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IRRADIATION BEHAVIOR OF ALUMINUM-BASE FUEL DISPERSIONS

M. M. Martin
A. E. Richt
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OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION • FOR THE U.S. ATOMIC ENERGY COMMISSION

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IRRADIATION BEHAVIOR OF ALUMINUM-BASE FUEL DISPERSIONS

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ABSTRACT

Miniature fuel plates were irradiated to determine factors that may influence the performance of aluminum-clad aluminum-base UAl_x and U_3O_8 dispersions in the Advanced Test Reactor (ATR) and the High Flux Isotope Reactor (HFIR). Our primary goal was to establish whether UAl_x- and U_3O_8-bearing fuel plates could be irradiated to fission densities greater than 1.8×10^{21} fissions/cm^3 without failure. The experiment, designated PM capsule 1, consisted of 18 uninstrumented single-core test plates and 3 instrumented double-core plates. It was irradiated in the permanent beryllium reflector region of HFIR for 11,613 MWd. The principal experimental variables were initial void content of the fuel dispersions as affected by the type of fuel dispersoid, dispersoid concentration, and fabrication procedure and the fuel dispersoid particle size. Two grades of U_3O_8, burned and high fired, and arc-cast UAl_x were used at dispersoid concentrations that corresponded to current HFIR and ATR loadings and 25% increases in these loadings.

Postirradiation examination of the test fuel plates showed all to be in excellent condition, without evidence of either actual or incipient failure. Maximal fuel core swelling of 8.8 vol % occurred in a UAl_x-bearing plate irradiated to a ^{235}U burnup of 1.8×10^{21} fissions/cm^3 at 63 to 80°C. The swelling behavior of the aluminum-clad U_3O_8-Al and UAl_x-Al dispersions, however, depended primarily upon the fission density and the void contents of their cores during irradiation; it was independent of the fuel dispersoid size distribution of less than 44 μm particles. Variations in fuel dispersoid concentration and fuel plate fabrication procedures can alter the void content of these dispersions and, consequently, affect significantly their irradiation-induced swelling behavior. A mathematical model considering final fission density and initial void content predicts the degree of final plate swelling adequately for most engineering applications.

INTRODUCTION

The fuel elements now used in the more advanced thermal research reactors, such as the High Flux Isotope Reactor (HFIR) and the Advanced

Test Reactor (ATR), have operated satisfactorily. In 1970, however, the ATR was not reactivity limited, and operation of this facility would be less costly if the lifetime of the fuel core could be extended. The deterrent to the use of the fuel for extended lifetimes was the absence of irradiation data demonstrating successful performance at the higher burnups. To obtain the needed information on irradiation-induced swelling, we irradiated and evaluated a series of miniature fuel plates in the HFIR. The purpose of this experiment, designated PM Capsule 1, was to establish whether UAl_x- and U_3O_8-bearing fuel plates could be irradiated to fission densities greater than 1.8×10^{21} without failure. Of secondary importance was demonstrating the irradiation performance of a clad Al_2O_3 dispersion in aluminum.

This report summarizes our findings and compares our data to those previously published.[1,2] Since our earlier work on irradiation of miniature test specimens had indicated that the swelling behavior of both UAl_x and U_3O_8 aluminum-base dispersions could be correlated with a simple mathematical model, we have compared our new data with the model to verify its validity. The model, for use in predicting the swelling of individual plates on an engineering basis, is based on the observation that the void content of the as-fabricated dispersion is a major factor in the swelling of irradiated fuels. A subsequent review[3] of all other available data supported this conclusion and led us to believe that we could predict within ±1 vol % the swelling of either type of dispersion by simply assuming that (1) the dispersion swells at a rate of 6.3 vol % per 10^{21} fissions/cm^3, and (2) all voids initially present in the dispersion are available to accommodate this swelling. We show that the original model was satisfactory for engineering estimates but required the inclusion

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[1]M. M. Martin and A. E. Richt, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1969*, ORNL-4470, pp. 94–97.

[2]A. E. Richt and M. M. Martin, *Fuels and Materials Development Program Quart. Progr. Rep. Dec. 31, 1969*, ORNL-4520, pp. 266–72.

[3]A. E. Richt, *Fuels and Materials Development Program Quart. Progr. Rep. Sept. 30, 1969*, ORNL-4480, pp. 218–19.

of an empirical correction before the new swelling data could be correlated accurately.

GENERAL DESCRIPTION OF TEST PLATES

The experiment consisted of 18 uninstrumented single-core test plates and three instrumented double-core plates. The clad cores are two-phase systems of fuel particles dispersed in a metallic matrix. The void content and the size distribution of the fuel particles in the single-core test plates are the principal variables. Table 1 lists the plates and presents their pertinent differing characteristics. The type of dispersoid, its concentration in the dispersion, and fabrication differences directly affect the void content of the cored plates.[4]

Design

Figures 1 and 2 present the configuration and dimensional requirements of the two types of plates. The principal variables were investigated with the single-core plates. The double-core plates[5] permit installation of thermocouples between the two cores and served only to provide temperatures during the irradiation from which the temperatures of the single-core plates were calculated.

Materials

The cladding and core frame material for both types of plates as well as the separator material that housed the thermocouples in the double-core plate was aluminum alloy 6061. As noted above, the cored region of the plates contained particular dispersoids in a matrix of Alcoa 101 aluminum. The dispersed compounds include two grades of U_3O_8

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[4]M. M. Martin and W. R. Martin, *Fabrication Voids in Aluminum-Base Fuel Dispersions*, ORNL-4611 (October 1970).

[5]V. A. Walker, M. J. Graber, and G. W. Gibson, *ATR Fuel Materials Development Irradiation Results — Part II*, IDO-17157 (June 1966).

Table 1. Pertinent Core Attributes of Fuel Plates Irradiated in HFIR in PM Capsule 1

Plate	Dispersoid[a]				Void Content of Dispersion (vol %)	Uranium Loading	
	Powder Blend	Type	Concentration (wt %)	Concentration (vol %)		Total (g)	Fissile Density (g $^{235}U/cm^3$ of core)
11-3[b]	PB-33	Arc-cast UAl_x	53.2	31.7	3.1	1.088	0.00
21-4	PB-33	Arc-cast UAl_x	54.0	31.9	4.5	1.089	0.00
31-1[c]	PB-33	Arc-cast UAl_x	54.8	32.2	5.8	1.088	0.00
12-3	PB-01	High-fired U_3O_8	47.1	21.7	4.1	1.187	1.41
22-4	PB-04	Burned U_3O_8	49.8	23.8	8.2	1.187	1.43
32-4[b,d]	PB-32	Arc-cast UAl_x	51.4	30.3	1.3	1.085	1.31
01-02-2[e]	PB-01	High-fired U_3O_8	47.1	21.6	4.4	2.607	1.40
13-4	PB-01	High-fired U_3O_8	40.1	17.5	3.0	0.953	1.14
23-1	PB-04	Burned U_3O_8	42.0	19.1	6.4	0.954	1.15
33-4	PB-35	Arc-cast UAl_x	53.7	31.6	4.6	1.087	0.00
14-3	PB-32	Arc-cast UAl_x	52.9	30.6	4.2	1.086	1.32
24-3[b]	PB-32	Arc-cast UAl_x	52.2	30.3	3.3	1.085	1.31
34-2	PB-36	Arc-cast UAl_x	53.2	30.9	4.3	1.089	1.32
03-04-1[e]	PB-01	High-fired U_3O_8	44.2	19.9	3.7	2.403	1.29
15-4	PB-32	Arc-cast UAl_x	62.8	38.6	6.6	1.344	1.67
25-4[c]	PB-32	Arc-cast UAl_x	64.1	37.3	8.4	1.345	1.69
35-4	PB-36	Arc-cast UAl_x	63.0	38.7	7.3	1.342	1.66
16-2	PB-11	High-fired U_3O_8	39.9	17.3	2.8	0.954	0.00
26-3	PB-34	High-fired U_3O_8	39.9	17.4	2.3	0.954	0.00
36-2	PB-31	Fused α-Al_2O_3	24.4	18.1	1.1	0.000	0.00
05-06-1[e]	PB-01	High-fired U_3O_8	51.3	24.3	5.4	2.973	1.58

[a]Matrix material is Alcoa 101 aluminum powder, powder blend designation PB-30.

[b]Plates hot rolled to final gage to lower core void content.

[c]Plates cold rolled an additional 20% in thickness to increase core void content.

[d]Plates hot pressed after hot rolling to further lower core void content.

[e]Instrumented plates.

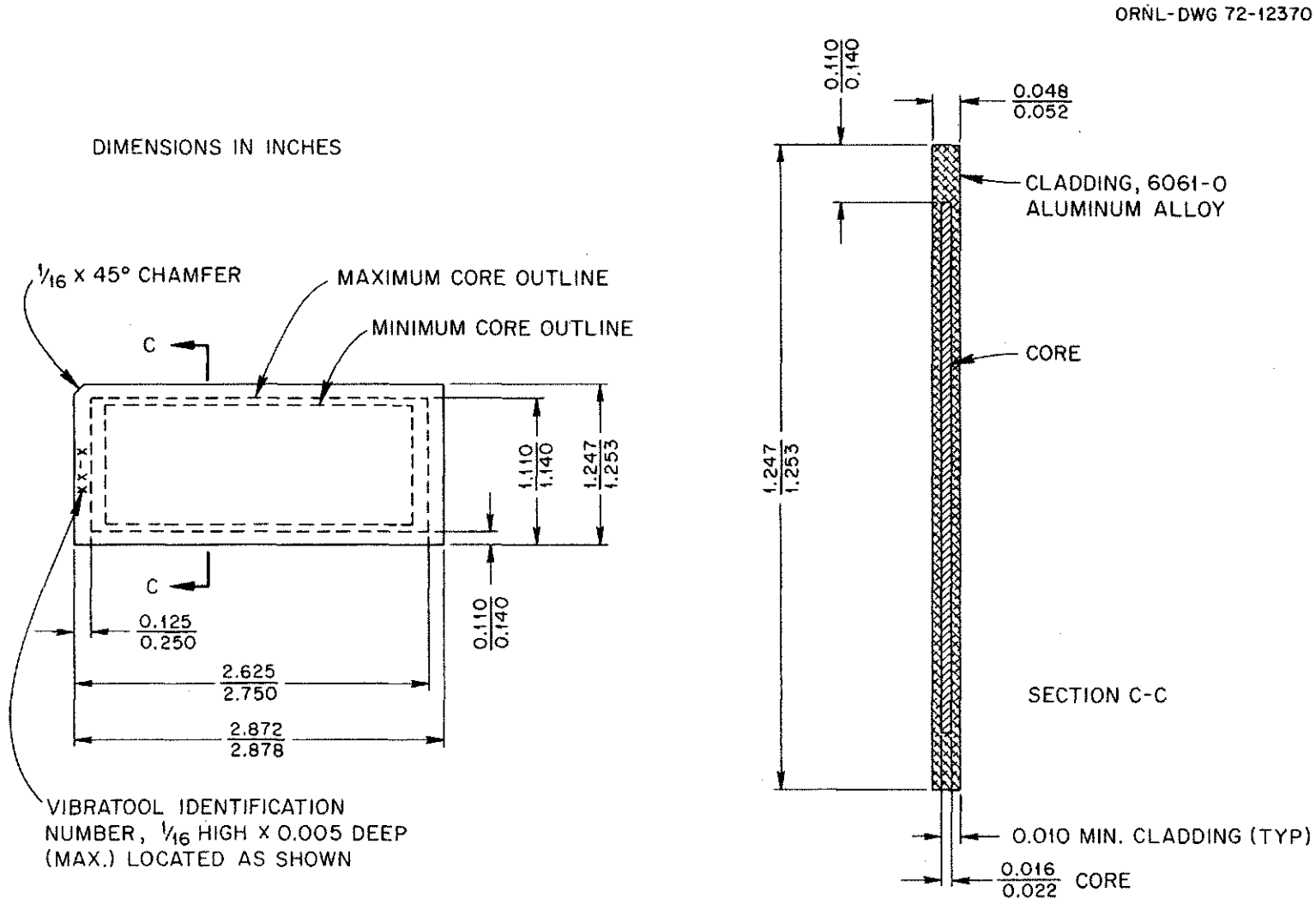

Fig. 1. Single-Core Test Plate Configuration and Dimensional Requirements.

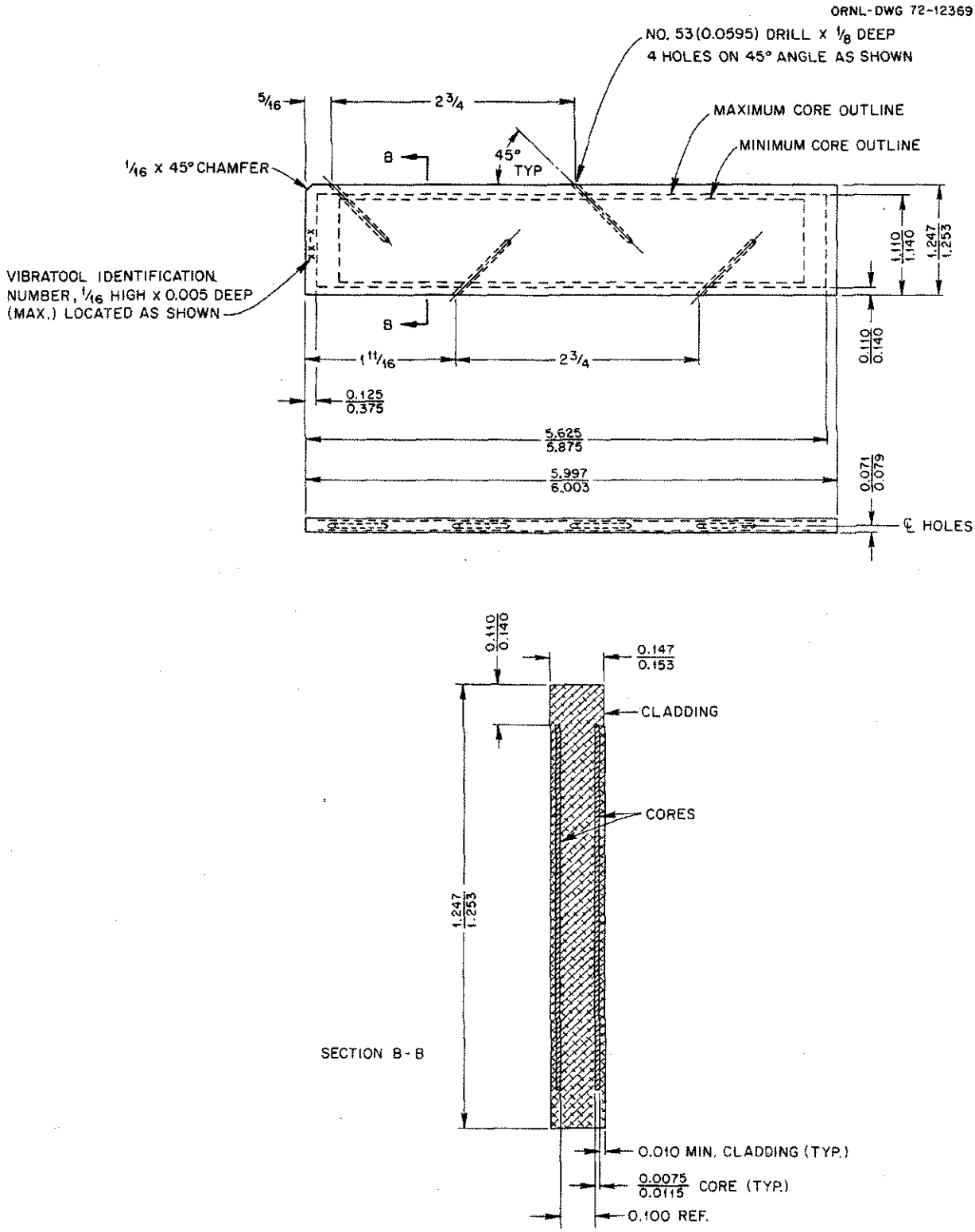

Fig. 2. Double-Core Instrumented Plate Configuration and Dimensional Requirements. Locations for thermocouple installation are shown. All dimensions are in inches.

(burned and high fired), arc-cast UAl_x, and fused α-Al_2O_3. The latter material was simply an abrasive alumina that may be added to the filler section of HFIR inner and outer annulus fuel plates to reduce blistering during fabrication.[6] Characteristics of the dispersoids and aluminum powder are given in Table 2.

The high-fired U_3O_8, identified as PB-01 in Table 2, is the oxide being used in HFIR.[7] Blends PB-11 and PB-34 are comparable materials to PB-01 but are depleted in ^{235}U isotope to 0.2 wt %; the latter also contains 39 wt % of particles that are smaller than 44 μm. Densities of the high-fired oxides, determined in a pycnometer with toluene, were usually greater than 98% of theoretical. Closed porosity, due to sintering, is found metallographically in this material. The burned U_3O_8 is made simply by burning uranium machining chips. The burning operation is an early stage of the manufacturing process to produce high-fired U_3O_8. The low density of burned U_3O_8 (PB-04 in Table 2), 7.6 g/cm^3, and low crushing strength[8] less than 1 g, indicate a highly friable particle containing considerable closed porosity. In comparison, individual 105 to 149-μm-diam particles of high-fired U_3O_8 exhibited a relatively high crushing strength of 4 g. To achieve the particular size distribution, both the as-supplied high-fired and burned particles were separated into known mesh fractions and then recombined in the desired proportions. The powders were sieved up to four times to effect a stable separation.

The uranium-aluminum intermetallics described in Table 2 were prepared by arc-casting, crushing, and sieving. Their major crystalline constituent is UAl_3, as determined by x-ray diffraction. All, however, contain significant amounts of UAl_2 and UAl_4 phases. Blend PB-32 is a similar material to that used[9] in the ATR. Removal of particles larger

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[6]M. M. Martin, *Fuels and Materials Development Program Quart. Progr. Rep. Dec. 31, 1969,* ORNL-4520, pp. 282–86.

[7]W. J. Werner and J. R. Barkman, *Characterization and Production of U_3O_8 for the High Flux Isotope Reactor,* ORNL-4052 (April 1967).

[8]M. M. Martin and W. R. Martin, *Fabrication Voids in Aluminum-Base Fuel Dispersions,* ORNL-4611 (October 1970).

[9]G. W. Gibson, *The Development of Powdered Uranium-Aluminide Compounds for Use as Nuclear Reactor Fuels,* IN-1133 (December 1967).

Table 2. Characterization of Core Component Powders

Type of Material	Supplier	Original Type or Lot Number	Prepared Powder Blend	Uranium Content, wt %		Particle Size		Toluene Density (g/cm^3)	Detected Impurity Element	
				Total	^{235}U	Principal Range (μm)	Fines <44 μm (wt %)		Major[a]	Minor[b]
High-fired U_3O_8	Union Carbide Corp., Nuclear Division, Y-12 Plant	30-2616	PB-01	84.5	93.2	44–88	10	8.243	None	None
		1-3315	PB-11	85.0	0.2	44–105	9	8.301	None	Fe
		1-3315	PB-34	85.0	0.2	44–88	39	8.304	None	Fe
Burned U_3O_8	Y-12 Plant	30-2615	PB-04	84.3	93.2	44–88	11	7.647	None	Si
Arc-cast UAl_x	Atomics International	128269	PB-32	71.7	93.1	44–149	24	6.477	O	C
		128269	PB-36	71.4	93.1	0–53	97	6.439	O	C, N
	Oak Ridge National Laboratory	71-D	PB-33	70.3	0.2	44–149	21	6.312	O	C, Cu, N, W
		71-D	PB-35	70.7	0.2	0–44	100	6.330	None	C, H, O, W
Atomized 101 Al	Aluminum Company of America	6911-93	PB-30			0–62	90	2.707	O	Fe, Ga, Mg, Si
Fused Al_2O_3	Norton Company	38	PB-31			0–88	55	3.916	Fe, Ga, Na	Ca, Zn

[a]Represents range between 0.1 and 1.0 wt %.

[b]Represents range between 0.01 and 0.1 wt %.

than 53 μm from PB-32 produced PB-36. Blends PB-33 and PB-35 correspond to PB-32 and PB-36, respectively, but are depleted in ^{235}U isotope to 0.2 wt %. Additionally, they were homogenized by annealing at reduced pressure (about 0.001 torr) and 600°C for 72 hr. The UAl_x particles have crushing strengths[8] more than 10 times that of the two grades of U_3O_8.

Fabrication Procedures

The general fabrication techniques used for the single-core test plate and the double-core instrumented plate have been reported.[10] The essential steps are

1. weighing and blending the component powder for each fuel compact,
2. cold pressing at 30 tsi to form the green compacts,
3. degassing the compacts at 500°C and less than 0.05 torr for 1 hr to remove pressing lubricant and absorbed gases,
4. cleaning the frames, cover plates, and separators for the rolling billets by etching with aqueous Oakite 160 solution,
5. assembling the degassed compacts into the frames and welding on cover plates to form the rolling billet,
6. cladding by hot roll bonding at 490°C to a reduction in thickness of of 84%,
7. annealing at 490°C for 1 hr to soften and also to test for blistering,
8. cold rolling to a reduction in thickness of 20% (total reduction in thickness is 87.5%),
9. heat treating to the "0" temper for alloy 6061, and
10. finishing operations — core location, plate shearing, machining, cleaning, and inspections for dimensional requirements, fuel homogeneity, and ultrasonic nonbond indications.

A significant deviation to the procedures described in ref. 10 is the cleaning of the billet components with aqueous Oakite solution. This development[11] eliminates the need for alclad cover plates and frame

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[10] M. M. Martin, W. J. Werner, and C. F. Leitten, Jr., *Fabrication of Aluminum-Base Irradiation Test Plates*, ORNL-TM-1377 (February 1966).

[11] J. H. Erwin, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1967*, ORNL-4170, pp. 179–81.

materials. We also modified the fabrication of the instrumented plates. Our new technique, which is cheaper and less time-consuming, employs a five-layer billet (cover plate/cored frame/separator/cored frame/cover plate), which is evacuated before roll bonding.

In Table 1, footnotes b, c, and d identify fuel plates that were fabricated with significantly different procedures than those given above. These intentional modifications yielded plates with increased or decreased void contents. For example, the direct hot rolling to final gage employed for plates 11-3, 32-4, and 24-3 lowered their normal void contents by 22, 22, and 31%, respectively. Additionally, plate 32-4 was pressed twice between hot steel platens at 475°C and 19,000 psi for a total duration under load of 120 sec. This operation further reduced the void content from 3.3 to 1.3 vol % (a change of 60%). In contrast, plates 31-1 and 25-4 were hot rolled to only 79% reduction in thickness. The necessary 40% cold reduction in thickness imparted to these plates to obtain the desired gage resulted, respectively, in 29 and 27% increases in their normal void concentrations.

DESCRIPTION OF IRRADIATION FACILITY AND PM CAPSULE 1 EXPERIMENT

General

The irradiation of PM capsule 1 was conducted in the VXF-17 position of HFIR. Figure 3 illustrates the orientation of the experiment in this permanent beryllium reflector hole with respect to the reactor vessel. A typical cross section through the test assembly near the reactor midplane is shown in Fig. 4. Coolant water from the primary reactor system flows in the 0.1-in. channel gaps between the fuel plates and also around the test assembly.

The S tube shown inside the reactor vessel in Fig. 3 connects the test assembly to the junction box on top of the reactor vessel. It protects the stainless-steel-sheathed thermocouples between the test assembly and the vessel's head from the highly turbulent water flow within the vessel and is also a convenient handle for the test assembly. Within the

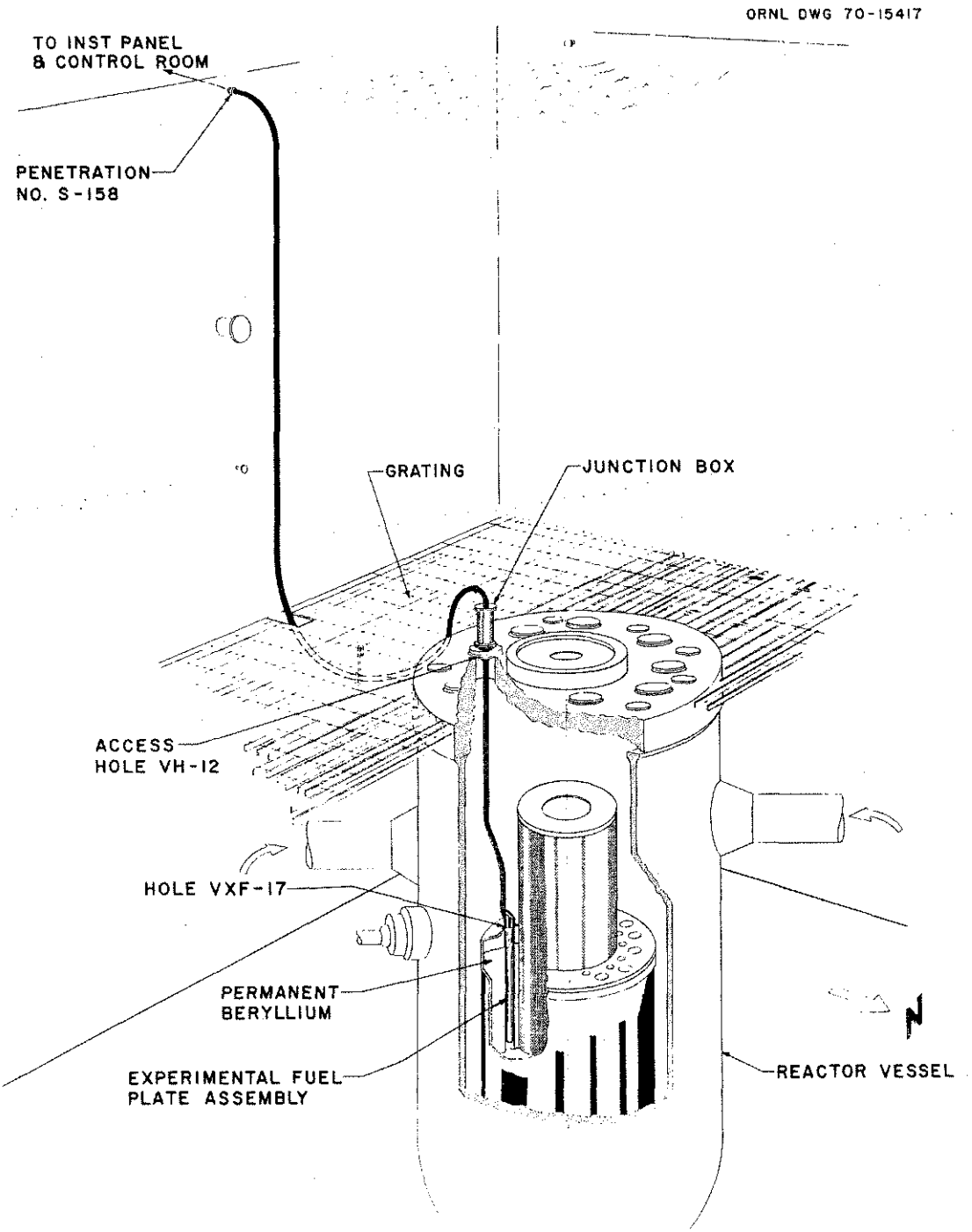

Fig. 3. Orientation of PM Capsule 1 Experiment in HFIR.

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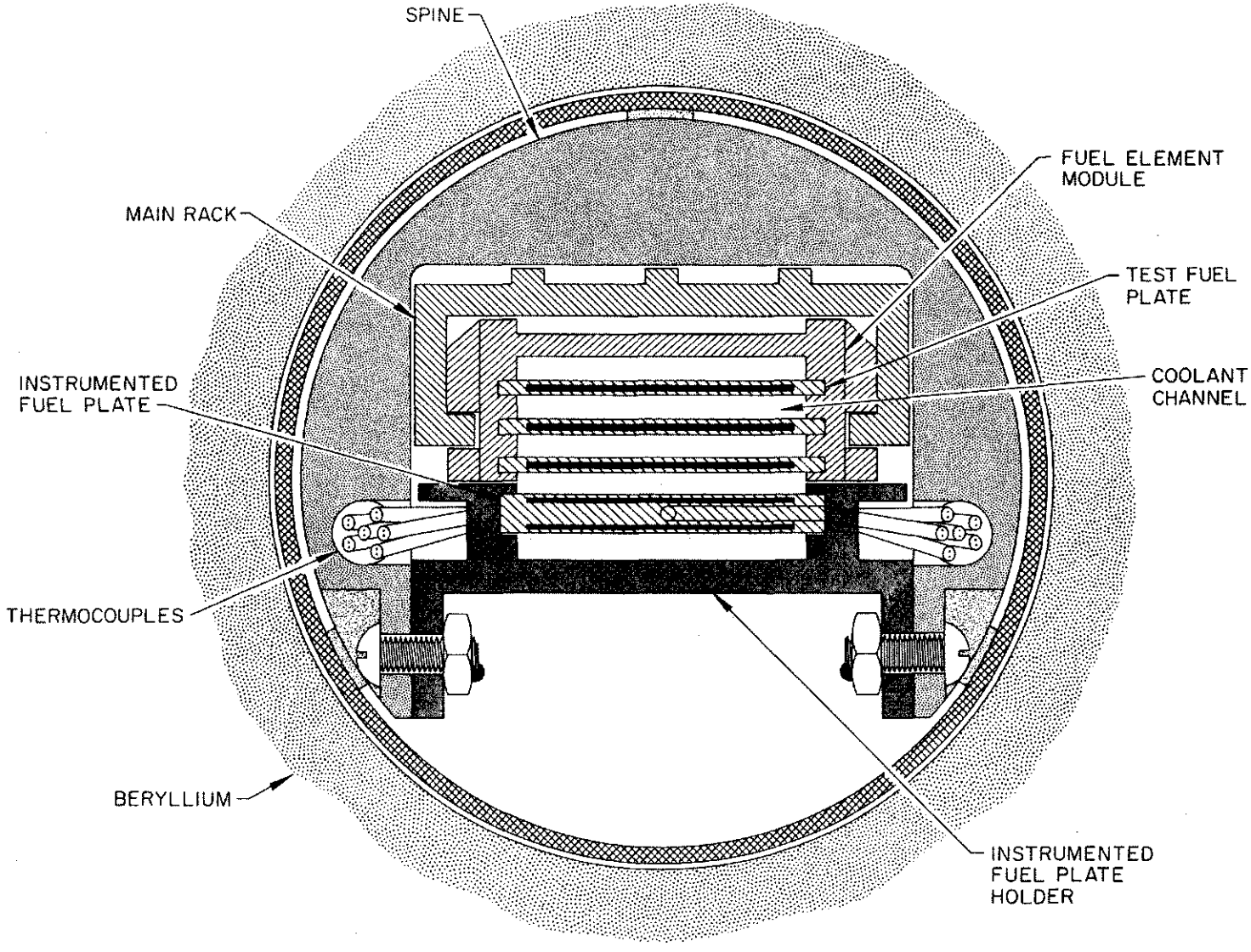

Fig. 4. Section Through Test Assembly.

junction box, the thermocouples are coupled to extension wires, which are taken to the experimenter's control room for monitoring the instrumented fuel plate temperatures.

Components of the in-core test assembly are shown in Fig. 5. The instrumented fuel plates cannot be seen in Fig. 5, but they are a part of the circular piece from which the stainless-steel-sheathed thermocouples protrude. They are contained in a channel directly below the recessed plate (instrumented plate holder) bolted to the circular housing. Since the thermocouple ends have an interference fit in the instrumented plates, all become inseparable from the S tube after assembly. The bayonet-appearing piece contains the 18 test plates in six removable modules. It is inserted into but not attached to the circular component containing the instrumented fuel plates. Thus, the test plates could be rearranged within their modules or new ones substituted while the bayonet was outside the reactor vessel. This option, however, was not exercised during the irradiation of PM capsule 1 experiment.

The two-number indexing scheme for the reference plate numbers in Table 1 identifies the location of the plates in the VXF-17 reflector hole. The first digit gives the radial column (0 to 3) with respect to the center of the active reactor core; the second indicates the vertical orientation from top to bottom (1 to 6); the third number denotes the particular plate from a group of like composition and fabrication history. The unperturbed thermal-neutron flux in VXF-17 is greatest near the 03-04 instrumented fuel plate and smallest for both 31 and 36 test plates, since the vertical flux pattern is nearly symmetrical about the horizontal midplane of the reactor. As determined from flux-wire measurements,[12] the average unperturbed thermal neutron flux within about 4 in. of the reactor midplane in the VXF-17 position is 3.1×10^{14} neutrons cm^{-2} sec^{-1}.

To cool the experiment, primary reactor water flows downward through the top of the test assembly and by the fuel plate faces at 50 fps, which is the prototypical condition for ATR and HFIR. The total pressure drop

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[12] J. H. Swanks, ORNL, private communications, April 6 and October 30, 1970.

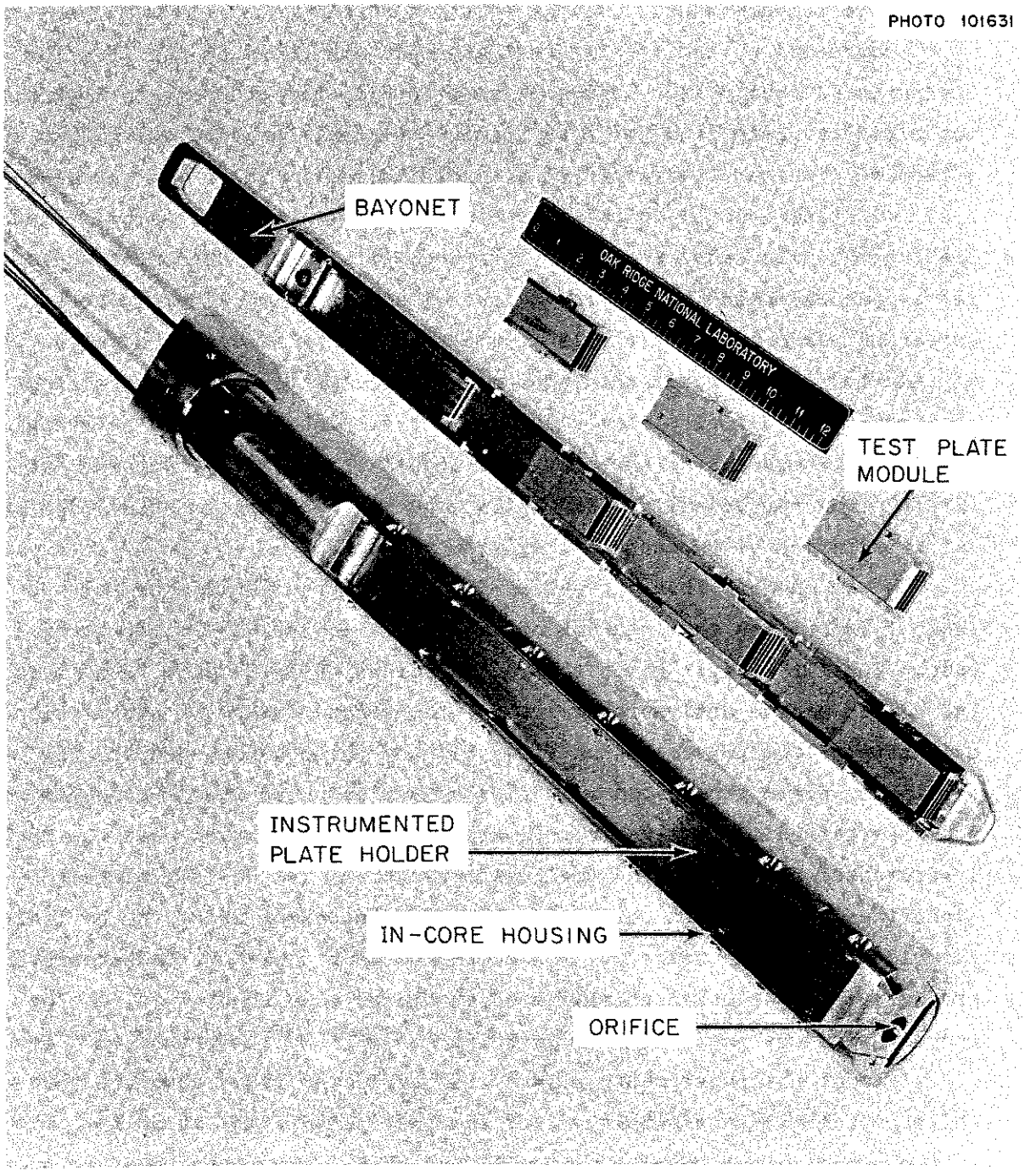

Fig. 5. Separable Components of Test Assembly Used in Flow Test.

across the VXF-17 hole is 109 psi, with an outlet pressure of 539 psia. This pressure drop and an orifice, shown at the end of the circular component of the test assembly in Fig. 5, establish the designed flow rate. The orifice is simply a round hole in an aluminum plate; it functions only as a restriction at the outlet of the coolant channels formed by the modular arrangement of test plates and the instrumented fuel plates. Flow in miscellaneous channels within and around the test assembly must, by design, bypass the orifice. Since the numerous flow paths make numerical analysis of the flow rate quite difficult, the diameter of the orifice was established experimentally to guarantee 50 ± 10 fps of water passing the fuel plate surfaces.

Fabrication and Quality Assurance

Figure 6 shows the distinct fabrication and testing stages and the approvals, reviews, and quality assurance audits that the experiment was subjected to before insertion into VXF-17 reflector hole of HFIR. It is beyond the scope of this report to present the details of these operations other than those given previously in the section describing the test plates, irradiation facility, and PM capsule 1 experiment. The procedures and specifications used to fabricate and build the experiment complied with RDT Standards F2-2T and F2-4T and ORNL Standard Procedures SPP-39, "Quality Assurance Program."

From initiation of an experimental plan through insertion into HFIR, PM capsule 1 was constructed with a minimum of difficulties. In view of the complexity of the network shown in Fig. 6, the smoothness of our progression was somewhat surprising. The reasons behind the marked ease in building and irradiating PM capsule 1 must lie with the particular expertise of the people cited in the acknowledgments, good initial planning, and our adoption of RDT Standard F2-2T and its implementation during all phases of the work.

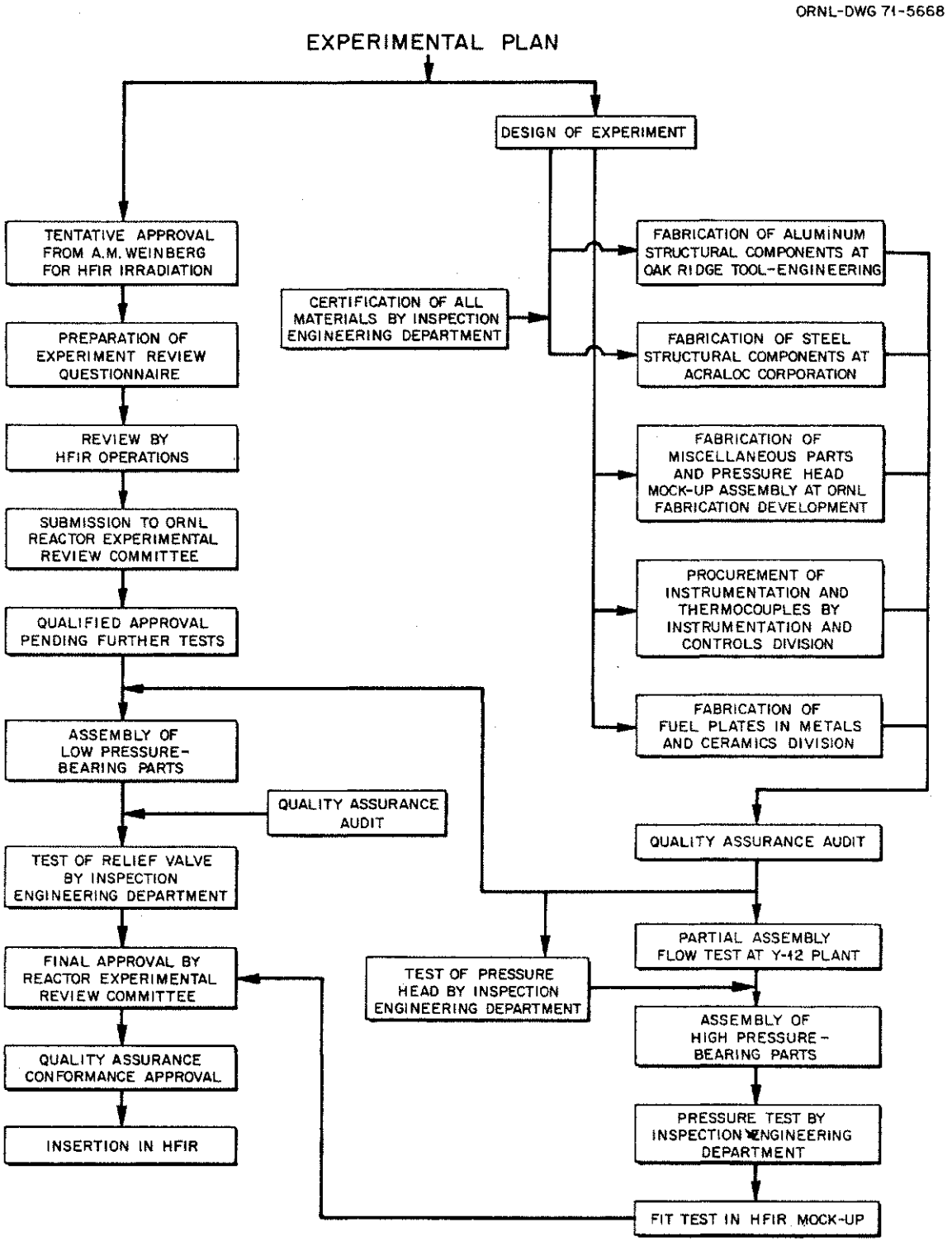

Fig. 6. Fabrication and Quality Assurance Network for PM Capsule 1.

Irradiation History

Irradiation of PM capsule 1 began January 11, 1971, and was terminated on schedule, on May 13, 1971. This period encompassed five reactor cycles (numbers 58 through 62), in which only four shutdowns occurred; all were scheduled for refueling the reactor. Because of the reactor's excellent plant factor of 95%, the experiment received the maximal exposure of 11,613 MWd of full-power operation at 100 MW.

During the period of irradiation, the center-line temperature of each of the three instrumented fuel plates was recorded continuously at four equidistant axial position. Figure 7 presents the chronological temperature histograms, recorded daily at 8:00 AM, from 4 of the 12 thermocouples. In this plot, the vertical lines represent either a shutdown or a startup of the reactor. At all times, thermocouple 9, which was located near the top of plate 05-06, indicated the maximum temperature for the experiment. Conversely, thermocouple 1 in the top of plate 01-02 near the coolant inlet end of the test assembly always gave the minimum temperature indication of the experiment. Thermocouples 6 and 7 were positioned symmetrically 0.75 in. above and below the reactor horizontal core midplane in plate 03-04. Since the thermal neutron flux distribution was also symmetric about the midplane, the single profile shown in Fig. 7 adequately represents their responses. The continuous temperature indications from the remaining eight thermocouples fell between those of thermocouples 1 and 9.

In general, the profiles shown in Fig. 7 depict three constant center-line core temperatures (within about ±2°C) for the entire duration of each cycle. These measured temperatures, however, decreased an average of 4.4°C with each succeeding cycle. The isothermal condition results primarily from the burnup of ^{235}U being balanced by a steady increase in neutron flux with time at the test location (i.e., the shielding of neutrons from the test assembly is continuously lessened during the cycle as the reactor control rods are withdrawn to maintain the reactor power). The reduction of the ^{235}U content of the instrumented fuel plates and the

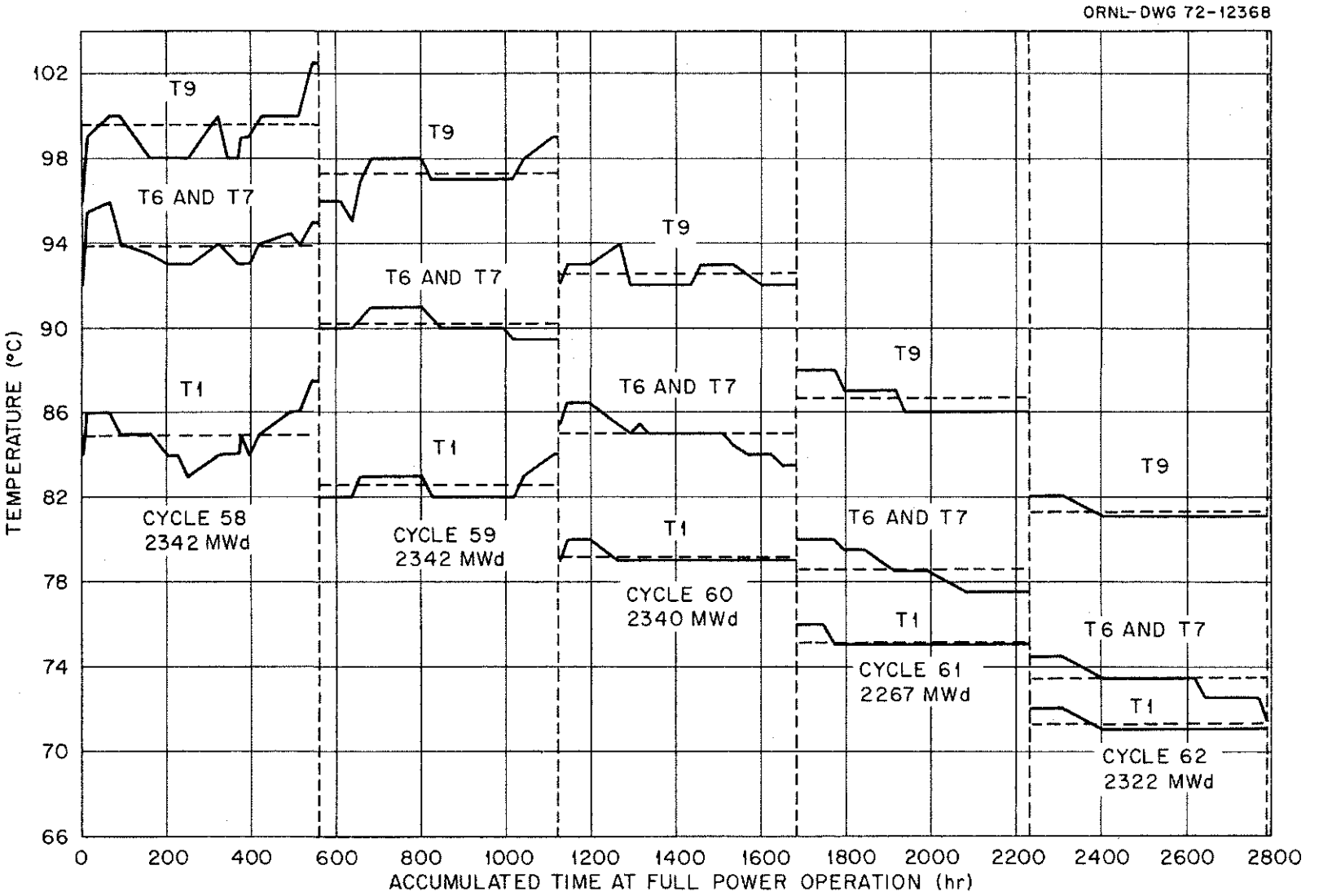

Fig. 7. Temperature Histogram of Selected Thermocouples from PM Capsule 1 Irradiations. Dashed lines indicate average cycle temperatures.

lower beginning-of-life neutron flux account for the downward stepwise changes in the measured temperatures for the next cycle.

Table 3 lists the total burnups in fissions per cubic centimeter of fuel core and the beginning-of-life, maximum, and end-of-life temperatures for each plate. The temperatures for the test plates were derived primarily from the direct center-line measurements of the instrumented

Table 3. Pertinent Center-Line Temperatures and Total Burnups for the Instrumented and Test Plates Irradiated in HFIR for 11,613 MWd

Plate	Core Composition (wt %)	Fissile Density ($g\ ^{235}U/cm^3$ of core)	Center-Line Temperature,[a] °C			Total Burnup[a,c] ($fissions/cm^3$)
			Beginning of Life	Maximum[b]	End of Life	
						$\times 10^{21}$
11-3	53 arc-cast UAl_x	0.00	49	49	49	0.004
21-4	54 arc-cast UAl_x	0.00	49	49	49	0.004
31-1	55 arc-cast UAl_x	0.00	49	49	49	0.004
12-3	47 high-fired U_3O_8	1.41	84	89	71	(1.98)
22-4	50 burned U_3O_8	1.43	81	86	68	(1.97)
32-4	51 arc-cast UAl_x	1.31	76	80	63	(1.79)
01-02-2	47 high-fired U_3O_8	1.40	(87, 91)	(89, 93)	(72, 74)	1.8, 2.0
13-4	40 high-fired U_3O_8	1.14	82	84	63	(1.81)
23-1	42 burned U_3O_8	1.15	80	83	62	(1.81)
33-4	54 arc-cast UAl_x	0.00	50	51	49	0.004
14-3	53 arc-cast UAl_x	1.32	88	92	69	(2.02)
24-3	52 arc-cast UAl_x	1.31	86	90	68	(1.92)
34-2	53 arc-cast UAl_x	1.32	84	88	67	(1.95)
03-04-1	44 high-fired U_3O_8	1.29	(94, 94)	(94, 94)	(71, 72)	2.1, 2.0
15-4	63 arc-cast UAl_x	1.67	92	98	78	(2.22)
25-4	64 arc-cast UAl_x	1.69	90	96	76	(2.17)
35-4	63 arc-cast UAl_x	1.66	88	94	74	(2.14)
16-2	40 high-fired U_3O_8	0.00	57	58	51	0.004
26-3	40 high-fired U_3O_8	0.00	56	57	51	0.004
36-2	24 α-Al_2O_3	0.00	53	54	51	0.00
05-06-1	51 high-fired U_3O_8	1.58	(97, 95)	(101, 99)	(80, 78)	2.7, 2.4

[a]Measured values are contained within parentheses; all others are calculated.

[b]Maximum temperature noted after exposure of 544 hr.

[c]For comparison, values ranging from 0.23 to 1.24×10^{21} were found in actual HFIR fuel elements by A. E. Richt, R. W. Knight, and G. M. Adamson, Jr., ORNL-4714 (December 1971).

plates. The Dittus-Boelter equation[13] for the heat transfer coefficient together with the initial fissile concentrations, an inlet coolant temperature of 49°C, a coolant velocity of 50 fps, and an estimation of the perturbed thermal neutron flux based on the actual burnups were also used in arriving at the test plate temperatures. The burnups were determined from measured changes[14] in the isotopic content of uranium contained in the fuel cores.

POSTIRRADIATION EXAMINATION OF THE TEST PLATES

All 18 test plates were inspected visually in the HFIR pool approximately 24 hr after shutdown of the experiment. The plates were free of warpage and distortion and generally appeared to be in excellent condition. No further examination of the three instrumented plates (01-02, 03-04, and 05-06) was attempted since the presence of the thermocouples rendered these plates virtually useless for more detailed analysis.

The test plates were allowed to cool for approximately three weeks in the pool and then transferred to the High Radiation Level Examination Laboratory (HRLEL) hot cells for detailed examination and evaluation. Examination consisted primarily of visual inspection of the plate surfaces for indications of structural damage or defects, determination of the plate weights and densities both before and after chemical removal of corrosion-product films, analytical determination of the specimen burnups, and metallographic examination.

In general, the examinations showed all 18 test plates to be in excellent condition. The density and weight-loss data indicated that the plates had increased less than 0.002 in. in thickness (including the oxide film formed on the plate surfaces during irradiation) and that corrosion had reduced the thickness of the cladding of these plates by less than

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[13]W. L. Badger and J. T. Banchero, *Introduction to Chemical Engineering*, McGraw-Hill, New York, 1955, p. 132.

[14]ASTM Designation E-244-6, "Standard Method of Test for Atom Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometric Method)," *1969 Book of ASTM Standards*, Part 30, American Society for Testing and Materials, Philadelphia, May 1969.

0.0002 in. Extensive visual and metallographic examination of sections from each plate revealed no indications of an actual or incipient structural failure of any of the test plates. Consequently, we conclude that all 18 plates performed quite satisfactorily under the imposed conditions (i.e., burnup levels of 1.8 to 2.2×10^{21} fissions/cm^3 at irradiation temperatures of 60 to 98°C). However, we did observe slight but significant differences in the swelling of the various fuel dispersions. As shown in Fig. 8, plates containing the UAl_x dispersions consistently swelled more than plates containing the burned or high-fired U_3O_8 dispersions when irradiated to comparable burnup levels. We also observed some rather unexpected changes in the microstructure of some of the fuel plates. These effects are discussed in greater detail in later sections of this report.

Swelling Behavior of UAl_x Dispersions

Fuel core swelling results for the seven fuel plates that contained the enriched UAl_x-Al dispersions are summarized in Table 4. These plates were irradiated as shown in Table 3 to burnups ranging between 1.79 and 2.22×10^{21} fissions/cm^3, resulting in fuel core volume increases of from 4.1 to 8.8%. Correlation of these data with fuel core fission density can be misleading. If these volume changes are plotted as a function of the fuel core fission density, as shown in Fig. 8, it would appear that the swelling of these dispersions is decreasing with increasing burnup. Such conclusion, of course, is completely erroneous. However, the swelling of these plates was approximately inversely proportional to the initial void content of the fuel dispersions. Thus, if fabrication voids are effective in accommodating swelling during irradiation, the behavior shown in Fig. 8 becomes more plausible. For example, if one compares the measured core volume increases with those predicted by our swelling model (which assumes swelling of 6.3 vol % per 10^{21} fissions/cm^3 and that voids are effective in accommodating swelling), one finds that each of these seven dispersions actually swelled about 1.2 ± 0.2 vol % less that expected (Fig. 9). As will be discussed later, there appears to be a

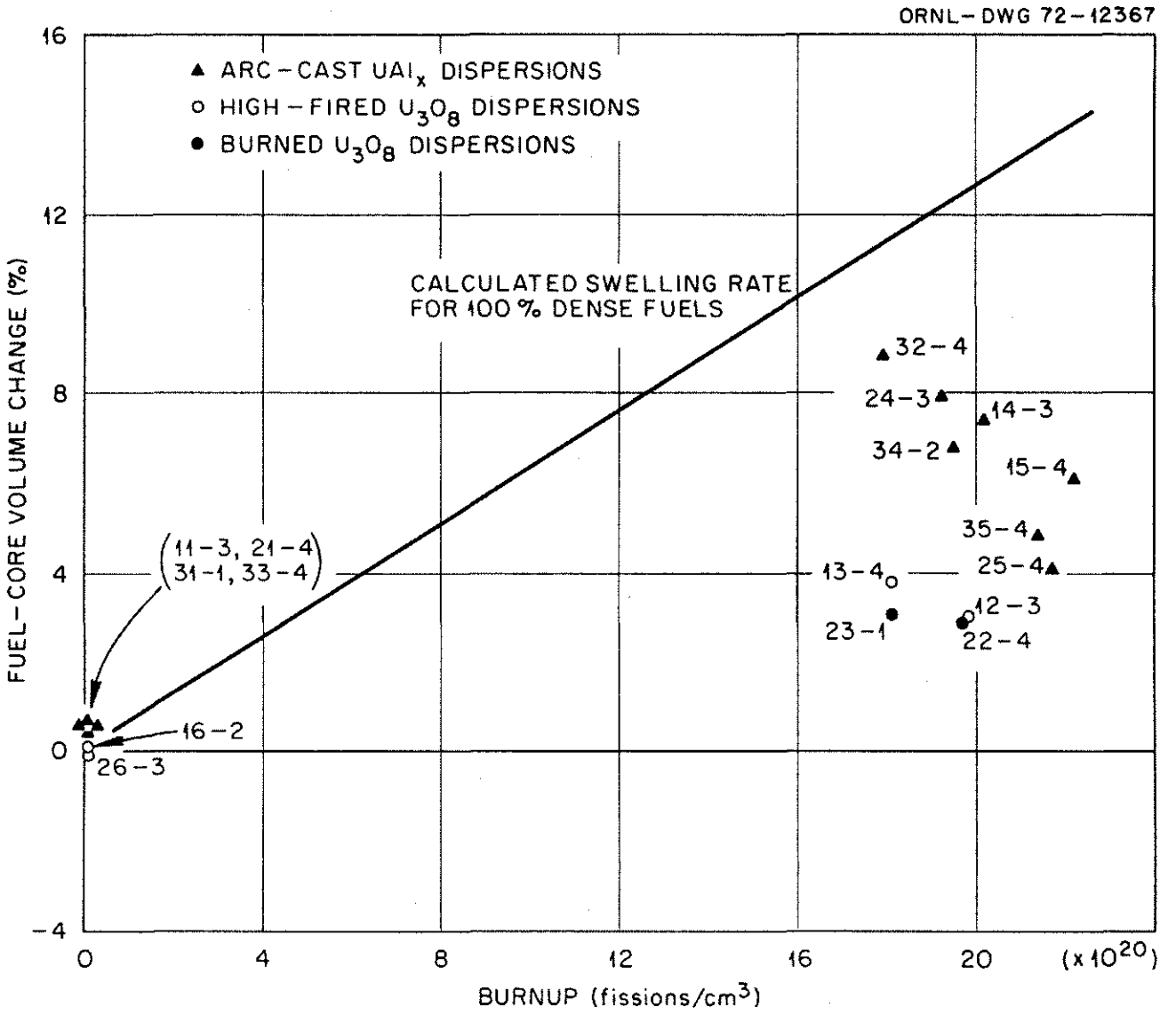

Fig. 8. Summary of Fuel-Core Swelling Data for Miniature Fuel Plates Irradiated in the ORNL PM-1 Experiment. All data points are identified by the fuel-plate identification number.

Table 4. Summary of Fuel Core Swelling Results for Enriched UAl_x Dispersions Irradiated in the ORNL PM-1 Experiment

Plate	UAl_x Loading (wt %)	Fines Content (vol %)	Fabrication Voids (vol %)	Fuel Core Swelling, %		
				Measured	Predicted[a]	Difference
32-4[b]	51	24	1.3	8.8	10.0	−1.2
24-3	52	24	3.3	7.9	8.9	−1.0
14-3[c]	53	24	4.2	7.4	8.6	−1.2
34-2	53	97	4.3	6.8	8.1	−1.3
15-4	63	24	6.6	6.1	7.4	−1.3
35-4	63	97	7.3	4.8	6.2	−1.4
25-4[c]	64	24	8.4	4.1	5.3	−1.2

[a] 6.3×10^{-21} times burnup (fissions/cm^3) minus voids (vol %).

[b] Plate was hot pressed after hot rolling to reduce the initial void content of the fuel core.

[c] Plates cold-rolled an additional 20% to increase the initial void content of the fuel cores.

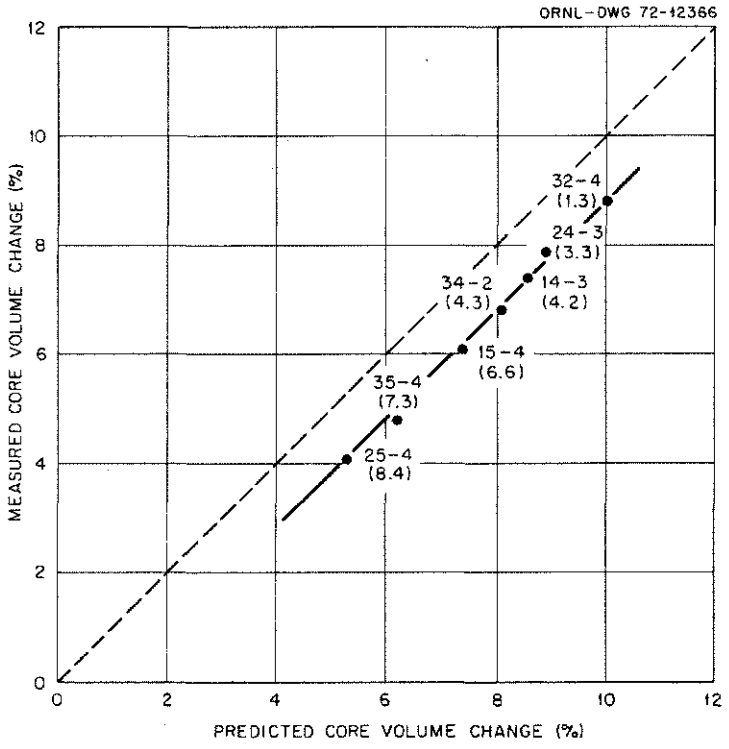

Fig. 9. Comparison Between Predicted and Measured Core-Volume Changes for Enriched UAl_x Dispersions. Data points are identified by plate number with initial void content given in parenthesis.

plausible explanation for the relatively constant discrepancy between the predicted and measured swelling values for these UAl_x-Al dispersions; however, for the following discussions it will be convenient to merely assume that the relatively good agreement between the predicted and measured swelling values proves that the swelling of these UAl_x dispersions is a function of both the initial void content and the fission density.

Influence of Fuel Loading

Previous work[15] at ORNL has shown that the as-fabricated void content of aluminum-base dispersions generally increases with increasing fuel loadings. One would therefore expect that dispersions containing higher UAl_x loadings would swell less than dispersions containing a lower loading at comparable fission densities. As shown in Table 4, this appears to be true since the three plates that contained about 63 wt % UAl_x (15-4, 25-4, and 35-4) actually swelled less than the four plates that contained about 52 wt % (14-3, 24-3, 32-4, and 34-2), even though the higher loaded plates had slightly higher fission densities. The relatively good agreement between the predicted and measured swelling values for all seven plates, however, indicates that the differences in the swelling behavior of these dispersions are a result of differences in the initial void content of as-fabricated fuel cores and not a result of the fuel loading *per se*.

Influence of Fabrication Schedule

The fabrication procedure used in manufacturing aluminum-base fuel plates can also affect the void content of the fuel dispersion.[15] For example, if the rolling schedule is altered to increase the relative amount of cold reduction, the void content of the fuel core will be increased somewhat above normal levels. If the relative amount of cold reduction is reduced or eliminated entirely, the void content of the fuel core will be somewhat less than normal. One would therefore expect plates

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[15]M. M. Martin and W. R. Martin, *Fabrication Voids in Aluminum-Base Fuel Dispersions*, ORNL-4611 (October 1970).

fabricated with a greater degree of cold rolling to swell less, while plates fabricated with a lesser degree of cold rolling to swell more than those plates fabricated by normal practice.

Comparison of the data in Table 4 shows this assumption to be correct. For example, the fuel cores of plates 32-4, 24-3, and 14-3 (all of which contained about 52 wt % UAl_x) swelled 8.8, 7.9, and 7.4% after burnups of 1.79, 1.92, and 2.02×10^{21} fissions/cm^3, respectively. Thus, the core of plate 32-4, which was hot pressed after rolling to reduce the void content of the dispersion to 1.3 vol % swelled more than that of plate 24-3 (which was fabricated by the normal rolling practice, resulting in about 3.3 vol % voids) even though the latter plate was exposed to a slightly higher burnup level. The core of plate 14-3, which was cold rolled an additional 20% to increase the void content to 4.2 vol %, swelled less than that of either plate 24-3 or 32-4, even though it was subjected to the highest fission density. A similar conclusion can be drawn from comparison of the swelling of plates 15-4 and 25-4, which contained about 63 wt % UAl_x.

These results clearly show that the *swelling behavior of UAl_x dispersions in aluminum can be affected by variations in the fuel-plate fabrication procedures.* However, the relatively good agreement between the predicted and measured swelling values for these five plates again indicates that the differences in the swelling behavior are a result of differences in the initial void content that result from the fabrication variations.

Influence of Fuel Fines

Current specifications limit the amount of fuel fines (i.e., particles < 44 μm in diameter or −325 mesh) to 10 and 25% of the total loading for the HFIR and ATR fuel plates, respectively. These limitations were established rather arbitrarily on the basis of a theoretical prediction that irradiation damage in dispersion fuels increases with decreasing fuel particle size. However, there is considerable economic incentive to permit the use of higher fines contents providing, of course, that such dispersions will perform satisfactorily during irradiation.

Consequently, two plates containing a large fraction of fuel fines (i.e., 97% of the total fuel loading) were irradiated in this experiment to permit comparison with plates containing the standard amount of fines.

Table 4 shows that the dispersions containing the high precentage of fuel fines (34-2 and 35-4) swelled slightly less than those containing the normal amount of fines (24-3 and 15-4) at both UAl_x loadings. Again, however, the good agreement between the predicted and measured swelling values for these four plates indicates that the differences in swelling result from differences in the initial void content of the dispersions and not a result of differences in the fines content *per se*.

Behavior of Depleted UAl_x Dispersions

Four fuel plates containing UAl_x dispersions depleted in ^{235}U were irradiated in the PM-1 experiment. These plates were included primarily to determine if the voids within the fuel cores were stable during irradiation when little or no fission product swelling would be expected. As shown in Table 5, the dispersions in all four plates swelled within ±0.1% of 0.6%. We believe the measured swelling values are accurate to ±0.2% and therefore these depleted UAl_x dispersions actually swelled.

At present, we have no completely satisfactory explanation for these small volume increases. However, these results indicate that the voids within the depleted UAl_x dispersions are stable during irradiation at relatively low temperatures (≈ 50°C) since one would expect that the cores would decrease in volume if the voids were being eliminated by sintering. This may not be true at higher irradiation temperatures or at high fission rates such as occur in enriched UAl_x dispersions. Other irradiation tests,[16] for example, have shown that enriched UAl_x dispersions actually can decrease significantly (as much as 7%) in volume when irradiated to low burnup levels at 130 to 180°C.

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[16]G. O. Hayner and K. C. Sumpter, *Reactor Engineering Branch Annual Report Fiscal Year 1969*, IN-1335, pp. 58–67.

Table 5. Summary of Fuel Core Swelling Results for Depleted UAl_x-Al Dispersions

Plate Number	UAl_x Loading (wt %)	Fines Content (wt %)	Fabrication Voids (vol %)	Fuel Core Swelling, %		
				Measured	Predicted	Difference
11-3	53	21	3.1	0.6	−3.1[a]	+3.7[a]
21-4	54	21	4.5	0.6	−4.5[a]	+5.1[a]
31-1	55	21	5.8	0.5	−5.8[a]	+6.3[a]
33-4	54	100	4.6	0.7	−4.6[a]	+5.3[a]

[a]Valid only if fabrication voids are annealed out during irradiation.

Comparison of PM-1 Results with Previous Tests

Before the PM-1 experiment, we had conducted only limited irradiation tests on UAl_x dispersions. However, in 1968 we irradiated[17] three UAl_x fuel plates in the G-12 loop of the ETR as part of a cooperative program with the Idaho Nuclear Corporation. As shown in Table 6, these plates were irradiated to somewhat lower burnup levels and at considerably higher temperatures than the plates in the PM-1 experiment. Although the fuel loadings, dispersoid composition, and irradiation temperatures differed considerably from those in the PM-1 test, the fuel core swelling results for the three G-12 plates were in good agreement with those from the PM-1 plates in that these dispersions also swelled about 0.8 to 1.4 vol % less than would be predicted by our swelling model. Thus it appears that the swelling behavior of UAl_x dispersions is independent of both the dispersoid composition and irradiation temperatures between 80 and 185°C.

Swelling Behavior of U_3O_8 Dispersions

Comparison Between Burned and High-Fired U_3O_8

At comparable fuel loadings, the cores of fuel plates containing burned U_3O_8 generally have significantly higher fabrication void contents

[17]M. M. Martin and A. E. Richt, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1969*, ORNL-4470, pp. 94-97.

Table 6. Summary of Fuel Core Swelling Data for Enriched 49 wt % UAl_x Dispersions Irradiated in the G-12 Loop of the ETR

Plate	Fabrication Voids (vol %)	Estimated Irradiation Temperature, °C		Burnup (fissions/cm³)	Fuel Core Swelling, %		
		Maximum	Minimum		Measured	Predicted	Difference
				(× 10^{21})			
0-24-959[a]	2.5	185	145	1.69	6.7	8.1	−1.4
0-24-964[b]	4.3	185	160	1.57	4.8	5.6	−0.8
0-24-965[b]	4.3	180	140	1.77	6.0	6.9	−0.9

[a]Solid-state reacted UAl_x (73.7 wt % U), 10% fines.

[b]Arc-cast UAl_x (75.2 wt % U), 10% fines.

than those containing high-fired U_3O_8. One would therefore expect that the burned U_3O_8 dispersions would swell less than high-fired U_3O_8 dispersions when irradiated to comparable burnup levels. As shown in Table 7, the high-fired dispersion did indeed swell slightly more than the burned U_3O_8 dispersion at both fuel loadings (i.e., plate 13-4 vs plate 23-1, and plate 12-3 vs plate 22-4).

Influence of Fuel Loading

As mentioned previously, the void content of aluminum-base dispersions generally increases with increasing fuel loadings. Consequently, one would expect that dispersions containing higher U_3O_8 loadings would swell less than those containing a lower loading when irradiated to comparable fission densities. The data in Table 7 also appears to confirm this conclusion. For example, plate 23-1 (which contained 42 wt % burned U_3O_8) swelled slightly more than plate 22-4 (50 wt % burned U_3O_8) even though the higher loaded plate sustained more fissions per unit volume. Similarly, plate 13-4 (which contained 40 wt % high-fired U_3O_8) swelled slightly more than plate 12-3 (47 wt % high-fired U_3O_8) even though the latter plate also was exposed to a slightly higher fission density. Again, however, it should be noted that the differences in the swelling behavior of both the burned and high-fired dispersions appear to be related to differences in the initial void content of the dispersions and not to differences in the fuel loading *per se*.

Behavior of Depleted U_3O_8 Dispersions

Two fuel plates containing high-fired U_3O_8 dispersions depleted in ^{235}U were included in the PM-1 experiment to determine if the voids within the fuel cores were stable during irradiation. As shown in Table 7, the volume of the cores of these plates did not change significantly as a result of irradiation. Consequently, we conclude that voids within the U_3O_8 dispersions are stable during irradiation at temperatures of 50 to 60°C. Again, however, this may not be true at higher irradiation temperatures or for enriched U_3O_8 dispersions where the fission rate within the fuel particles would be much higher. For example, fuel core volume

Table 7. Summary of Fuel Core Swelling Results on U_3O_8-Al Dispersions. Irradiation temperatures and burnup are given in Table 3.

Plate Number	Dispersoid Loading (wt %)	Fines Content (wt %)	Fabrication Voids (vol %)	Fuel Core Swelling, %		
				Measured	Predicted[a]	Difference
			Burned U_3O_8-Al Dispersions			
23-1	42	11	6.4	3.1	5.0	−1.9
22-4	50	11	8.2	2.9	4.2	−1.3
			High-Fired U_3O_8-Al Dispersions			
13-4	40	10	3.0	3.8	8.5	−4.7
12-3	47	10	4.1	3.0	8.4	−5.4
16-2[b]	40	9	2.8	0.1	−2.8[c]	+2.9[c]
26-3[b]	40	39	2.3	−0.1	−2.3[c]	+2.2[c]

[a] 6.3×10^{-21} times burnup (fissions/cm^3) minus voids (vol %).

[b] These plates contained U_3O_8 fuel particles that were depleted in ^{235}U and therefore were subjected to relatively low fission burnups.

[c] These numbers valid only if the fabrication voids anneal out during irradiation and the cover plates and matrix collapse to fill the volume.

decreases of 1 to 2% have been observed[18] when enriched U_3O_8 dispersions were irradiated to very low burnup levels at approximately 130°C.

Comparison of the PM-1 Results with Previous Tests

Before the PM-1 experiment, we had irradiated[19,18] a number of miniature fuel plates containing aluminum-base dispersions of burned or high-fired U_3O_8 in the G-12 loop of the ETR. Fuel core swelling results for these plates are summarized in Table 8. As would be expected from consideration of the differences in the initial void content, the burned U_3O_8 dispersions generally swelled less than the high-fired U_3O_8 dispersions at comparable fission densities. However, as shown in Fig. 10, the burned dispersions rather consistently swelled about 1 vol % less than predicted by our swelling model, while the high-fired dispersions swelled about 3 vol % less than expected. In comparison, in the PM-1 experiment the burned U_3O_8 dispersions swelled about 2 vol % less than expected, while the high-fired dispersions swelled about 5 vol % less than predicted. Possible explanations for the discrepancy between the measured and predicted swelling values for these dispersions and the differences between the G-12 and PM-1 results will be presented later.

IRRADIATION-INDUCED MICROSTRUCTURAL CHANGES

Metallographic examinations of the test plates from the PM-1 experiment indicated all to be in excellent physical condition. No evidence of blisters, core-cladding separation, matrix cracking, or any other type of structural defect that could be considered indicative of an actual or incipient failure of a fuel plate was apparent in any of the sections of these fuel plates. Consequently, we conclude that all fuel plates in this experiment performed quite satisfactorily under the imposed test conditions.

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[18] A. E. Richt and M. M. Martin, *Fuels and Materials Development Program Quart. Progr. Rep. Dec. 31, 1969*, ORNL-4520, pp. 266–72.

[19] M. M. Martin and A. E. Richt, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1969*, ORNL-4470, pp. 94–97.

Table 8. Summary of Fuel Core Volume Changes for U_3O_8-Al Dispersions Irradiated in the G-12 Loop of the ETR

Plate	Dispersoid Loading (wt %)	Fines Content (wt %)	Fabrication Voids (vol %)	Estimated Irradiation Temperature, °C		Burnup (fissions/cm^3)	Fuel Core Swelling, %		
				Maximum	Minimum		Measured	Predicted	Difference
						($\times 10^{21}$)			
				High-Fired U_3O_8-Al Dispersions					
0-67-974	40	10	3.2	205	170	1.79	4.7	8.0	−3.3
0-67-982	40	25	3.1	195	160	1.80	5.1	8.3	−3.2
0-67-986	40	51	3.0	200	165	1.81	6.2	8.4	−2.2
0-56-899	50	3	3.3	150	125	1.17	1.4	4.1	−2.7
0-56-957	50	3	3.2	150	125	2.24	7.6	11.0	−3.4
				Burned U_3O_8-Al Dispersions					
0-3-893	45	3	7.3	175	135	1.55	2.0	2.6	−0.6
0-68-997	49	11	8.1	175	135	1.97	3.6	4.4	−0.8
0-68-1638	49	26	8.0	220	170	1.27	0.4	0.1	+0.3
0-68-1633	49	26	7.8	215	165	1.27	0.4	0.2	+0.2
0-68-1642	49	26	5.9	210	160	1.27	1.7	2.2	−0.5
0-68-1605	49	26	8.5	165	125	1.99	3.0	4.1	−1.1
0-68-1607	49	54	8.5	170	130	1.95	3.1	3.9	−0.8
0-56-894	53	3	8.9	130	100	1.36	−1.4	−0.3	−1.1

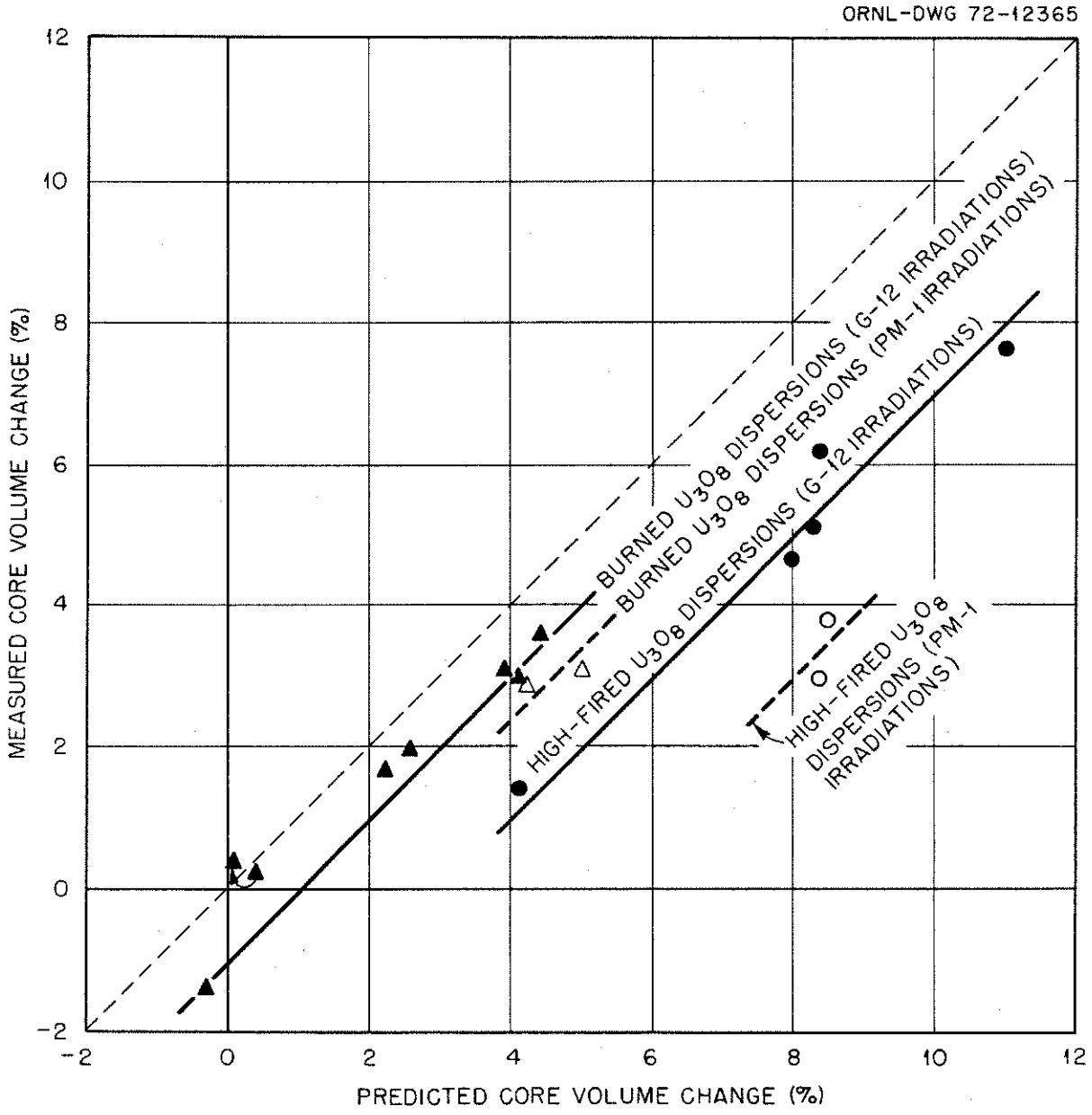

Fig. 10. Comparison Between Measured and Predicted Core Volume Changes for Burned and High-Fired U_3O_8 Dispersions Irradiated in the ETR G-12 and ORNL PM-1 Experiments.

The microstructure of the fuel dispersion in many of the uranium-bearing test plates, however, differed considerably from that before irradiation. This was not unexpected since it has been known for several years that irradiation can alter the microstructure of aluminum-base fuel dispersions. The exact nature of these microstructural changes is still not completely understood; however, they appear to be related to at least two different mechanisms: (1) chemical reactions between the fuel particles (i.e., UAl_x or U_3O_8) and the aluminum matrix material, and (2) changes in the chemical and physical composition of the fuel particles as a result of the generation of fission products. Since these microstructural changes may have some relationship to the swelling behavior of the different types of dispersions during irradiation, a short summary of the metallographic observations appears to be warranted.

Typical examples of the microstructural changes that occurred as a result of the irradiation of the enriched UAl_x fuel plates in the PM-1 experiment are shown in Figs. 11 and 12. As apparent in Fig. 11, the voids and cracks initially present within the fuel particles of the as-fabricated dispersion almost completely disappeared during irradiation. This, of course, is consistent with the behavior predicted by our swelling model, which assumes that all voids initially present within the dispersion will eventually be eliminated by swelling of the fuel particles during irradiation. It is also perhaps interesting to note that there are no clearcut indications of fission-gas bubbles within the fuel particles of the irradiated plate, even at these relatively high burnup levels (about half the total uranium atoms fissioned). This observation is consistent with other irradiation testing results,[20] which indicate that diffusion and agglomeration of fission gases in UAl_x compounds are relatively slow at these irradiation temperatures.

Figure 12 also shows that the microstructure of the fuel particles is altered significantly during irradiation. Initially the UAl_x particles consist primarily of a mixture of UAl_3 with lesser amounts of UAl_4 and

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[20] G. W. Gibson, *The Development of Powdered Uranium-Aluminide Compounds for Use as Nuclear Reactor Fuels*, IN-1133 (December 1967).

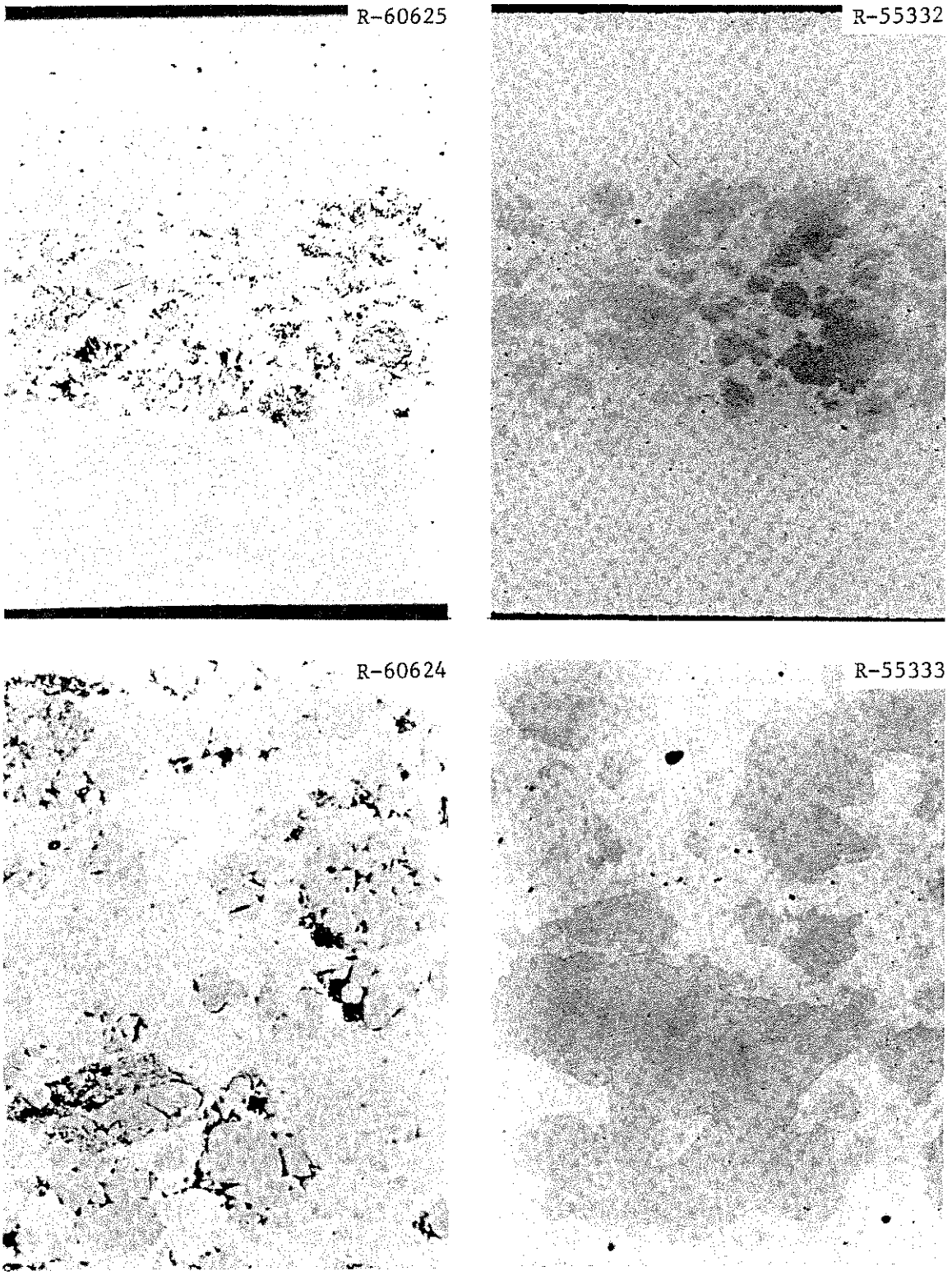

Fig. 11. Typical Microstructure of Enriched UAl_x Fuel Plates Unirradiated (Left; plate 14-2) and Irradiated in the PM-1 Experiment (Right; plate 14-3). Plate cross section (top), 75×; fuel dispersion (bottom), 250×. As polished.

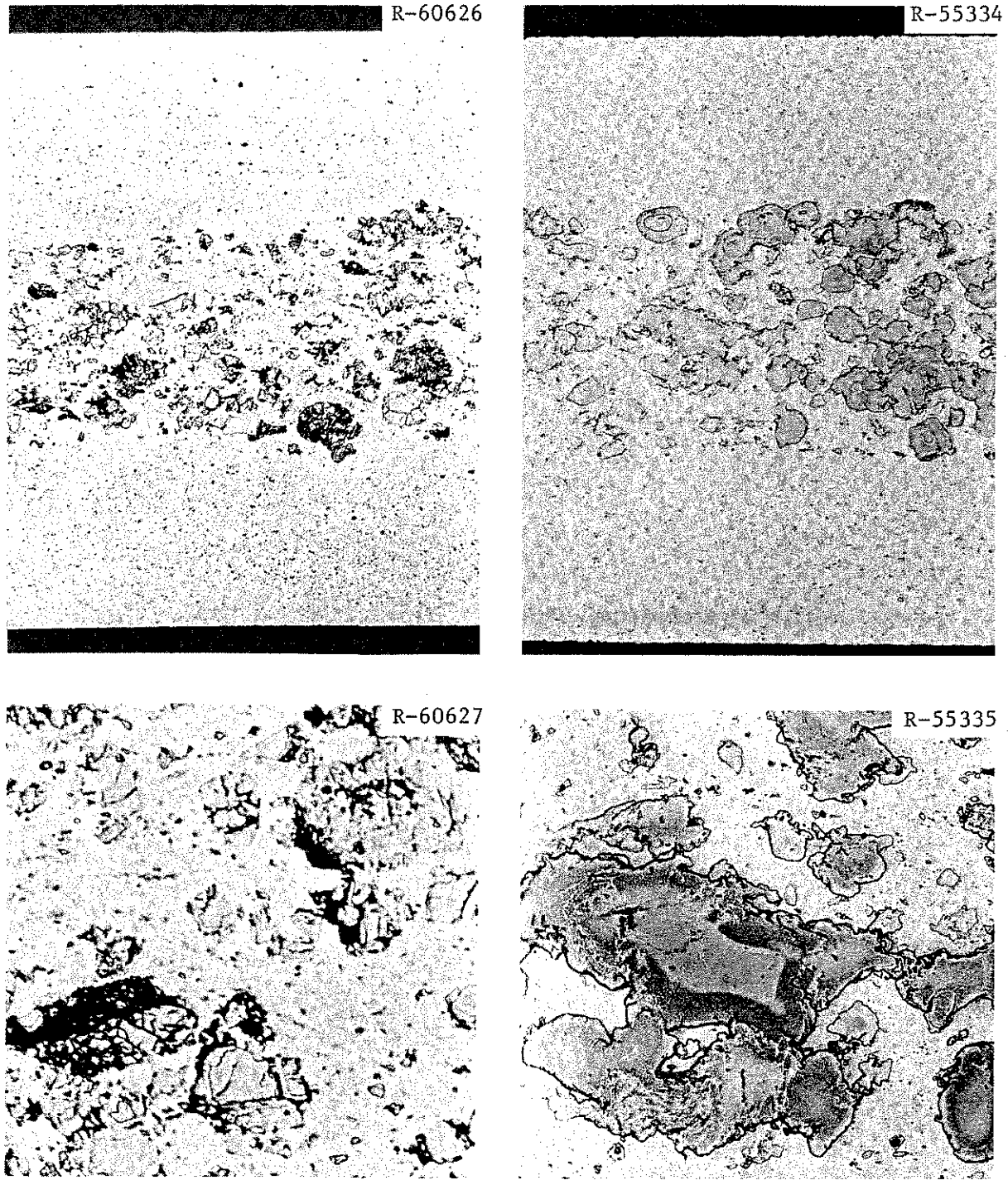

Fig. 12. Typical Microstructures of Enriched UAl_x Fuel Plates Unirradiated (Left; plate 14-2) and Irradiated in the PM-1 Experiment (Right; plate 14-3). Plate cross section (top), 75×; fuel dispersion (bottom), 250×. Etched with 1% HF, 10% HNO_3, 89% H_2O.

possible traces of UAl_2 and free aluminum. After irradiation, three or four phases appear to be present, and the overall morphology of the particle is quite different. Many of the fuel particles in the irradiated dispersion also exhibit flowlike patterns, which suggest that the normally hard, brittle UAl_x particles deformed plastically during irradiation. The overall significance of these microstructural changes is not known; however, from prior observations[21] we suspect that some of the UAl_3 initially present within the fuel particles has been converted to UAl_4 by reaction with the aluminum matrix during irradiation.

As mentioned previously, the cores of the fuel plates that contained the depleted UAl_x dispersions swelled about 0.6% as a result of irradiation in the PM-1 experiment. We initially thought that this small volume increase might be a result of transformation of some of the UAl_3 in these fuel particles to UAl_4. However, as shown in Figs. 13 and 14, these plates showed little or no significant differences in the microstructure before and after irradiation. Since a slight degree of transformation would be difficult to detect metallographically, we cannot entirely eliminate the possibility that such a transformation is responsible for the slight volume increase; however, such an explanation appears to be unlikely.

Typical examples of the microstructural changes that occurred during irradiation of the enriched burned and high-fired U_3O_8 dispersions in the PM-1 experiment are shown in Figs. 15 and 16, respectively. As expected, both types of fuel particles reacted with the aluminum matrix during irradiation, with the extent of reaction being greater for the burned U_3O_8 dispersions. The extent of reaction in both types of dispersions, however, was considerably less than was observed in similar fuel plates

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[21]A. E. Richt, C. F. Leitten, Jr., and R. J. Beaver, "Radiation Performance and Induced Transformations in Aluminum-Base Fuels," pp. 469–88 in *Research Reactor Fuel Element Conference, September 17–19, 1962, Gatlinburg, Tennessee,* TID-7642, Book 2 (1963).

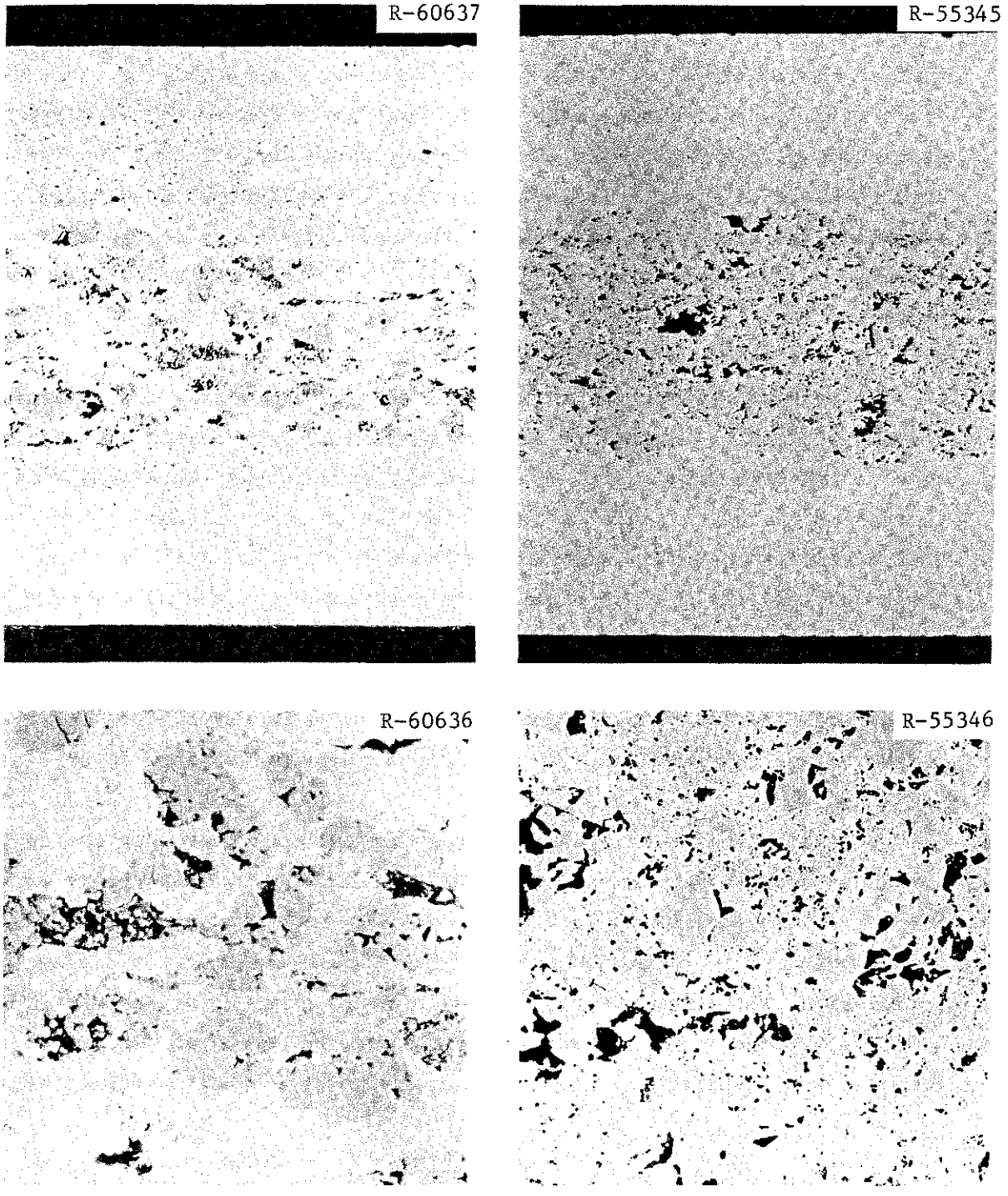

Fig. 13. Typical Microstructures of Depleted UAl_x Fuel Plates Unirradiated (Left; plate 21-2) and Irradiated in the PM-1 Experiment (Right; plate 21-4). Plate cross sections (top), 75×; fuel dispersion (bottom), 250×. As polished.

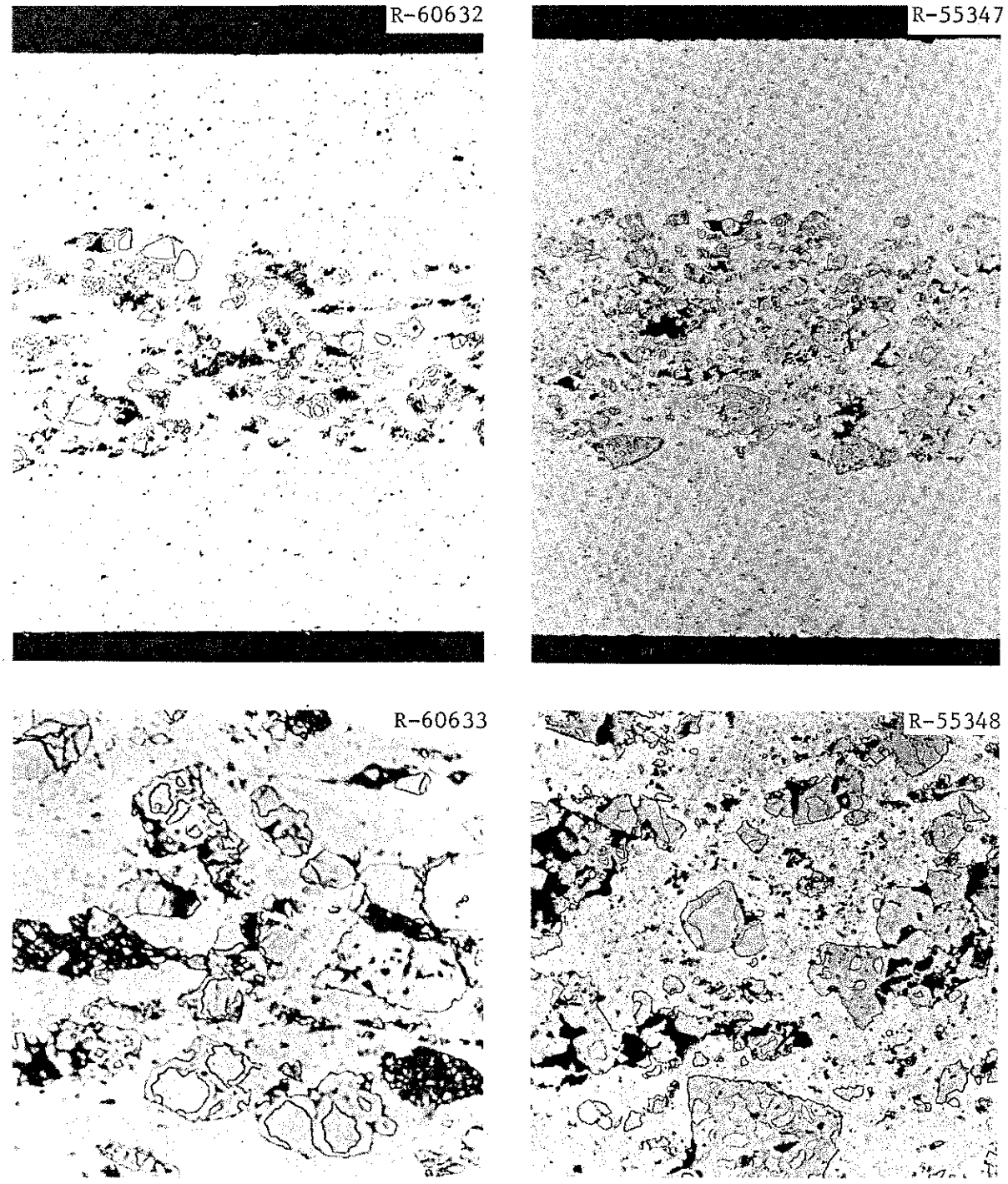

Fig. 14. Typical Microstructures of Depleted UAl_x Fuel Plates Unirradiated (Left; plate 21-2) and Irradiated in the PM-1 Experiment (Right; plate 21-4). Plate cross sections (top), 75×; fuel dispersion (bottom), 250×. Etched with 1% HF, 10% HNO_3, 89% H_2O.

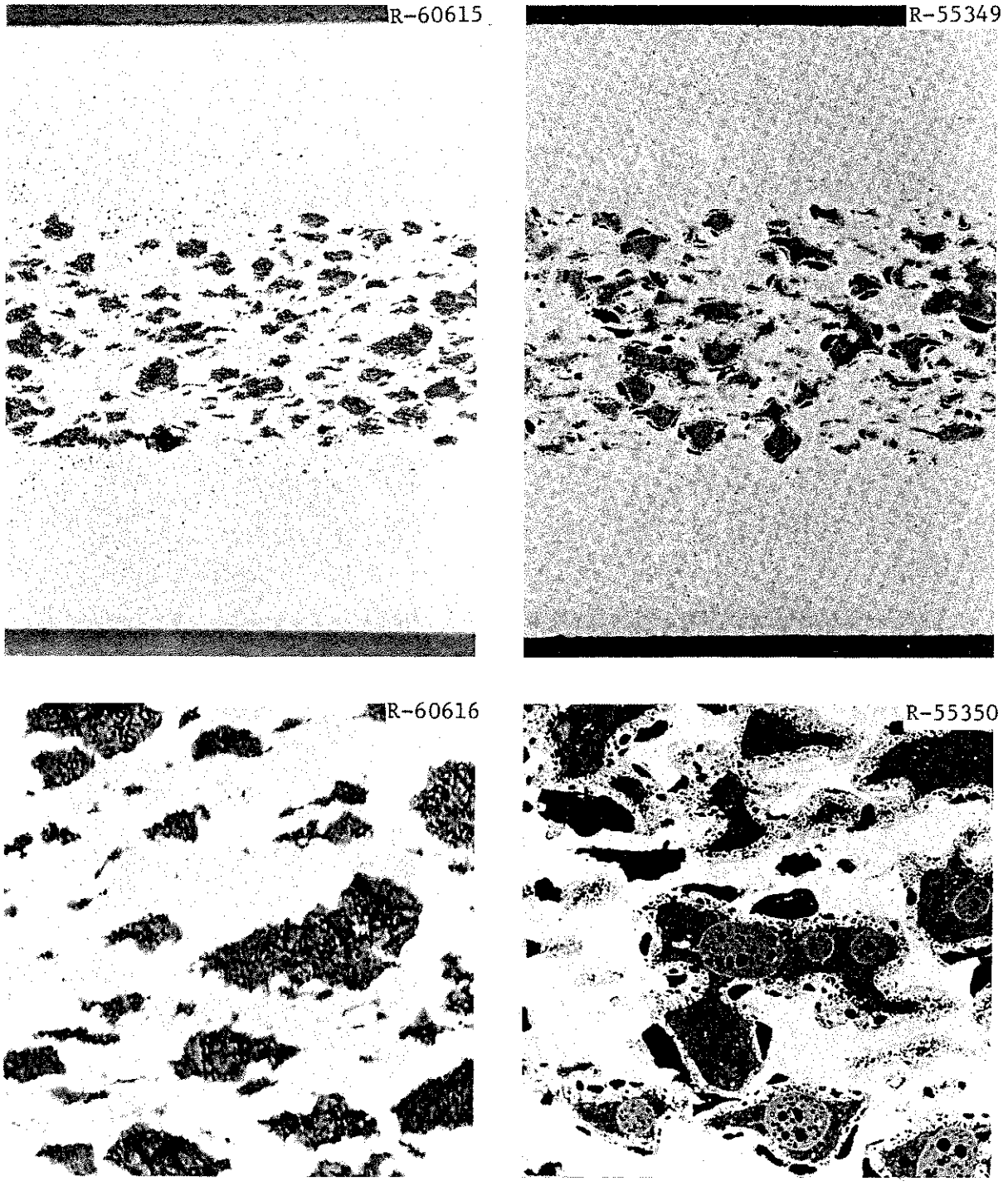

Fig. 15. Typical Microstructures of Burned U_3O_8 Fuel Plates Unirradiated (Left; plate 22-1) and Irradiated in the PM-1 Experiment (Right; plate 22-4). Plate cross section (top), 75×; fuel dispersion (bottom), 250×. As polished.

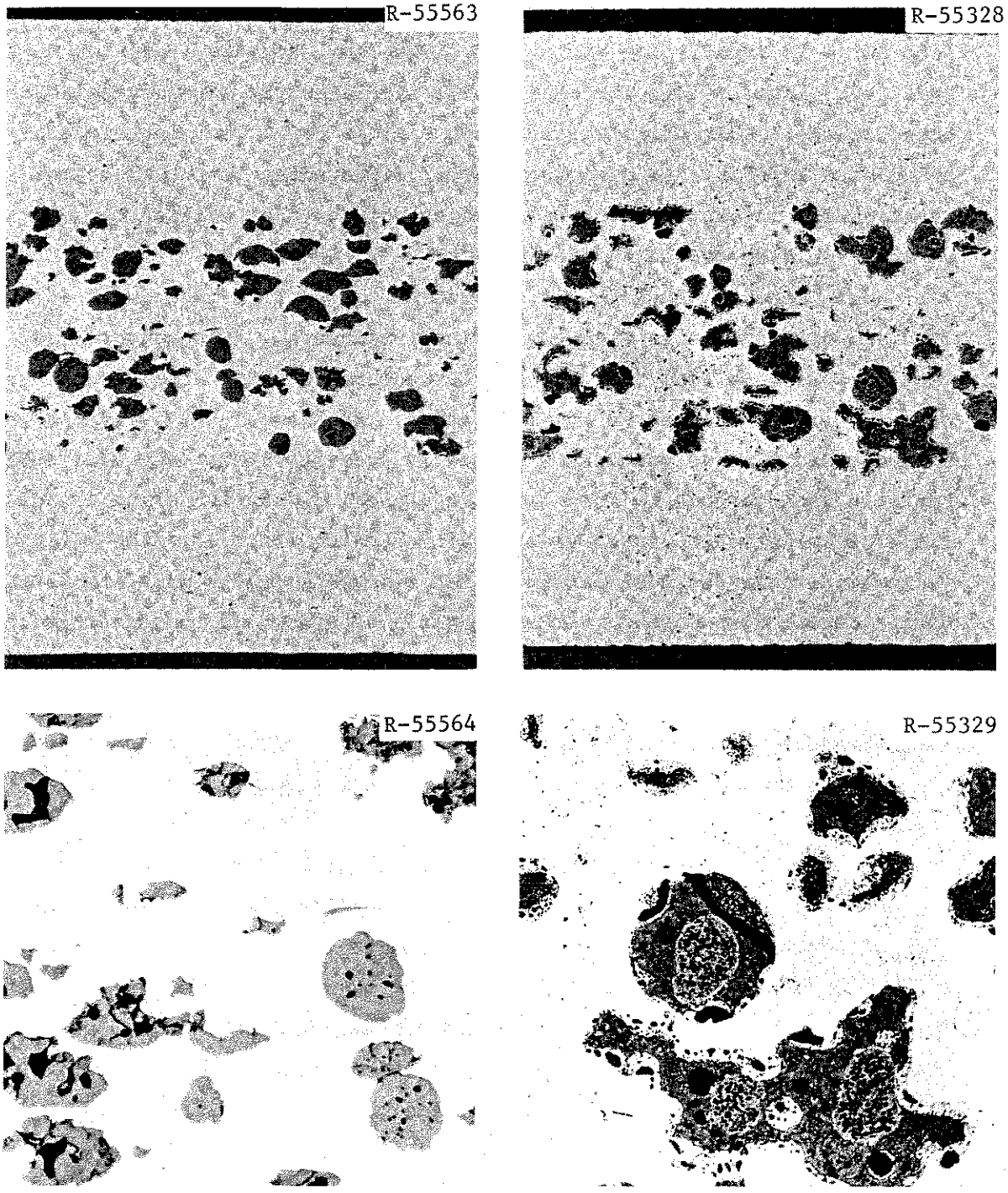

Fig. 16. Typical Microstructure of Enriched, High-Fired U_3O_8 Fuel Plate Unirradiated (Left; plate 13-2) and Irradiated in the PM-1 Experiment (Right; plate 13-4). Plate cross section (top), 75×; fuel dispersion (bottom), 250×. As polished.

from our G-12 irradiation tests.[22,23] The less extensive reaction in the PM-1 test plates is attributed to their significantly lower irradiation temperatures. This is consistent with earlier data[24] on high-fired oxide irradiated in HFIR. As discussed later, differences in the extent of reaction may account for the discrepancy between the swelling behavior of the U_3O_8 dispersions in the PM-1 and G-12 tests.

Fuel plates in PM-1 that contained the high-fired U_3O_8 depleted in ^{235}U did not swell significantly during irradiation. Consequently, we were surprised to find that the microstructure of these dispersions had been significantly altered by irradiation. As shown in Fig. 17, the numerous small voids and cracks initially present within each U_3O_8 fuel particle agglomerated to form one large spherical cavity during irradiation. Examination of these plates after etching (Fig. 18) also revealed indications that the fuel particles had reacted slightly with the aluminum matrix during irradiation. Areal anaylsis of groups of photomicrographs showed that the overall void content of the core of the irradiated fuel plates was essentially the same as that of identical unirradiated plates. Thus irradiation apparently does not affect the overall void content of the dispersion but does redistribute the smaller voids into a few large cavities.

Attempts to identify the phases present after irradiation of the depleted and enriched dispersions by x-ray diffraction techniques were unsuccessful since only peaks corresponding to the aluminum matrix material were found. This could mean that the other phases were not present in quantities sufficient for detection or that they were amorphous. A limited electron-microprobe analysis on one of the irradiated (enriched) high-fired U_3O_8 dispersions, however, indicated that at least some of

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[22]M. M. Martin and A. E. Richt, *Metals and Ceramics Div. Annu. Progr. Rep. June 30, 1969*, ORNL-4470, pp. 94–97.

[23]A. E. Richt and M. M. Martin, *Fuels and Materials Development Program Quart. Progr. Rep. Dec. 31, 1969*, ORNL-4520, pp. 266–72.

[24]A. E. Richt, R. W. Knight, and G. M. Adamson, Jr., *Postirradiation Examination and Evaluation of the Performance of HFIR Fuel Elements*, ORNL-4714 (December 1971), pp. 50–53.

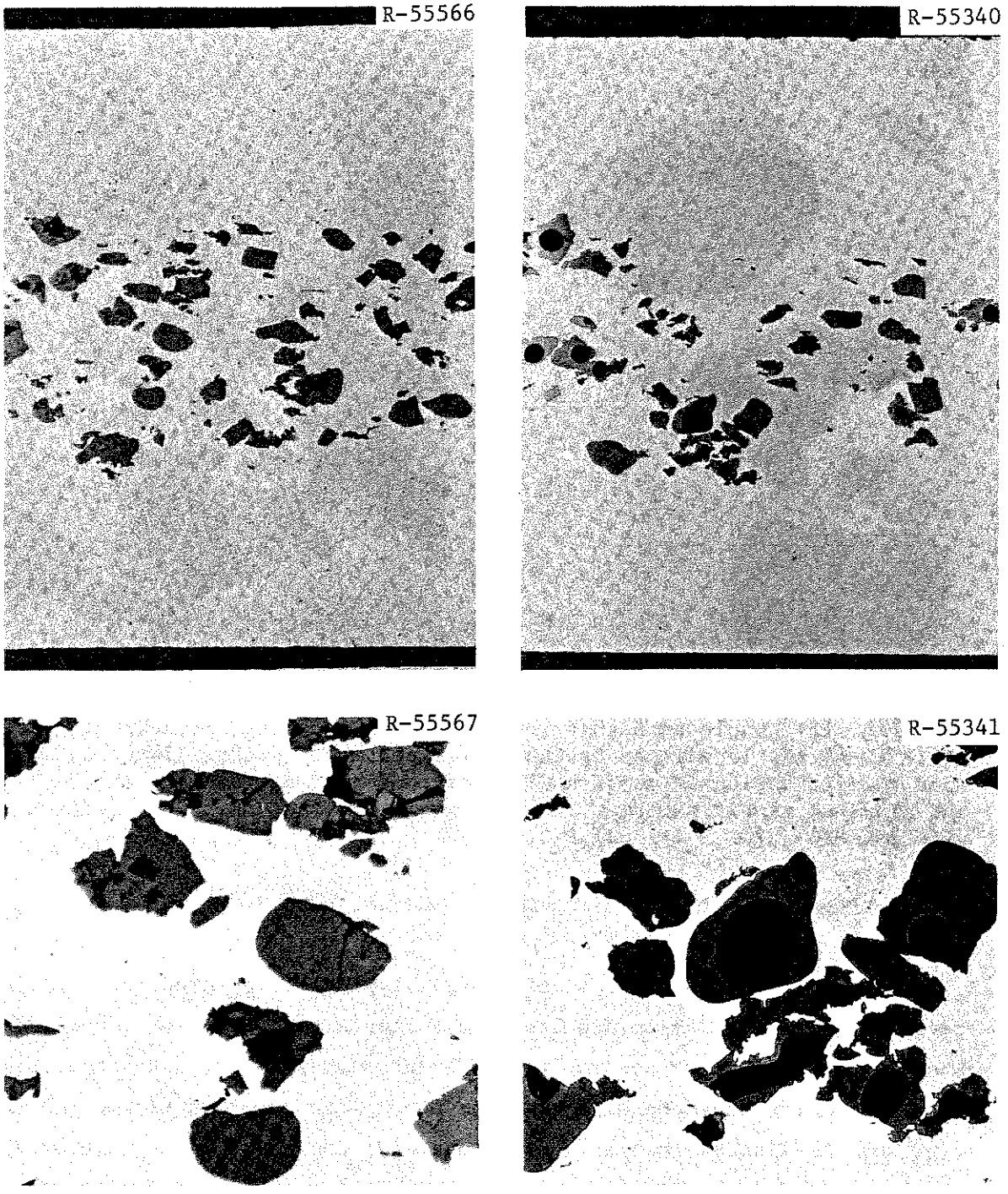

Fig. 17. Typical Microstructure of Depleted High-Fired U_3O_8 Fuel Plates Unirradiated (Left; plate 16-4) and Irradiated in the PM-1 Experiment (Right; plate 16-2). Plate cross section (top), 75×; fuel dispersion (bottom), 250×. As polished.

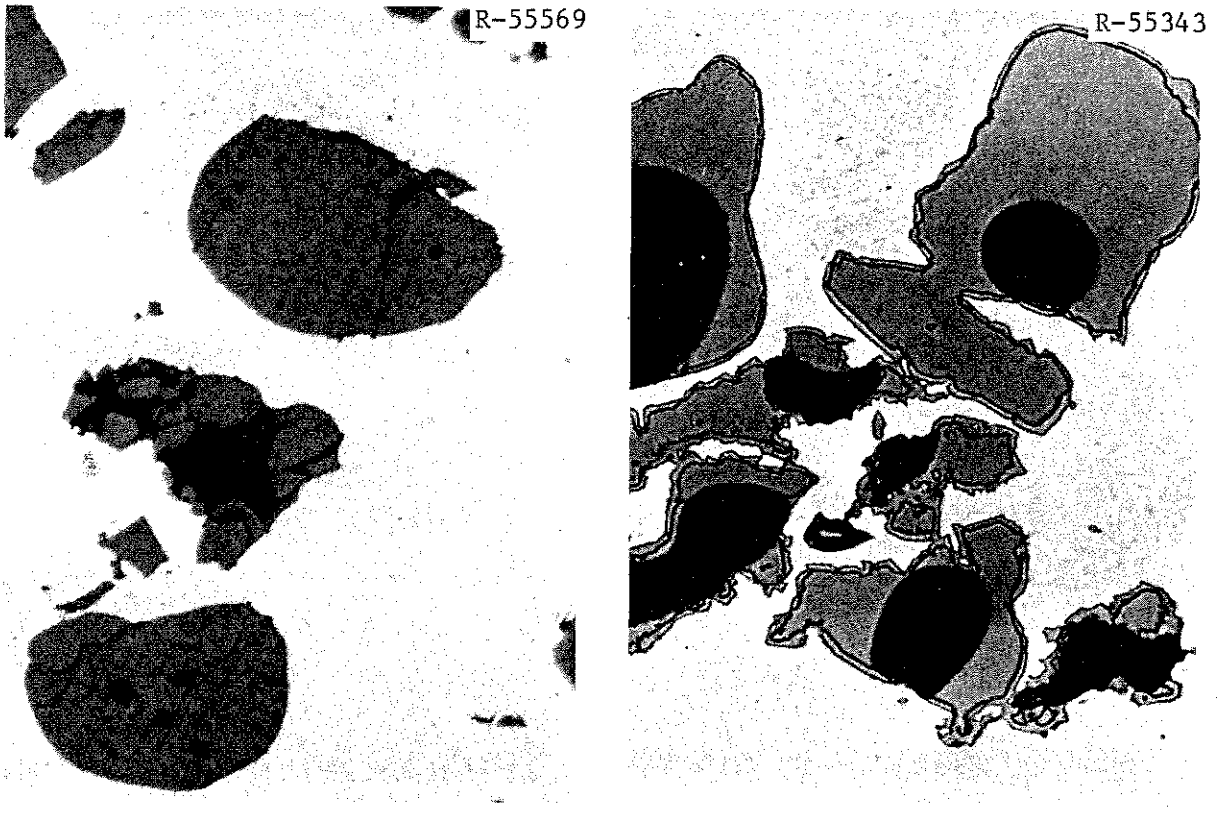

Fig. 18. Depleted High-Fired U_3O_8-Al Dispersion Unirradiated (Left; plate 16-4) and Irradiated in the PM-1 Experiment (Right; plate 16-2). Note redistribution of voids and indications of reaction between the U_3O_8 and aluminum matrix material in the irradiated specimen. 750×. Etched with 20% HNO_3, 20% H_2O_2, 60% H_2O.

these phases are a result of chemical reaction between the U_3O_8 and the aluminum matrix.

As shown in Fig. 19, a scan across a typical irradiated fuel particle revealed that the uranium and aluminum contents varied in an inverse, step-like manner corresponding to different phases in the microstructure. Such variations would be expected from incomplete reaction between the U_3O_8 and the aluminum matrix. Unfortunately, programmatic restrictions prevented us from determining either the absolute concentration of uranium and aluminum or the relative or absolute concentration of various fission-product species in these different phases. Such data might prove useful in understanding the effects of the reaction upon the swelling behavior of these dispersions.

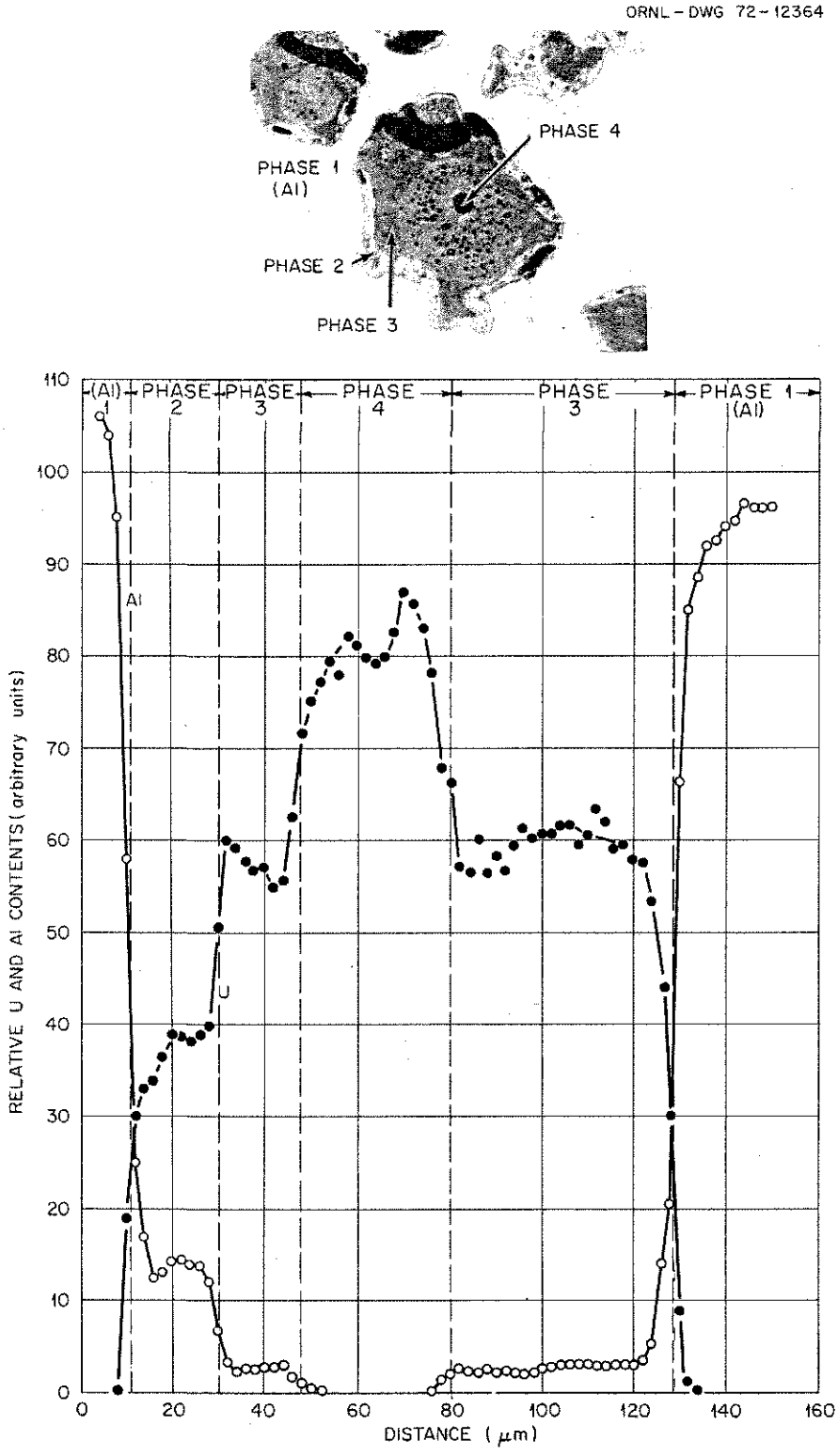

Fig. 19. Relative Uranium and Aluminum Distribution Across a Typical Fuel Particle of an Irradiated U_3O_8 Fuel Dispersion

Comparison of the microstructures of the depleted (Fig. 17) and enriched (Fig. 16) U_3O_8 fuel plates might suggest that the voids within the enriched U_3O_8 fuel particles also migrate to form large spherical cavities early during irradiation and that these cavities subsequently fill up with fission products. However, the microprobe analysis shows that the suspected filled-in region contains the highest uranium content of the various phases present in the microstructure. Thus such an assumption is unwarrented.

PERFORMANCE OF α-Al_2O_3 DISPERSION

Plate 36-2, which contained the 24 wt % α-Al_2O_3 dispersion, exhibited no deleterious effects after irradiation to perturbed thermal neutron fluences of about 1.5×10^{21} neutrons/cm^2. Density measurements showed that the dispersion had swelled less than 0.05% as a result of irradiation. As shown in Fig. 20, no indications of structural damage were observed. Consequently, we conclude that the α-Al_2O_3 dispersions will perform satisfactorily in the filler section of the HFIR fuel plates.

ACCURACY OF MODEL TO PREDICT SWELLING OF DISPERSION FUELS DURING IRRADIATION

General Considerations

Our model for predicting swelling of aluminum-base dispersion fuels can be described mathematically by the simple equation

$$\Delta V/V = (6.3 \times 10^{-21})B - V_v \quad , \tag{1}$$

where

$\Delta V/V$ = predicted change in core volume (%),

B = burnup (fissions/cm^3 of fuel core),

V_v = void content of fuel core (vol %).

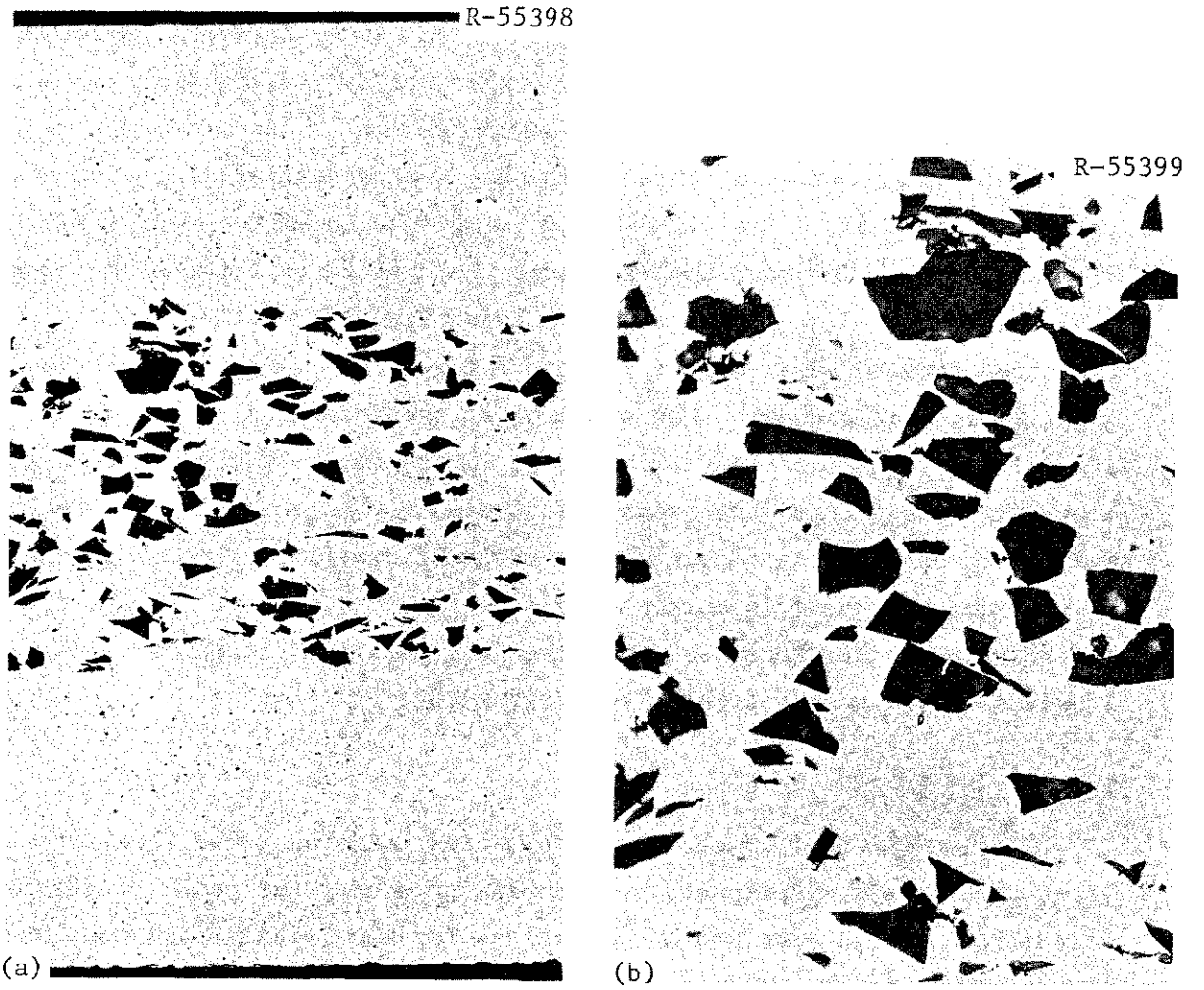

Fig. 20. Microstructure of Plate 36-2 After Irradiation for 11,613 MWd in the VXF-17 Position of the HFIR. (a) Plate cross section, 100×; (b) Al_2O_3-Al dispersion core, 250×. As polished.

The theoretical swelling rate of 6.3×10^{-21} vol % per fission per cm^3 represents the calculated volume change for ^{235}U fission in uranium[25] and urania.[26]

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[25]J. A. Brinkman, "Fundamentals of Fission Damage," pp. 1–12 in *Nuclear Metallurgy,* Vol. 6 (A symposium on effects of irradiation on fuel and fuel elements, Chicago, Nov. 4, 1959), IMD Special Report Series No. 9, The Metallurgical Society of American Institute of Mining, Metallurgical, and Petroleum Engineers, New York.

[26]A. R. Olsen, D. A. Douglas, Y. Hirose, J. L. Scott, and J. W. J. W. Ullmann, "Properties and Prospects of Thoria-based Nuclear Fuels," pp. 289–310 in *Proc. Brit. Ceram. Soc.* 7, The British Ceramic Society, Shelton House, Shelton, Stoke-on-Trent, England, February, 1967.

Results of our G-12 and PM-1 irradiation tests, however, have shown that enriched UAl_x, burned U_3O_8, and high-fired U_3O_8 dispersions rather consistently swell about 1, 1 to 2, and 3 to 5 vol % less, respectively, than would be predicted by this equation. Such discrepancies, of course, raise questions as to the validity of using this model to predict swelling of these fuels. However, the fact that the difference between the measured and predicted swelling values is essentially a constant for each of the three types of dispersions is rather convincing evidence that the model is basically sound. For example, if fabrication voids were not effective in accommodating swelling, or if the fuel actually swelled at a rate different from that assumed in the model, an essentially constant difference between the measured and predicted swelling values for a relatively large number of specimens would be highly unlikely. It therefore appears that these discrepancies arise from some additional factor(s). A discussion of such possible factors follows.

Fabrication Voids

Consideration of numerous possibilities leads us to believe that at least part of the disprepancy between the predicted and measured swelling values results from the use of low values for the initial void content of the plate cores. This, in turn, results in a corresponding overestimation of the predicted swelling values for the dispersions. Unfortunately, there is no known method to directly determine the void content of the core within this type of fuel plate. Consequently, the void content must be calculated from relationships that require rather precise knowledge of the true densities of the cladding, fuel (i.e., UAl_x or U_3O_8), and aluminum matrix materials (the method used in these calculations is described in greater detail in the Appendix). Relatively small errors in the density of these constituents can result in significant errors in the calculated void contents. For example, an error of only 0.01 g/cm^3 in the density of the cladding material could result in an error of 1 vol % in the calculated void content of the dispersion in these small fuel plates.

Although the density of the cladding can be determined rather precisely by measurements on scrap sections from the as-fabricated fuel

plates, one cannot unambiguously define the true density of either the fuel compound or the aluminum matrix. One also cannot use theoretical values for the densities of these two constituents because their specific composition can vary considerably. We therefore chose to use toluene-pycnometric values obtained from measurements on the powders used to make the compacts. The powders, however, undoubtedly contained at least some small amount of closed porosity. Consequently, the actual void content of these dispersions must have been at least slightly greater than the calculated values.

The error in the calculated void content is probably greatest for the burned U_3O_8 dispersions, less for the UAl_x dispersions, and least for the high-fired U_3O_8 dispersions. For example, if one uses a theoretical (x-ray) value of 8.337 g/cm^3 instead of the toluene-pycnometric value of 7.647 g/cm^3 for the density of the burned U_3O_8 fuel particles, the calculated void content of the as-fabricated dispersions increases from about 6–8 vol % to about 8–10 vol %. Thus the burned U_3O_8 dispersions in the PM-1 tests may actually have contained about 2 vol % more voids than we report. Similar calculations indicate that the actual void contents of the high-fired U_3O_8 dispersions may have been about 0.5 vol % more than the reported values.

One cannot apply the above reasoning to the UAl_x-bearing dispersions because of the difficulty and expense of arriving at a theoretical density for UAl_x. However, as will be discussed later, there is reason to believe that these dispersions actually contained about 1 vol % more voids than reported.

Chemical Reactions

A second possible source of the discrepancies between the measured and predicted swelling values for these dispersions may be the effects of the chemical reaction between the fuel particles and the aluminum matrix material during irradiation. If the volume of the reaction products differs from that of the reactants, the overall change in the volume of the dispersion could be greater or less than would be expected from fission-product swelling alone. Since we know so little about the nature

of the reactions in the *irradiated* dispersions, it is difficult to evaluate how significant this factor may be. However, if one is willing to assume that the reactions that occur in the irradiated plates are similar to those that occur in out-of-reactor tests, it appears that chemical reactions would have little effect upon the swelling behavior of UAl_x but possibly a major effect upon the swelling behavior of U_3O_8 dispersions.

Reactions in the UAl_x-Al dispersions are generally thought to consist of the transformation of any UAl_2 and UAl_3 initially present within the fuel particles to the more stable UAl_4 compound.[27] Although the relative amounts of UAl_2, UAl_3, UAl_4, and free aluminum in UAl_x can very widely depending upon the manner of production, the UAl_x powders used to make the PM-1 and G-12 fuel plates closely correspond to the UAl_3 compound both structurally and stoichiometrically. Consequently, in estimating the magnitude of the dimensional changes that result from these reactions, it will be convenient to assume that all the uranium in these dispersions was initially present as UAl_3 and that the UAl_3 completely transforms to UAl_4 by reaction with the aluminum.

Calculations based upon theoretical values of 6.662, 6.011, and 2.698 g/cm^3 for the densities of UAl_3, UAl_4, and aluminum, respectively, indicate that complete transformation in a 63 wt % UAl_3-Al dispersion should result in a volume decrease of about 0.3%. The assumed reaction is

$$63.00 \text{ g } UAl_3 + 37.00 \text{ g Al} \rightarrow 68.38 \text{ g } UAl_4 + 31.62 \text{ g Al, or} \quad (2a)$$

$$9.46 \text{ cm}^3 + 13.71 \text{ cm}^3 \rightarrow 11.37 \text{ cm}^3 + 11.72 \text{ cm}^3\text{, or} \quad (2b)$$

$$23.17 \text{ cm}^3 \rightarrow 23.09 \text{ cm}^3 \quad . \quad (2c)$$

Similar calculations indicate that complete reaction in the 53 wt % UAl_x-Al dispersions should result in only a 0.2% volume decrease. Thus, this reaction could cause the UAl_x dispersions to swell slightly less

[27] J. L. Gregg, R. S. Crouse, and W. J. Werner, *Swelling of UAl_3-Al Compacts*, ORNL-4056 (January 1967), pp. 4–6.

than expected during irradiation; however, the net reduction in swelling would not be sufficient to account for the differences between the measured and predicted swelling values.

Volume changes associated with chemical reactions between U_3O_8 and aluminum, however, could have a very significant effect upon the swelling behavior of the U_3O_8 dispersions. Out-of-reactor tests[28] indicate that aluminum reduces U_3O_8, forming UAl_4 and Al_2O_3. Calculations based upon theoretical values of 8.337, 6.011, 3.965, and 2.698 g/cm^3 for the densities of U_3O_8, UAl_4, Al_2O_3, and aluminum, respectively, indicate that complete reaction in a 50 wt % U_3O_8 dispersion should result in a volume decrease of about 8.2%. The assumed reaction is

$$50.00 \text{ g } U_3O_8 + 50.00 \text{ g Al} \rightarrow 61.75 \text{ g } UAl_4 + 16.32 \text{ g } Al_2O_3 + 21.93 \text{ g Al, or} \quad (3a)$$

$$6.00 \text{ cm}^3 + 18.52 \text{ cm}^3 \rightarrow 10.27 \text{ cm}^3 + 4.12 \text{ cm}^3 + 8.13 \text{ cm}^3\text{, or} \quad (3b)$$

$$24.53 \text{ cm}^3 \rightarrow 22.52 \text{ cm}^3 \quad . \quad (3c)$$

Similar calculations indicate that complete reaction in a 40 wt % U_3O_8 dispersion should result in a volume decrease of about 6.0%.

This chemical change could cause the U_3O_8 dispersions to swell considerably less than might be expected during irradiation and, as will be discussed later, appears to be at least partially responsible for the discrepancy between the predicted and measured swelling values for these dispersions.

Predicted Swelling in UAl_x Dispersions

In general, our PM-1 and G-12 irradiation tests have shown that the UAl_x dispersions consistently swell about 0.8 to 1.4 vol % less than would be predicted by our swelling model. From a practical standpoint, the

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[28]R. C. Waugh and R. J. Beaver, *Recent Developments in the Powder Metallurgy Application of Uranium Oxides to Aluminum Research Reactor Fuel Elements*, ORNL-CF-57-9-60 (Sept. 16, 1957).

discrepancy between the measured and predicted swelling is essentially insignificant. Consequently, it appears that this model can predict the swelling behavior of UAl_x with an accuracy sufficient for most engineering applications, particularly since the model seems to be conservative and slightly overestimates the amount of swelling. However, there are reasons to believe that the model is capable of predicting the swelling behavior of these dispersions with an even greater accuracy than indicated by these results. Errors in the calculation of the void content of these dispersions could easily account for the discrepancies. For example, if the UAl_x powder used to fabricate the UAl_x dispersions in the PM-1 experiment contained 4% closed pores, the actual void content of these dispersions would have been about 1.2 to 1.4 vol % greater than that calculated from the pycnometric densities. This, in turn, would lower the predicted swelling values about 1.2 to 1.4% and essentially eliminate the discrepancy.

Chemical reactions between the fuel particles and the aluminum matrix would be expected to have little or no effect upon the swelling behavior of UAl_x dispersions. The reaction appears to be only about 50% complete in the irradiated plates. Thus, the transformation probably could account for a difference of only 0.1 to 0.2% between the measured and predicted values.

The discrepancies for the UAl_x dispersions irradiated in the G-12 tests could also be explained similarly. The UAl_x powder used to manufacture those plates was different from that used for the PM-1 plates and probably contained a different concentration of closed pores. Hence the error in the calculated void content may have been somewhat greater or less than that for the PM-1 dispersions. Consequently, we believe that the discrepancy between the measured and predicted swelling values for the UAl_x dispersions irradiated in our tests can be explained by errors in calculating the void content of the as-fabricated dispersions. If this is true, then the accuracy of the model to predict swelling of the UAl_x dispersions would appear to be limited only by the accuracy of the determination of the void content of the dispersions.

Other irradiation tests[29] by the Idaho Nuclear Corporation, however, have shown differences as large as ±3 vol % between the measured and predicted swelling values for UAl_x dispersions. Since we do not know how the void contents of these dispersions were calculated, we do not know if these larger discrepancies can also be attributed to errors in the void content. However, as mentioned previously, small errors in the density of the cladding, matrix material, or the fuel compound can result in rather large errors in the calculated void content. If nominal values were used for the densities of these constituents, errors as great as ±3 vol % could easily occur. Since there appears to be no method of precisely determining the void content of these dispersions, we suggest that workers in this field arrive at some standardized method for calculating the void content of UAl_x-Al dispersions. This would permit easier comparison of swelling data from different facilities.

Predicted Swelling of U_3O_8 Dispersions

Results of our PM-1 and G-12 irradiation tests have shown that burned and high-fired U_3O_8 dispersions generally swell about 1–2 and 3–5 vol % less, respectively, than would be predicted by our basic swelling model [Eq. (1)]. Thus it appears that this model could be used for a rather conservative estimate of the swelling behavior of these dispersions. Such estimates should be satisfactory for most engineering applications, particularly since the model overestimates the swelling. For some applications, however, more precise estimates may be required, and the basic swelling model is obviously not satisfactory.

In contrast to the results on the UAl_x-Al dispersions, we lack a completely satisfactorily explanation for the discrepancies between the measured and predicted swelling values for the U_3O_8 dispersions. As mentioned previously, there is reason to believe that the actual void content of the high-fired and burned U_3O_8 dispersions may have been about 0.5 and 2 vol % greater, respectively, than the reported values. If

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[29] M. F. Marchbanks, *Metallurgy and Materials Science Branch Annual Report Fiscal Year 1970*, IN-1437 (November 1970), pp. 38–53.

this is true, then the high-fired dispersions actually swelled about 2.5–4.5 vol % less (instead of 3–5 vol % less) than predicted by the model, while the burned U_3O_8 dispersions swelled about 0–1 vol % *more* (instead of 1–2 vol % *less*) than the revised prediction. Thus, errors in determining the initial void content of these dispersions could account for most of the discrepancies in the results for the burned U_3O_8 dispersions, but only for a relatively insignificant part of the discrepancies for the high-fired dispersions.

Chemical reaction between the fuel particles and the aluminum matrix material during irradiation could be a major cause for these discrepancies. As mentioned previously, complete reaction between the U_3O_8 and aluminum could cause these dispersions to swell as much as 6 to 8 vol % less than would be predicted by the model. Since metallographic results indicate that the reaction generally is about 50% complete in the irradiated plates, one therefore might expect that these dispersions would swell about 3 to 4 vol % less than predicted by the model. This reasoning, of course, is in relatively good agreement with the experimental results for the high-fired dispersions, which swelled about 2.5 to 4.5 vol % less than the revised prediction. However, experimental results on the burned U_3O_8 dispersions fail to support this assumption in that these dispersions swelled about 0–1 vol % *more* than the revised prediction, whereas the reaction should have caused them to swell at least 3 to 4 vol % less. Furthermore, such reactions cannot account for the differences in the swelling behavior of both the burned and high-fired dispersions in the PM-1 and G-12 tests. The fuel plates in the G-12 tests were irradiated at higher temperatures and had reacted more extensively than those in the PM-1 tests. Consequently, one would expect that the dispersions in the G-12 tests would swell less than those in the PM-1 tests at comparable burnup levels. However, the experimental results show that both types of dispersions actually swelled more in the G-12 tests.

Modified Swelling Model

Our inability to account for the discrepancies between the measured and predicted swelling values for the burned and high-fired U_3O_8 dispersions raises considerable questions as to the validity of our model to

predict the swelling behavior of these dispersions. However, if the model is modified to include an additional term, as shown below, one can obtain relatively good agreement (within about ±1.5 vol %) between the measured and predicted swelling values for dispersions irradiated in both the PM-1 and G-12 tests. The equation for the new model follows:

$$\Delta V/V = (6.3 \times 10^{-21})B - V_v - C \tag{4}$$

where

$\Delta V/V$ = predicted change in core volume (%)

B = burnup (fissions/cm^3 of fuel-core)

V_v = void content of fuel-core (vol %)

C = an arbitrary constant of 3.8 or 0.8 for the high-fired or burned U_3O_8 dispersions, respectively

Such good agreement, or course, is rather convincing evidence that the swelling model is basically sound (i.e., that these dispersions actually swell at the rate assumed in the model and that fabrication voids are effective in accommodating swelling). Consequently, the question then arises as to what the C term physically represents.

Although we cannot completely account for differences between the swelling behavior of the burned and high-fired dispersions, we believe that this term represents a change in the volume of the dispersion resulting from chemical reaction between the U_3O_8 and aluminum. The value of C, therefore, would vary as a function of both the fuel loading and the extent of reaction. Since the extent of reaction appears to depend upon the type of oxide particle (i.e., burned or high-fired U_3O_8), the irradiation temperature, and probably the time at temperature, it is not surprising that the value of C would vary somewhat depending upon the type of dispersion and the irradiation conditions. Since we know little about the nature or kinetics of this reaction during irradiation, it would be presumptuous to assume that one could calculate a theoretical value for C. Consequently, the modified swelling model appears to be capable of predicting the swelling behavior of U_3O_8 dispersions only under irradiation conditions similar to those for which the value of C has been experimentally determined.

CONCLUSIONS

From the results of the PM-1 and G-12 irradiation experiments, we conclude the following:

1. Dispersions of UAl_x in aluminum perform quite satisfactorily to burnup levels of 1.8 to 2.2×10^{21} fissions/cm^3 at irradiation temperatures of 60 to 90°C and to lower fission densities at temperatures up to 180°C.

2. Dispersions of U_3O_8 in aluminum perform quite satisfactorily to burnup levels of 1.5 to 2.4×10^{21} fissions/cm^3 at irradiation temperatures of 60 to 120°C and to lower fission densities at temperatures up to 200°C.

3. The swelling behavior of both UAl_x and U_3O_8 dispersions during irradiation is primarily a function of both the fission density and the initial void content of the as-fabricated dispersion.

4. Since fabrication voids generally increase with increasing fuel loadings, a higher loaded fuel plate will generally swell less than a lower loaded fuel plate of the same type when irradiated to equivalent fission densities.

5. Variations in the fuel plate fabrication procedures can significantly affect the void content and hence the swelling behavior of both UAl_x and U_3O_8 dispersions.

6. At comparable fuel loadings, plates containing burned U_3O_8 dispersions generally have a higher void content than fuel plates containing either high-fired U_3O_8 or UAl_x dispersions. Consequently, fuel plates containing burned U_3O_8 dispersions will generally swell less when irradiated to equivalent fission densities.

7. Irradiation-induced swelling of both UAl_x and U_3O_8 dispersions is independent of the fuel fines content (i.e., the fraction of fuel particles <44 μm in diameter).

8. Our model can predict the irradiation-induced swelling behavior of both UAl_x and U_3O_8 dispersions with an accuracy adequate for most engineering applications. More precise estimates of the swelling behavior of U_3O_8 dispersions, however, require experimental determination of a

correction that depends upon the type of fuel compound, the irradiation temperature, and perhaps the rate of fissioning.

9. A fused α-Al_2O_3 dispersion in aluminum exhibited no deleterious effects after exposure to perturbed thermal-neutron fluences of about 1.5×10^{21} neutrons/cm^2.

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APPENDIX

Calculation of Fabrication Voids

The density, D, of the fuel dispersions in the fabricated test plates was obtained from conventional pycnometer techniques and the expression

$$D = \frac{W_c}{(W_a - W_s)/\rho_s - (W_a - W_c)/\rho_{Al}} \quad ,$$

where

ρ_{Al} = density of aluminum cladding, g/cm^3,
ρ_s = density of solution, g/cm^3,
W_c = weight of fuel dispersion, g,
W_a = weight of plate in air, g,
W_s = weight of plate in solution, g.

The concentration, V_v, of fabrication voids in the completed fuel plates was then calculated from the measured density of the fuel dispersion and the theoretical density, ρ_t, for the dispersions:

$$V_v \equiv 100(1 - D/\rho_t) \quad ,$$

where

$$\rho_t \equiv 1/\left[x_f/\rho_t + (1 - x_f)/\rho_m\right] \quad ,$$

and

ρ = toluene density, g/cm^3,
x = weight fraction in fuel dispersion.

The subscripts t, f, and m denote theoretical, fuel, and matrix attributes, respectively.

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