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THE IRRADIATION OF
ALUMINUM ALLOY-CLAD
THORIA-URANIA PELLETS

by

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TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	5
INTRODUCTION	5
IRRADIATION	7
POSTIRRADIATION EXAMINATION	11
General Observations	11
Dimensional and Volumetric Measurements	12
Metallography	15
Fission Gas Release	29
DISCUSSION OF RESULTS	32
CONCLUSIONS	34
ACKNOWLEDGMENTS	35
REFERENCES	35

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Cross Section of Unirradiated Specimen Showing $\text{ThO}_2\text{-UO}_2$ Pellets Lead-bonded to the Aluminum Alloy Cladding	6
2.	Longitudinal Section of One Fuel Tube of Unirradiated Specimen	6
3.	Irradiation Specimen Partially Withdrawn from Aluminum Capsule	7
4.	Specimen DG-1 after 5800-MWd/T Burnup	11
5.	Specimen ANL-35-70 after 28,500-MWd/T Burnup	12
6.	Specimen ANL-35-69 after 33,200-MWd/T Burnup	13
7.	Specimen Volume Increase as a Function of Burnup	14
8.	Specimen Volume Increase as a Function of $\int_{T_s}^{T_0} kd\theta$	14
9.	Transverse As-cut Sections and Tube Diameter Measurements of Specimen ANL-35-69	16
10.	Transverse As-cut Sections and Tube Diameter Measurements of Specimen ANL-35-70	17
11.	Polished Section of Bottom Pellet in Specimen DG-9, $\int_{T_s}^{T_0} kd\theta = 5 \text{ W/cm, 5000-MWd/T Burnup}$	17
12.	Polished Section of Bottom Pellet in Specimen DG-12, $\int_{T_s}^{T_0} kd\theta = 11 \text{ W/cm, 11,000-MWd/T Burnup}$	18
13.	Polished Section of Bottom Pellet in Specimen DG-2, $\int_{T_s}^{T_0} kd\theta = 17 \text{ W/cm, 28,400-MWd/T Burnup}$	18
14.	Polished Section of Bottom Pellet in Specimen ANL-35-65, $\int_{T_s}^{T_0} kd\theta = 42 \text{ W/cm, 28,100-MWd/T Burnup}$	18

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
15.	Polished Section Through Bottom Pellet in the Unfailed Tube of Specimen ANL-35-69, $\int_{T_s}^{T_0} kd\theta = 43 \text{ W/cm}$, 33,200-MWd/T Burnup	19
16.	Polished Section Through Bottom Pellet in the Unfailed Tube of Specimen ANL-35-70, $\int_{T_s}^{T_0} kd\theta = 50 \text{ W/cm}$, 28,500-MWd/T Burnup	19
17.	Partial Section Through Right Tube of ANL-35-69, 2.22 cm from Bottom of Specimen.	20
18.	Partial Section Through Left Tube of ANL-35-70, 8.25 cm from Bottom of Specimen.	20
19.	Partial Section Through Right Tube of ANL-35-70, 6.35 cm from Bottom of Specimen.	21
20.	Partial Section Through Right Tube of ANL-35-70, 1.90 cm from Bottom of Specimen.	21
21.	Longitudinal Section Through a Bottom Pellet in ANL-35-67 .	22
22.	Longitudinal Section Through a Bottom Pellet of ANL-35-67 .	23
23.	Boundary Between Two-phase Dendritic Structure (Right) and Columnar Grains (Left) in the Center of an ANL-35-70 Tube	24
24.	Light-gray Phase in Region One-third the Distance from Center to Edge of ANL-35-70	24
25.	Pipe of Light-gray Phase in Extension from Center Region in Specimen ANL-35-70.	25
26.	Light-gray Phase Adjacent to Central Void above Dendritic Structure at Bottom of Pellet in Specimen ANL-35-70.	25
27.	Longitudinal Section Through Bottom Pellet of ANL-35-67. . .	26
28.	Longitudinal Section Through Bottom Pellet in Unfailed Tube of ANL-35-70.	26
29.	Unirradiated ThO_2 -12.7w/o UO_2	27

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
30.	Transverse Section Through Defect in ANL-35-69	27
31.	Cladding Failure in ANL-35-70.	28
32.	Upper Cladding Failure in ANL-35-69	28
33.	Apparatus for Puncturing Specimens for Fission Gas Collection.	29
34.	Fission Gas Release as a Function of Burnup	30
35.	Fission Gas Release as a Function of $\int_{T_s}^{T_0} kd\theta$	30

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Postirradiation Uranium Isotopic Analyses	8
II.	Irradiation Summary of Aluminum Alloy-clad Thoria-urania Pellets	10

THE IRRADIATION OF ALUMINUM ALLOY-CLAD THORIA-URANIA PELLETS

by

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ABSTRACT

Specimens consisting of $\text{ThO}_2\text{-UO}_2$ pellets lead-bonded to extruded Al-1w/oNi alloy cladding were irradiated to maximum burnups of 33,200 MWd/T of oxide and to peak heat

ratings of $\int_{T_s}^{T_0} k d\theta = 63 \text{ W/cm}$. The specimens were pro-

tototype BORAX-V fuel plates. The $\text{ThO}_2\text{-UO}_2$ solid solutions were formed by air-firing cold-pressed mixtures of ThO_2 and U_3O_8 to a final oxygen-to-metal atom ratio of about 2.05 to 1. Fuel compositions irradiated were 6.36, 12.7, and 25.6w/o UO_2 in ThO_2 . Two specimen failures were attributed to fuel thermal expansion in areas of thin-walled cladding. The hyperstoichiometry of the fuel with respect to oxygen was believed to have caused a lower fuel thermal conductivity and abnormally large amounts of recrystallization, grain growth, and fission gas release. Fission gas release ranged from 4.6 to 81.2% of the theoretical gas generated.

INTRODUCTION

Based in part on the good irradiation performance of aluminum alloy clad lead-bonded $\text{ThO}_2\text{-UO}_2$,⁽¹⁾ pellets of a solid solution of $\text{ThO}_2\text{-6.36w/oUO}_2$, lead-bonded in Al-1w/oNi alloy cladding, were chosen as the fuel for the BORAX-IV reactor experiment. No data were available, however, on the ultimate potential of such a fuel or fuel assembly, or on the fission gas release characteristics of $\text{ThO}_2\text{-UO}_2$ pellets.

To determine the burnup limitations on the BORAX-IV fuel assembly and gather more irradiation data on the $\text{ThO}_2\text{-UO}_2$ fuel, small sections of the standard extruded X8001 aluminum alloy BORAX-IV tube plate containing fuel pellets were irradiated directly in the Materials Test Reactor-Engineering Test Reactor (MTR-ETR) process water. Irradiating the specimens directly in the cooling water alleviated the problem of undesirably high cladding temperatures incurred in previous NaK capsule irradiations.⁽¹⁾

The irradiation specimens were 13.34 cm long and consisted of two tubes, each of 7.62-mm OD and a nominal 0.51-mm wall, joined with a 4.06-mm-wide web, and having thick webs along the outer edges, giving the specimen a total width of 2.38 cm. The tubes in 14 of the specimens contained 5.84-mm-diameter pellets of ThO_2 -6.36w/o UO_2 , having an average geometric density of 7.00 gm/cc, or 69.4% of theoretical density. Two specimens contained ThO_2 -12.7w/o UO_2 pellets having an average geometric density of 8.14 gm/cc, and two specimens contained ThO_2 -25.6w/o UO_2 pellets having an average geometric density of 8.45 gm/cc. These latter densities correspond to 80.1 and 82.4% of theoretical, respectively. In all compositions, the uranium was 93.2% enriched in U^{235} . The 3.30-mm annulus between the pellets and cladding was filled with lead. Figure 1 shows a cross section of an unirradiated specimen. Since the construction of the specimen was identical to that of the full-size fuel plate, the end closures were made with the silicon bonding technique, the top pellet in each tube was aluminum, and each tube contained a helium-filled gas space at the top which could be punctured to collect the escaped fission gases. Figure 2 shows a longitudinal section of a fuel tube of a typical unirradiated specimen.

Figure 1. Cross Section of Unirradiated Specimen Showing ThO_2 - UO_2 Pellets Lead-bonded to the Aluminum Alloy Cladding

Figure 2. Longitudinal Section of One Fuel Tube of Unirradiated Specimen

The fabrication of the fuel pellets and fuel assembly has been described previously in detail.^(2,3) Briefly, the pellets were mixtures of $\text{ThO}_2\text{-U}_3\text{O}_8$, cold-pressed at from 914 to 1055 kg/cm^2 . The pellets were then air-sintered in a gas-fired kiln at 1700 to 1750°C for approximately 16 hours. Although the specimen compositions quoted on page 6 are based on complete reduction of the U_3O_8 to UO_2 , X-ray diffraction analyses indicated that all the U_3O_8 was not reduced to UO_2 , but probably to some oxide with an oxygen-to-metal ratio of 2.3 to 1.⁽²⁾ Under these conditions, the solid solution that is formed has an oxygen-to-metal ratio in the vicinity of 2.05 to 1.⁽⁴⁾

IRRADIATION

The specimens were irradiated in water-cooled, flow-through capsules made of Type 6061 aluminum alloy. The capsules were of split construction and were held together at each end by an aluminum rivet which also held the specimen and full-length monitor wires in the capsule. The ends of the capsules were machined so that they would nest together and align the water coolant channels for unimpeded flow. Figure 3 shows a partially assembled capsule and the water-coolant channel around the specimen.

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Figure 3. Irradiation Specimen Partially Withdrawn from Aluminum Capsule

Preirradiation calculations to determine irradiation parameters were made with the intent of achieving high burnups at heat ratings that would be insufficient to cause cladding burnout or central melting of the fuel. The maximum desired cladding surface heat flux was 205 W/cm^2 , which is equivalent to an integrated thermal conductivity value of 37 W/cm for $\text{ThO}_2\text{-6.36w/oUO}_2$. The integrated thermal conductivity is defined as

$$\int_{T_s}^{T_0} k d\theta, \text{ where } k \text{ is the thermal conductivity of the fuel, } \theta \text{ is the temperature coordinate, and } T_s \text{ and } T_0 \text{ are the fuel pellet surface and center temperatures, respectively.}$$

The calculated maximum unperturbed thermal flux necessary to obtain these conditions was $1.6 \times 10^{14} \text{ n/cm}^2$.

The specimens were irradiated in four groups, each group consisting of a vertical column of five specimens. The residence time in the reactor was varied for each column to obtain a range of burnups without exceeding the calculated maximum-permissible ratings in any one column. Three columns were irradiated in the MTR for periods of 169, 200, and 266 effective full-power days of irradiation. The fourth column was irradiated in the ETR for 148 effective full-power days. Specimens DG-2 and DG-10 were in the reactor for irradiation periods totaling 348 and 421 effective full-power days, respectively. The column that was irradiated in the MTR for 266 days and contains specimens DG-10 and ANL-35-65, -66, -67, and -70 was inadvertently placed in a position of high flux, about $4.0 \times 10^{14} \text{ n/cm}^2$, unperturbed, for its last cycle in the reactor. The effect of this high flux, more than twice that desired, will be discussed in following sections of this report.

The burnups, and consequently the heat ratings, that were attained by the specimens were computed from a combination of mass spectrometric analyses for the uranium isotopes in selected specimens, and the thermal neutron flux as indicated by the 15.24-cm-long aluminum-cobalt alloy monitor wires that were close to the specimen in each capsule. Table I lists the uranium isotopic percentages found in the analyzed samples. The burnup

Table I

POSTIRRADIATION URANIUM ISOTOPIC ANALYSES

Specimen No.	U^{233} , w/o	U^{234} , w/o	U^{235} , w/o	U^{236} , w/o	U^{238} , w/o
DG-2	23.2 ± 0.2	2.63 ± 0.03	48.3 ± 0.1	16.7 ± 0.1	8.9 ± 0.1
DG-9	1.77 ± 0.02	1.04 ± 0.02	89.65 ± 0.05	1.66 ± 0.02	5.88 ± 0.04
DG-12	7.28 ± 0.05	1.24 ± 0.02	79.19 ± 0.07	5.61 ± 0.06	6.69 ± 0.03
ANL-35-65	20.38 ± 0.07	2.19 ± 0.01	55.20 ± 0.06	13.87 ± 0.04	8.37 ± 0.06
ANL-35-67	3.05 ± 0.02	1.05 ± 0.02	84.82 ± 0.05	4.53 ± 0.03	6.54 ± 0.03
ANL-35-69L	1.79 ± 0.01	1.02 ± 0.01	86.56 ± 0.05	3.87 ± 0.03	6.76 ± 0.05
ANL-35-69R	1.68 ± 0.02	1.03 ± 0.01	85.82 ± 0.03	4.27 ± 0.01	7.21 ± 0.02
ANL-35-70L	0.98 ± 0.01	1.03 ± 0.01	87.94 ± 0.04	3.83 ± 0.02	6.23 ± 0.03
ANL-35-70R	1.17 ± 0.01	1.06 ± 0.01	87.07 ± 0.04	4.32 ± 0.03	6.39 ± 0.03

was calculated from the isotopic analyses by the method of Kittel and Paine⁽⁵⁾ which takes into account the production of U^{235} by neutron capture in U^{234} and the depletion of U^{235} by fission and capture, if a constant quantity of U^{238} is assumed. The relationship between the analyzed burnup, the U^{235} 2200-m/sec fission cross section of 582 barns, and the monitor-indicated flux established a basis for calculating effective perturbation factors for each composition. This effective perturbation factor was then used with the monitor-indicated flux for each specimen to obtain the burnup value. No assumption was made that the monitors saw unperturbed fluxes, since the reaction rate depends only upon the product of the monitor flux and the experimentally-derived effective perturbation factor. The greatest uncertainty in this method arises from the precision of the effective perturbation factor determination. This uncertainty, determined from three of four 6.36w/o UO_2 specimens, was about $\pm 7\%$. All burnups were calculated both as megawatt-days per ton (MWd/T) of mixed oxide and as total fissions per cm^3 of mixed oxide.

The initial heat output from each specimen was calculated from the monitor-indicated flux and the effective perturbation factor by using a U^{235} fission cross section of 582 barns and an energy release in the fuel of 180 MeV per fission. A corresponding average pellet surface temperature was calculated by using standard heat-transfer equations, the initial heat output, and a water coolant temperature of $45^\circ C$. Internal fuel temperatures were not calculated because the uncertainty in fuel thermal conductivity made such a calculation impractical. Instead, the integrated thermal con-

ductivity, $\int_{T_s}^{T_0} k d\theta$, was determined for peak conditions in each specimen.

For all specimens, except those placed in the high flux for one cycle, peak conditions occurred during the initial part of the irradiation when the maximum amount of U^{235} was present. Calculations indicated that although the U^{233} contribution to the heat output became significant at the end of the irradiation (for example, about 14% of the total in DG-2), the total heat output at the end of the irradiation from U^{235} and U^{233} was less than the initial heat output from U^{235} alone.

In the specimens that experienced the high flux for one cycle (DG-10 and ANL-35-65, -66, -67, and -70), peak heat output conditions were achieved during this cycle from the fission of both U^{233} and U^{235} . The neutron fluxes that these specimens experienced during this cycle could not be ascertained directly from the flux monitors, however. The higher neutron flux was determined from a flux plot of an adjacent reactor position during the same reactor cycle and by knowing the approximate peak flux of the reactor position in question.

The burnup contribution resulting from fission of U^{233} in each specimen was calculated from Taraba's work,⁽⁶⁾ and the U^{233} heat output was calculated on the basis of a fission cross section of 527 barns⁽⁷⁾ and an approximate energy release of 180 MeV per fission.

The $\int kd\theta$ values were calculated by using Robertson's method.⁽⁸⁾ The parameter $\mathcal{K}$ (the inverse diffusion length for neutrons in the fuel) was computed for each ThO_2-UO_2 composition using Westcott's values⁽⁹⁾ for average cross sections, $\bar{\sigma}$, at 40°C, and Taraba's values⁽¹⁰⁾ for the ratio of average flux to surface flux, as a function of the radius and macroscopic absorption cross section of the fuel.

Table II summarizes the irradiation data and results for all specimens. The nomenclature refers to arbitrarily-set "left" and "right" tubes (LT and RT).

Table II
IRRADIATION SUMMARY OF ALUMINUM ALLOY-CLAD ThO_2-UO_2 PELLETS

Specimen No.	UO ₂ w/o	Effective Full-power Days of Irradiation	Burnup		Fuel Surface Temp, T _s , °C	$\int_{T_s}^{T_0} kd$ W/cm	Cladding Surface Heat Flux, W/cm	Diameter Change, %								Volume Increase, %	Gas Release, % Theoretical	
			MWd/T Oxide	Fiss/cc x 10 ²⁰				LT ₁	LT ₂	LT ₃	LT ₄	RT ₁	RT ₂	RT ₃	RT ₄		LT	RT
DG-1	6.36	121	5,800	1.2	70	8	42	5.6	0.7	0.0	0.0	5.3	0.7	0.0	-6.9	0.22	(c)	(c)
DG-2	6.36	348	28,400	6.0	95	17	95	0.7	2.9	-0.2	0.6	-0.8	-0.9	0.0	-1.1	1.08	17.4	16.0
DG-3	6.36	200	20,600	4.4	105	20	108	0.2	-7.9	0.1	-6.5	0.0	-8.0	0.0	0.0	1.14	12.8	(c)
DG-5	6.36	200	19,000	4.0	100	18	97	0.2	0.1	0.6	0.0	-0.9	0.9	0.8	0.2	2.00	13.9	14.3
DG-7	6.36	200	11,600	2.5	75	10	54	-0.1	0.2	0.3	-0.1	-0.1	0.3	0.3	0.4	0.90	10.5	10.4
DG-9	6.36	169	5,000	1.1	60	5	26	0.3	-0.1	0.2	-1.0	-0.9	-0.1	0.1	-0.4	0.61	4.6	0.3
DG-10	6.36	435	26,600	5.7	135 ^(a)	30 ^(a)	160 ^(a)	1.1	0.4	0.6	0.7	0.8	0.9	0.5	1.2	0.81	45.4	41.1
DG-11	6.36	169	12,200	2.6	80	12	66	0.5	0.4	0.6	0.3	0.0	-0.2	0.1	0.1	0.92	7.4	0.1
DG-12	6.36	169	11,000	2.3	80	11	60	-0.4	0.1	0.5	0.1	0.4	-0.1	-0.2	-0.2	0.91	8.6	9.7
DG-13	6.36	169	6,000	1.3	60	6	31	-0.7	0.0	0.3	-0.8	0.2	0.2	0.2	0.0	0.61	9.7	7.6
ANL-35-63	6.36	148	21,500	4.6	130	29	157	1.2	0.3	0.4	0.3	0.8	0.4	-0.4	-0.2	0.98	16.4	17.0
ANL-35-64	6.36	148	19,900	4.2	115	24	131	0.5	0.5	0.3	0.0	0.5	0.2	-1.6	1.3	0.83	16.3	2.1
ANL-35-65	6.36	266	28,100	6.0	170 ^(a)	42 ^(a)	227 ^(a)	0.6	0.2	0.4	-0.2	-0.5	0.5	0.3	0.0	0.81	43.4	30.2
ANL-35-66	6.36	266	24,800	5.3	200 ^(a)	52 ^(a)	280 ^(a)	0.4	1.1	1.2	0.2	2.3	0.7	1.8	0.3	1.14	(c)	50.7
ANL-35-67	12.7	266	23,400	5.8	240 ^(a)	63 ^(a)	355 ^(a)	3.6	1.5	1.4	0.6	1.6	1.2	0.9	0.0	1.71	47.4	49.2
ANL-35-68	12.7	148	24,300	6.0	145	32	180	1.0	0.9	1.2	-0.3	1.8	0.9	0.7	0.2	1.21	22.4	28.9
ANL-35-69	25.6	148	33,200	8.5	190	43	268	4.8	3.2	1.0	0.5	3.4	2.8	2.2	0.8	(d)	(d)	48.0
ANL-35-70	25.6	266	28,500	7.3	240 ^(a)	59 ^(a)	356 ^(a)	7.8	2.5	2.5	0.4	1.9	1.2	1.6	1.4	(b)	(d)	81.2

^a Estimated last cycle conditions.

^b Not determined because of condition of specimen.

^c Sample lost.

^d Cladding failure.

POSTIRRADIATION EXAMINATION

General Observations

The capsule containing specimen DG-1 was damaged after a burnup of 5800 MWd/T and was returned to Argonne for hot-cell examination. It

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Figure 4. Specimen DG-1 after 5800-MWd/T Burnup. The lower three-fourths of the specimen, where the cladding is lighter-colored, contains the fuel pellets. Positions at which tube diameters were measured are indicated.

was subsequently found that although the silicon-bonded end closures were severely bent, no leaks had developed and the gaseous fission products were contained within the cladding. Figure 4 shows the appearance of specimen DG-1 after its return to Argonne. The coloration of the cladding, light next to the fuel and dark where there is no fuel, was typical of all the specimens. The dark areas were a pale rust color, which is indicative of the iron oxide in the reactor cooling water. The area next to the fuel, however, appeared to be covered by a loosely adhering corrosion product that showed a tendency to slough off easily as seen on specimen ANL-35-70 (Figure 5).

After 266 effective full-power days of irradiation in the MTR, the column containing specimens DG-10 and ANL-35-65, -66, -67, and -70 was returned to Argonne because of a suspected leaking specimen indicated by fission product activity in the reactor process water. The leak appeared to have developed during the cycle that these specimens were in the high flux position. After their return to Argonne, the specimens were tested for leaks by immersing them in water, drawing a partial vacuum over the water, and observing for bubbles rising from leaking areas. A 6.35-mm-long longitudinal fissure was found in this manner in the bottom of the left tube of specimen ANL-35-70 which contained

ThO₂-25.6w/oUO₂. Figure 5 shows this defect. Because of the optical arrangement in the hot cell, the specimen in Figure 5 is shown as a mirror image. All other specimens in this group were sound.

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Figure 5. Specimen ANL-35-70 after 28,500-MWd/T Burnup. A small longitudinal failure is indicated. A light-colored corrosion product covering the fuel section has partially flaked off. (Photograph is a mirror image.)

The four locations measured, which are indicated in Figure 4, included three in the fuel section and one in the gas space of each tube. The accuracy of the measurements was about ± 0.05 mm, or $\pm 0.6\%$ of the diameter. Diameter changes of less than 0.6% , therefore, are not considered significant. The data are given in Table II. The diameter changes indicate the significant fact that the greatest expansion occurred at the bottom of the fuel section while the gas space at the top was only slightly affected. This is especially true for the two specimens that ruptured near the bottom of the fuel section (specimens ANL-35-69 and ANL-35-70). Maximum diametral increases at the points of failure were 3.6 and 7.9% , respectively.

While these specimens were being examined, the column of specimens irradiated in the ETR and containing specimens DG-2 and ANL-35-63, -64, -68, and -69 was routinely returned to Argonne for an interim examination. It was found that the left tube of specimen ANL-35-69 contained a longitudinal crack running almost the entire length of the tube. Unlike the failure which occurred in the MTR, this defect was not detected at the ETR. This defect was considerably larger than that in specimen ANL-35-70, and it was accompanied by a loss of 14.35 gm of cladding, lead, and fuel. Reactor records showed that this column had once been removed because of a suspected leaking specimen but was replaced and irradiated for an additional four cycles. If the rupture did occur when suspected, the additional irradiation period may have caused the gross loss of material and the enlargement of the defect. The resulting fissure is shown in Figure 6 (also in a mirror image).

Dimensional and Volumetric Measurements

Diameter measurements were made with a standard micrometer at four locations along the length of each tube and compared to preirradiation measurements at the same locations.

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Figure 6. Specimen ANL-35-69 after 33,200-MWd/T Burnup. Long longitudinal failure is indicated. (Photograph is a mirror image.)

The volume change induced by irradiation was obtained directly from pre- and postirradiation immersion measurements in CCl_4 . All specimens, except the two ruptured ones, increased in weight from 76 to 182 mg in a total weight of approximately 64 gm. This weight increase, probably from the adherence of corrosion products, made it impractical to calculate a volume change from the measured density change by assuming no weight change, as is occasionally done. Calculations based on this assumption gave inherently lower volume changes than those determined directly. The per cent volume change, determined directly, is plotted as a function of burnup and $\int_{T_s}^{T_0} k d\theta$ in Figures 7 and 8, respectively, for all specimens except ANL-35-69 and ANL-35-70, which were ruptured. Disregarding the one anomalous point for specimen DG-5, which may be due to an erroneous measurement, a relationship is suggested between the burnup and the increase in specimen volume. At these burnups, however, the volume increases are more likely attributable to thermal expansion of the fuel creating permanent changes in the lead bonding and aluminum cladding. The thermal dependence of the vol-

ume expansion is shown in Figure 8 where the volume increases are plotted against the maximum $\int_{T_s}^{T_0} k d\theta$ rating.

The overheated specimens, DG-10 and ANL-35-65, -66, and -67, are shifted to the right of the trend exhibited by the other specimens in Figure 8. An estimate of the initial heat ratings for these specimens brings them into better alignment at the same volume-expansion values. The increased thermal expansion of these specimens was probably balanced by some measure of oxide densification and shrinkage which would explain the low values for DG-10 and ANL-35-65. At higher heat ratings, the overriding thermal expansion would ultimately lead to failure, as in ANL-35-70, and the significant diameter increases in ANL-35-67.

106-6691

Figure 7

Specimen Volume Increase as a Function of Burnup. Numbers refer to specimen identification given in Table II. Overheated specimens are underlined.

Figure 8

Specimen Volume Increase as a Function of $\int_{T_s}^{T_0} k d\theta$. Numbers refer to specimen identification given in Table II. Overheated specimens are underlined.

106-6692

Unlike the full-size BORAX-IV fuel plates,⁽¹¹⁾ the gas spaces at the top of the prototype irradiation specimens did not collapse. The collapse in the full-size plate was attributed to overstressing of thin wall sections. Although the test specimens were made from the same fuel plate extrusions, the test specimens did not collapse because they were not irradiated in a pressurized system. The operating pressure of the BORAX-IV reactor was 21.1 kg/cm^2 , whereas the MTR and ETR reactors operate between 2.8 and 3.9 kg/cm^2 .

Metallography

Selected specimens for metallographic examination were sectioned by a low-speed abrasive cutoff machine.⁽¹²⁾ The pieces were mounted in cold-setting epoxy resin and polished through $1/4$ -micron diamond abrasive. The sample preparation and metallographic techniques have been described in detail elsewhere.^(13,14)

The metallographic specimens could not be etched successfully in a warm solution of saturated NH_4F , which had been used with effect on hot-pressed $\text{ThO}_2\text{-UO}_2$ specimens. It was therefore necessary to rely on the absence of pores in grains and their presence in grain boundaries to determine the nature and extent of recrystallization.

Three transverse sections through specimens ANL-35-69 and ANL-35-70 showed that a vertical temperature gradient existed during irradiation, as evidenced by more extensive recrystallization and larger central voids in the bottom pellets. The position at which these sections were made is shown in Figures 9 and 10, together with the resulting as-cut macrostructures and measured diametral increases. The temperature gradient was attributed to the fact that these specimens were irradiated above the reactor midplane so that the bottom pellets were in a higher flux. Only one section, through the bottom pellets, was made on the remaining specimens. Of the specimens selected for sectioning (DG-2, DG-9, DG-12, ANL-35-65, ANL-35-67, ANL-35-69, and ANL-35-70), specimens DG-12 and ANL-35-67 were irradiated below the reactor midplane, and specimen ANL-35-65 was irradiated at the reactor midplane. The as-polished macrostructures of these specimens at comparable positions are shown in Figures 11 through 16. Only one tube is shown for each specimen since the oxide in the second tube was used for the burnup analysis.

The transverse sections show the normal cracking at very low heat ratings, a change in coloration at slightly higher heat ratings, and, finally, columnar recrystallization and central void formation at the highest heat ratings. Specimen ANL-35-65 was expected to exhibit a central void at

$$\int_{T_s}^{T_0} k d\theta = 42 \text{ W/cm}$$

similar to that in ANL-35-69, but the section may

have been taken below the void since ANL-35-65 was irradiated at the reactor midplane. Specimen ANL-35-67 is not shown because a major portion of oxide was lost during preparation, leaving only the very outer edge. This edge showed evidence that the specimen had undergone extensive columnar recrystallization, substantiating the approximate value of

$$\int_{T_s}^{T_0} k d\theta = 63 \text{ W/cm calculated for the high flux cycle.}$$

28648

2.5X

28647

2.5X

28646

2.5X

Figure 9. Transverse As-cut Sections and Tube Diameter Measurements of Specimen ANL-35-69

28651

2.5X

28650

2.5X

28649

2.5X

Figure 10. Transverse As-cut Sections and Tube Diameter Measurements of Specimen ANL-35-70

31836

4.5X

Figure 11

Polished Section of Bottom Pellet in
Specimen DG-9, $\int_{T_s}^{T_0} k d\theta = 5 \text{ W/cm}$,
5000-MWd/T Burnup

31837

4.5X

Figure 12

Polished Section of Bottom Pellet
in Specimen DG-12, $\int_{T_s}^{T_0} kd\theta =$
11 W/cm, 11,000-MWd/T Burnup

Figure 13

Polished Section of Bottom Pellet
in Specimen DG-2, $\int_{T_s}^{T_0} kd\theta =$
17 W/cm, 28,400-MWd/T Burnup

31835

4.5X

31838

4.5X

Figure 14

Polished Section of Bottom Pellet
in Specimen ANL-35-65, $\int_{T_s}^{T_0} kd\theta =$
42 W/cm, 28,100-MWd/T Burnup

29771

7X

Figure 15

Polished Section Through Bottom Pellet in the Unfailed Tube of Specimen ANL-35-69, $\int_{T_s}^{T_0} kd\theta = 43 \text{ W/cm}$, 33,200-MWd/T Burnup

Figure 16

Polished Section Through Bottom Pellet in the Unfailed Tube of Specimen ANL-35-70, $\int_{T_s}^{T_0} kd\theta = 59 \text{ W/cm}$, 28,500-MWd/T Burnup. Light-gray region in the center is a two-phase dendritic structure.

29289

7X

The observed recrystallization was unlike that previously seen in hot-pressed $\text{ThO}_2\text{-UO}_2$ specimens.⁽¹⁴⁾ Generally, in the present specimens, a central void formed which was surrounded by a densified area containing large spherical or radially-oriented pores. Beyond this region, toward the pellet edge, the voids were smaller and more numerous. At higher heat ratings (i.e., $\int_{T_s}^{T_0} kd\theta > 40 \text{ W/cm}$), the central void became larger, the densified region extended closer to the pellet surface, a semblance of large radial columnar grains was apparent, small columnar grains were present behind the larger grains, the circumferential crack was closer to the pellet surface, and the oxide outside the crack was essentially unaffected. The range of recrystallization phenomena between $\int_{T_s}^{T_0} kd\theta = 42 \text{ W/cm}$ and 59 W/cm is shown in Figures 17 through 20.

Figure 17

Partial Section Through Right
Tube of ANL-35-69, 2.22 cm
from Bottom of Specimen.

$$\int_{T_s}^{T_0} kd\theta = 43 \text{ W/cm.}$$

29776

35X

Figure 18

Partial Section Through Left
Tube of ANL-35-70, 8.25 cm
from Bottom of Specimen.

$$\int_{T_s}^{T_0} kd\theta = 42 \text{ W/cm.}$$

29781

35X

Figure 19

Partial Section Through Right
Tube of ANL-35-70, 6.35 cm
from Bottom of Specimen.

$$\int_{T_s}^{T_0} kd\theta = 46 \text{ W/cm.}$$

29777

35X

Figure 20

Partial Section Through Right
Tube of ANL-35-70, 1.90 cm
from Bottom of Specimen.

$\int_{T_s}^{T_0} kd\theta = 59 \text{ W/cm.}$ Two-
phase dendritic structure is at
the right.

29779

35X

The slight discrepancy in the appearance and heat ratings of specimens ANL-35-69 and ANL-35-70 (Figures 17 and 18, respectively) is believed attributable to the former's longer irradiation under steady conditions where the retained fission gases have contributed to the porosity. Specimen ANL-35-70 was originally at a lower flux, and the major structural changes took place during the one cycle when it was in a very high flux. With these circumstances, the gases escaped and did not form large voids.

Nowhere in the dense region of large columnar grains were transverse lenticular voids found. Such voids are generally accepted as being part of the mechanism forming columnar grains.⁽¹⁵⁾ In only one specimen, a longitudinal section of ANL-35-67 (part of which is shown in Figure 21), were transverse, almost lenticular voids found in the small columnar region. The large columnar grains were about twice the size of columnar grains observed in previous hot-pressed $\text{ThO}_2\text{-UO}_2$ specimens, where lenticular voids at the heads of columnar grains were present. Figure 22 is an axial view of the columnar grains shown in Figure 21. The grains have a stable boundary condition which can be regarded as an indication of grain growth after the initial grain formation.

EI-429

150X

Figure 21. Longitudinal Section Through a Bottom Pellet in ANL-35-67. Area is transition region between equiaxed grains (left) and large columnar grains (right). Note lenticular void configurations in center.

A light-gray second phase was found in some of the microstructures of specimens ANL-35-69 and ANL-35-70, both of which contained ThO_2 -25.6w/o UO_2 . The phase was near the central void at various elevations in

EI-94

13X

Figure 22. Longitudinal Section Through a Bottom Pellet of ANL-35-67. Section is not through centerline, and view is axial down columnar grains. Note grain boundary configurations.

both specimens and formed the matrix of a two-phase dendritic structure at the base of the central void in ANL-35-70. The dendritic structure can be seen in Figures 16 and 20. The dendrites are evidently the result of solidification of a molten zone. The gray phase extended outward from the center by way of cracks and grain boundaries (as shown in Figure 23) and was found as far as half the distance from the center to the edge of the pellet. Figures 24 and 25 show this phase as it appeared in areas away from the center. Figure 26 shows the phase as it appeared higher in the specimen in an area that does not appear to have melted.

Attempts to identify the gray phase yielded inconclusive results. One possibility was that the phase was aluminum from the aluminum pellet at the top of the fuel column. A small

sample of the dendritic area was chipped out and analyzed by emission spectroscopy. It was reported to contain 25 w/o aluminum and 0.4 w/o calcium. Both of these constituents were thought to be impurities introduced by dissolving the oxide sample in a Pyrex beaker with a solution containing hydrofluoric acid. The spectral region that was analyzed, however, did not include other elements that also would be present from the dissolution of glass. Therefore, the hypothesis that the aluminum came from alumina in the Pyrex beaker could be neither proved nor disproved. The aluminum pellet from the top of the fuel column was then removed and examined. Because of its poor surface quality both before and after irradiation, it was not possible to determine conclusively that part of the pellet had melted. It was found, however, that the central void in the oxide was not continuous up to the pellet, and that a polished section of the pellet did not have the same appearance as the gray second phase.

If the aluminum pellet was the source of the gray second phase, a dendritic structure or free aluminum should also have been present at the base of the central void in specimen ANL-35-67. This specimen achieved an estimated maximum $\int_{T_s}^{T_0} k d\theta$ rating of 63 W/cm at the top of the fuel

31800

250X

Figure 23. Boundary Between Two-phase Dendritic Structure (right) and Columnar Grains (left) in the Center of an ANL-35-70 Tube. Second phase extends into cracks and grain boundaries of the columnar grains.

31796

250X

Figure 24. Light-gray Phase in Region One-third the Distance from Center to Edge of ANL-35-70

31799

250X

Figure 25. Pipe of Light-gray Phase in Extension from Center Region in Specimen ANL-35-70

29764

250X

Figure 26. Light-gray Phase Adjacent to Central Void above Dendritic Structure at Bottom of Pellet in Specimen ANL-35-70. Material in central void (top) is epoxy resin.

in the same high flux position in which specimen ANL-35-70 reached an estimated rating of 59 W/cm at the bottom of the fuel. Specimen ANL-35-67 was irradiated below the midplane of the reactor, and specimen ANL-35-70 was irradiated above the midplane. With this situation, the aluminum pellet in specimen ANL-35-67 had a greater probability of melting than did the aluminum pellet in specimen ANL-35-70. But a longitudinal section through a bottom pellet of specimen ANL-35-67 (Figure 27) showed no two-phase structure or free aluminum. Therefore, the gray phase is not believed to be aluminum. A comparison of the section in Figure 27 with a similar one from specimen ANL-35-70 (Figure 28) shows the results of the different heat ratings at the base of the fuel tubes. The degree of convex "dishing" is directly related to the heat rating, as is the columnar recrystallization.

Figure 27. Longitudinal Section Through Bottom Pellet of ANL-35-67. No two-phase material is present at the base of central void.

Figure 28. Longitudinal Section Through Bottom Pellet in Unfailed Tube of ANL-35-70. Two-phase material is at the base of central void.

The gray second phase was observed only in specimens containing ThO_2 -25.6w/o UO_2 fuel, and its appearance may be dependent on composition. Unfortunately, none of this material was available in the unirradiated condition for examination. The microstructure of an unirradiated, unbonded 12.7w/o UO_2 pellet (Figure 29) was free of any second phase. Coring has been ruled out as the cause of the gray phase because of its occurrence in areas that were not melted. Also, coring was not found in a ThO_2 -30w/o UO_2 specimen that melted during a transient irradiation test in the TREAT

reactor.⁽¹⁶⁾ Although the nature of the gray phase had not been established, it is felt that it is native to the fuel material and not from an external source.

Figure 29. Unirradiated ThO_2 -12.7w/o UO_2 . No second phase is evident. Unetched.

The cladding of specimens ANL-35-69 and ANL-35-70 proved to be of uneven thickness, and it was in the thinner regions in which the specimen failures occurred. Figures 30 and 31 show the defects and the thin-wall

Figure 30

Transverse Section Through Defect in ANL-35-69. Note reduced cladding thickness in the region of failure. The fuel was lost during irradiation.

29291

7X

Figure 31

Cladding Failure in ANL-35-70. Fuel was partially lost during irradiation.

claddings of the two specimens. Figure 32 shows the failure in ANL-35-69 near the top of the specimen where the oxide is beginning to show waterlogging effects. The failures appear to have been ductile type fractures and not related to cladding burnout. It is not evident if corrosion played a contributing role.

29738

50X

Figure 32. Upper Cladding Failure in ANL-35-69. Fuel (bottom) has begun to show effects of waterlogging.

Fission Gas Release

The out-of-cell fission gas collecting and measuring apparatus consisted of a known volume containing two tilting McLeod gages and a Wallace-Tiernan dial gage for measuring pressure, and two pressure-reducing 300-cc bulbs. The in-cell puncturing apparatus (shown in Figure 33) consisted of an aluminum vacuum chamber that completely contained the specimen. Two piercing needles, each aligned with the gas space of one of the fuel tubes, entered the chamber through double O-ring compression seals. The needles were driven through the cladding, while a static vacuum of less than 10^{-2} torr was maintained in the system. The escaped gases diffused throughout the evacuated system, and the resulting pressure rise was measured after equilibrium had been reached. For roughly half the specimens, a 0.5-cc aliquot of the gas was taken directly for mass spectrometric analysis of the stable and long-lived isotopes of krypton and xenon. For the rest of the specimens, all the escaped fission gas was statically trapped onto activated charcoal, cooled to liquid nitrogen temperature. The gas was later desorbed from the charcoal, passed over uranium turnings at 800°C , and retrapped onto a series of three activated charcoal traps, cooled to liquid nitrogen temperature, while a dynamic vacuum was maintained in the system. The purification step was added to remove the helium and air filler gas from low-burnup specimens to allow a more sensitive analysis for the fission gases. The charcoal traps were desorbed, and the fission gas was transferred by a Toeppler pump to an evacuated known volume of the system, where the pressure rise was measured and the volume of gas determined. An aliquot sample of the gas was then taken for mass spectrometric analysis.

EI-207

Figure 33. Apparatus for Puncturing Specimens for Fission Gas Collection

The mass spectrometric analysis contained the following isotopes: Xe^{131} , Xe^{132} , Xe^{134} , Xe^{136} , Kr^{83} , Kr^{84} , Kr^{85} , Kr^{86} . Additional fission yields of 5.8% for U^{235} and 5.6% for U^{233} were added to the total theoretical yields of the above isotopes to account for neutron capture by Xe^{135} and formation of Xe^{136} . The total fission yields used in calculating the theoretical gas yields were 31.4% for U^{235} fission and, where significant, 33.2% for U^{233} fission.(17)

The gas results, in terms of per cent of theoretical gas release, are included in Table II and are plotted as functions of burnup and

$\int_{T_s}^{T_0} k d\theta$ in Figures 34 and 35, respectively. A gas-release value from

each tube of the specimen that was analyzed is included in the table. Figures 34 and 35, however, exclude those values that were obviously in error as a result of sampling or analyzing techniques. There were three such erroneous values of a total of 30 samples, and neither their presence nor absence in the figures is deemed significant.

106-6690

Figure 34. Fission Gas Release as a Function of Burnup. Numbers refer to specimen identification given in Table II. Overheated specimens are underlined.

106-6693

Figure 35. Fission Gas Release as a Function of $\int_{T_s}^{T_0} k d\theta$. Numbers refer to specimen identification given in Table II. Overheated specimens are underlined.

The gas release as a function of burnup strongly reflects the thermal flux, and hence the heat rating, that was necessary to achieve the burnup in the prescribed irradiation period. This is best exemplified by specimen DG-2, which was irradiated to 28,400 MWd/T over a comparatively long period of time at a low heat rating, and which released no more gas than specimens having 30% lower burnup in shorter times at slightly higher heat ratings. The plot of gas release versus heat rating is not at a constant time. However, the release of gas, even in one reactor cycle, is shown to be strongly temperature-dependent no matter what the irradiation period.

The specimens that received a high flux for their last reactor cycle released abnormally large amounts of fission gas compared to their low-flux counterparts. Gas release ranged from 4.6 to 48.0% in specimens which were not overheated, and from 30.2 to 81.2% in overheated specimens. Metallographic examination showed that the overheated specimens had extensive columnar recrystallization. Such recrystallization below the melting point is attributed to oxide sublimation and condensation with accompanying large release of fission gas. Specimen ANL-35-70, which experienced melting, released the largest amount of gas, 81.2% of theoretical. Recrystallization or melting occurring spontaneously after an appreciable burnup would be expected to release a large percentage of the gas already generated.

DISCUSSION OF RESULTS

A search for the cause of the two specimen failures made the following facts apparent: (1) both failed specimens contained 25.6w/oUO₂, 93.2% enriched; (2) local cladding surface-heat fluxes exceeded 265 W/cm²; (3) the specimens were both irradiated above the reactor midplane; (4) the specimens failed near the bottom of the assembly where maximum diametral expansion occurred; (5) a vertical temperature gradient existed, the bottom of the specimen being the hottest; (6) the failures occurred in regions of thin-walled cladding, a result of a poor extrusion; and (7) a specimen containing 12.7w/oUO₂, 93.2% enriched, irradiated below the reactor midplane (the top end was then the hottest) with a maximum local surface heat flux of 355 W/cm², did not fail.

The immediate cause of failure is attributed to excessive internal pressure in areas of weak cladding. The internal pressure was probably caused by a combination of fuel thermal expansion and released fission gases. In specimens irradiated above the reactor midplane, the greatest radial expansion was at the bottom of the specimen. It is possible that the radial strain in the lower portion of the specimen could not be absorbed by a vertical movement of the soft lead in the annulus because of restraint by cooler and more rigid lead above and the end of the assembly below. The radial expansion would therefore continue with subsequent cladding failure at weak areas. The failures took the form of longitudinal cracks which tapered into regions of lower stress in cooler sections of the assembly, as would be the case if the proposed hypothesis is correct.

The specimen containing 12.7w/oUO₂ and with a maximum local surface heat flux of 355 W/cm² probably did not fail because the plastic lead could partially absorb the radial fuel expansion by moving vertically into the unoccupied free space at the top of the tube. Since a metallographic section was not taken in this region, it is not known whether or not the cladding was thin-walled. This specimen exhibited a significant volume expansion and a diametral expansion in the lower portions of both fuel tubes; the absence of a failure suggests that a thin cladding section did not exist in this specimen or failure might have occurred. Likewise, specimen ANL-35-66, with an estimated maximum cladding heat flux of 280 W/cm², did not fail. This specimen was irradiated in an essentially flat flux during the high flux cycle. A severe vertical temperature gradient in the lead would then not exist, and no local pressure-point would result from restriction of the lead.

The effect of fuel composition on the specimen failures came indirectly from the increased amount of fissionable material in the 25.6w/oUO₂ specimens. Higher heat fluxes were obtained from these specimens when they were irradiated in the same irradiation column as the specimens having lower UO₂ content.

The lowest heat rating at which columnar grain growth and central void formation were observed was in a ThO_2 -12.7w/o UO_2 specimen at an $\int_{T_s}^{T_0} kd\theta$ rating of about 40 W/cm. This value compares well with a value of 41 W/cm for ThO_2 -10w/o UO_2 from the work of Kittel and Handwerk.⁽¹⁾ However, this value is low compared to a more recent value for ThO_2 -10w/o UO_2 , between 54 and 67 W/cm, found by the authors at considerably higher surface temperatures.⁽¹⁴⁾ (The range of 54 to 67 W/cm is quoted because the limited number of specimens in that investigation precluded a more accurate determination.) The difference can probably be traced to the fabrication methods and consequent differences in final oxygen-to-metal ratios. The present pellets and those of Kittel and Handwerk were a cold-pressed and air-fired mixture of ThO_2 - U_3O_8 . The pellets giving the higher values of $\int_{T_s}^{T_0} kd\theta$ were a ThO_2 - UO_2 mixture, hot-pressed in graphite molds.

An average value of $\int_{T_s}^{T_{cg}} kd\theta = 23 \text{ W/cm}$ (where T_{cg} is the temperature at which columnar grain growth is just discernible) was calculated for specimens ANL-35-67, ANL-35-69, and ANL-35-70, the only specimens exhibiting columnar recrystallization where sections were made. The individual values for these specimens were 22, 24, and 23 W/cm, respectively. Since the greatest part of the recrystallization in ANL-35-67 and ANL-35-70 took place during the last overheating cycle, the values of $\int_{T_s}^{T_{cg}}$ could not be correlated with the irradiation period. The good agreement between specimens may be fortuitous. A value of $\int_{T_s}^{T_m} kd\theta = 54 \text{ W/cm}$ (where T_m is the melting temperature of the oxide) was calculated for ANL-35-70, the limit of the two-phase region being taken as the extent of melting. These values for columnar growth and melting are lower than corresponding values for UO_2 (about 65 W/cm⁽¹⁸⁾ and 81 W/cm,⁽¹⁹⁾ respectively). The pellet surface temperature, T_s , in the UO_2 experiments was estimated at between 300 and 350°C, compared to 190 to 240°C in the present experiment. Considering the lower vapor pressure, higher melting point, and comparable thermal conductivity of ThO_2 , it would be expected that recrystallization and melting in ThO_2 - UO_2 solid solutions would correspond to higher $\int kd\theta$ values than for those of UO_2 . The lower values can be explained by either the low oxide density or the excess oxygen present from incomplete reduction of U_3O_8 to UO_2 in the air-firing process.

Excess oxygen in the fuel would enhance grain growth and account for the large grains observed in the hotter specimens. These large recrystallized grains contained no transverse voids that had been observed in the hot-pressed $\text{ThO}_2\text{-UO}_2$ pellets.⁽¹⁴⁾ Almost all voids, right up to the edge of the central cavity, were either spherical in shape or radially oriented. Only in the transition region between equiaxed and columnar grains in one specimen were small columnar grains noted with transverse lenticular voids. Since excess oxygen reduces the thermal conductivity, large thermal gradients would be expected in the fuel. A large thermal gradient could account for the absence of lenticular voids since these voids could move rapidly to extinction. The probability of finding them in an area where the temperature was above the minimum required for movement would be low. It could also be reasoned that oxygen-enhanced grain growth after recrystallization would obliterate the original grain boundaries and the lenticular voids. The presence of stable lateral boundaries of the columnar grains can be considered an indication of grain growth.

The fission gas release from these specimens is considered high for the burnups and heat ratings that were attained. This is attributed to the low bulk density of the oxide (about 70 to 80%) and to the excess oxygen present in the fuel.

The extensive loss of fuel from the failed specimens does not directly contradict previous findings of excellent corrosion resistance by $\text{ThO}_2\text{-UO}_2$ fuels.^(20,21)

The previous tests were at maximum $\int_{T_s}^{T_0} k d\theta$ ratings of 15 W/cm, and no melting, recrystallization, or central void formation was observed. The much higher heat ratings of our failed specimens would make extensive water-logging a primary cause for loss of fuel.

CONCLUSIONS

1. Lead bonding performs its intended task of improving fuel-cladding gap conductivity, but its use with low strength, thin-walled cladding may result in swelling and eventual rupture of the fuel rod.
2. Fabricating solid solutions of $\text{ThO}_2\text{-UO}_2$ by air-firing cold-pressed compacts of $\text{ThO}_2\text{-U}_3\text{O}_8$ produced a fuel containing excess oxygen. The hyperstoichiometric oxide possesses the undesirable irradiation properties of lower thermal conductivity, greater recrystallization, and significant fission gas release.

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