3

[a]Time average value.

[b]Sintering boundary not delineated.

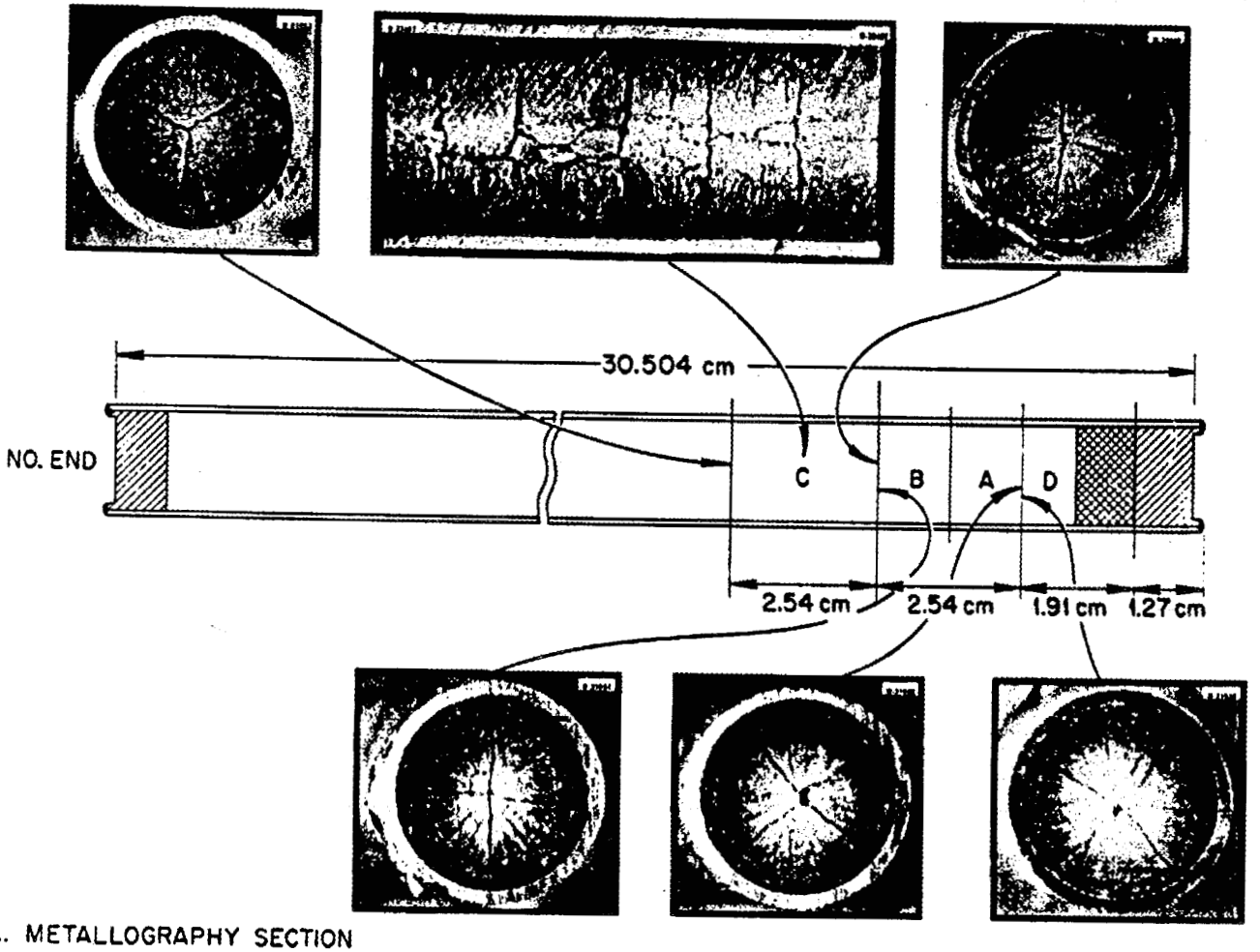
ORNL-DWG 65-2474



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION

Fig. 13. ETR Group I, Capsule 18 – Macroscopic Views of Selected Specimen Locations, As Cut.

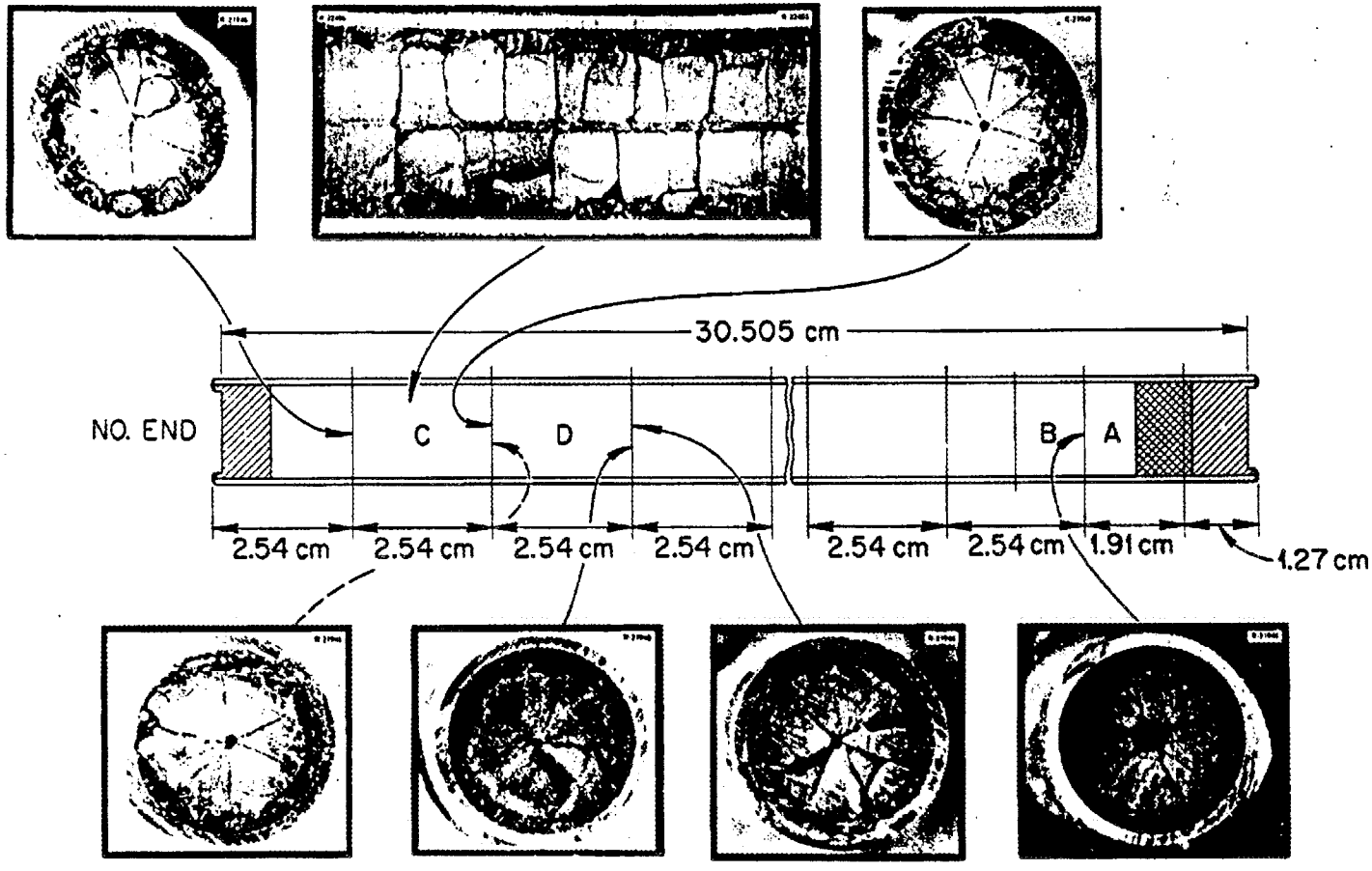
ORNL-DWG 65-2473



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE

Fig. 14. ETR Group I, Capsule 10 – Macroscopic Views of Selected Section Locations, As Cut.

ORNL-DWG 65-2470



A. METALLOGRAPHY SECTION
B. BURNUP SECTION
C. LONGITUDINAL SECTION
D. SPARE LONGITUDINAL SECTION

Fig. 15. ETR Group I, Capsule 8 — Macroscopic Views of Selected Section Locations, As Cut.

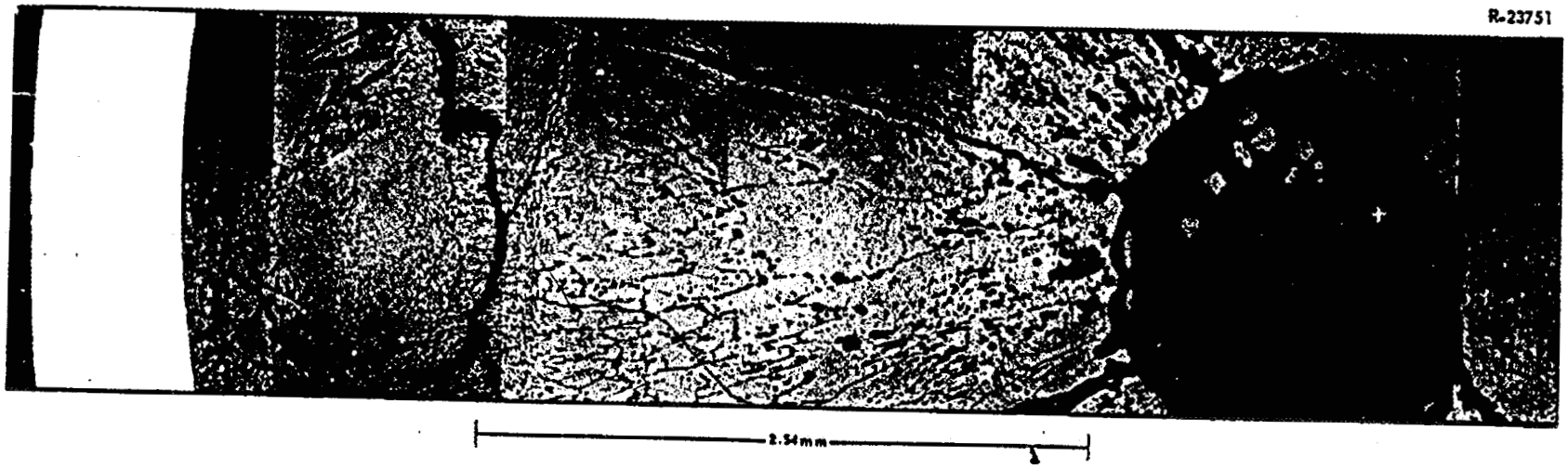
never over 50% of full power. Since this would have produced an $\int k d\theta$ very close to the 54 w/cm defined by experiment L-1-B for columnar grain growth it is entirely possible that during this time sufficient sintering and grain growth occurred to raise the effective thermal conductivity, develop a central void, and prevent melting. Whether or not this is the case, it is apparent that at a linear heat rating in excess of 1000 w/cm the maximum performance capabilities of these vibratorily compacted sol-gel fuels have not been reached.

The calcining atmosphere appears to influence the fission-gas release rates. This conclusion will need verification from the lower rated rods in the MTR-III group which contain the same fuels but are still under irradiation. Other experiments at Chalk River[13] have shown an effect on grain growth in hydrogen-sintered and air-sintered ThO_2-UO_2 pellets when subjected to short-time rabbit type irradiation tests, with the air-fired material showing more grain growth.

The microstructures on the ETR-I rods, shown in Figs. 16, 17, and 18, are similar to those developed in UO_2 fuel rods at high heat ratings.

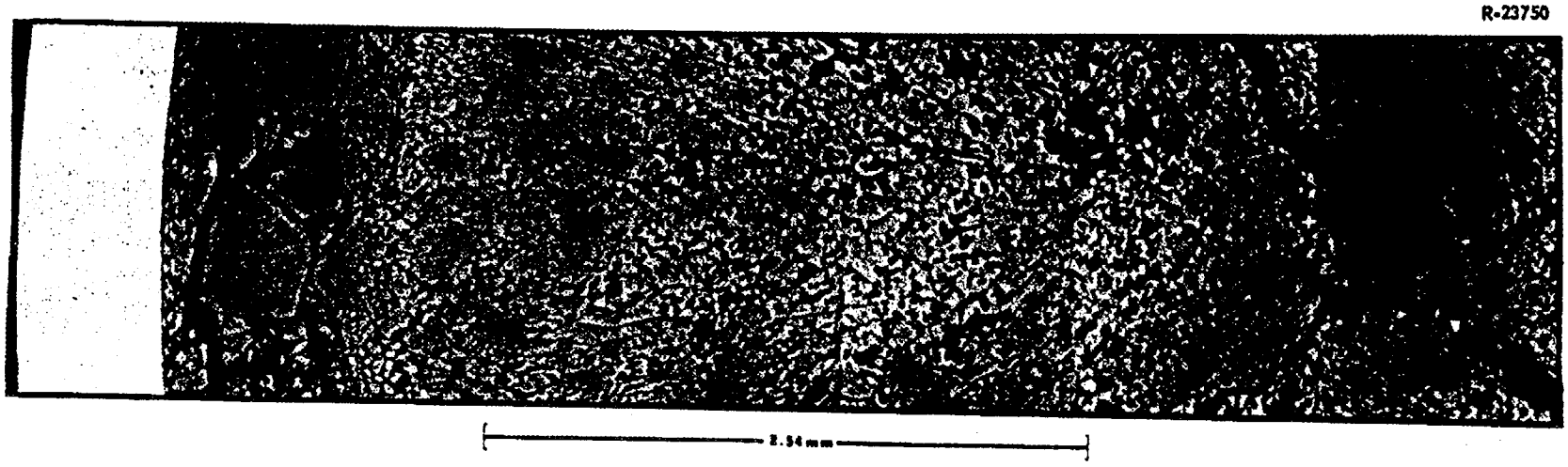
Although the maximum performance characteristics have not been defined, it would appear that sufficient data are available for a preliminary comparison of ThO_2-UO_2 and UO_2 fuels. The most common method of comparison for bulk fuels of this type is the use of the $\int k d\theta$ values for specific microstructural changes.[14] A review of the very extensive literature on UO_2 experiments will show that there are considerable differences of opinion among the various investigators on the precise values of $\int k d\theta$ for various structural changes. These differences develop primarily from the treatment of the fuel-to-cladding conduction evaluation, the interpretation of the extent of melting, and the thermal conductivity value chosen for temperatures below about 500°C. The temperatures for equiaxed grain growth (1500°C) and columnar grain growth (1700°C) in UO_2 are, however, fairly well established but they have not been investigated for ThO_2-UO_2 fuels as yet.

Because of the differences in UO_2 experimental interpretations and the sparsity of data on ThO_2-UO_2 fuels an absolute comparison is impossible. However at ORNL vibratorily compacted fuel rods of both UO_2 and ThO_2–6% UO_2 have been tested in the pressurized loop at essentially identical conditions.



CAPSULE 18

Fig. 16. Composite Photomicrograph of Vibratorily Compacted Sol-Gel ThO_2-UO_2 Calcined in Air and Irradiated to 4.28×10^{20} fissions/cm^3 at a Linear Heat Rating of Approximately 1,000 w/cm.



CAPSULE 10

Fig. 17. Composite Photomicrograph of Vibratorily Compacted Sol-Gel ThO_2-UO_2 Calcined in Ar–4% H_2 and Irradiated to 4.26×10^{20} fissions/cm^3 at a Linear Heat Rating of Approximately 1000 w/cm.

Fig. 18. Composite Photomicrograph of Vibratorily Compacted Sol-Gel ThO_2-UO_2 Calcined in H_2 and Irradiated to 4.46×10^{20} fissions/cm^3 at a Linear Heat Rating of Approximately 1000 w/cm.

The UO_2 work has been reported[15–16] and the data listed in Table 5 were derived from these reports. All $\int k d\theta$ values are quoted to the fuel surfaces which should have been at essentially the same temperature.

The most obvious difference is the lack of void formation in the ThO_2-UO_2 loop rods at $\int k d\theta$ values where significant voids were formed in the UO_2 rods. To illustrate void formation in ThO_2-UO_2 the ETR-I rods which operated at lower surface temperatures but at higher power levels and to much higher burnups have been included in this table. The extent of columnar grain growth is also fairly well defined in postirradiation microstructures and again the $\int k d\theta$ values show the ThO_2-UO_2 material to be better in the comparable loop exposures. The extent of equiaxed grain growth or sintering is difficult to determine in these powder compacts but it would appear that similar temperatures and $\int k d\theta$ values apply to ThO_2-UO_2 and UO_2 fuels.

Any attempt to place a quantitative value on the superiority of ThO_2-UO_2 compacts from these data would be highly speculative. However, Rao[13] at Chalk River has compared 3-min rabbit tests of UO_2 and ThO_2-UO_2 using pressed and sintered pellets. He has concluded that similar microstructural changes in ThO_2–1% UO_2 compared with UO_2 will require a 10% higher power output. Such short time tests have proven to be good guides in the past and from the data reported here it appears that his estimate may prove to be conservative. Irradiation tests at higher linear heat ratings are planned and additional out-of-pile data are being obtained including thermal simulation and thermal conductivity measurements. These out-of-pile tests are needed to remove some of the uncertainties associated with evaluation of the microstructures from in-pile tests.

Mixed Progeny and Remotely Fabricated Sol-Gel ThO_2-$^{233}UO_2$ Fuel Tests

We have done very little testing with mixed progeny fuels but three fuel rods containing sol-gel ThO_2 mixed with PuO_2 before calcining and tamp-packed into fuel rods have been examined after exposure at linear heat ratings up to 245 w/cm to burnups of 29,000 Mwd/tonne (Th+Pu). The fission-gas release rates were less than 5% and the microstructures were similar to ThO_2-UO_2 exposed under the same conditions.

Table 5. Comparison of UO_2 and ThO_2-UO_2 Vibratorily Compacted Fuel Rods

Experiment		Fuel Material[a]	Fuel Density (% TD)	Burn Up Mwd/tonne Heavy Element	Inside Surface Temperature of Cladding (°C)	Linear Heat Rating (w/cm)	$\int_s^c k d\theta$[b] (w/cm)	$\int_s^v k d\theta$ (w/cm)	$\int_s^{cg} k d\theta$ (w/cm)	$\int_s^{eg} k d\theta$ (w/cm)	^{85}Kr Release (%)
ORR Loop[c]	7N1	UO_2	87.1	4,780	353	434		32.5	20.7		12.4
	7O1	UO_2	87.1	5,140	361	465		34.8	21.1		47.0
	7P1	UO_2	86.9	6,290	383	566		41.2	20.0		72.0
	8N1	UO_2	85.6	4,810	347	404		30.9	22.8	17.5	77.0
	8O1	UO_2	85.8	5,040	351	423		31.3	21.2	16.1	25.0
	8P1	UO_2	85.5	6,880	387	578		40.8	23.9	20.3	25.0
ORR Loop	L1A	ThO_2–6% UO_2	85.2	1,600	341	381	30.3	No Void		20.5	2.3
	L1B	ThO_2–6% UO_2	84.1	2,100	368	499	39.7	No Void	36.6	17.7	
	L1C	ThO_2–6% UO_2	84.1	1,730	348	410	32.6	No Void		20.7	3.9
ETR-I	18	ThO_2–5% UO_2	88.1	20,400	211	870		63.0	35.8	34.7	38.0
	10	ThO_2–5% UO_2	89.4	20,000	209	865		66.4	24.6	19.8	28.0
	8	ThO_2–5% UO_2	85.1	22,000	218	914		61.3	28.2	25.3	21.0

(a) UO_2 fuel was arc-fused crushed and vibratorily compacted; oxygen-to-uranium ratio 2.002:2.003; UO_2 enriched 5 to 6%. ThO_2-UO_2 fuel was sol-gel material crushed and vibratorily compacted; UO_2 enriched 93%.

(b) $\int_s^c$ is center to surface of fuel; $\int_s^v$ is void to surface of fuel; $\int_s^{cg}$ is the limit of columnar grain growth to surface of fuel; and $\int_s^{eg}$ is the limit of discernible equiaxed grain growth to surface of fuel.

(c) V. O. Haynes, W. C. Thurber, and E. L. Long, Jr., "Fuel Irradiation Tests," pp. 61–77, Maritime Reactor Program Ann. Progr. Rept. Nov. 30, 1963, USAEC Report ORNL-3775, Oak Ridge National Laboratory.

The semiremotely fabricated, vibratorily compacted rods containing sol-gel ThO_2-$^{233}UO_2$ (ETR Group III) have been in the ETR for four months and appear to be performing satisfactorily.

Summary

A rather extensive series of irradiation tests, over the past five years has shown that sol-gel vibratorily compacted ThO_2–5% UO_2 fuel rods perform as well as similar arc-fused ThO_2-UO_2 fuel rods and rods containing pressed and sintered pellets of ThO_2-UO_2 at burnups up to 80,000 Mwd/tonne. There has been no evidence of breakaway swelling or sudden increases in fission-gas release in these rods at linear heat ratings between 300 and 350 w/cm ($\int k d\theta$ 32–38 w/cm).

Although the maximum performance characteristics have not been determined, sol-gel ThO_2–5% UO_2 vibratorily compacted fuels have been operated at linear heat ratings of 1000 w/cm ($\int k d\theta$ of 100 w/cm) to burnups over 20,000 Mwd/tonne with no evidence of swelling, some void formation, but no central melting and with fission-gas release rates less than 30%.

It appears that ThO_2-base fuels with low UO_2 contents will permit significantly higher power densities than similar metal-clad UO_2 fuels and that vibratory compaction is a satisfactory rod fabrication technique.

THORIUM-CONTAINING PYROLYTIC-CARBON-COATED PARTICLE FUELS

Several irradiation tests of thorium-bearing fuels in the form of carbides and oxides have been conducted in support of high-temperature gas-cooled reactor design requirements. The fuel has all been in the form of small particles coated with pyrolytic carbon for fission-product retention; both loose beds of particles and graphite matrix elements have been irradiated. Most ORNL irradiations of HTGR fuels to date have utilized high concentrations of uranium to obtain accelerated burnup and experience with thorium is limited. Much of the testing of thorium-bearing fuels has been related to pebble-bed reactor application, first for the PBRE concept[17] and later for the German AVR.[18] Some recent tests have been in support of the TARGET[19] reactor fuel concept.

Coated-Particle Capsule Irradiation Tests

In the development of these fuels it has proven desirable first to evaluate the particles separately and then the fuel element assembly. Significant results of irradiation tests on unsupported coated particles at ORNL have been described in detail,[20,21] except for the results of the more recent tests involving coated oxide particles. The test facilities have included static capsules inserted in the Low-Intensity Test Reactor and the Materials Test Reactor, sweep capsules in the Oak Ridge Research Reactor, and, for coated oxide particles only, the ORR loop No. 1 facility.[3]

The results of static tests have generally corroborated those of sweep tests. Results from sweep facilities are rather emphasized in this discussion because in these tests a continuous account of the behavior of the test material is provided by analysis of the helium gas that is passed through the capsule. This gas is monitored continuously for total activity and sampled periodically for fission-gas analysis. Descriptions of the coated particles, the techniques used, and their characterization in coating application have been reported in detail.[22–25] Information on performance tests involving multilayer coatings on uranium and uranium-thorium is listed in Table 6 and the characterization of coatings in Table 7. The results are summarized below.

Coated Carbide Particles.

The following conclusions were drawn from extensive testing of coated, high-density UC_2 and $(Th,U)C_2$ fuel particles with nominal particle diameters of 200 μ; (1) multilayer coatings are superior to the monolithic coatings tested; (2) the performance of monolithic coatings with average thicknesses of 50 to 75 μ is substantially inferior to that for 100-μ-thick coatings; (3) spheroidal particles generally perform better than irregularly shaped particles with sharp edges and protrusions; (4) in experiments in which no coatings have ruptured during irradiation, the fission-gas release, expressed as the ratio of the release rate to the birth rate (R/B) for ^{88}Kr, has been of the order of 10^{-5} to 10^{-7} and can be accounted for by the uranium contamination in the coatings as determined before irradiation by alpha-counting; and

Table 6. Irradiation Test Conditions and Fission-Gas Release Data for Unsupported Coated Fuel Particles with Multilayer Pyrolytic-Carbon Coatings

Experiment	Sample Designation	Type of Fuel Particles	Coating Structure	Test Temperature (°C)	Burnup (at. % heavy metal)	Ratio of R/B for ^{88}Kr
C1-11	GA-310	$(Th,U)C_2$	Triplex[a]	1120[b]	14.7	3.6×10^{-6}
B9-10	NCC-AD	UC_2	Duplex[c]	1150	2.6	2.4×10^{-6}
B9-11	NCC-208-2	$(Th,U)C_2$[d]	Duplex	1120	10.0	1.3×10^{-6}
B9-14	GA-309	UC_2	Triplex	1370	20.6	1.6×10^{-5}
C1-15	GA-314	$(Th,U)C_2$[e]	Triplex	1400	8.9	3.5×10^{-5}
B9-15	GA-309	UC_2	Triplex	1370	18.7	4.0×10^{-5}
B9-16	NCC-216	UC_2	Duplex	1370	18.8	1.0×10^{-5}
B9-17	OR-138	UC_2	OR-2D[f]	1370	14.9	1.1×10^{-5}
B9-19	OR-206	$(Th,U)O_2$[g]	OR-2D	1200	0.6	2.3×10^{-6}
B9-20	OR-201	UO_2	OR-2D	1370	4.2	1.5×10^{-6}
A9-2	OR-298	UO_2	OR-2D	1400	12.0[i]	2.0×10^{-7}
Loop 1-14[j]	OR-206	$(Th,U)O_2$[g]	OR-2D	1370	2.7[j]	2.5×10^{-5}

[a]W. O. Harms, "Carbon-Coated Carbide Particles as Nuclear Fuels," in Modern Ceramics – Some Principles and Concepts, ed. by J. Hove and W. Riley, John Wiley & Sons, Inc., New York (in press); D. W. Dayton, W. V. Goeddel, and W. O. Harms, "Ceramic-Coated Particle Nuclear Fuels," A/Conf.28/P/235, paper presented at the 3rd United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964; Final Report, Graphite-Matrix Fuels Development for the AVR, p. 8, USAEC Report GA-4695, General Atomic, October 1963.

[b]Periodic thermal cycles were applied between 925 and 1315°C.

[c]R. A. Reuter, "Duplex Carbon-Coated Fuel Particles," Nucl. Sci. Eng. 20(2), 219–226 (1964).

[d]Thorium-to-uranium ratio 0.6:1.0.

[e]Thorium-to-uranium ratio 2.1:1.0.

[f]Two-layer coatings. Ref: W. O. Harms and D. B. Trauger, Fabrication Variables, Performance, and Cost Considerations for HTGR Coated-Particle Fuels, USAEC Report ORNL-TM-1123, Oak Ridge National Laboratory, April 1965. Properties are described in Table 7.

[g]Particles contain 8 wt % UO_2 and were prepared by the ORNL sol-gel process.

[h]Test terminated due to thermocouple failure.

[i]Test still in progress.

[j]This test is described in a later section of this report. About 9 months of irradiation was accumulated at conditions indicated.

Table 7. Characterization of OR-2D Coatings Deposited from Methane and Listed in Table 6.

Sample Designation	OR-138	OR-182	OR-201	OR-205	OR-206	OR-298
Type of Fuel Particle	UC_2	$(Th,U)O_2$	UO_2	ThO_2	$(Th,U)O_2$	UO_2
Average Fuel-Particle Diameter, μ	186	217	229	243	206	302
Inner Layer						
Coating temperature, °C	1400	1400	1400	1400	1400	1400
CH_4 flow rate, $cm^3\ min^{-1}cm^{-2}$	0.17	0.15	2.8	0.83	0.83	0.16
Coating thickness, μ	40	55	40	50	35	70
Outer Layer						
Coating temperature, °C	1700	1800	1800	1800	1800	1900
CH_4 flow rate, $cm^3\ min^{-1}cm^{-2}$	0.17	0.33	0.27	0.17	0.17	0.10
Coating thickness, μ	72	45	80	80	63	65

(5) the performance to substantial burnup of coated $(Th,U)C_2$ particles with thorium to uranium ratios up to 2.1:1 has been comparable to that of similarly coated UC_2 particles.

Coated Oxide Particles.

The highest burnup demonstrated, at this writing, for loose coated oxide particles is 12 at. % heavy metal at 1400°C in a sweep capsule (experiment A9-2). This test has operated for about 9 weeks and, by comparison with results from other tests in this type of facility, the fission-gas release has been constant and very low. Thermocouple failure occasioned termination of a previous sweep capsule test (experiment B9-20) on coated UO_2 particles after a burnup of 4.2 at. % heavy metal at 1370°C. None of the coatings were ruptured in this test as the result of irradiation, and metallographic examination revealed only minor damage by wedge-shaped fractures, none of which extended beyond one-half the thickness of the inner coating. Sol-gel $(Th,U)O_2$ particles containing 8% UO_2 and coated with a two-layer coating have been irradiated at 1200°C to a short burnup in experiment B9-19 and for an extended time in the ORR loop No. 1.

Typical photomicrographs of unirradiated and irradiated particles of both uranium and uranium-thorium fuels are shown in Figs. 19 and 20.

Fueled Graphite Irradiation Performance

Sweep Capsule Tests.

Evaluation of a reactor fuel element must take into account irradiation test performance. Although it is desirable to test in actual power-reactor service, no suitable facility yet exists for in-service testing of HTGR fuels. The irradiation capsule and loop tests now employed in evaluating spherical fuel elements offer advantages for instrumentation and control and for measurement of fission-gas release as a function of irradiation experience.

Most ORNL capsule irradiations have been conducted in the ORR Poolside Facility, which provides the reference conditions, including fission power density, temperature, and a sweep atmosphere of helium. Other capsules in core positions have been used to obtain high burnup.

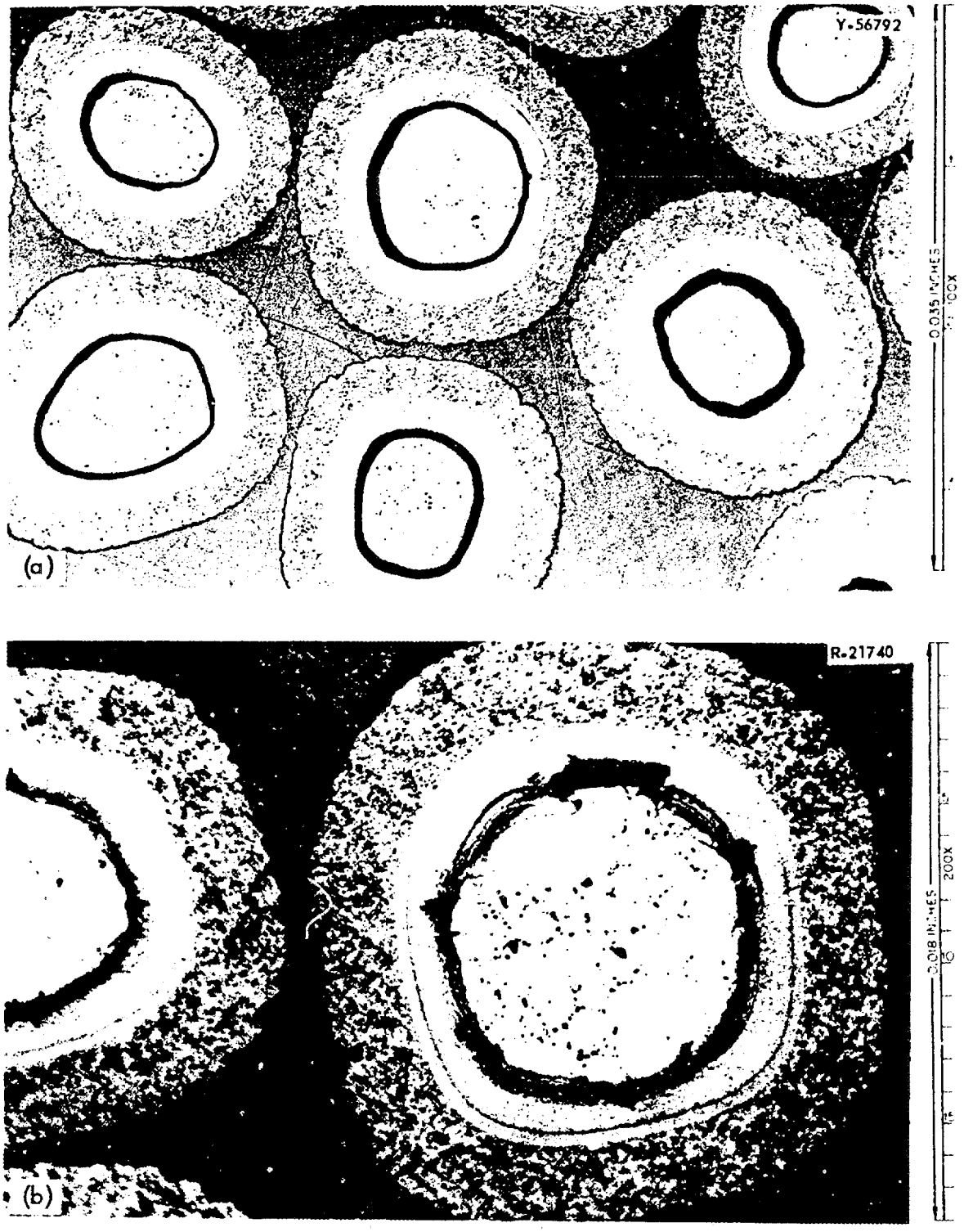


Fig. 19. Pyrolytic-Carbon-Coated UO_2 Particles from Batch OR-201. (a) Unirradiated. (b) Irradiated to 4.2 at. % U burnup at 1370°C in Capsule B9-20. As polished.

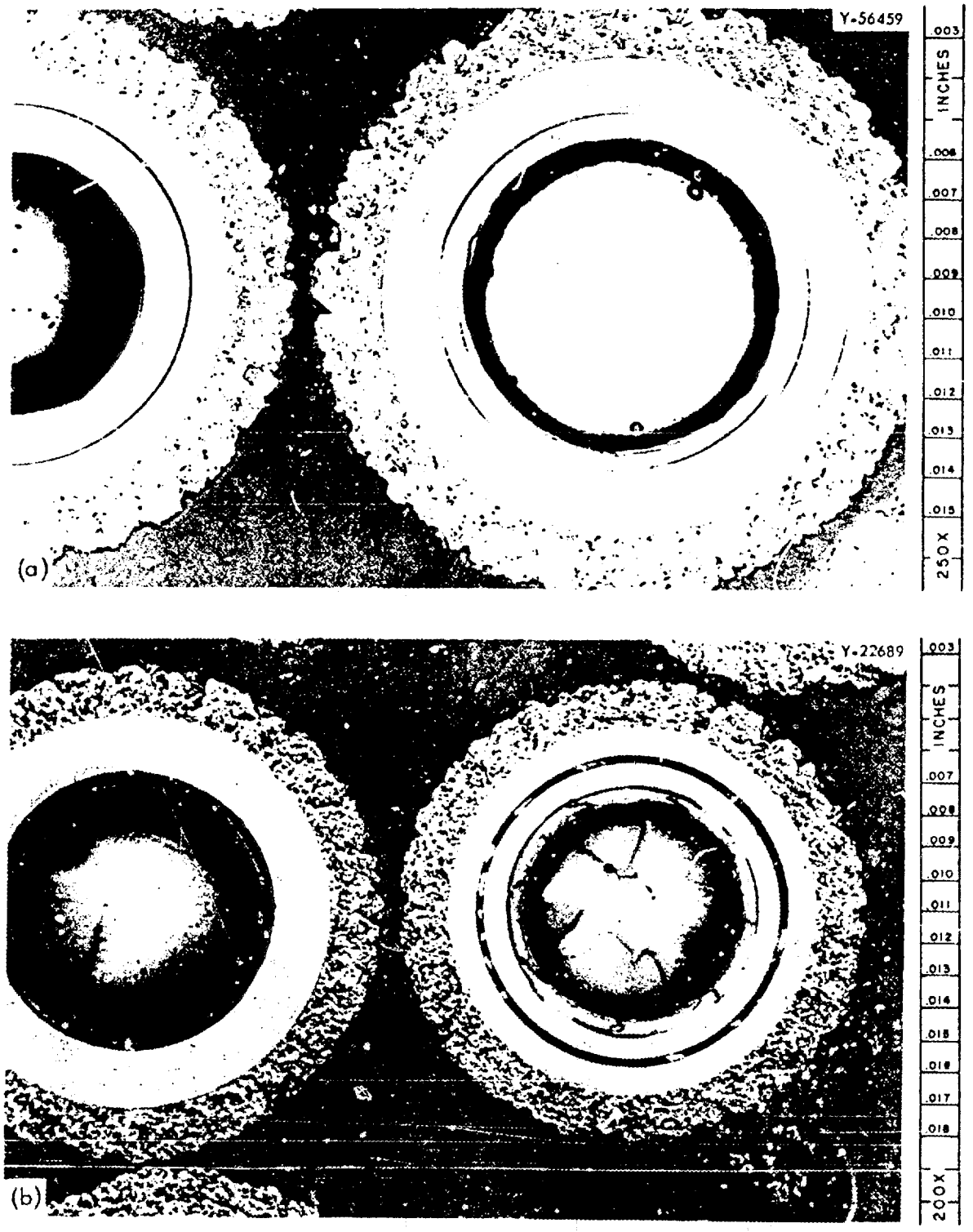


Fig. 20. Pyrolytic-Carbon-Coated $(Th,U)O_2$ Particles from Batch OR-162. (a) Unirradiated. (b) Irradiated to 0.25 at. % heavy metal burnup at 1200°C in Capsule 01-S. Note the bright, metallic-appearing fragments at the interface and within the inner coating after irradiation. As polished.

The design of the poolside irradiation capsule is shown in the schematic diagram of Fig. 21 and the irradiation test facilities have been described in detail.[3]

Specially equipped hot cells are used for examination of the spheres following irradiation. The examinations include a gross gamma scan, visual and photographic observations, dimensional measurements, crushing and impact strength measurements, burnup determinations, metallography, and radiochemical analysis of parts from the graphite structures. The apparatus and techniques used for the examinations were described in a recent publication.[26]

Irradiation tests of spherical fuel elements such as that shown in Fig. 22 have been conducted principally to investigate the effect of fabrication variables. These findings have been summarized recently,[27] and have been reported in greater detail in ORNL progress reports.[28–31]

Operating conditions and fission-gas release during irradiation for the spheres used in several tests and representative of both uranium and thorium-uranium elements are given in Table 8. It is notable that only one sphere showed a ratio of fission-gas release rate to birth rate, R/B, significantly in excess of 10^{-3}. This sphere, GA-VS11-2, fabricated as shown in Table 8 showed evidence of excessively high temperatures in the matrix and probably operated at a much higher temperature than was estimated because of lack of bonding and the development of a gap, which was observed in postirradiation examination. Relative fractional release of noble fission gases is illustrated in Fig. 23, which shows little difference between the molded and machined shells. A somewhat wider variation is apparent for different elements having machined shells, however, this probably indicates variation in the effectiveness of bonding between the matrix and shell. The shell bonding difficulty may not be inherent in the design, since these were all experimental elements, but this does represent an important problem area. Sphere 3M-VS16 showed, at the start, very low fission-gas release but the release increased steadily with burnup. This behavior is attributed to progressive failure of monolithic-laminar coatings with increasing burnup (see above).

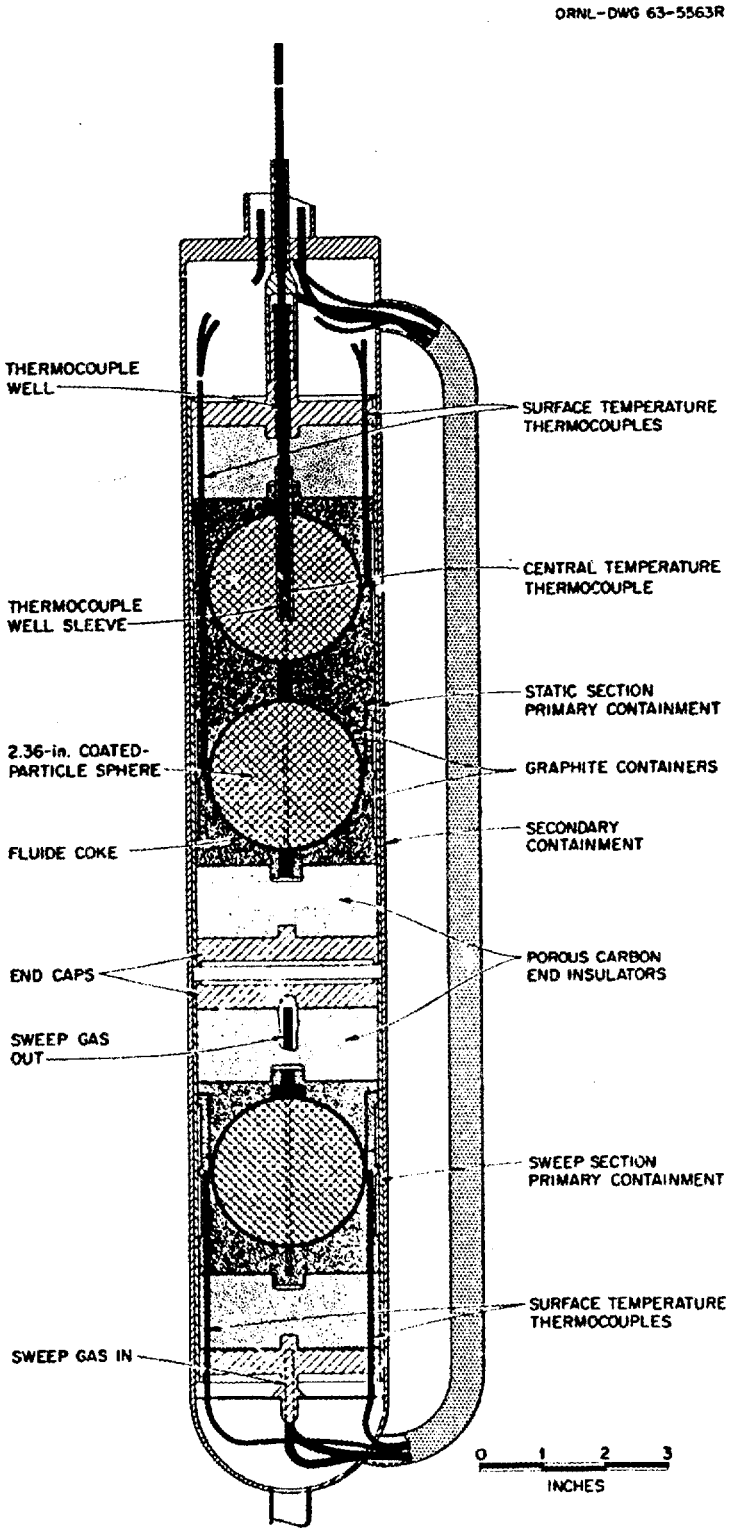


Fig. 21. ORR Poolside Capsule for Testing 6-cm-diam Fueled Graphite Spheres.

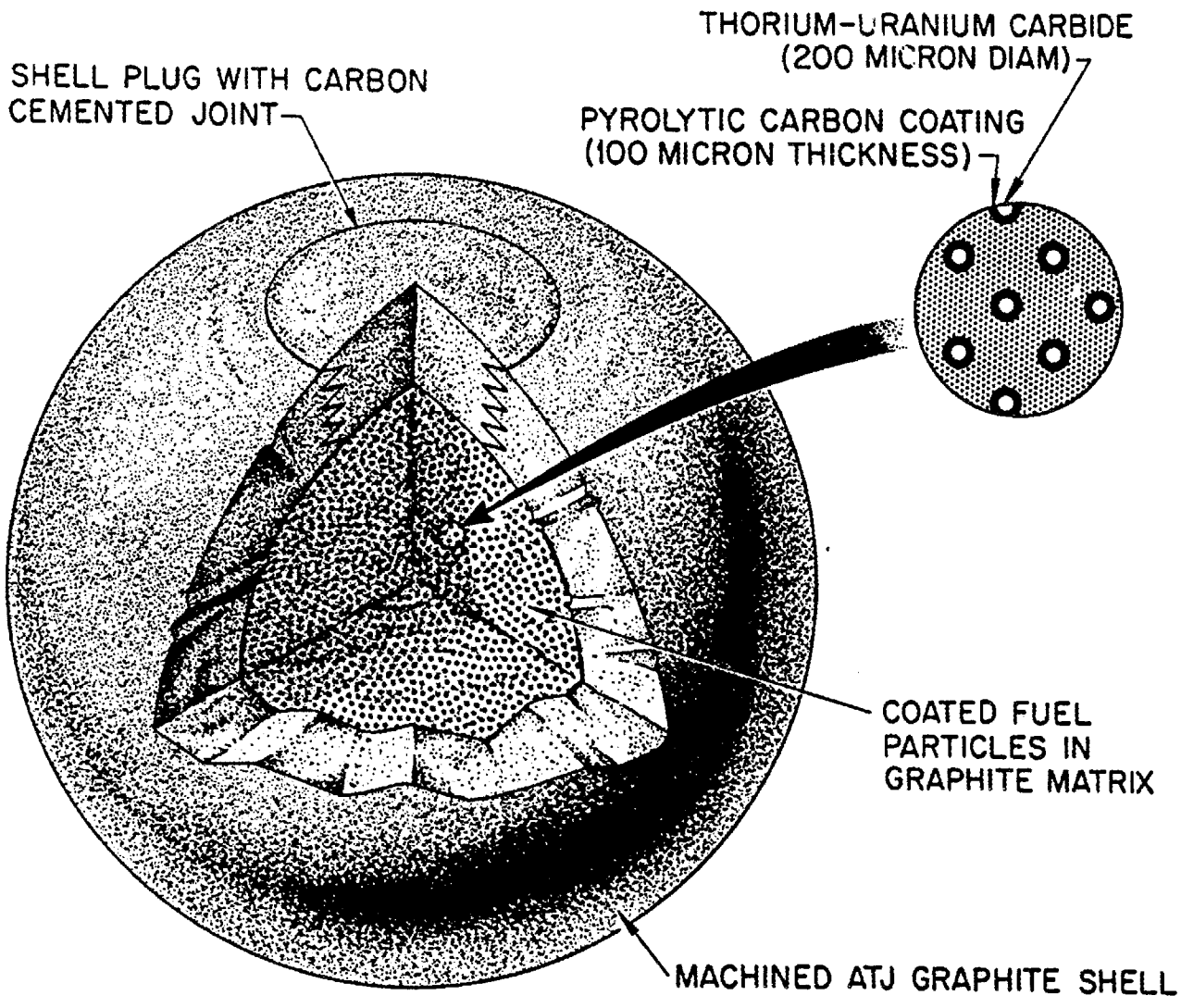


Fig. 22. Schematic of a Typical Coated-Particle Fuel Element.

Table 8. Operating Conditions and Fission-Gas Release Data for 6-cm-diam ORR-Irradiated Fuel Spheres Containing Coated Carbide Fuel Particles, Poolside Capsules

Type of Shell	Sphere	Shell Thickness (cm)	Insert Fabrication	Unfueled Shell	Particle Type	Particle Coating	Burnup (at. % heavy metal)	Operating Time (days)	Average Power Density (w/cm^3)	Average Temperature, °C Surface	Average Temperature, °C Center Estimated	R/B ^{86}Kr	R/B ^{133}Xe
												$\times 10^{-4}$	
Machined	GA-VS11	1.1	Hot pressed at 750°C, baked at 1800°C	Type ATJ	$(Th,U)C_2$	Triplex	3.2	27	30.0	930	1370	1 to 43[a]	1 to 43[a]
	GA-VS15	0.8	Hot pressed at 750°C, baked at 1800°C	Type ATJ	$(Th,U)C_2$	Triplex	2.0	92	18.6	910	Not determined	2.5	6.0
	GA-VS16[a]	0.8	Warm pressed at 100°C, baked at 1800°C	Type ATJ	$(Th,U)C_2$	Triplex	2.3	96	18.6	920	1370	2.0	4.0
	NCL-VS1	1.1	Mold injection baked at 1400°C	Type ATJ	$(Th,U)C_2$	Duplex	1.5	46[c]	18.6	920	1320	2.0	4.0
	ORNL-VS3	0.8	Warm pressed at 160°C, baked at 1000°C	Type ATJ	UC_2	Duplex	1.2	62	16.0	850	1180	0.2	0.3
Molded	GA-VS12[d]	0.63	Warm pressed at 150°C	Hot pressed at 750°C, baked at 1800°C	$(Th,U)C_2$	Triplex	4.3	146	30.0	920	Not determined	3.0	2.5
	GA-VS12[d]	0.63	Warm pressed at 150°C	Hot pressed at 750°C, baked at 1800°C	$(Th,U)C_2$	Triplex	2.7	87	30.0	940	Not determined	2.0	2.5
	3M-VS16[b]	0.63	Not known	Not known	$(Th,U)C_2$	Laminar	3.0	140	18.6	910	1270	0.01 to 1[a]	0.01 to 1[a]

[a] R/B increased steadily throughout irradiation.

[b] Similar spheres.

[c] Test terminated by leak in capsule.

[d] Similar spheres.

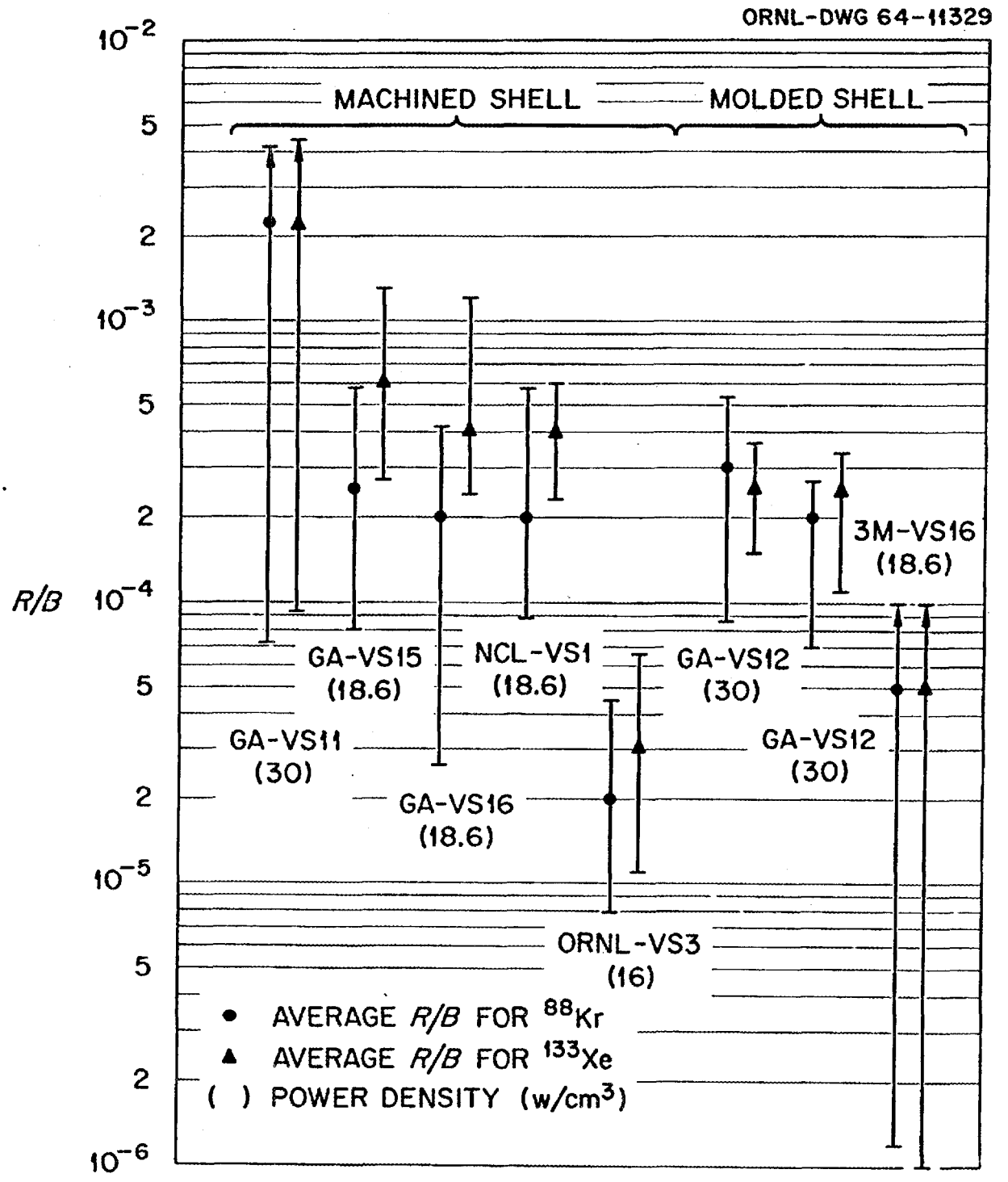


Fig. 23. R/B Values of ^{88}Kr and ^{133}Xe for 6-cm-diam Fuel Spheres Irradiated in ORR Poolside Facility.

No significant difference is apparent for the fission-gas release behavior of uranium and thorium fuels. These release rates are considered quite satisfactory for pebble-bed reactor service. Observations concerning release of solid fission products have been more limited, but again no difference has been observed between uranium and thorium-bearing coated particle fuels.

In-Pile Loop Test of Thorium-Uranium Oxide Pyrolytic-Carbon-Coated Particle Fuel Element

Perhaps the most significant test by ORNL of thorium-bearing fuel for HTGR reactors has been conducted in a recirculating loop, ORR No. 1.[3] The irradiation assembly designated as experiment 14 contains two cylindrical fueled-graphite elements with loose coated particles. This type of fuel element is of interest for advanced reactor designs.[19]

The design of the fuel elements for experiment 14 is shown in Fig. 24. The two cylindrical fuel elements are aligned one above the other in the loop. They differ in that the coated particles in the upper element are contained within an annulus between a tube and a rod of type ATJ graphite. The lower element consists of a solid graphite rod with five longitudinal holes containing loose coated particles located symmetrically around the axis of the rod. The lower element is contained within a sealed stainless steel capsule and the upper element is vented to the helium coolant in the loop via a porous nickel plug. The upper capsule is instrumented with nine thermocouples, one of which is located at the axial center of the fuel. The lower capsule is uninstrumented, but the heat generation rate is so low that its temperature is not much higher than the outlet gas temperature so no instrumentation is necessary. The stainless steel container is required to protect the fuel element from damage during insertion into and removal from the loop.

The fuel used in experiment 14 is comprised of a mixture of two batches of pyrolytic-carbon-coated particles fabricated at ORNL and designated as OR-205 and -206. The particles for both batches were prepared by the sol-gel technique. The particles in batch 205 consist of pure ThO_2, whereas those in batch 206 contain approximately 8 wt % UO_2

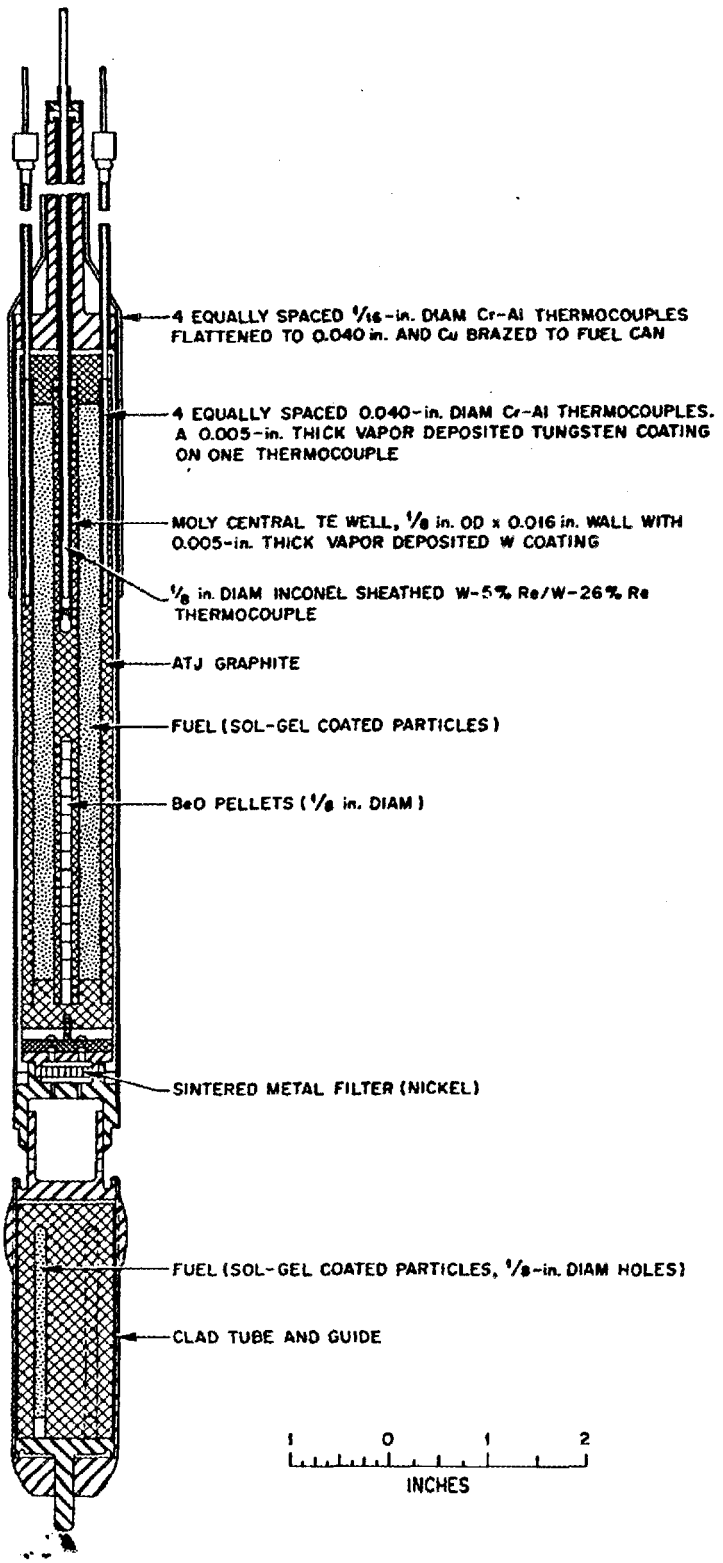


Fig. 24. Fuel Assembly for Experiment 14 in ORR Loop No. 1.

(93% ^{235}U). A detailed description of these coated particles is given in Table 7. The mixture was prepared by blending 53.415 g from batch OR-206 with 10.551 g from batch OR-205; it was then poured into the individual compartments.

The fuel burnup in the $(Th,U)O_2$ particles in the upper vented region is estimated to be 2.7 at. % heavy metal after approximately 9 months of operation. Since initially only 7.2% of the heavy metal in these particles was ^{235}U, the ^{235}U consumption, including fission and conversion to ^{236}U, is 44 at. %. Burnup of fuel in the lower, sealed element which contains the same fuel but was in a lower neutron flux is estimated to be 2.2 at. % heavy metal.

The temperature in the upper region has been maintained at 1370°C in the center of the fuel and 1040°C in the surrounding graphite fuel support tube. The fuel temperature of the uninstrumented lower region is estimated to be 700°C.

Equilibrium concentrations of noble gas fission products in the loop coolant have been nearly constant and indicate R/B values for the upper region of approximately

^{85m}Kr	1.9×10^{-5}
^{87}Kr	1.0×10^{-5}
^{88}Kr	2.5×10^{-5}
^{135}Xe	1.5×10^{-5}
^{133}Xe	1.4×10^{-5}

The contaminant gases CO, CO_2, and H_2 have tended to build up slowly but continuously in the loop coolant, although the rate has decreased with time. The contaminants have been controlled by replacement of the coolant about once every 3 to 4 weeks when the concentration of CO and CO_2 reached approximately 125 and 75 ppm by volume, respectively. Approximately 30% Ne was added to the helium to lower the thermal conductivity of the coolant. This has made it possible to maintain the central fuel temperature in the vented region at 1370°C despite the high consumption of ^{235}U.

The element has been removed from the loop for postirradiation examination, but has not yet been opened. It is significant that the residual radioactivity of the loop did not increase measureably over

the period of this test, as determined by external gamma scanning. Analysis of samples from exposed surfaces near the compressor and on the fuel element support also indicate very low activity from metallic fission products.

Eight-Ball Static Capsule Test

The eight-ball capsule contains eight 1.5-in.-diam spheres placed inside individual graphite containers and assembled inside a graphite tube with end insulators. The graphite tube assembly is inserted in stainless steel primary and secondary containment tubes; this tube assembly makes up the capsule which is tested in the F-1 position of the ORR core. A fifth and final test of this type, F1-8B-5, has been irradiated to higher fuel burnup than any other ORNL test of thorium-uranium carbide HTGR fuel elements. The fueled spheres used in this experiment are described in Table 9. Heavy metal fuel burnup in this capsule was approximately 25 at.% with approximately 45% for ^{235}U during 9 months exposure in the ORR. None of the spheres showed visible damage; overall linear shrinkage for spheres having machined shells was approximately 1% with approximately 2% for molded shells and elements. Observation of the coated particles indicates performance about as expected from previous high burnup tests of uranium carbide particles.

Summary

Limited test data for thorium-uranium HTGR fuels at low thorium concentration or low burnup show no observable difference from uranium fuel performance for comparable irradiations. Overall performance for these coated particle fuels has been favorable when compared with reactor service requirements where release to birth ratio (R/B) of 10^{-4} for noble gases has been considered acceptable. Both thorium-uranium carbide and oxide fuels have been tested; results for oxides, although limited, have been excellent.

Table 9. Description of Fueled Spheres and Average Operating Conditions for Eight-Ball Irradiation Capsule F1-8B-5

Position	Sphere Designation	Fuel Loading: Total U (g)	Fuel Loading: ^{235}U (g)	Fuel Loading: Th (g)	Unfueled Shell: Thickness (in.)	Unfueled Shell: Type	Average Temperature: Surface (°C)	Average Temperature: Center (°C)	Estimated Burnup (at. % Heavy Metal)	Estimated Burnup (Fissions/cm³)[b]
										$\times 10^{19}$
1 (top)	3M-S13-10	1.58	0.29	1.00	0.25	molded	786	973[c]	23	3.3
2	GA-S14-2A[d]	1.46	0.28	6.27	0.20	ATJ	869		25	3.6
3	NCF-S24-L3	1.58	0.29	1.00	0.00		902		26	1.5
4	3M-S13-7	1.58	0.29	1.00	0.20	ATJ	886		27	3.8
5	NCF-S25-V10	1.58	0.29	1.00	0.00		854		26	1.5
6	3M-S13-14	1.58	0.29	1.00	0.25	molded	841		25	3.6
7	GA-S14-3A[e]	1.46	0.28	6.27	0.20	ATJ	808		23	3.4
8 (bottom)	3M-S13-9	1.58	0.29	1.00	0.20	ATJ	773		22	2.9
B_4C Specimens										
Top TE 12							580			
Top TE 13							590			
Bottom TE 14							630			

[a]Fabricators: GA, General Atomic Division of General Dynamics; 3M, the 3M Company; NCF, Carbon Products Division of Union Carbide.

[b]Calculated on basis of fueled-core volume.

[c]Only the top sphere had a central thermocouple.

[d]The shell of this sphere was cracked prior to irradiation by dropping 24 times from a height of 4 meters on to a bed of 1.5-in.-diam graphite spheres.

[e]This sphere was dropped 25 times without cracking under the same conditions as (d).

THORIUM-BASE METAL FUELS

The potential advantage of high fuel density available with metallic fuels has long been recognized, but has not been exploited for power reactors because fuel swelling and growth phenomena can cause fuel element failures at relatively low burnup. As part of the studies on the use of nuclear reactors for desalination, a review of the available irradiation data on uranium, thorium, and their alloys has been made. Recent developments in metallic fuel technology, the lower fuel temperatures of interest in such reactors; the low fuel cycle costs for metallic fuel; and probable fabrication economies with large reactor complexes suggest that metallic fuels should again be considered.

There has been very little work reported on thorium and thorium alloy irradiations. However, several experiments on thorium metal have indicated that it has considerably greater dimensional stability during irradiation than uranium. Thorium, which has an isotropic body-centered cubic crystal structure, is not subject to growth effects and the attendant internal stresses that develop in the anisotropic orthorhombic crystals of uranium.

Early data on thorium irradiation experiments are collected by Bauer et al.[32] An Argonne National Laboratory investigation of thorium-uranium alloys has been summarized by Kittel et al,[33] who irradiated small specimens (0.14-in. diam × 0.875 in. long) of a number of thorium-uranium alloys at temperatures up to 1000°C and burnups to 10 at. %. Swelling for all alloys increased from approximately 1% per at. % burnup at lower temperatures to about 2.5% per at. % burnup at 650°C. At higher temperatures the swelling rate increased somewhat, reaching a value of 6% per at. % burnup at 800°C. Volume increases were linear with burnup and independent of uranium content. The data are plotted in Fig. 25. Specimens containing in excess of 25% U became warped and distorted, but those containing 20% U, or less, did not show significant distortion or surface roughness. In a few cases, excessive swelling was observed as a result of locally high temperatures at the top end of the specimen. The authors attribute the excellent behavior to the fact that chillcast specimens were used. Uranium particles thus were very thin so that fission products were largely trapped in the thorium matrix.

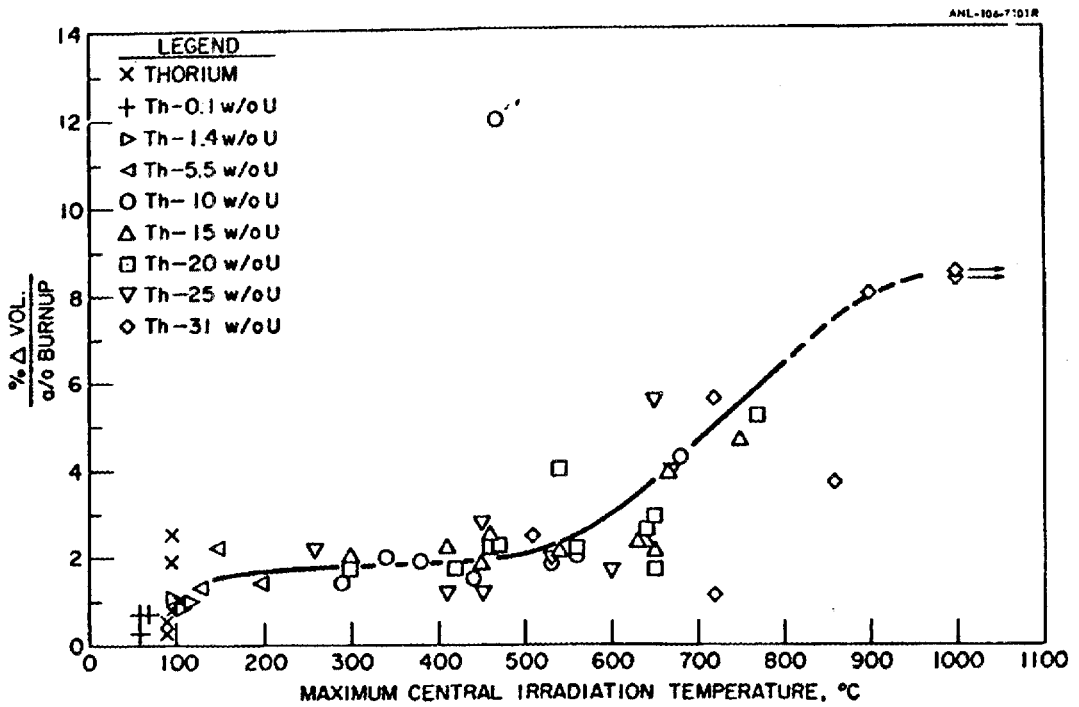


Fig. 25. Effect of Irradiation Temperature on the Swelling Rate of Thorium and Thorium-Uranium Alloys. Ref: J. H. Kittel et. al. Effects of Irradiation on Thorium and Thorium Alloys, USAEC Report ANL-5674, Argonne National Laboratory, April 1, 1963.

Data on the swelling of specimens of cast and swaged Th–11 wt % U irradiated to 1.5 at. % burnup at temperatures up to 650°C were reported by Battelle[34] and by Atomics International.[35] Specimens irradiated below 590°C decreased in density at the rate of 3% per at. % burnup. Specimens irradiated at higher temperatures showed greater density decreases even at burnups as low as 0.2%, but evidence of considerable overheating was also observed. Slugs of Th–7 1/2 wt % U, 3/4 in. in diameter and clad in stainless steel with a 25-mil-thick annulus of sodium, were used as fuel in Core II of the Sodium Reactor Experiment.[36] The maximum coolant outlet temperature was 490°C and maximum fuel central temperatures were near 650°C. Thorium fuel elements were operated to exposures of 5260 Mwd/tonne, and satisfactory operation was achieved,[37] but no data on examination of the fuel are yet available.

Hanford is irradiating three 8-in.-long test elements of Zircaloy-2 clad annular tubes (1.75 in. OD, 1.05 in. ID) of Th–2 1/2 wt % U–1 wt % Zr alloy in the ETR[38] high-temperature pressurized water loop. Preliminary

data at the maximum measured exposure of 9300 Mwd/tonne show an 0.9% volume increase. Maximum fuel operating temperatures have ranged between 460 and 585°C, and specific power between 45 and 69 w/g (ref. 39). The irradiations are being continued.

From these data it is apparent that thorium metal fuels are not subject to the excessive "cavitation" swelling reported for uranium metal in the 400 to 550°C temperature range.[40–42] However, the effects of fabrication variables and alloying or dispersion hardening are not known. In addition, the fuel temperature at which the %ΔV/at. % burnup increases rapidly needs to be defined. The compatibility of the metallic fuels with possible lower cost cladding materials must also be investigated. In general the successful utilization of metallic fuels, particularly thorium-base fuels, appears feasible, but their performance will depend on the adequate control of a number of variables. Further research and development is required to define the requirements necessary to achieve optimum performance.

ACKNOWLEDGMENTS

The efforts of many people are represented in the work reviewed in this paper, and it is impossible to give full credit to each. In the bulk oxide fuel irradiations program special note is due to J. W. Ullmann and S. D. Clinton of the Chemical Technology Division for preirradiation planning and calculations and from the Metals and Ceramics Division to W. S. Ernst, J. W. Tackett and R. W. McClung for fuel rod fabrication, welding, and nondestructive testing; E. J. Manthos for metallography; Y. Hirose, a foreign visitor from the Hitachi, Ltd., Japan, for postirradiation calculations; and to W. C. Thurber and S. A. Rabin who supervised most of the early experiments. In the coated particle fuel irradiation program three Laboratory divisions are involved. H. C. McCurdy, in the Reactor Division, G. M. Watson, in the Reactor Chemistry Division, and P. Patriarca, in the Metals and Ceramics Division, supervised most of the recent effort. Special note is due to R. L. Beatty for coating studies, F. L. Carlsen for evaluation of spheres, C. O. Smith for impact studies, O. Sisman and F. R. McQuilkin for irradiation tests, J. L. Scott for thermal conductivity studies, and E. L. Long for metallography.

1. D. E. Ferguson, O. C. Dean, and D. A. Douglas, "The Sol-Gel Process for the Remote Preparation and Fabrication of Recycle Fuels," paper presented at the 3rd United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, August 31–September 9, 1964.

2. J. L. Matherne et al., Summary of the Kilorod Project – A Semiremote 10 kg/day Demonstration of $^{233}UO_2$-ThO_2 Fuel-Element Fabrication by the ORNL Sol-Gel Vibratory-Compaction Method, USAEC Report ORNL-3681 (in press), Oak Ridge National Laboratory.

3. D. B. Trauger, Some Major Fuel-Irradiation Test Facilities at the Oak Ridge National Laboratory, USAEC Report ORNL-3574, Oak Ridge National Laboratory, April 1964.

4. S. A. Rabin, pp. 102–128, Status and Progress Report for Thorium Fuel Cycle Development for Period Ending December 31, 1962, USAEC Report ORNL-3385, Oak Ridge National Laboratory.

5. R. G. Wymer and D. A. Douglas, Status and Progress Report for Thorium Fuel Cycle Development for Period Ending December 31, 1963, USAEC Report ORNL-3611 (in press), Oak Ridge National Laboratory.

6. S. A. Rabin, M. F. Osborne, S. D. Clinton, and J. W. Ullmann, "Thorium Fuel Cycle Irradiation Program at the Oak Ridge National Laboratory," p. 643 in Proceedings of the Thorium Fuel Cycle Symposium, Gatlinburg, Tennessee, December 5–7, 1962, USAEC Report TID-7650, Bk II, AEC Technical Information Division.

7. S. A. Rabin, S. D. Clinton, and J. W. Ullmann, "Irradiation of Non-sintered ThO_2-UO_2 and ThO_2-PuO_2 Fuel Rods for Power Reactor Applications," paper presented at the Symposium on Powder Filled Uranium Dioxide Fuel Elements, Worcester, Massachusetts, November 5–6, 1963.

8. R. G. Wymer and D. A. Douglas, Status and Progress Report for Thorium Fuel Cycle Development for Period Ending December 31, 1964, USAEC Report ORNL-3831 (in press) Oak Ridge National Laboratory.

9. W. D. Kingery, J. Am. Ceram. Soc. 4(12), 617 (1959).

10. S. A. Rabin et al., Irradiation Behavior of High Burnup ThO_2–4.45% UO_2 Fuel Rods, Oak Ridge National Laboratory Report (in preparation).

11. T. D. Anderson, "Effects of High Burnup on Bulk UO_2 Fuel Elements," Nucl. Safety 6(2), 164–169 (Winter 1964–65).

12. J. R. MacEwan and V. B. Lawson, J. Am. Ceram. Soc. 45(1), 42–46 (1962).

13. S. V. K. Rao, Investigation of ThO_2-UO_2 as a Nuclear Fuel, Canadian Report AECL-1785, July 1963.

14. J. A. L. Robertson, $\int k d\theta$ in Fuel Irradiations, Canadian Report CRFD-835, April 1959.

15. M. F. Osborne and E. L. Long, Jr., Post Irradiation Examination of Maritime-ORR Loop Experiments 5, 7 and 8, USAEC Report ORNL-TM-921 Oak Ridge National Laboratory, Oct. 1964.

16. V. O. Haynes, W. C. Thurber, and E. L. Long, Jr., "Fuel Irradiation Tests," pp. 61–77, Maritime Reactor Program Ann. Progr. Rept. Nov. 30, 1963, USAEC Report ORNL-3775, Oak Ridge National Laboratory.

17. Staff, Oak Ridge National Laboratory, Conceptual Design of the Pebble Bed Reactor Experiment, USAEC Report ORNL-TM-201, Oak Ridge National Laboratory, May 1962.

18. C. Cautius, "The AVR High-Temperature Reactor," Atom und Strom, 11, 33–38 (May 1964).

19. Design Study Report for TARGET: A 1000-Mw(e) High-Temperature Gas-Cooled Reactor, USAEC Report GA-4706, General Atomic, March 1964.

20. P. E. Reagan, F. L. Carlsen, and R. M. Carroll, "Fission Gas Release from Pyrolytic-Carbon-Coated Fuel Particles During Irradiation," Nucl. Sci. Eng., 18(3) 301–308 (1964).

21. P. E. Reagan et al, "Fission-Gas Release from Coated Particles," pp. 115–126, GCRP Semiann. Progr. Rept. Sept. 30, 1964, USAEC Report ORNL-3731, Oak Ridge National Laboratory.

22. W. O. Harms, "Carbon-Coated Carbide Particles as Nuclear Fuels," in Modern Ceramics – Some Principles and Concepts, ed. by J. Hove and W. Riley, John Wiley & Sons, Inc., New York (in press).

23. R. W. Dayton, W. V. Goeddel, and W. O. Harms, "Ceramic-Coated Particle Nuclear Fuels," A/Conf.28/P/235, paper presented at the 3rd United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964.

24. Final Report, Graphite-Matrix Fuels Development for the AVR, p. 8, USAEC Report GA-4695, General Atomic, October, 1963.

25. J. C. Bokros, The Structure of Pyrolytic Carbon Deposited in a Fluidized Bed, USAEC Report GA-5163, General Atomic, July 1964.

26. D. R. Cuneo, E. L. Long, Jr., and H. E. Robertson, "Hot Cell Techniques for the Evaluation of Fueled Graphite Specimens," pp. 2456–256 in Proceedings of the 12th Conference on Remote Systems Technology, American Nuclear Society, New York, 1964.

27. W. O. Harms and D. B. Trauger, Fabrication Variables, Performance, and Cost Considerations for HTGR Coated-Particle Fuels, USAEC Report, ORNL-TM-1123, Oak Ridge National Laboratory, April 1965.

28. J. L. Scott et al., "Preparation of Fueled-Graphite Irradiation Test Specimens," pp. 326–341; V. A. DeCarlo et al., "Irradiation Tests," pp. 344–354, GCRP Semiann. Progr. Rept. Sept. 30, 1963, USAEC Report ORNL-3523, Oak Ridge National Laboratory.

29. J. L. Scott and C. Michelson, "Preparation of Fueled-Graphite Irradiation Specimens," pp. 217–228; V. A. DeCarlo et al., "Irradiation Tests," pp. 230–240; D. R. Cuneo et al., "Postirradiation Examinations of Fueled-Graphite Spheres," pp. 246–268, GCRP Semiann. Progr. Rept. Mar. 31, 1964, USAEC Report ORNL-3619, Oak Ridge National Laboratory.

30. V. A. DeCarlo and F. R. McQuilkin, "Fueled-Sphere Irradiations in ORR Poolside Facility," pp. 58–70; D. R. Cuneo et al., "Postirradiation Examinations of Fueled-Graphite Spheres," pp. 78–95, GCRP Semiann. Progr. Rept. Sept. 30, 1964, USAEC Report ORNL-3731, Oak Ridge National Laboratory.

31. V. A. DeCarlo and F. R. McQuilkin, "Fueled-Sphere Irradiation in ORR Poolside Facility," and D. R. Cuneo et al., "Postirradation Examination of Fueled-Graphite Spheres," GCRP Semiann. Progr. Rept. Mar. 31, 1965, USAEC Report ORNL-3807 (in press) Oak Ridge National Laboratory.

32. A. A. Bauer, M. S. Farkas, and V. W. Stoohok, Effects of Nuclear Reactions on Metallic Materials, USAEC Report REIC-29, Battelle Memorial Institute, September 1963.

33. J. H. Kittel, J. A. Horak, W. F. Murphy and J. H. Paine, Effects of Irradiation on Thorium and Thorium Alloys, USAEC Report ANL-5674, Argonne National Laboratory, April 1, 1963.

34. J. E. Gates, G. E. Lamale, and R. F. Dickerson, The Examination and Evaluation of Irradiated Thorium-11 wt % Uranium Specimens, USAEC Report BMI-1334, Battelle Memorial Institute, April 16, 1959.

35. B. R. Hayward and L. E. Wilkinson, Radiation Behavior of Metallic Fuels for Sodium Graphite Reactors, USAEC Report NAA-SR-3411, Atomics International, August 15, 1959.

36. E. N. Pearson, SRE Systems and Components Experience, Core II, USAEC NAA-SR-MEMO-9003, Atomics International, November 15, 1963.

37. Atomics International - Quarterly Technical Progress Report - AEC Unclassified Programs, January-March 1964, USAEC Report NAA-SR-9700, Atomics International, May 19, 1964.

38. M. M. Hendrickson, and J. E. Greer, et al., Research and Development Programs Executed for the Division of Reactor Development, Quarterly Progress Report April-June 1963, USAEC Report HW-78388, Hanford Atomic Products Operations.

39. Hanford Atomic Products Operations - Quarterly Progress Report, Division of Reactor Development, April-June 1964, USAEC Report HW-83298, Hanford Atomic Products Operations.

40. R. D. Leggett, T. K. Bierlein, and B. Mastel, Irradiation Behavior of High Purity Uranium, USAEC Report HW-79559, Hanford Atomic Products Operations, November 1963.

41. R. S. Barnes, R. G. Bellamy, B. R. Butcher, and P. G. Mardon, "The Irradiation Behavior of Uranium and Uranium Alloy Fuels," A/Conf. 28/P/145, paper presented at the 3rd United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1964.

42. C. L. Angerman and G. R. Caskey, Jr., "Swelling of Uranium by Mechanical Cavitation," J. Nucl. Mater. 18(2) 182-96 (1964).