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A HIGH-TEMPERATURE IRRADIATION
AND POST-IRRADIATION ANALYSIS
OF URANIUM NITRIDE FUEL

MASTER

E. D. Albrecht, A. J. Rothman, J. D. Lee,
J. M. Johnson, and W. N. Hayes, Jr.

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Lawrence
Radiation
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Lawrence Radiation Laboratory
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LIVERMORE

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A HIGH-TEMPERATURE IRRADIATION AND POST-IRRADIATION ANALYSIS OF URANIUM NITRIDE FUEL

Abstract

A fission-heated, gas-bonded, binary-gas-controlled irradiation capsule containing eight uranium nitride specimens has been irradiated in the Battelle Research Reactor for eight cycles. The median specimen temperature ranged from 1307 to 1417°C during the irradiation, and the average burnup was 2.3×10^{20} fissions/cc. Two specimens each of four different microstructures, including fine-grain (30 μ), 96-percent-dense; coarse-grain (105 μ), 97-percent-dense; single-crystal, 100-percent-dense; and fine-grain (30 μ), 78-percent-dense fuel were irradiated for a total of 3558 hr. The 9.6-percent-enriched specimens were nominally 0.750 in. in length and 0.200 in. in outside diameter and had a 0.100-in. -diam axial hole. Each specimen was composed of from one to three segments; the lower segment of the single-crystal specimens was an undrilled, solid piece. The fuel specimens were clad in vented W-25.5Re that had a wall thickness of 0.020 in. To prevent decomposition of the uranium nitride, a partial nitrogen pressure was maintained during the irradiation by the external heating of U_2N_3 powder located at the bottom of the capsule. Neutron radiographs were made of the capsule before irradiation and at the conclusion of each cycle in an attempt to acquire

progressive swelling data during the irradiation. An extensive post-irradiation examination was made that included measurement of physical dimensions such as lengths, inside diameters and outside diameters, density, metallography, chemical analyses for composition, and radiochemical analyses for determination of uranium burnup. The only significant changes observed in the microstructure were the creation of fine porosity at the grain boundaries of all polycrystalline specimens and the preferential alignment of this porosity in those grain boundaries lying parallel to a specimen surface. The single-crystal specimens did not exhibit this porosity except for rare occurrences along subgrain boundaries near the surface. Analyses of retained fission gas by mass spectrometric techniques have shown that nearly 100 percent of the fission gas generated has been retained in each specimen, regardless of microstructure. The fine-grain (30 μ), 96-percent-dense UN swelled the greatest amount at 1417°C and to a lesser extent at 1307°C, revealing an implied temperature dependence. The fine-grain (30 μ), 78-percent-dense and single-crystal, 100-percent-dense UN showed the least swelling under the conditions of this irradiation.

Objective

The objective of the high-temperature nuclear-fuels irradiation experiment was to obtain swelling data on uranium nitride

fuel in support of the reactor designs developed in the Space Electric Power Program at the Lawrence Radiation Laboratory.

Introduction

Preliminary studies led to the selection of UN as the prime fuel candidate for the Space Electric Power Program's reactor designs. From considerations of cycle thermodynamics, one develops the thesis that "hotter is better," and so it was established that a high-temperature fuel irradiation program was needed. And, since fuel swelling is a major problem in most reactor designs, it was given prime consideration.

Various theories of fuel swelling—or at least theories that relate to fuel swelling—have been propounded.^{1,2} Some studies and analyses on bubble migration in solids that relate to fuel swelling have been made.³⁻⁶ Even so, no satisfactory comprehensive theory of fuel swelling exists.

Some of the above studies suggest variables that may be important in influencing fuel swelling. If we assume that swelling is caused by the growth and migration of bubbles,⁶ it is clear that diffusion of gas atoms and/or gas bubbles will affect swelling behavior. In turn, the diffusion rates should be affected by the thermodynamic activity of the fuel. In previous experiments, no specific attempts have been made to fix the activity. In this experiment, a fixed pressure of nitrogen over the UN was maintained by heating at the U_2N_3 source. Ultimately

it was planned to change the uranium activity to other fixed values in future experiments.

There is excellent experimental evidence that bubbles migrate in a temperature gradient.⁷ But other experiments show that bubbles tend to be trapped at defects, especially at grain boundaries. Thus, we were interested in fuel with no grain boundaries (single crystals); would the bubbles migrate out of the fuel without impedance by boundaries? We also wished to determine the effect of few grain boundaries (coarse grain) and many grain boundaries (fine grain).

Once the fuel and temperature range of interest were determined, it was then necessary to establish certain desired conditions for the fuel samples to be irradiated. These conditions are specified below:

- (1) Several microstructures were to be compared, including single-crystal, large-grain high-density, small-grain high-density, and approximately 75-percent-dense uranium nitride.
- (2) Specimen surface temperature from 1350 to 1800°C.
- (3) Radial temperature profile limited to less than 50°C.
- (4) Accurate temperature control and measurement.

- (5) Up to 6 percent total uranium burnup.
- (6) Annular specimen design to be used to permit fission gas release and inner wall (center-line) temperature measurement. Solid specimens to follow, permitting annular versus solid specimen comparison.
- (7) Nitrogen cover gas to be maintained at a fixed partial pressure and temperature to keep constant uranium thermodynamic activity and to prevent UN dissociation.
- (8) Optimum specimen design to be free-standing or clad specimens that would allow approximately 30 percent unrestrained radial and axial growth.
- (9) Specimen inner-to-outer-radius burnup ratio to be greater than 0.8 to approximate fast reactor conditions.
- (10) Retention of fission gas from individual sample of secondary interest.
- (11) Eight or more individual samples per capsule.

Battelle Memorial Institute, Columbus, Ohio, was selected to design, construct, and irradiate two capsules and to fabricate the fuel samples for each under contractual agreement with LRL. The selection of BMI was based primarily upon their extensive experience in the high-tempera-

ture fuel irradiation field and the programmatic requirement for UN swelling data at the earliest possible date.

The first irradiation capsule was designated LRL-1 and was of the fission-heated type, requiring no external heaters surrounding the fuel. The gas-bonded, binary-gas-controlled capsule contained two specimens each of four different microstructures. Capsule LRL-2 was of a similar design, but its fabrication lagged that of LRL-1 by sufficient time to permit design changes for the purpose of correcting inadequacies revealed during the irradiation of the first capsule. The irradiation of capsule LRL-2 was delayed because the LRL Space Electric Power Program was cancelled by the AEC. Had this program continued, our plan would have been to irradiate an LRL-designed, gas-bonded and binary-gas-controlled capsule in a higher power reactor such as the GETR. This was to have been followed by a liquid-bonded, binary-gas-controlled capsule and finally by a capsule using the annular heat-pipe concept of temperature control.

This report will be limited to a description of capsule LRL-1 design, its irradiation, post-irradiation examination and other related topics, as well as the reporting and discussion of irradiation results.

Experimental Techniques

The experimental techniques, calculations and methods used to determine the capsule design, fabricate fuel and cladding, irradiate the capsule,

calculate specimen temperatures and perform the post-irradiation examination are described in detail in the following paragraphs.

CAPSULE DESIGN

In conjunction with the AEC's High Temperature Fuels program, BMI, Columbus had designed and irradiated capsules containing up to six clad specimens in the 1600 to 2000°C range.⁸ Because of this experience, and LRL's immediate need for fuel swelling data, the BMI high-temperature capsule was selected as the design basis for a capsule to be altered to the specific needs of this program. A diagrammatic description of this capsule is shown in Fig. 1.

The clad fuel specimens were suspended end to end on a molybdenum and tungsten support structure along the centerline of the vertical capsule. This assembly was sealed in a molybdenum shell that formed the primary container. A helically wound nichrome heater having magnesium oxide insulation and sheathed in stainless steel was attached to the bottom end of the primary container to permit the external heating of depleted U_2N_3 powder to 750°C. At a fixed temperature, the equilibrium $U_2N_3 = 2UN + 1/2 N_2$ maintained a fixed N_2 pressure of 6 Torr in the capsule. Argon was used to fill the remaining space in the primary container to minimize heat conduction. The predominate mechanism of heat transfer from the fuel was by thermal radiation. A 1-in.-diam stainless-steel shell formed the outside or secondary containment. A smaller gas annulus between the primary and secondary containers was used to permit temperature selection and control during irradiation by varying the composition of the helium-nitrogen gas mixture, i. e., changing the conductivity of the gas mixture.

Temperatures were monitored by W-5Re (tungsten-5 percent rhenium)

versus W-26Re thermocouples, each leg of which was composed of eight strands of 0.003-in.-diam wire. One thermocouple was placed in a well receptacle in the top specimen cladding, permitting centerline temperature determination of only that specimen. All specimens had surface temperature monitored by similar thermocouples tied to their cladding as shown in Fig. 2. The hot junction was simply a square knot. A similar thermocouple was inserted in the U_2N_3 container to sense the temperature of that mass. Five chromel vs alumel thermocouples were attached to the outside of the primary container in the control gas gap.

The thermocouples from within the primary containment were insulated in BeO insulators and brought through a header assembly. Epoxy was used to make the final gas-containing seal well above the elevated temperature zone.

Assuming that the fuel enrichment and density were to remain reasonably constant from one capsule to another, the capsule temperature could be varied by simple alteration of the primary and secondary gas gap dimensions. This would permit the same specimen configuration and arrangement for temperatures from 1200 to 2000°C.

FUEL FABRICATION

Again, BMI, Columbus was selected for fuel fabrication for the reasons of experience and immediate availability of fabrication facility. The uranium nitride fuel was to have certain variables in configuration, which are listed below in four categories:

1. Single-crystal, 100 percent dense

Fig. 1. Diagrammatic description of irradiation capsule LRL-1.

Fig. 2. Typical clad specimen from capsule LRL-1 showing vent hole, positioning wires and W-5Re vs W-26Re thermocouple attachment. The smallest divisions on the accompanying scale are 1/32-in.

2. Large-grain ($150\ \mu$), 100 percent dense
3. Fine-grain ($25\ \mu$), 100 percent dense
4. Fine-grain ($25\ \mu$), 75 percent dense

Duplicate specimens approximating each of these four types were irradiated in capsule LRL-1.

Depleted and 93-percent-enriched uranium metal were blended in an induction furnace for 20 min and the resulting 10-percent-enriched metal was cast in copper molds. Part of this product was fabricated into 0.25-in.-diam rods for consumable arc melting under nitrogen, and the remainder was rolled and nibbled for making UN powder by the metal-gas-reaction process.

Procedures for the preparation of single-crystal UN basically consisted of reacting a consumable electrode of uranium in about 15 atm of nitrogen pressure, and, by arc-melting procedures, drip melting the UN into a water-cooled crucible. Melting conditions were adjusted to produce UN with a U_2N_3 surface or grain boundary film to insure complete reaction. Heat treatment at 1650°C under a pressure of less than 1×10^{-3} Torr for 8 hr was used to remove U_2N_3 from the grain boundaries. At this point, large single grains were extracted from the casting and used as stock for the single-crystal specimens.

The polycrystalline specimens were produced by powder metallurgical methods. Chunks of uranium approximately 1/4 in. on a side were reacted with nitrogen at approximately one atmosphere pressure and 850°C . U_2N_3 was produced by this process and spalled from the surface, yielding finely divided powder. A subsequent heat treatment at 1300°C under vacuum permitted conversion to UN. The UN powder was cold pressed into cylinders that were in turn placed in graphite-lined tantalum molds, which were electron-beam welded closed under a pressure of 2×10^{-5} Torr or less. This material was then isostatically hot pressed at 1650°C and 10,000 psi. The fine-grained, high-density materials were produced by this procedure. A lower pressing pressure was used to produce the 78-percent-dense material.

Fine-grained, high-density UN was then shipped to LRL and annealed at 2350°C under 100 Torr nitrogen pressure for 115 hr to produce the large-grained UN, having an average grain diameter of $105\ \mu$.

All specimens were ground into right circular cylinders 0.200 in. in diameter and from 1/4 to 3/4 in. long. A 0.095-in.-diam hole was machined by EDM drilling along the centerline and this was honed to a final diameter of 0.100 in. The single crystals were stacked in three pieces, nominally 1/4 in. in length, with the bottom segment remaining a solid, undrilled piece. All other specimens were composed of one or two pieces to make up their nominal 3/4 in. length. The loading order for capsule LRL-1 is given in Table 1.

Table 1. Specimen loading order for capsule LRL-1. Density is based upon the percentage of theoretical density of 14.32 g/cm².

Specimen	Microstructure
1-C Top	Fine grain (30 μ), high density (96%)
2-A	Single crystal, high density (100%)
3-B	Single crystal, high density (100%)
4-D	Fine grain (30 μ), low density (78%)
5-E	Fine grain (30 μ), low density (78%)
6-G	Fine grain (30 μ), high density (96%)
7-H	Coarse-grain (105 μ), high density (97%)
8-I Bottom	Coarse grain (105 μ), high density (97%)

CLADDING

W-25.5Re material purchased from Chase Copper and Brass Company as 1/4-

in.-diam rod was used as the stock for the claddings. The rods were bored by EDM drilling and then honed to remove the layer of carbon contamination and damaged surface caused by the spark discharge. In the honing operation, 0.002 in. of stock was removed from the tube wall and a mechanically polished surface was produced. Six claddings were used in the mechanically polished condition, while two were electropolished on the inside surface to a mirror finish. The thermocouple well for the top specimen and the bottom and top end plugs were machined from the same material. The bottom specimen surface was separated from the cladding by a 0.030-in.-thick depleted UN disc inserted for the purpose of better distinguishing the fuel during neutron radiograph measurements. Electron-beam welding was employed to weld the end plugs to the cladding tubes, thereby encapsulating the UN fuel. A single tapered vent hole, 0.010 in. in diameter on the inside and 0.030 in. in diameter on the outside, was located near the top of each encapsulated specimen.

IRRADIATION

Capsule LRL-1 was irradiated in the Battelle Research Reactor for a total of 3558 hr. A 3 in. $\times$ 3 in. core position was used for the eight-cycle irradiation. The open pool-type reactor operates at 2 mW, with a peak flux in excess of 1×10^{13} nV thermal. A continuous record of control gas composition and pressure, as well as temperature detection from each of the 15 thermocouples, was made during irradiation. A solid-state flux monitor with continuous current readout was installed on the outside of the capsule

opposite the bottom specimen and moved to the top specimen near the end of each cycle to provide a relative axial flux profile of the capsule.

NEUTRONIC AND THERMAL ANALYSIS

Prior to irradiation of Capsule LRL-1, independent neutronic and thermal analyses were made at LRL and BMI to test the analytical methods employed, check capsule design and reduce uncertainties in the predicted fission rates and temperatures of the fuel specimens.

Neutronic analysis of the capsule was performed using a P_3 flux perturbation cell code that used specimen loading, capsule specifications and an unperturbed flux profile provided by BMI.

Subsequently, a thermal analysis was performed using the fission rates calculated above, capsule specifications, and a two-dimensional (R and Z) nonlinear (T^4 radiation and T^2 conduction) heat transfer code called HTRZ.⁹ The analyses included determination of the relative temperature profile of a single specimen and absolute temperature profiles of the total capsule under various power levels and gas gap compositions. The resulting capsule profiles were then compared with measured capsule temperatures. The above analysis has been completely described by Lee.¹⁰

During the first irradiation cycle it became obvious that the specimen-clad thermocouples were not measuring actual specimen temperatures, since the hot junctions were so large compared to the size of the annulus they were placed in, and because of the steep temperature gradient in this annulus. This made direct

temperature measurement of specimens 2-A through 8-I impossible. Fortunately, the top specimen, 1-C, also had a centerline thermocouple which appeared to record reasonable temperatures throughout the irradiation. For this reason it was necessary to calculate temperature histories of the remaining seven specimens during the eight irradiation cycles.

The temperature histories were calculated from available information including capsule specifications, one relative neutron flux profile measurement for each cycle except cycle 2, and the temperature history of the top specimen, 1-C, as recorded by its centerline thermocouple. Assumptions made were that the properties and dimensions of the materials did not change during the irradiation; that the relative neutron flux profile was constant during a cycle and changed between cycles; and that variations in the top specimen's measured temperature during a cycle were caused by variations in reactor power and these variations were experienced by all specimens.

Power profiles for each cycle were calculated by weighting the specimen powers in cycle 1 by the ratio of the specimen relative flux in the cycle of interest to the same specimen's cycle 1 relative neutron flux. Detailed calculations of the power profile have been reported by Lee.¹⁰

The second step of the analysis was to run a heat transfer problem for each different power profile to determine the effect of varying the relative neutron flux profile on the specimen train temperature profile. This heat transfer analysis is also further described by Lee¹⁰ and the results are given in Table 2.

Table 2. Calculated specimen temperature profile (°C).

Specimen No.	Cycle						
	1	3	4	5	6	7	8
1-C	1415	1390	1382	1370	1350	1371	1341
2-A	1485	1467	1470	1450	1430	1447	1417
3-B	1487	1483	1483	1472	1445	1466	1438
4-D	1436	1439	1439	1431	1404	1427	1407
5-E	1441	1459	1458	1455	1437	1454	1438
6-G	1469	1503	1500	1499	1490	1496	1492
7-H	1409	1460	1455	1454	1440	1443	1450
8-I	1417	1487	1476	1471	1465	1463	1469

Step three in the analysis relates the temperature history of each specimen to that measured for specimen 1-C, weighted by the calculated temperature profiles for each irradiation cycle. This was done for each cycle using the following relationship:

$$T_n = \frac{(TM_1 - 50)(TC_n - 50)}{(TC_1 - 50)} + 50$$

where

n = specimen number

TM_1 = measured temperature of specimen 1-C (°C)

TC = Table 2 calculated temperatures (°C)

50 = temperature of heat sink (°C).

The results of the temperature history calculation based upon the measured temperature history of specimen 1-C are given in Table 3. Average (mean) and median temperatures for each cycle and for the entire eight-cycle irradiation were calculated.

NEUTRON RADIOGRAPHY

A system of examining specimens by neutron radiography was used during this irradiation. Prior to irradiation and at the conclusion of each BRR cycle, capsule LRL-1 was moved into a reactor pool position where the capsule was interposed between a collimated neutron beam and an indium foil. Variation in neutron attenuation of the capsule caused the indium to activate nonuniformly and an image of the capsule and its contents was formed on the foil. A photographic film was then brought into contact with the decaying foil to transfer the image. The film was not exposed to the highly radioactive specimen and therefore not fogged by the radiation.

This technique was used in an attempt to observe the progression of swelling from one cycle to another, and thereby establish a swelling rate. Unfortunately, due to the presence of excessive amounts of molybdenum, tungsten, and rhenium in the capsule, the specimen image was not sharp and definitive dimensions were not readily obtained.

Table 3. Calculated specimen temperatures (°C).

Specimen No.		Cycle								Overall
		1	2	3	4	5	6	7	8	
1-C	Average	1381	1352	1302	1351	1327	1290	1268	1232	1317
	Median	1417	1377	1297	1357	1307	1287	1287	1237	1307
2-A	Average	1449	1429	1372	1436	1401	1364	1337	1302	1388
	Median	1487	1447	1367	1437	1377	1357	1357	1307	1377
3-B	Average	1449	1431	1386	1449	1425	1379	1356	1321	1401
	Median	1487	1447	1377	1457	1407	1377	1377	1327	1397
4-D	Average	1401	1382	1343	1403	1387	1340	1318	1292	1359
	Median	1437	1397	1337	1407	1367	1337	1337	1297	1357
5-E	Average	1401	1382	1360	1432	1408	1370	1345	1321	1376
	Median	1437	1397	1357	1427	1387	1367	1367	1367	1377
6-G	Average	1431	1411	1404	1464	1450	1421	1383	1369	1415
	Median	1467	1427	1397	1467	1427	1417	1407	1377	1417
7-H	Average	1371	1352	1363	1421	1408	1391	1335	1331	1369
	Median	1407	1375	1357	1427	1387	1387	1357	1337	1367
8-I	Average	1381	1362	1391	1441	1421	1399	1351	1349	1385
	Median	1417	1377	1387	1447	1397	1397	1367	1377	1387

POST-IRRADIATION EXAMINATION

At the conclusion of the final irradiation cycle (No. 8), capsule LRL-1 was transferred to the BMI Hot Cells where the primary containment was pierced by drilling and the fission gases released during irradiation were pumped into glass collection vials of known volume that were later analyzed by mass spectrometry for xenon and krypton and their isotopic compositions.

The capsule was then cut to about 19 in. in length and shipped in a lead-lined cask to LRL for the remaining post-irradiation analyses, which were performed at the β - γ Hot Cell facility described by Albrecht, et al.¹¹

Capsule disassembly began with the removal of the secondary stainless-steel containment by cutting, with a motorized pipe cutter, just below the header. The tubular vessel was easily slipped off, revealing the primary molybdenum container, which was removed by the same procedure. Figure 3 shows the disassembled capsule.

The specimen train was then disassembled from bottom to top by first removing a sleeve and then a clad specimen and its attached thermocouple. All parts were carefully numbered and placed in specially designed storage boxes for each specimen.

Each clad specimen was photographed at 4X with its thermocouple in place using a "windowless" macro-camera, described

Fig. 3. The primary and secondary containment removed from the buffered specimen train of capsule LRL-1 (upper photo), and remote disassembly of the specimen train (lower photo).

by Albrecht, et al.¹² The thermocouples were removed and each clad specimen was again photographed on its circumference at 0, 90, 180 and 270° and on its top and bottom ends. A 10X photograph of the vent hole was made as well as higher magnification photos of cladding cracks, weld porosity and other features of probable interest.

To establish comparative information on cladding dimensions before and after irradiation, diameter measurements were made with micrometers accurate to 0.0002 in. near the top, center and bottom of each clad specimen at 0 and 90° on the circumference. Each measurement was repeated three times. Cladding length measurements were similarly made. Each clad specimen was then weighed to establish if a significant weight change had occurred during irradiation.

The ends of the W-25.5Re cladding were removed by sawing through the upper void space near the vent hole and through the depleted UN disk adjacent to the bottom of the specimen with a 3-in.-diam, 0.006-in.-thick diamond saw running at slow speed, described by Albrecht, Stearns, and Wittmayer.¹³ The internal surface of the cladding ends and the exposed fuel ends were then photographed.

At this point in the examination, the internal diameter of each fuel specimen was measured at 0, 60 and 120° on the circumference and at five positions approximately 1/8 in. apart along the length of the specimens. These measurements, accurate to 0.0002 in., were repeated a second time to establish reproducibility.

Each specimen was then chucked in a specially designed diamond saw where the 0.020-in.-thick cladding was slit length-

wise at 120° intervals on the circumference. The three pieces of cladding were then photographed on interior and exterior surfaces. Photographs were also made of the surface of all pieces of each fuel specimen.

Outside diameter measurements were made at 0, 60 and 120° on the circumference of each fuel segment at from two to five positions along the length to determine the diametral changes resulting from irradiation. Each measurement was repeated at least once as a reproducibility check. Length measurements were also made at this time.

The next step in post-irradiation examination was the determination of specimen density. Each piece of the one-, two- or three-segment specimen was weighed on a semi-microbalance and its volume was determined several times by use of a mercury pycnometer described by Albrecht, et al.¹⁴ The density calculated from these weights and volume measurements was then compared to the pre-irradiation density to establish the gross volumetric swelling of the fuel. At the conclusion of the volume measurements, each segment of a specimen was ultrasonically cleaned and examined to be certain that its surface did not retain mercury. The surface condition was recorded photographically.

Figure 4 shows diagrammatically the sequence of cladding removal and sectioning of the specimens for metallographic and chemical analyses. A longitudinal section was removed from each specimen to permit observation of the microstructure along its length related to the variation in temperature and burnup along the specimen. Wafers were then removed

Fig. 4. Diagrammatic sequence of cladding removal and sectioning of the fuel specimen for metallographic and chemical analyses.

for chemical analyses and adjacent to each of them a wafer was cut for metallography. The remainder of the specimen was stored for future undetermined use. Each of the 9 to 11 wafers cut from the nominally 3/4-in.-long specimen were photographed on each side for possible future reference.

Metallographic specimens were mounted in glass-filled diallylphthalate using a remote mounting press and were then ground in a two-stage grinding sequence using 12.5- μ SiC and 9- μ Al_2O_3 abrasive on 12 in. Lapmaster lapping machines. After ultrasonic cleaning, the specimens were polished on Syntron vibratory pol-

ishers using 1 μ diamond abrasive on silk or nylon cloth. The specimens were again ultrasonically cleaned and the entire surface was photographed in the as-polished condition at 50X magnification. Some areas were also photographed at 200X and 400X. The specimens were then chemically stain etched using 10 ml nitric, three drops hydrofluoric, 30 ml lactic acid etchant and subsequently all areas were re-photographed at the same magnification used in the as-polished condition. The metallographic facility and procedures have been described in detail by Albrecht and Johnson.¹⁵

At least three samples from each specimen were analyzed for total uranium content by solution analytical chemistry. All analyses were run from two to five times for each sample. The procedure followed for this controlled-potential coulometric analysis is a slightly modified version of tentative ASTM Method D-217, which is based upon the method of Booman and Holbrook described elsewhere.¹⁶ The controlled-potential coulometer is described by Harrar and Behrin,¹⁷ and the cell assembly by Harrar.¹⁸

A bomb combustion technique was used for the total nitrogen, xenon and krypton determinations. This method, developed for the simultaneous determination of carbon, hydrogen and nitrogen, has been described by J. W. Frazer.¹⁹ The system used for UN was modified by the omission of a halogen trap and furnaces used for carbon and hydrogen analyses. Other modifications included a larger (100 mg) sample size, higher firing temperature (1000°C), and longer time (24 hr) to allow for complete sample combustion. The evolved gases were then pumped into bombs of predetermined volume that were attached to a mass spectrometer for total nitrogen and isotopic analyses of xenon and krypton.

Fuel burnup was determined by radiochemical analysis where gamma spectroscopy was made on samples that were dissolved in nitric acid and extensively diluted. A carefully calibrated planar Ge(Li) detector²⁰ was used and the data analyzed by the GAMANAL computer code.^{21,22,23} Corrections to the activity were made for the effect of variations in flux during the irradiation, which was long in relation to the half-life of the investi-

gated species. ⁹⁵Zr was used as the primary fission indicator with ¹⁴⁴Ce, which has the advantages of a longer half-life, being used as a secondary fission indicator. Uranium isotopic ratios were determined by standard mass-spectroscopic techniques, while the total uranium content was determined by controlled potential coulometric analysis described earlier.

Representative samples from the cladding on each specimen were mounted and metallographically prepared for inspection in the as-polished, chemically etched or cathodically etched condition. The microstructures were examined and features of interest were photographed.

Two W-5Re vs W-26Re thermocouples, which were attached to the cladding surface during irradiation, were examined metallographically to determine if the bead knot sintered to the cladding and to see if any other microstructural changes took place (such as grain growth or appearance of a second phase).

The centerline thermocouple was left intact and calibrated at 1100°C in an argon-filled, platinum-wound furnace. The activity level of the thermocouple was sufficiently low and swipe-free for the calibration to be performed in a hood outside the hot cells. A platinum vs platinum 10-percent-rhodium standard thermocouple; the unirradiated W-5Re vs W-26Re thermocouple of interest; and the irradiated thermocouple were bound together with beads intimate, but not in contact, and inserted into the argon-filled furnace tube. The free ends were attached to copper extension wire, and these cold junctions were immersed in ice water during the comparative emf readings.

Experimental Results

MACROSCOPIC OBSERVATIONS

Disassembly of the capsule presented no particular difficulties, as both the secondary and primary containers were removed with ease. The primary molybdenum container was highly discolored, apparently caused by minor oxidation during irradiation. The five chromel vs alumel thermocouples were intact and in their proper positions at the conclusion of the irradiation. The specimen train, including the molybdenum buffers, was completely intact with all parts bright and apparently unreacted, and all W-5Re vs W-26Re specimen thermocouples were operational and correctly positioned. A single thermocouple running to the U_2N_3 body at the bottom of the specimen train was also in proper position and operational. The auxiliary heater placed about the U_2N_3 failed during the eighth and final cycle of irradiation; the cause of this failure was not investigated.

Each specimen cladding developed from one to three longitudinal cracks (apparently caused by diametral fuel swelling), resulting in rupture stresses. These cracks varied in length and width in direct relation to the amount of fuel swelling. A typical cladding failure is shown in Fig. 5. The exterior cladding surface remained clean during irradiation and the vent holes were completely open. The W-5Re vs W-26Re thermocouples had sintered to the W-25.5Re cladding in several spots on the higher-temperature specimens.

After the cladding was removed, it was seen that all eight specimens had re-

tained their original configuration. No breakage, spalling, splintering, powdering, crumbling or macroscopic cracks were observed in the fuel. All fuel specimens were readily removed from their cladding with no bonding or adherence, indicating no significant reaction. It was observed later, however, that a layer of UN approximately 0.001 in. thick was deposited on the inner cladding surface of both single crystal specimens 2-A and 3-B and one high-density, fine-grained (6-G) and one high-density, coarse-grained specimen (7-H) in the hottest locations in the capsule. The complementary specimens to Nos. 6-G and 7-H, which were in positions 1-C and 8-I in the capsule, and two fine-grain, low-density specimens, 4-D and 5-E, did not exhibit this UN deposit on the cladding. The 0.030-in.-thick depleted UN disk used for delineation of the bottom edge of each specimen in neutron radiography was sintered to the fuel in specimen 6-G only, the specimen with the hottest thermal history. Specimen 6-G was composed of two segments that were also sintered together during irradiation. No other multiple-segment specimens showed any sintering or bonding between segments.

MICROSCOPIC CHANGES

Careful microscopic comparison of pre- and post-irradiation fuel microstructures were made for each of the four varieties of UN that were irradiated.

The fine-grain, low-density (78 percent theoretical) microstructure was originally composed of polycrystalline, high-density

Fig. 5. Typical cladding failure of specimen 6-G, showing the low ductility of the W-25.5Re cladding used in this capsule. The photo at the left shows the entire clad specimen, while the photo at the right shows a closeup of cracks at 20X magnification (smallest divisions on the scale are 1/32 in.).

UN particles that had rather sharp, angular surface features and that were sintered together in a hot-pressed compact that retained discontinuous angular voids at the particle boundaries. Figure 6 shows a typical pre-irradiated micro-

structure. The only significant change observed in the irradiated material was the collection of spherical porosity, approximately 0.5 to 1 μ in diameter, at the grain boundaries as shown in Fig. 7. This porosity was heavily concentrated

in grain boundaries parallel to the surfaces of the specimen; the concentration was greatest near the inside specimen surface. The grain diameter of this material was from 10 to 40 μ , while the particle size varied from 60 to 100 μ . The grain diameter and particle size were not altered during the irradiation.

Fig. 6. Typical pre-irradiated, 78-percent-dense, fine-grained UN. (200X mag.)

Minor amounts of a second phase, identified as U_2N_3 , were observed and were attributed to fabrication rather than irradiation environment.

Fine-grained, high-density (96 percent theoretical) UN having an average original grain diameter of 30 μ was the second of the four microstructures irradiated. A

Fig. 7. Specimen 5-E. Post-irradiated, low-density, fine-grained UN showing collection of spherical porosity at the grain boundaries. (200X mag.)

Fig. 8. Typical pre-irradiated, 96-percent-dense, fine-grained UN.

photomicrograph of a typical pre-irradiated microstructure is shown in Fig. 8. Again, rather angular voids were present at the grain boundaries, which accounts for the density of the UN being nominally 4 percent less than theoretical. It should be noted in Fig. 8 that the voids are discontinuous and do not delineate the grains.

Figure 9 shows the irradiated fine-grained, high-density microstructure. Two major changes in microstructure occurred within this material. The first was the collection of spherical porosity, approximately 0.5 to $1.0\ \mu$ in diameter, at the grain boundaries. The porosity was most predominant in grain boundaries parallel to the

Fig. 9. Specimen 6-G. High-density, fine-grained UN after irradiation showing spherical porosity collected at the grain boundaries, with the greatest concentration in grain boundaries parallel to the surfaces of the specimen.

the specimen surfaces, and the concentration was again greatest at the inner surface. The second change was that longitudinal cracks appeared near the center of the top of specimen 1-C; the cracks extended radially 0.003 to 0.007 in. into the UN from the outside surface. The complementary specimen, 6-G, which has a higher irradiation temperature history and exhibited greater swelling,

did not show evidence of cracking. This may be related to the presence of the centerline thermocouple well in specimen 1-C.

The coarse-grained, high-density (97 percent theoretical) UN is shown in its unirradiated condition in Fig. 10. The original average grain diameter of $105\ \mu$ resulted from the annealing of the fine-grained UN described earlier. This heat

(a) Stain-etched, 50X mag.

(b) As-polished, 50X mag.

(c) Stain-etched, 200X mag.

(d) As-polished, 200X mag.

Fig. 10. Typical pre-irradiated, 97-percent-dense, coarse-grained UN.

treatment caused the voids to become more equiaxed and to relocate at the triple points of the grain boundaries. A slight densification from 96 to 97 percent theoretical also resulted during the anneal. Figure 11 shows the post-irradiation microstructure. Nearly all grains are completely delineated by spherical

porosity approximately 0.5 to $1.0\ \mu$ in diameter at the grain boundaries. This collection of porosity is a repeat of that previously described for the fine-grained high- and lo-density UN. The porosity located in the grain boundaries parallel to the specimen surface is nearly continuous, due to its abundance. By contrast,

(a) Stain-etched, 50 $\times$ mag.

(b) As-polished, 50 $\times$ mag.

(c) Stain-etched, 200 $\times$ mag.

(d) As-polished, 200 $\times$ mag.

Fig. 11. Specimen 7-H. High-density, coarse-grained UN after irradiation showing a collection of spherical porosity in the grain boundaries, with the highest concentration in those boundaries parallel to the surface of the specimen.

Fig. 12. Typical single-crystal UN, pre-irradiated and stain etched, showing no porosity or other distinguishable features. (50X mag.)

porosity in grain boundaries perpendicular to the surface are more sparsely distributed. The extensive porosity weakened the grain boundaries and thus caused the extraction of entire grains from the microstructure during metallographic specimen preparation.

Finally, the single-crystal UN with a density of 100 percent theoretical is shown in the pre-irradiated condition in Fig. 12. The pre-irradiated microstructure reveals no voids or porosity, no grain or sub-grain boundaries, and no cracks or fissures. The post-irradiation microstructure shown in Fig. 13 reveals no significant change from that of the pre-irradiated UN. No porosity is seen in the bulk crystal at magnifications above 400X. Some very fine porosity of a spherical nature, approximately 0.1 to 0.5 μ in diameter, has collected on sub-grain boundaries extending into the crystal from the external surface. No other

changes in the microstructure were observed.

Figure 14 shows the microstructure of the W-25.5Re cladding before irradiation. This material was single phase and cold worked with heat-affected zones near the welds at the bottom and top of the cladding. Figures 15 through 17 show typical microstructures of the cladding after irradiation. The changes include creation of a stable microstructure of equiaxed grains through recrystallization and grain growth with the average grain diameter of 30 μ . A second phase (sigma) was precipitated from solution and occurred in the grain boundaries, especially at the triple points, indicating that the composition of the cladding was richer in rhenium than the amount that could be held in solution at the temperature of irradiation. There was no indication of any significant chemical reaction between the fuel and the cladding during this irradiation.

Electron microprobe examination revealed that no detectable uranium was present in the cladding and confirmed the presence of a second phase (sigma) having a higher rhenium concentration than the primary tungsten-rhenium solid solution.

DIMENSIONAL CHANGES

Neutron radiographs made before irradiation and at the conclusion of each cycle are shown in Fig. 18. Diameters of the clad specimens were measured from the neutron radiographs at the top, middle and bottom of the cladding as well as lengths of the fuel and cladding. Measurements were made at BMI by use of an x-ray film reader with a micrometer-driven cross hair. These readings are

shown in Table 4. The radiographs were re-measured at LRL by use of a two-coordinate comparator at 10X magnification and these measurements are given in Table 5. When scanning these measurements from beginning to final irradiation cycle it becomes evident that both the

measurements made by BMI and LRL have sufficient random error associated with them to make the progressive swelling measurements uninterpretable. The presence of large amounts of molybdenum, tungsten, and rhenium in the capsule contributed greatly to the poor resolution of

(a) As-polished, longitudinal section, 50X mag.

(b) As-polished, cross-section, 50X mag.

(c) Stain-etched, 200X mag.

(d) As-polished, 400X mag.

Fig. 13. Specimens 2-A and 3-B, single-crystal UN. The post-irradiation microstructure shows no change in optically observed microstructure other than a collection of small spherical porosity along the subgrain boundaries, which rarely occurred.

Fig. 14. W-25.5Re cladding before irradiation showing worked structure, chemically etched. (200X mag.)

Fig. 15. W-25.5Re cladding after irradiation, showing precipitation of sigma phase. As-polished. (200X mag.)

Fig. 16. W-25.5Re cladding after irradiation, showing sigma phase precipitation at triple-points in equiaxed grains of annealed microstructure. Chemically etched. (200X mag.)

Fig. 17. W-25.5Re cladding after irradiation, showing precipitation of sigma phase at grain boundaries and within the grains. Cathodically etched. (400X mag.)

the neutron radiographs. Another contribution to the error was the intensity variation within one film and from one film to another. For these reasons, and because the actual changes

in dimensions during complete irradiation were small, it appears that the dimensional changes due to fuel swelling were no greater than the error limits of the method.

Fig. 18. Radiographs of capsule LRL-1.

Table 4. BMI measurements of LRL-1 specimens from neutron radiographs. Top, middle and bottom (T, M, B) cladding diameters and fuel lengths (L) are shown in inches.

Specimen ^a		Pre-irrad ^b	Pre-irrad	After Cycles No.							
				1	2	3	4	5	6	7	8
1-C	T	0.2417 (17)	0.2400	0.2399	0.2398	0.2420	0.2409	0.2423	0.2421	0.2407	0.2398
	M	0.2416 (13)	0.2425	0.2425	0.2399	0.2441	0.2392	0.2414	0.2448	0.2462	0.2447
	B	0.2420 (10)	0.2395	0.2428	0.2388	0.2412	0.2444	0.2406	0.2413	0.2406	0.2389
	L	0.7358	0.7427	0.7446	0.7375	0.7421	0.7402	0.7409	0.7364	0.7350	0.7345
2-A	T	0.2413 (13)	0.2398	0.2403	0.2375	0.2401	0.2454	0.2406	0.2424	0.2404	0.2407
	M	0.2414 (13)	0.2395	0.2386	0.2362	0.2422	0.2453	0.2399	0.2442	0.2456	0.2416
	B	0.2415 (20)	0.2371	0.2387	0.2360	0.2397	0.2443	0.2409	0.2414	0.2444	0.2429
	L	0.7158	0.7175	0.7260	0.7246	0.7265	0.7194	0.7228	0.7235	0.7197	0.7202
3-B	T	0.2410 (13)	0.2403	0.2373	0.2391	0.2388	0.2445	0.2402	0.2408	0.2415	0.2406
	M	0.2410 (12)	0.2406	0.2378	0.2410	0.2408	0.2450	0.2410	0.2423	0.2412	0.2401
	B	0.2410 (10)	0.2385	0.2380	0.2394	0.2402	0.2432	0.2405	0.2415	0.2404	0.2390
	L	0.7043	0.7107	0.7237	0.7165	0.7133	0.7134	0.7112	0.7127	0.7124	0.7155
4-D	T	0.2415 (17)	0.2387	0.2393	0.2300	0.2398	0.2453	0.2392	0.2390	0.2423	0.2407
	M	0.2416 (15)	0.2378	0.2404	0.2386	0.2410	0.2427	0.2390	0.2392	0.2421	0.2448
	B	0.2414 (12)	0.2407	0.2417	0.2402	0.2424	0.2428	0.2391	0.2412	0.2416	0.2394
	L	0.7465	0.7445	0.7494	0.7432	0.7510	0.7396	0.7403	0.7337	0.7415	0.7450
5-E	T	0.2423 (16)	0.2400	0.2387	0.2399	0.2400	0.2424	0.2402	0.2399	0.2410	0.2395
	M	0.2413 (14)	0.2403	0.2380	0.2385	0.2401	0.2443	0.2412	0.2420	0.2455	0.2471
	B	0.2412 (12)	0.2413	0.2412	0.2405	0.2414	0.2444	0.2410	0.2390	0.2416	0.2403
	L	0.7895	0.7862	0.7831	0.8960	0.7861	0.7862	0.7880	0.7951	0.7733	0.7897
6-G	T	0.2413 (12)	0.2378	0.2387	0.2392	0.2406	0.2466	0.2389	0.2437	0.2432	0.2408
	M	0.2413 (11)	0.2407	0.2380	0.2386	0.2412	0.2470	0.2482	0.2486	0.2472	0.2496
	B	0.2410 (10)	0.2400	0.2412	0.2377	0.2394	0.2456	0.2390	0.2420	0.2431	0.2427
	L	0.7505	0.7565	0.7831	0.7590	0.7607	0.7574	0.7578	0.7604	0.7549	0.7542
7-H	T	0.2412 (14)	0.2396	0.2383	0.2407	0.2411	0.2456	0.2390	0.2411	0.2419	0.2408
	M	0.2412 (16)	0.2411	0.2406	0.2401	0.2433	0.2453	0.2420	0.2422	0.2430	0.2433
	B	0.2415 (12)	0.2384	0.2406	0.2401	0.2423	0.2450	0.2395	0.2408	0.2414	0.2407
	L	0.7324	0.7118	0.7153	0.7055	0.7155	0.7115	0.7285	0.7183	0.7231	0.7221
8-I	T	0.2404 (05)	0.2408	0.2390	0.2378	0.2414	0.2432	0.2405	0.2411	0.2398	0.2404
	M	0.2405 (06)	0.2417	0.2374	0.2391	0.2428	0.2450	0.2390	0.2398	0.2444	0.2410
	B	0.2402 (01)	0.2417	0.2419	0.2379	0.2407	0.2467	0.2413	0.2392	0.2402	0.2409
	L	0.7510	0.7443	0.7328	0.7431	0.7558	0.7510	0.7464	0.7464	0.7517	0.7552

^aSpecimen order from top of capsule.

^bPre-encapsulation micrometer measurements with two digits in () showing last two figures of measurement at 90° orientation.

Table 6 shows comparative post- and pre-irradiation weights of the clad and unclad specimens. The minor milligram discrepancies in the total clad specimen weights are attributed to sintering of the thermocouple wires to the cladding and the subsequent loss or gain of particles of the cladding or when the thermocouple was removed from the specimen. The three longitudinal cladding cuts discussed earlier in the description of cladding re-

moval frequently resulted in cuts or scores in the fuel. These cuts explain the small amount of weight loss in each fuel specimen.

Outside and inside diameters and lengths of each specimen segment were made repeatedly as described earlier in this report. The fractional changes in lengths, diameters, and densities are given in Table 7. Actual data and calculations for a representative sample, No. 1-C, are given in Appendix A.

Table 5. LRL measurements of LRL-1 specimens from neutron radiographs. Top middle and bottom (T, M, B) cladding diameter, fuel length (FL), and cladding length (CL) are shown in inches.

Specimen ^a		Measured pre-irrad	Radiograph pre-irrad	After cycles No.								Measured post-irrad
				1	2	3	4	5	6	7	8	
1-C	T	0.242	0.243	0.239	0.236	0.243	0.246	0.240	0.245	0.246	0.244	0.242
	M	0.242	0.246	0.249	0.243	0.246	0.246	0.249	0.251	0.254	0.249	0.247
	B	0.242	0.244	0.246	0.238	0.244	0.252	0.238	0.244	0.249	0.244	0.244
	FL	0.736	0.737	0.741	0.732	—	0.736	0.739	0.743	0.732	0.738	0.737
	CL	1.020	1.017	1.019	1.017	—	1.019	1.020	1.023	1.019	1.022	1.021
2-A	T	0.241	0.240	0.243	0.242	0.249	0.249	0.237	0.248	0.246	0.245	0.242
	M	0.241	0.245	0.245	0.246	0.250	0.250	0.242	0.245	0.250	0.244	0.244
	B	0.242	0.242	0.245	0.246	0.245	0.246	0.239	0.244	0.242	0.244	0.244
	FL	0.716	0.718	0.720	0.712	0.719	0.715	0.721	0.722	0.725	0.719	0.715
	CL	1.020	1.009	1.010	1.012	1.008	1.013	1.011	1.012	1.016	1.008	1.020
3-B	T	0.241	0.241	0.242	0.243	0.245	0.241	0.243	0.252	0.243	0.245	0.243
	M	0.241	0.241	0.244	0.242	0.242	0.249	0.243	0.242	0.250	0.247	0.244
	B	0.241	0.247	0.246	0.240	0.242	0.248	0.242	0.247	—	0.247	0.241
	FL	0.704	0.709	0.715	0.711	0.709	0.707	0.707	0.718	0.715	0.713	0.726
	CL	1.025	1.021	1.020	1.020	1.022	1.022	1.020	1.021	1.022	1.022	1.027
4-D	T	0.242	0.239	0.245	0.244	0.245	0.242	0.246	0.249	0.243	0.245	0.242
	M	0.242	0.244	0.245	0.248	0.244	0.246	0.249	0.250	0.247	0.246	0.245
	B	0.241	0.245	0.245	0.241	0.243	0.245	0.245	0.247	0.242	0.248	0.243
	FL	0.747	0.741	0.734	0.744	0.743	—	0.748	—	—	—	0.749
	CL	1.028	1.021	1.022	1.024	1.024	1.019	1.024	1.022	1.023	1.021	1.029
5-E	T	0.242	0.241	0.245	0.245	0.245	0.242	0.241	0.251	0.247	0.245	0.242
	M	0.242	0.243	0.244	0.241	0.244	0.248	0.245	0.250	0.244	0.249	0.246
	B	0.241	0.245	0.242	0.243	0.242	0.247	0.240	0.244	0.250	0.249	0.242
	FL	0.790	0.778	0.790	0.778	0.781	0.778	0.787	0.786	0.778	0.782	0.792
	CL	1.036	1.027	1.029	1.025	1.029	1.028	1.028	1.028	1.033	1.029	1.033
6-G	T	0.241	0.241	0.241	0.244	0.245	0.247	0.242	0.243	0.249	0.244	0.242
	M	0.241	0.243	0.243	0.247	0.243	0.250	0.251	0.251	0.249	0.256	0.254
	B	0.241	0.240	0.243	0.243	0.244	0.248	0.244	—	0.247	0.250	0.244
	FL	0.751	0.749	0.758	0.754	0.755	0.751	0.761	0.763	0.758	0.755	0.751
	CL	1.021	1.016	1.022	1.019	1.020	1.017	1.019	1.022	1.020	1.017	1.022
7-H	T	0.241	0.239	0.245	0.238	0.243	0.245	0.246	0.238	0.240	0.245	0.242
	M	0.241	0.245	0.247	0.243	0.243	0.250	0.245	0.243	0.248	0.250	0.248
	B	0.241	0.243	0.244	0.243	0.244	0.253	0.246	0.242	0.243	0.247	0.243
	FL	0.732	0.707	0.709	0.710	0.716	0.712	0.720	0.722	0.721	0.713	0.738
	CL	1.032	1.027	1.027	1.028	1.028	1.035	1.034	1.030	1.035	1.028	1.035
8-I	T	0.240	0.241	0.243	0.238	0.244	0.247	0.241	0.246	—	0.244	0.242
	M	0.240	0.240	0.243	0.241	0.242	0.249	0.248	0.246	—	0.246	0.244
	B	0.240	0.241	0.245	0.243	0.240	0.243	0.247	0.245	—	0.245	0.243
	FL	0.751	0.746	0.744	0.746	0.747	0.752	0.753	—	—	0.750	0.758
	CL	1.025	1.023	1.022	1.023	1.023	1.025	1.034	1.029	1.031	1.032	1.029

^aSpecimen order from top of capsule.

CHEMICAL ANALYSES

The results from chemical analyses for uranium, nitrogen, xenon, and krypton are presented in Table 8. Multiple uranium analyses were run for each specimen. No result below 92.0 percent or above 96.0 percent was accepted unless it could be duplicated. On a statistical basis,

the uranium results are not significantly different from the theoretical value for pure UN of 94.44 percent. The overall mean is 93.94 percent uranium. Uranium analysis was performed according to ASTM Method D-217, and all calculations were based upon an average atomic weight of 237.7 for uranium.

Table 6. Pre- and post-irradiation weights.

Specimen	Clad specimen weight, g		Fuel weight, g	
	Pre-irradiation	Post-irradiation	Pre-irradiation	Post-irradiation
1-C	10.298	10.293	3.948	3.934
2-A	9.803	9.807	4.237	4.098
3-B	9.794	9.798	4.105	4.029
4-D	8.953	8.955	3.247	3.097
5-E	9.137	9.137	3.427	3.406
6-G	9.693	9.694	4.034	3.980
7-H	9.428	9.430	3.736	3.686
8-I	9.462	9.479	3.842	3.833

Table 7. Post-irradiation dimensional changes capsule 1.

Specimen No.	Material	Median temp (°C)	% meas. vol increase	% theoreti ρ decrease	% diam increase	% length increase
1-C	F.G. high ρ	1307	4.8	4.7	2.0	0.1
2-A	Single crystal	1377	3.5	3.7	0.7	1.4
3-B	Single crystal	1397	2.7	4.0	0.4	1.7
4-D	F.G. low ρ	1357	3.6	3.0 ^a	0.7	0.3
5-E	F.G. low ρ	1377	4.6	3.1 ^a	0.9	0.4
6-G	F.G. high ρ	1417	12.0	11.4	3.1	0.1
7-H	C.G. high ρ	1367	5.4	7.6	2.0	0.6
8-I	C.G. high ρ	1387	5.2	5.8	1.5	1.6

^aBased upon dry density pre-irradiation measurements at BMI. All others based upon wet pre-irradiation density at BMI. Wet = CCl₄. F.G. = fine grain size; C.G. = coarse grain size; ρ = density.

The nitrogen and retained fission gas analyses were troubled by the long distance separating the final Toepler pump from an intermediate mercury diffusion pump within the hot cell. Many of the first analyses were lost due to this pumping problem while others were incomplete and nitrogen content could not be determined. Since very small quantities of material were available for analyses, few specimens were large enough to permit duplicate runs. This accounts for unre-

ported analyses in those cases where specimens were lost. When duplicate analyses were made, they were in close agreement. The 19 nitrogen determinations from mass spectrometer analyses that were made covered a range of 5.37 percent to 5.64 percent with a mean composition of 5.48 percent compared to 5.56 percent for the stoichiometric compound. These results are tabulated in Table 8.

Post-irradiation fission gas release data was obtained by BMI as described

Table 8. Chemical analyses of specimens from capsule LRL-1.

Specimen No.	1-C			2-A			3-B			4-D			
Sample No.	1-2	1-6	1-10	2-2	2-6	2-10	3-2	3-5	3-7	4-2	4-4	4-6	4-10
Uranium (%) ^b	93.3	94.2	93.7	92.7	93.7	93.4	93.5	94.0	93.8	94.3		94.0	94.3
(No. of determinations)	(2)	(3)	(2)	(3)	(4)	(3)	(2)	(4)	(3)	(2)		(2)	(2)
Nitrogen (%) ^c	5.45		5.48	5.42	5.37		5.54	5.54		5.40	5.40		5.49
Xe (tot) (ppm) ^c	7.0	7.1 ^a	7.3	7.8	7.8	7.6	7.8	S	6.9 ^a	5.0	8.1	S	8.3
Kr (tot) (ppm) ^c	0.8	1.0 ^a	1.0	1.4	0.8	1.0	1.0	A	1.0 ^a	1.6	1.2	A	1.0
Xe ¹³¹ (% tot)	10.5	11.5	11.6	11.8	11.3	11.6	11.5	M				M	
Xe ¹³² (% tot)	16.7	17.5	17.5	17.5	17.1	17.4	17.2	P	11.4	11.0	11.2	P	11.0
Xe ¹³⁴ (% tot)	31.7	31.2	31.1	31.0	31.4	31.3	31.3	L	17.4	17.1	17.0	L	17.1
Xe ¹³⁶ (% tot)	41.1	39.8	39.8	39.6	40.3	39.7	40.0	E	31.4	31.2	31.7	E	31.2
Kr ⁸³ (% tot)		15.7	15.3	15.7		17.3	15.7	L	39.8	40.7	40.1	L	40.7
Kr ⁸⁴ (% tot)	33.9	28.9	30.6	26.4	33.3	28.8	29.2	O	15.4	13.0	15.0	O	11.8
Kr ⁸⁵ (% tot)		55.4		9.9				S	29.5	27.5	26.5	S	29.0
Kr ⁸⁶ (% tot)	66.1		54.1	47.9	66.7	53.8	55.1	T		5.0	8.0	T	
									55.1	54.5	50.4		59.1

^aEstimate based upon partial gas sample.^bDetermined by solution analytical chemistry (controlled potential coulometry).^cDetermined by mass spectrometric analysis

earlier. Assuming equal gas release from each specimen and a total burnup of 2.3×10^{20} fissions/cc, it was calculated that 2.8 percent of the stable xenon isotopes and 2.2 percent of the stable krypton isotopes were released. The isotopic composition of these gases is given in Table 9.

Retained fission gas analyses were made on from one to three samples from each specimen by the bomb-combustion techniques described previously. The average analysis of 15 specimens was 7.3 ppm of xenon and 1.0 ppm of krypton to which an analytical error of ± 20 percent must be applied due to the small amount of the gas present. Therefore an attempt to make a material balance of retained and released fission gas vs calculated fission gas generated has failed due to an average yield in excess of 113 percent for the xenon and 123 percent for the krypton derived from retained fission gas analyses.

Fuel burnup was obtained from 24 samples radiochemically analyzed by gamma spectroscopy. Each specimen was analyzed for ⁹⁵Zr, ¹⁴¹Ce, ¹⁴⁴Ce, ¹³⁷Cs, ¹⁰³Ru and ¹⁰⁶Ru. The combined results of ⁹⁵Zr, ¹⁴¹Ce and ¹⁴⁴Ce were used in the final determination. Table 10 gives the results of the burnup analysis

Table 9. Isotopic composition of released fission gas.

Isotope	Percent
Xe-131	12.7
Xe-132	19.1
Xe-134	30.0
Xe-136	38.2
Total Xe:	100.0
Kr-83	13.3
Kr-84	26.5
Kr-85	12.0
Kr-86	48.2
Total Kr:	100.0

Table 8. (continued)

5-E			6-G					7-H				8-I			
5-2	5-7	5-11	6-2	6-4	6-6	6-7	6-9	7-2	7-5	7-8	7-10	8-2	8-4	8-7	8-11
94.8	94.2	93.7	92.0	94.2		94.5	94.8	93.6	94.3		94.3	94.7		94.1	94.4
(4)	(3)	(3)	(3)	(2)		(2)	(2)	(3)	(3)		(2)	(3)		(5)	(2)
S	5.60	5.47	S	S	5.44	5.64	S	S	S	5.52	S	S	5.52	5.47	S
A			A	A			A	A	A		A	A			A
M	6.0	7.1	M	M	7.0	7.9	M	M	M	7.8	M	M	7.5	7.2	M
P	0.6	1.0	P	P	1.0	0.6	P	P	P	1.0	P	P	0.8	1.0	P
L			L	L			L	L	L		L	L			L
E	10.3	11.1	E	E	11.4	11.3	E	E	E	11.5	E	E	11.4	11.5	E
	16.5	17.1			17.3	17.3				17.4			17.3	17.3	
L	31.7	31.3	L	L	31.1	31.3	L	L	L	31.2	L	L	31.2	31.2	L
O	41.4	40.5	O	O	40.2	40.1	O	O	O	39.8	O	O	40.0	40.1	O
S			S	S			S	S	S		S	S			S
T	6.2	11.3	T	T	14.9	28.0	T	T	T	15.1	T	T		16.5	T
	26.2	30.0			28.7					26.4			35.0	29.1	
										8.5					
	67.7	58.8			56.4	72.0				50.0			65.0	54.4	

Table 10. Burnup analyses. All analyses are growth corrected. ⁺f = fissions.

Spec. No.	Sample No.	⁸⁵ Kr			¹⁴¹ Ce			¹⁴⁴ Ce			3 Isotope average
		⁺ f/g × 10 ¹⁹	⁺ f/cc × 10 ²⁰	Specimen Ave. ⁺ f/cc × 10 ²⁰	⁺ f/g × 10 ¹⁹	⁺ f/cc × 10 ²⁰	Specimen Ave. ⁺ f/cc × 10 ²⁰	⁺ f/g × 10 ¹⁹	⁺ f/cc × 10 ²⁰	Specimen Ave. ⁺ f/cc × 10 ²⁰	
1-C	2	1.698	2.33		1.562	2.15		1.667	2.29		
	6	1.587	2.18		1.701	2.34		1.641	2.26		
	10	1.71	2.35	2.29	1.627	2.24	2.24	1.667	2.29	2.28	2.27
2-A	2	1.744	2.50		1.639	2.35		1.71	2.45		
	6	1.662	2.38	2.38	1.741	2.40	2.40	1.703	2.44	2.43	2.40
	10	1.572	2.25		1.641	2.35		1.68	2.41		
3-B	2	1.696	2.43		1.645	2.36		1.657	2.37		
	5	1.67	2.39	2.37	1.601	2.29	2.28	1.663	2.38	2.35	2.33
	7	1.598	2.29		1.534	2.20		1.599	2.29		
4-D	2	1.84	2.05		1.818	2.03		1.739	1.94		
	6	1.832	2.04	2.06	1.734	1.94	2.00	1.775	1.98	2.00	2.02
	10	1.879	2.10		1.83	2.04		1.870	2.09		
5-E	2	1.877	2.09		1.80	2.01		1.873	2.09		
	7	1.814	2.02	2.05	1.876	2.09	2.04	1.98	2.21	2.13	2.07
	11	1.337	2.05		1.805	2.01		1.800	2.01		
6-G	4	1.802	2.48		1.856	2.55		1.739	2.39		
	7	1.787	2.46	2.45	1.736	2.39	2.49	1.727	2.37	2.46	2.47
	9	1.757	2.42		1.836	2.52		1.902	2.62		
7-H	2	1.818	2.52		1.801	2.63		1.826	2.54		
	5	1.745	2.42	2.46	1.774	2.46	2.48	1.723	2.39	2.44	2.46
	10	1.752	2.43		1.699	2.36		1.716	2.38		
8-I	2	1.77	2.46		1.753	2.43		1.72	2.39		
	7	1.692	2.35	2.40	1.755	2.44	2.40	1.808	2.51	2.44	2.41
	11	1.731	2.40		1.675	2.33		1.753	2.43		

and the verification of the original estimate of total capsule burnup of 2.3×10^{20} fissions/cc.

Uranium isotopic ratios for all ir-

radiated samples were measured but are not included in the report because these analyses were not obtained for the pre-irradiated fuel.

THERMOCOUPLE CALIBRATION

A single thermocouple calibration point was measured at 1100°C, as described earlier. Additional points were not measured because the irradiated thermocouple failed when it was inserted into the furnace for the 1200°C calibration point. This failure was due to the loss of the inert furnace atmosphere and the rapid oxidation of the thermocouple during insertion. However, the one good calibration run indicates that the thermocouple was read-

ing within 15°C of the actual temperature, as shown in Table 11.

Table 11. Thermocouple calibration.

Thermocouple	Temperature ^a (°C)
Standard Pt vs Pt-10Rh	1104
Unirradiated W-5Re vs W-26Re	1107
Irradiated W-5Re vs W-26Re	1119

^aCold junction temperature 0°C.

Discussion

Paramount among the beginning goals of this investigation was the development of an understanding of the mechanism responsible for fuel swelling in a high-temperature irradiation environment. It was expected that whatever theory that evolved could be substantiated through experiment. At the conclusion of only one relatively low burnup irradiation, however, we are not in a position to generalize our results.

SWELLING OF UN

One of the major objectives of this irradiation was to compare variations in microstructure of the same basic fuel. Materials of high density (96 to 100 percent) were compared to those of low density (78 percent), and materials of high density were compared on a basis of grain size 30 μ , 105 μ and single crystal. It can be seen from Table 7 that the low-density, fine-grained UN showed less swelling than the high-density materials. This can reasonably be interpreted as accommodation of volume increase within

the self-contained voids of the fuel. The superiority of the low-density (78 percent theoretical) UN is quite clear when related to the fine-grain, high-density UN and the coarse-grain, high-density UN, but borderline when compared to the single crystal. Subsequently, it becomes obvious that the fewer the grain boundaries or the larger the grain size, the better the resistance to swelling exhibited in the high-density fuel. It is apparent in the photographs presented earlier in this report that porosity, presumably resulting from collection of fission gases, collects in the grain boundaries and is strikingly absent from the single-crystal microstructure.

Calculation reveals that a temperature drop of approximately 20°C was present between the centerline of the fuel and the cooler outside surface of each specimen. The temperature gradient within a specimen creates a driving force along the gradient, causing the fission gas to migrate in the direction of higher temperature, i. e., the centerline of the specimen. In the case of the polycrystalline uranium

nitride studied in this irradiation, the grain boundaries parallel to the cylindrical surface acted as barriers to the migration of fission gas and have become microscopically delineated by its collection and the concentrated formation of porosity in these grain boundaries.

Swelling was directly measured by comparing pre-irradiation dimensions to post-irradiation dimensions as described earlier. Of the many dimensional measurements made in the post-irradiation analysis, the lengths are subject to the greatest error because of indications that the original fuel segments, especially the single crystals, were not perfect right circular cylinders, i. e., both ends were not parallel. Therefore, post-irradiation lengths were measured on a basis of maximum length which, in the case of the shorter length three-segment crystals, could result in considerable error, as could the volume increases calculated from the lengths. The diametral increase is quite accurate and the volumetric increase was determined by immersion in mercury, where volumes can be measured accurately to 1 part in 3000.

The median temperature for each specimen was calculated as explained previously. From the information in Table 3, it can be seen that the duplicate pairs of fine-grain, low-density; coarse-grain, high-density; and single-crystal UN specimens were all irradiated within $\pm 20^\circ\text{C}$ of the median irradiation temperature for the capsule. For this reason, the diameter increase and volumetric swelling were comparable for both specimens of a particular microstructure. The fourth pair of duplicate specimens was of the fine-grain, high-density UN and

they were in the positions of minimum and maximum median irradiation temperature, 110°C apart. They show diametral increase of 2 and 3 percent respectively and volumetric increases of 4.5 and 11.0 percent. From this information it is apparent that temperature has a strong relation to swelling in the direction of increased swelling occurring at increased temperature.

As a result of a relatively constant reactor flux profile and nearly identical ^{235}U enrichment of the fuel specimens, the burnup for any one specimen was within 12 percent of the average measured burnup for all eight specimens. This variation is insignificant and cannot be related to any differences in the irradiated microstructures.

All of the eight claddings failed by cracking, including those where the diametral increase was negligible. Therefore it was concluded that the cladding offered no observable restraining effect on the fuel. Additional confirmation of this statement is rendered in the negligible length increases shown in Table 7. Essentially, no indication of a "tooth-paste" extrusion of the fuel was seen in the microstructural analysis or the length increase.

The dimensional changes and metallographic examination implied that fission gas retention might be considerably less in the single crystal than for the other microstructures due to the absence of porosity and grain boundaries in the irradiated specimens. However, as can be seen in Table 8, the fission gas retention in all samples was nearly identical, indicating that it was present on a sub-microscopic level within the structure of

the single crystal, and that swelling was possibly more closely related to the size of the porosity.

The UN fuel was chemically stable during the irradiation, as electron microprobe analysis gave no indication of free uranium or U_2N_3 in the fuel. No reaction between fuel and cladding was observed, suggesting that W-25Re cladding or the W-Re family of materials is acceptable from the standpoint of chemical stability and fuel-cladding compatibility.

COMPARISON OF RESULTS WITH PREVIOUS DATA

Uranium nitride irradiations have been made previously at Pratt and Whitney under the SNAP-50 program and have been described.²⁴ Oak Ridge National Laboratory has continued much of the Pratt and Whitney work and is now generating its own program. The SNAP-50 capsules

were irradiated under quite different conditions pertaining to cladding, nitrogen cover gas, temperature of irradiation and total burnup, so that little information was provided with which to compare the results of capsule LRL-1. Oak Ridge has irradiated capsules at somewhat higher temperatures and higher burnups. These capsules are in either in-pile or in post-irradiation analysis²⁵ at this writing and direct comparison of data is impossible.

Battelle Memorial Institute, Columbus, has irradiated individual UN specimens in various capsules in their high-temperature fuels program and intends to expand these studies to include a study of the effect of grain size on swelling in UN.²⁶ Unfortunately, the temperature of interest in this program is again not the same as or near that of capsule LRL-1 but much higher (approximately 1700°C), allowing little direct comparison of results.

Conclusions

The experimental results have shown that uranium nitride, enriched 9.6 percent and irradiated at about 1400°C for 3558 hours to a burnup of 2.3×10^{20} fission/cc, swelled at varying rates, depending upon the original microstructure of the fuel.

Uranium nitride having an original theoretical density of 78 percent swelled less than similar material having an original theoretical density of 96 percent.

Uranium nitride having original theoretical densities of 96 to 100 percent swelled at different rates, depending upon the original grain size of the fuel. Fine-grain, 30 μ -diameter swelled more than coarse-grain, 105 μ -diameter, and single-

crystal uranium nitride swelled least of the three grain sizes.

No significant differences in swelling of solid single crystals and single crystals with a 0.100-in.-diam axial hole were observed.

The results also show that the swelling rate of fine-grained, 96-percent-dense uranium nitride is related to irradiation temperature: the swelling rate is greater at higher temperatures.

About 98 percent of the xenon and krypton generated in the fuel were retained in all microstructures. This indicates that fission gas retention was not dependent upon grain boundary effects under the conditions of this irradiation.

In this experiment there did not appear to be any relationship between amount of retained fission gas and fuel swelling.

Tungsten-25.5-percent-rhenium cladding was chemically compatible with uranium nitride under the conditions described above.

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Appendix A

Sample Calculations and Tabulation of Post- and Pre-Irradiation Measurements from Specimen 1-C

Original Dimensions - BMI

UN Fuel

<u>O. D.</u>	<u>I. D.</u>	<u>Length</u>	<u>Weight</u>	<u>Density % Theoret</u>
0.2010 in.	0.100 in.	0.7358 in.	3.948 g	96.43 wet (CCl ₄) 97.15 dry

Cladding

<u>O. D. (in.)</u>	<u>Length</u>	<u>Weight</u>
0.2417 top at hole	1.020 in.	10.2979 g
0.2416 mid point		
0.2420 bottom		
0.2417 90° top		
0.2413 90° mid point		
0.2410 90° bottom		

Post-Irradiation Dimensions - LRL

Cladding

<u>O. D. (1) (in.)</u>	<u>O. D. (2) (in.)</u>	<u>O. D. (3) (in.)</u>	<u>O. D. (average) (in.)</u>
0.2431 top at hole	0.2415	0.2416	0.2412
0.2434 mid point	0.2459	0.2462	0.2452
0.2438 bottom	0.2438	0.2440	0.2439
0.2418 90° top	0.2425	0.2420	0.2421
----- 90° mid point	0.2468	0.2498	0.2483
0.2446 90° bottom	0.2440	0.2446	0.2444

<u>Length (1)</u>	<u>Length (2)</u>	<u>Length (3)</u>	<u>Length (average)</u>
1.0213 in.	1.0213 in.	1.0213 in.	1.0213 in.

Weight

10.29268 g

<u>Fuel</u>		
<u>I. D. (1) (in.)</u>	<u>I. D. (2) (in.)</u>	<u>I. D. (average) (in.)</u>
0.0999 top	0.1000	0.1000
0.1020 mid top	0.1013	0.1016
0.1022 mid	0.1023	0.1023
0.1022 mid bottom	0.1019	0.1020
0.1007 bottom	0.1007	0.1007
0.1003 60° left top	0.1003	0.1003
0.1008 60° left mid top	0.1010	0.1009
0.1020 60° left mid	0.1018	0.1019
0.1010 60° left mid bottom	0.1013	0.1012
0.1008 60° left bottom	0.1011	0.1009
0.1003 60° right top	0.1003	0.1003
0.1013 60° right mid top	0.1006	0.1010
0.1022 60° right mid	0.1023	0.1022
0.1018 60° right mid bottom	0.1012	0.1015
0.1008 60° right bottom	0.1004	0.1006
<u>O. D. (1) (in.)</u>	<u>O. D. (2) (in.)</u>	<u>O. D. (average) (in.)</u>
0.2032 top	0.2028	0.2030
0.2039 mid top	0.2038	0.2039
0.2061 mid	0.2060	0.2060
0.2065 mid bottom	0.2059	0.2062
0.2045 bottom	0.2043	0.2044
0.2028 60° left top	0.2030	0.2029
0.2033 60° left mid top	0.2038	0.2036
0.2072 60° left mid	0.2056	0.2064
0.2054 60° left mid bottom	0.2050	0.2052
0.2048 60° left bottom	0.2037	0.2042
0.2025 60° right top	0.2025	0.2025
0.2047 60° right mid top	0.2046	0.2046
0.2095 60° right mid	0.2101	0.2098
0.2100 60° right mid bottom	0.2098	0.2099
0.2048 60° right bottom	0.2035	0.2041

<u>Length (1)</u>	<u>Length (2)</u>	<u>Length (3)</u>	<u>Length (average)</u>	
0.7369 in.	0.7367 in.	0.7367 in.	0.7368 in.	
<u>Volume (1)</u>	<u>Volume (2)</u>	<u>Volume (3)</u>	<u>Volume (4)</u>	<u>Volume (average)</u>
0.2995 cc	0.2983 cc	0.2990 cc	0.2990 cc	0.2990 cc
<u>Density (1)</u>	<u>Density (2)</u>	<u>Density (3)</u>	<u>Density (4)</u>	<u>Density (average)</u>
13.13 g/cc	13.19 g/cc	13.16 g/cc	13.16 g/cc	13.16 g/cc

$$\frac{\text{measured density}}{\text{theoretical density}} = \frac{13.19}{14.32} \times 100 = 91.9\%$$

Weight

3.93370 g

Pre-Irradiation

<u>I. D.</u>	<u>O. D.</u>	<u>Length</u>	<u>Volume</u>
0.100 in.	0.2010 in.	0.7358 in.	0.01757 in. ³

Post-Irradiation

<u>I. D. (average)</u>	<u>O. D. (average)</u>	<u>Length (average)</u>	<u>Volume</u>
0.1012 in.	0.2051 in.	0.7368 in.	0.01842 in. ³

$$\Delta V = \frac{\text{Post-Vol} - \text{Pre-Vol}}{\text{Pre-Vol}} \times 100 = 4.83\%$$

$$\Delta(\text{O.D.})_{\text{av}} = \frac{\text{Post-O.D.} - \text{Pre-O.D.}}{\text{Pre-O.D.}} \times 100 = 2.04\%$$

$$\Delta(\text{wall thickness})_{\text{av}} = \frac{\left(\frac{\text{Post-O.D.} - \text{Post-I.D.}}{2} \right) - \left(\frac{\text{Pre-O.D.} - \text{Pre-I.D.}}{2} \right)}{\left(\frac{\text{Pre-O.D.} - \text{Pre-I.D.}}{2} \right)} \times 100 = 2.87\%$$

$$\Delta(\text{length})_{\text{av}} = \frac{\text{Post-Length} - \text{Pre-Length}}{\text{Pre-Length}} \times 100 = 0.14\%$$

$$\Delta\rho = \frac{\text{Pre-Density} - \text{Post-Density}}{\text{Pre-Density}} \times 100 = 4.67\%$$

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