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PRELIMINARY EVALUATION OF T-111 CLAD UN FUEL SPECIMENS FROM 2500-HOUR 1040° C (1900° F) LITHIUM LOOP TEST

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ABSTRACT

Uranium mononitride (UN) cylinders clad with tungsten-lined T-111, a tantalum alloy, were exposed at 1040°C (1900°F) to flowing lithium in a pumped lithium loop. Two fuel element specimens were removed after 2500 hours and evaluated. The specimens were compatible with flowing lithium based on appearance, weight change, chemistry, and metallography. Room temperature ductility tests of the T-111 cladding resulted in brittle, intergranular fractures. Ductility could be restored by heating at 1315°C (2400°F) for one hour in vacuum.

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SUMMARY

Uranium mononitride (UN) fuel specimens were exposed at 1040°C (1900°F) to flowing lithium in a pumped loop. The UN was clad with tungsten-lined T-111, a tantalum alloy. Two of the fuel element specimens were removed from the loop after 2500 hours and evaluated on the basis of chemistry, metallography, weight and dimensional changes, and cladding ductility.

The two fuel element specimens appeared to be in excellent condition after the test. Visual examination revealed no evidence of corrosion. Only minor weight changes were observed and no dimensional changes were noted. Except for a slight reduction in the oxygen content of the T-111, very little change in chemistry was observed as a result of the lithium exposure. No microstructural changes were noted in the UN, but some changes were observed in the T-111. Bands of fine, unidentified precipitates could be seen in the T-111 after the 2500-hour exposure. These precipitates are thought to be due to simple thermal aging rather than the result of the lithium exposure.

Although the T-111 appeared in excellent condition after the lithium exposure, room temperature ductility tests of the T-111 cladding resulted in brittle, intergranular fractures. The cause of the brittle behavior is not fully understood at this time, but one possible explanation is hydrogen embrittlement during the post-test lithium cleaning operation. Room temperature ductility could be restored by heating the T-111 cladding for one hour at 1315°C (2400°F) in vacuum.

INTRODUCTION

A fast spectrum, liquid-metal cooled reactor for space power applications is being investigated at the NASA-Lewis Research Center (ref. 1). Operating temperature for the reactor is to be about 980°C (1800°F) with the possibility of 1040°C (1900°F) hot spots. Lithium is to be used as the coolant. The fuel elements for the proposed reactor consist of uranium mononitride (UN) cylinders clad with T-111, a tantalum-based alloy containing eight weight percent tungsten and two weight percent hafnium. The T-111 is separated from the UN fuel by a thin layer of tungsten to prevent contact and possible reactions of the fuel and cladding. The results of isothermal tests in lithium filled T-111 capsules showed that these materials are chemically compatible with lithium up to at least 1260°C (2300°F). No compatibility tests, however, have been conducted on the fuel elements under the more severe conditions present in a pumped-lithium loop.

The purpose of this study was to test simulated fuel element specimens in a 1040 °C (1900 °F) lithium loop for times up to 7500 hours. These specimens consisted of UN cylinders clad with tungsten-lined T-111. Fabrication and evaluation of the fuel element specimens were to be done at the NASA-Lewis Research Center. The exposure of the fuel element specimens in a 1040 °C (1900 °F) lithium loop was performed under NASA Contract NAS3-6475 by General Electric Co.-Nuclear Systems Programs. The loop was successfully operated for the planned 2500-hour initial test period. During a planned shutdown, two of the three fuel element specimens were removed and two new fuel element specimens were installed. Operation of the loop was then resumed for another 5000 hours. The results of the initial evaluation of the two fuel element specimens removed from the loop after 2500 hours are presented in this report. The fuel element specimens were evaluated primarily on the basis of chemistry, metallography, weight and dimensional change, and cladding ductility.

LITHIUM LOOP DESCRIPTION

The 1040 °C (1900 °F) lithium loop used in this study was designed, fabricated, and operated by General Electric Co.-Nuclear Systems Programs (GE-NSP) under NASA Contract NAS3-6474. A detailed description of the loop and the operating history for the initial 2500 hours is available in References 2 to 6.

A schematic of the lithium loop and a cross section of the fuel specimen test section are shown in Figure 1. In addition to the fuel specimen test section, a tensile test specimen assembly (containing advanced tantalum and tungsten alloys) and a corrosion specimen assembly (containing T-111 and advanced tantalum alloys) are included in the loop. The entire loop, with the exception of the Cb-1Zr surge tank (not shown in fig. 1), was fabricated from T-111. The liquid lithium is pumped through the loop by an electromagnetic pump. A coil of T-111, heated by electrical resistance, is used to heat the lithium to the 1040 °C (1900 °F) operating temperature. The loop is contained in a 61 centimeter (24-in.) diameter vacuum chamber capable of a vacuum during loop operation in the 1.3×10^{-6} to 1.3×10^{-7} newton/meter^2 (10^{-8} to 10^{-9} torr) range. Three T-111 clad UN specimens are arranged in series in the fuel specimen test section as shown in Figure 1. Flow velocity of lithium in this section is 1.5 meters (5 ft.) per second. The fuel element specimens are centered in the test section by two molybdenum alloy (TZM) spacers. These spacers are included in the loop because the neutron reflectors of the proposed reactor are to be fabricated from the TZM alloy. It is hoped that this loop test will provide some information on the compatibility of the TZM alloy in a flowing lithium-T-111 system.

The three fuel element specimens, designated LT-1, LT-2, and LT-3 (see fig. 1) were exposed to the flowing lithium for 2500 hours at 1900 °F (1040 °C). Following a scheduled shutdown, two fuel element specimens (LT-1 and LT-3) were removed. Two new fuel specimens (LT-5 and LT-6) were installed and operation of the loop was resumed for an additional 5000 hours. Evaluation of the fuel element specimens is being done at the NASA-Lewis Research Center. Evaluation of all other loop components is being done at GE-NSP.

FUEL ELEMENT SPECIMEN FABRICATION

The fuel element specimens used in the loop test were fabricated at the NASA-Lewis Research Center. These specimens consisted of 94 percent dense UN cylinders clad with T-111. Details of the specimen design are shown in Figure 2.

The UN cylinders were produced by the Oak Ridge National Laboratory (ORNL) using a die pressing and sintering technique. The fabrication methods are described in detail in Reference 7. Analyses of the UN cylinders are listed in Table I. Note that there is some disagreement between vendor analysis and check analysis. The problems associated with obtaining good agreement of UN analytical results have been discussed previously (refs. 7 and 8).

The T-111 tubing (19mm OD x 1mm wall) (0.750" OD x 0.040" wall) for the cladding and the T-111 rod for the end caps were obtained from commercial vendors. Prior to fuel element assembly, all T-111 components were cleaned with acid (two parts HF, four parts HNO_3, one part H_2SO_4, two parts H_2O), thoroughly rinsed, and air dried. The T-111 components were then heated to 1090°C (2000°F) for one hour in vacuum (2.7 x 10^{-3} newton/meter^2 (2 x 10^{-5} torr) or better) to remove any volatile absorbed impurities.

Contact and possible reactions of the UN fuel with the T-111 cladding was prevented by a thin (0.13mm) (0.005 in.) layer of tungsten between the fuel and cladding. The tungsten liners for fuel element specimens LT-1, LT-2, and LT-3 were fabricated by gas-pressure bonding multiple wraps of tungsten foil (0.025mm) (0.001 in.) around a molybdenum mandrel. The finished tungsten liner was in the form of a thin-wall, free standing tungsten tube having very accurate dimensions.

Tungsten discs (0.25mm (0.010 in.) thick) were used to prevent contact of the UN fuel with the end caps. The UN fuel cylinders were axially positioned by dished tungsten washers. The purpose of the gap between the fuel and the end caps was to minimize heat input to the fuel during welding of the end caps to the cladding. Another purpose of the gap was to allow for thermal expansion differences between the fuel and cladding.

During assembly of the fuel element specimens, "white glove" handling techniques were used and every effort was made to prevent contamination. The female end cap of each specimen was welded to the cladding by electron beam welding. Then the assembly was annealed at 1315°C (2400°F) for one hour in vacuum (2.7 newton/meter^2 (2 x 10^{-5} torr) or better). The tungsten components and the UN fuel cylinders were positioned in the fuel element specimens, and the male end caps were electron beam welded in place. Finally, the assembled fuel element specimens were annealed at 1315°C (2400°F) for one hour in vacuum (2.7 x 10^{-3} newton/meter (2 x 10^{-5} torr) or better). The purpose of the post-weld anneal is to prevent grain boundary corrosion of the welds in a liquid metal environment. Following annealing, the fuel element specimens were checked for cracks and defects by a helium mass spectrometer leak detector and by visual examination.

POST TEST EVALUATION

General Observations

The two fuel element specimens (LT-1 and LT-3), removed from the lithium loop after 2500 hours at 1040°C (1900°F), appeared to be in excellent condition as can be seen in Figure 3. Visual examination revealed no discolorations or any other evidence of corrosion. No leaks or cracks were found in the specimens as determined by dye penetrant inspection and by a mass spectrometer leak detector. No dimensional changes were observed, and only minor weight changes were noted (see Table II). A slight weight loss on both specimens was measured at GE-NSP; whereas, a slight weight gain was observed when the specimens were weighed at NASA. The difference in the observed weights does not appear to be significant and is probably because of differences in the balances or weighing procedures. In addition, the maximum weight change observed (0.0033 gram) does not appear significant when compared to the overall weight of the fuel element specimen ($\sim$ 136 grams).

The male end cap was carefully cut from each fuel element specimen using a hacksaw. The two UN fuel cylinders in each specimen were removed easily and no evidence of any reaction between the tungsten liner and the UN fuel was observed. The tungsten liners, however, could not be removed from the T-111 claddings. These liners were loose prior to the exposure in the loop, but probably a slight amount of solid-state welding occurred between the liner and the cladding as a result of the long thermal exposure. All of the fuel element specimen components were examined, and they all appeared clean and bright (as shown in fig. 4).

Chemical Analysis

Samples of the T-111 end caps and T-111 cladding from specimens LT-1 and LT-3 were analyzed for interstitials and major constituents after 2500 hours in the lithium loop. The results are compared in Table III to similar material prior to testing. Except for a reduction in the oxygen content of the T-111, very little change in chemistry was observed as a result of the lithium exposure. The reduction in oxygen content is typical of metals exposed to liquid lithium. The variations in the tungsten and the hafnium contents are probably within the range of inhomogeneity and analytical inaccuracies.

Because of the problems in obtaining good analytical agreement on UN, no attempt was made at this time to analyze the UN from the fuel element specimens exposed to lithium for 2500 hours. The UN from all the specimens, along with representative samples of untested UN, will be analyzed at the same time upon completion of the entire (7500 hours) lithium loop test.

Metallography

The lithium-exposed fuel element specimens were examined metallographically and compared to an untested specimen. Figure 5 shows an end-cap weld area of specimens LT-1 and LT-3 and of the untested sample. The only noticeable change is that dark bands can be seen in the LT-1 and LT-3 specimens. At higher magnification (fig. 6), these bands appear as fine precipitates. These precipitates have not been identified, but they are probably the result of localized areas of inhomogeneity in the T-111. In addition to the bands of precipitates, some grain boundary precipitates can be seen in the T-111 of the tested samples (see fig. 6). Both types of observed precipitates are probably the result of thermal aging and not the result of the lithium exposure.

Examination of the T-111 cladding from the lithium-exposed specimens showed no evidence of lithium corrosion and no contamination of the T-111 by either the UN fuel or the tungsten liner. As can be seen in Figure 7, the T-111 cladding appeared in good condition after the lithium exposure except for the precipitates described previously.

A metallographic comparison (fig. 8) of the UN before and after testing showed no change as a result of the lithium exposure of the fuel element specimens. The 2500-hour test had no apparent effect on density, grain size, or overall appearance.

Ductility Tests

Ductility tests were conducted on the T-111 cladding from the lithium-exposed specimens and on the T-111 cladding from an untested specimen. Rings, approximately three millimeters (1/8 in.) wide, were cut from the specimens using a water-cooled silicon carbide abrasive saw. The cut surfaces were ground with 400-grit silicon carbide paper. The cladding ductility was determined by flattening the rings in a hydraulic compression machine at a ram speed of one inch (2.5cm) per minute. Rings from both lithium-exposed samples (LT-1 and LT-3) failed in a brittle manner. Conversely, the untested sample was very ductile and could be flattened completely with no evidence of any cracking. Examples of the ring ductility results are shown in Figure 9. The tungsten liner can be seen separating from the cladding of the rings cut from specimen LT-1 and LT-3. The intergranular nature of the brittle fractures can be seen in Figure 10.

In an attempt to find the cause of the brittle behavior of the lithium-exposed samples, scanning electron micrographs were made of the fractured surfaces. The intergranular fracture behavior is obvious in the fractographs shown in Figure 11. Small particles, as yet unidentified, can be seen on the grain boundaries, but there is no evidence of any continuous grain boundary film.

One possible explanation for the observed behavior is hydrogen embrittlement. Hydrogen could be produced by moisture reacting with trace amounts of lithium remaining on the samples after the post-test cleaning operation. Although the bulk analysis of the brittle samples indicated that the hydrogen content is less than one-half part per million, possibly the hydrogen content of the grain boundaries could be much higher. Additional evidence that the brittle behavior of the T-111 might be because of hydrogen is that annealing the embrittled samples for one hour at 2400°F (1315°C) in vacuum restored the room temperature ductility to the T-111. However, more work is necessary to establish the validity of the hydrogen embrittlement hypothesis.

SUMMARY OF RESULTS

Uranium mononitride (UN) fuel specimens clad with tungsten-lined T-111 have been exposed to lithium flowing at 1.5 meters (5 ft.) per second in a pumped loop at 1040°C (1900°F). Two fuel element specimens were removed from the loop after 2500 hours, and the initial evaluation of these two specimens has been completed. The major results of this evaluation are listed below.

1. The UN fuel specimens clad with tungsten-lined T-111 are compatible with flowing lithium for up to 2500 hours at 1040°C (1900°F) based on appearance, weight change, chemistry, and metallography.

2. The T-111 cladding from the fuel element specimens fractured in a brittle, intergranular manner when subjected to room temperature flattening tests. The cause of this brittle behavior is not known, but it might be because of hydrogen embrittlement from moisture reacting with trace amounts of lithium on the specimens. Heating the cladding for one hour at 1315°C (2400°F) in vacuum restored room temperature ductility.

3. Bands of fine precipitates were observed in the T-111 after exposure. The precipitates probably are due to thermal aging and not the result of the lithium exposure.

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TABLE I

CHEMICAL ANALYSES OF UN FUEL PELLETS USED IN FUEL ELEMENT SPECIMENS

Element	Vendor Analysis	Check Analysis
Uranium	94.56%	-
Nitrogen	5.31%	4.96% 4.97% 4.92%
Oxygen	1490ppm[a]	733ppm 913ppm 795ppm
Carbon	290ppm	203ppm 213ppm

(a) Parts per million, by weight.

TABLE II

WEIGHTS OF FUEL ELEMENT SPECIMENS BEFORE AND AFTER 2500-HOUR EXPOSURE TO FLOWING LITHIUM AT 1040°C (1900°F)

Fuel Specimen No.	Weight (Grams) Before Test	After Test	Change
LT-1[a]	135.9871	135.9901	+0.0030
LT-1[b]	135.9875	135.9853	-0.0022
LT-3[a]	135.5784	135.5817	+0.0033
LT-3[b]	135.5796	135.5769	-0.0027

(a) Weighed at NASA-Lewis Research Center
(b) Weighed at GE-NSP

TABLE III

CHEMICAL ANALYSIS OF FUEL ELEMENT CLADDINGS
AND END CAPS AFTER 2500-HOUR EXPOSURE
TO FLOWING LITHIUM AT 1040°C (1900°F)

	Parts Per Million (By Weight)				Weight Percent	
	C	O	N	H	W	Hf
T-111 Cladding						
Untested	61	72	22	1	7.70	2.22
LT-1	69	35	20	<0.5	7.92	2.09
LT-3	71	27	15	<0.5	7.63	2.20
T-111 End Caps						
Untested	24	68	12	1	7.51	2.19
LT-1	53	47	10	<0.5	7.88	2.14
LT-3	33	45	10	<0.5	7.55	2.02

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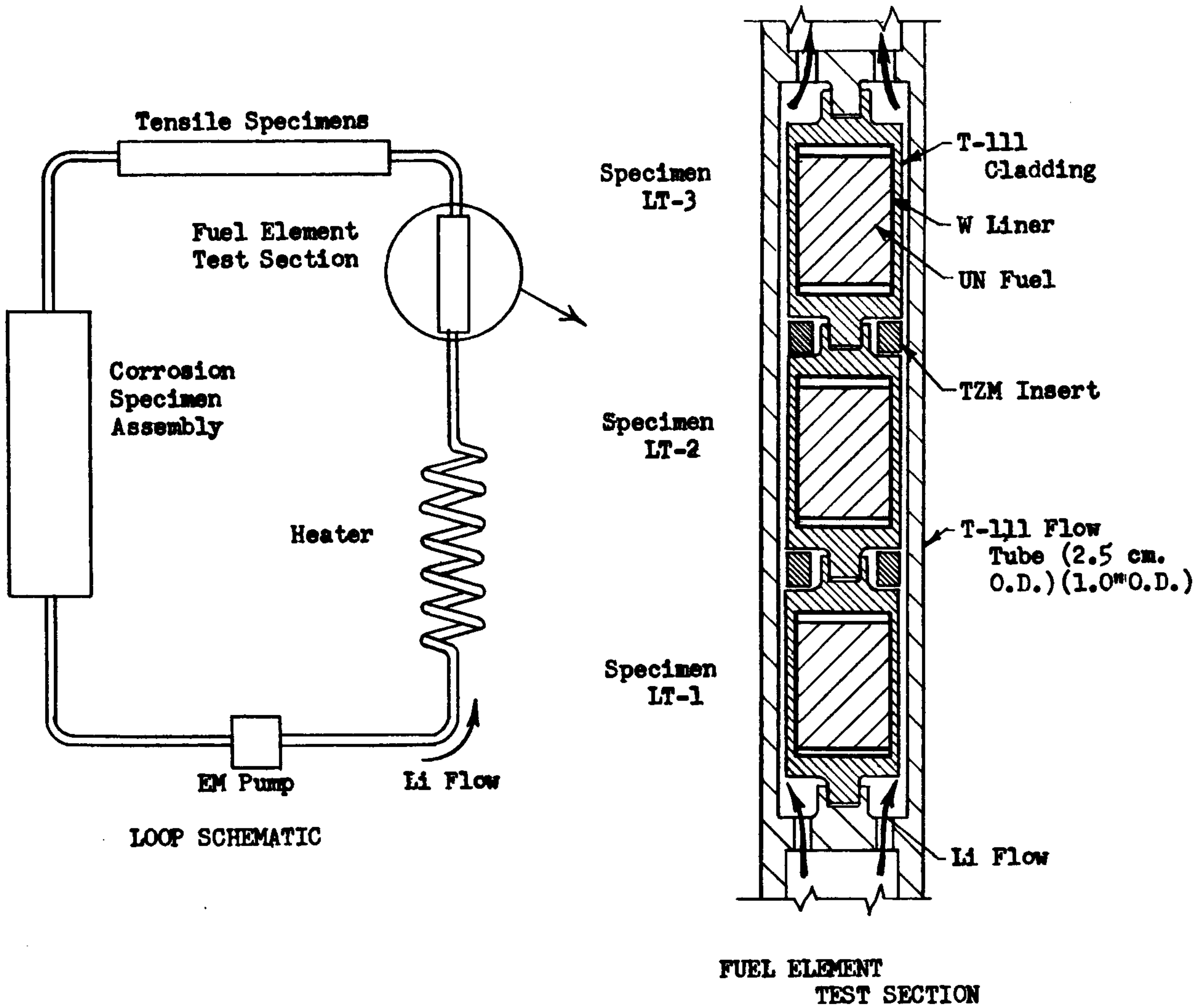

Figure 1 - Loop schematic and fuel element test section of 1040°C (1900°F) lithium loop.

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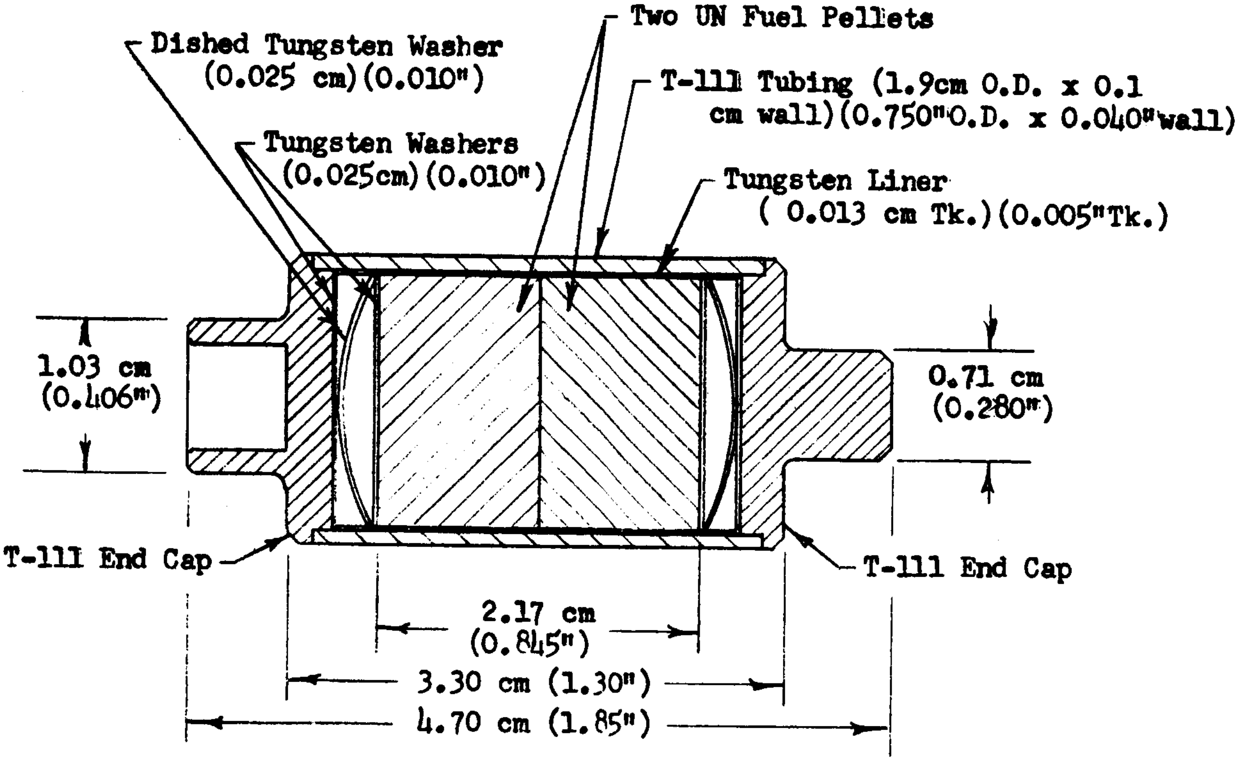

Figure 2 - Design of fuel element specimen used in 1040°C (1900°F) lithium loop test.

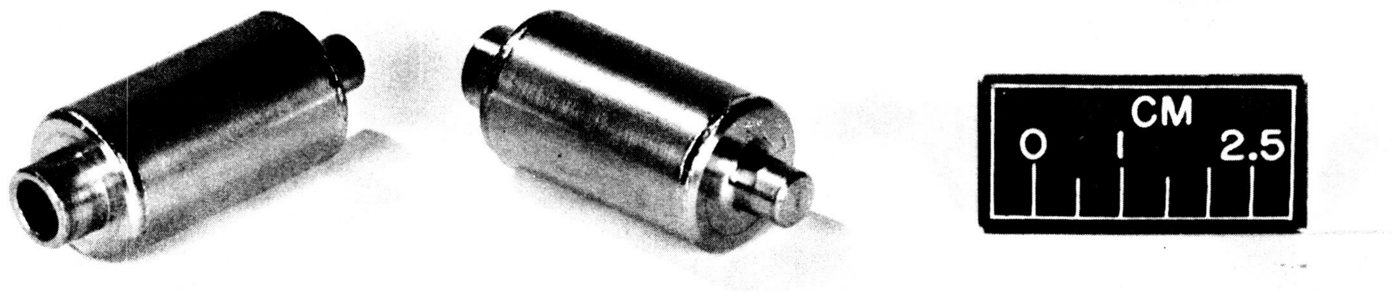

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Figure 3. - Fuel element specimens LT-1 and LT-3 after 2500 hours in 1040° C (1900° F) lithium loop.

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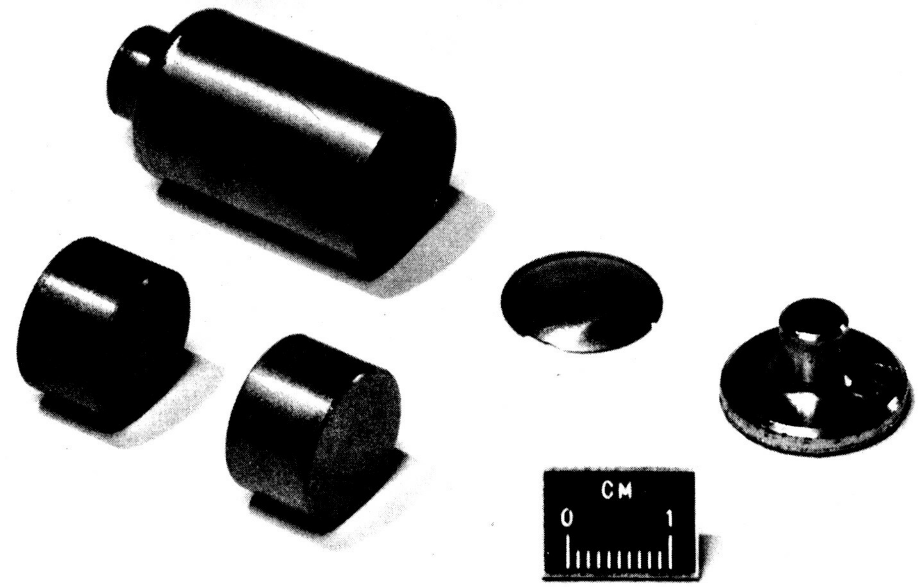

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Figure 4. - Fuel element specimen LT-1 disassembled after 2500 hours in 1040° C (1900° F) lithium loop.

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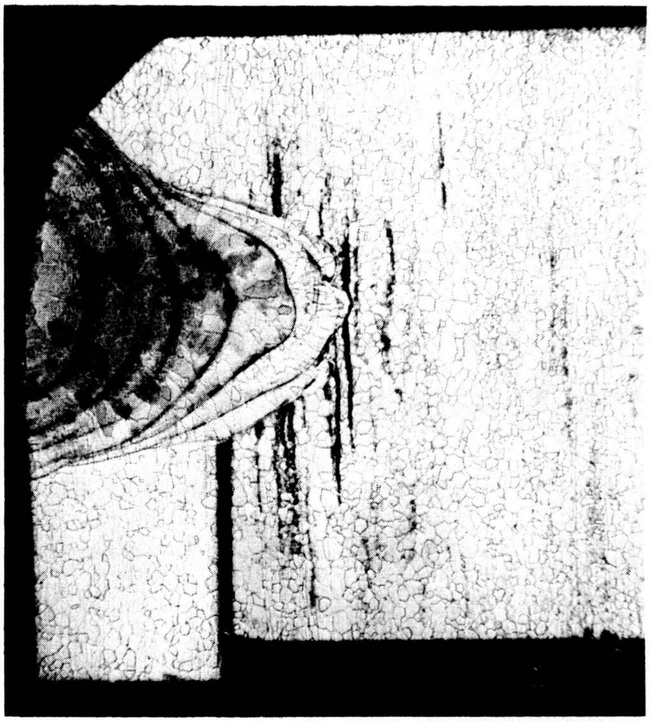
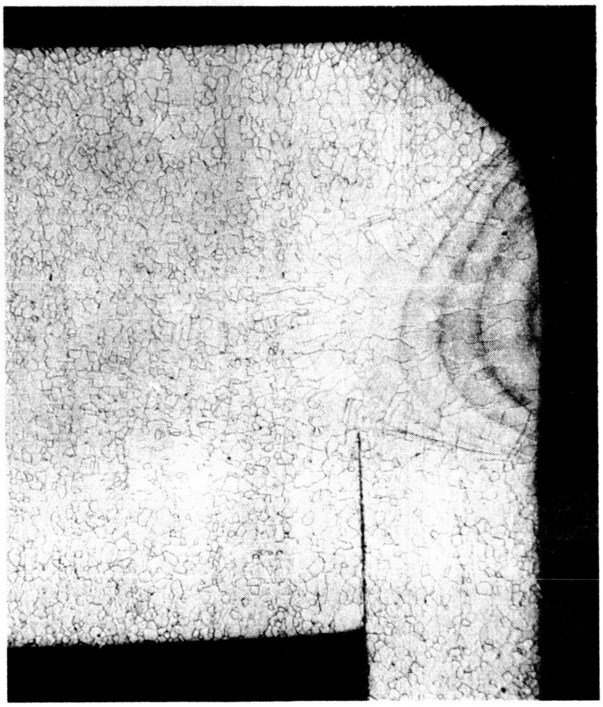


(a) Specimen LT-1.

(b) Specimen LT-3.



(c) Untested.

Figure 5. - End cap weld area of fuel element specimens in both the untested condition and after 2500 hours in 1040° C (1900° F) lithium loop. Magnification, X25. Etchant - 30 grams ammonium bifluoride, 50 ml nitric acid, 20 ml water.

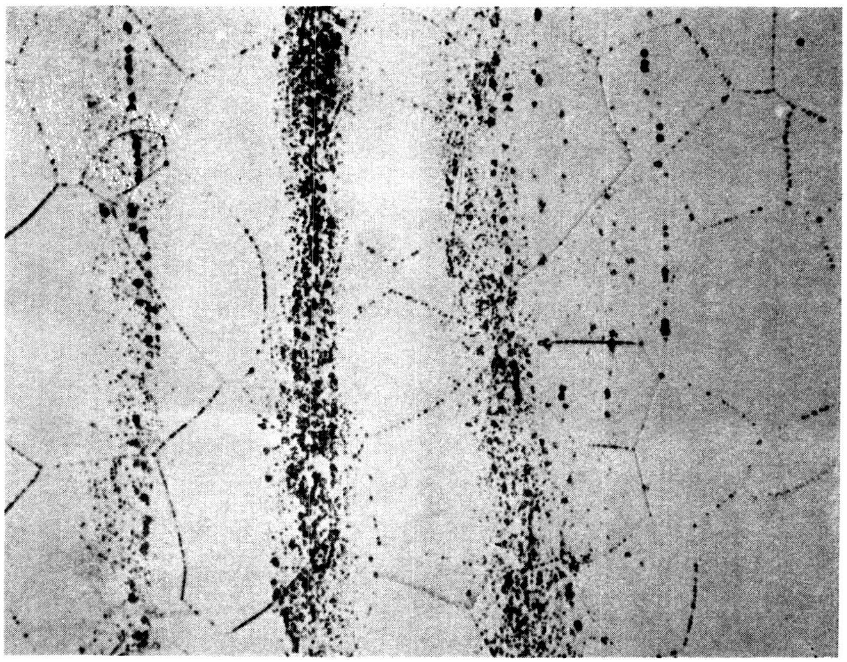


Figure 6. - End cap of fuel element specimen LT-3 after 2500 hours in 1040° C (1900° F) lithium loop. Note bands of precipitates and grain boundary precipitates. Magnification, X500. Etchant - 30 grams ammonium bifluoride, 50 ml nitric acid, 20 ml water.

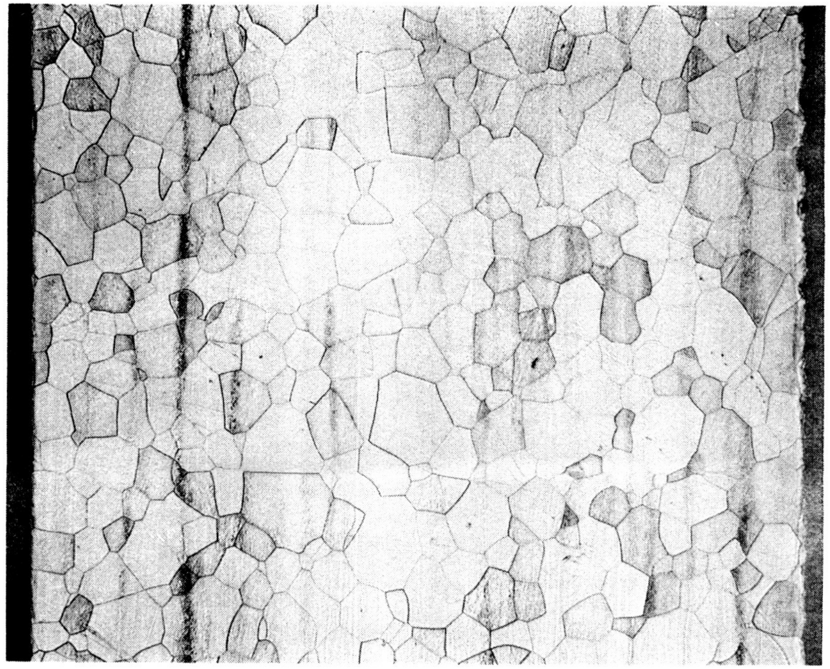


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Figure 7. - Longitudinal section of T-111 cladding from fuel element specimen LT-3 after 2500 hours in 1040° C (1900° F) lithium loop. Magnification, X100. Etchant - 30 grams ammonium bifluoride, 50 ml nitric acid, 20 ml water.

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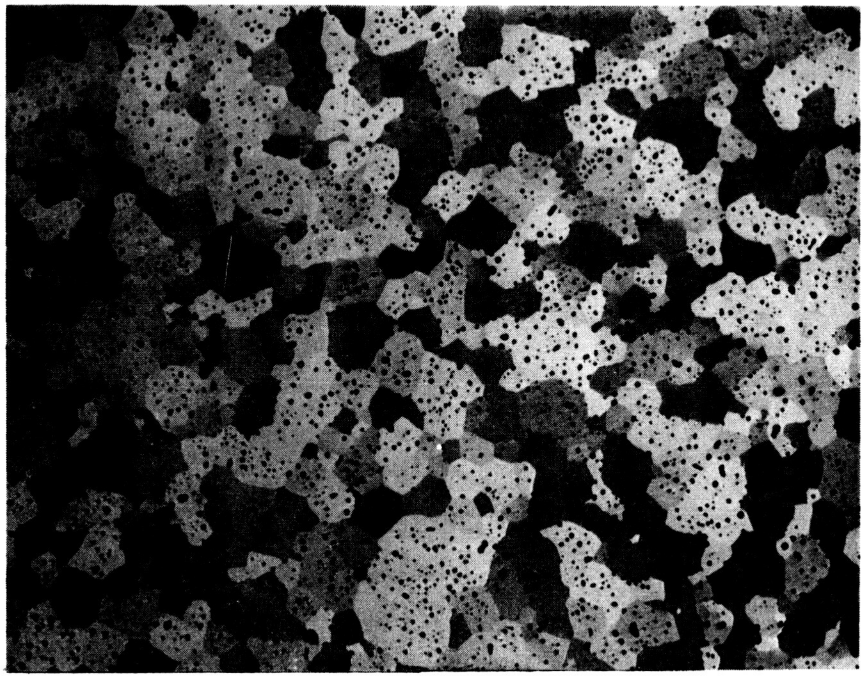
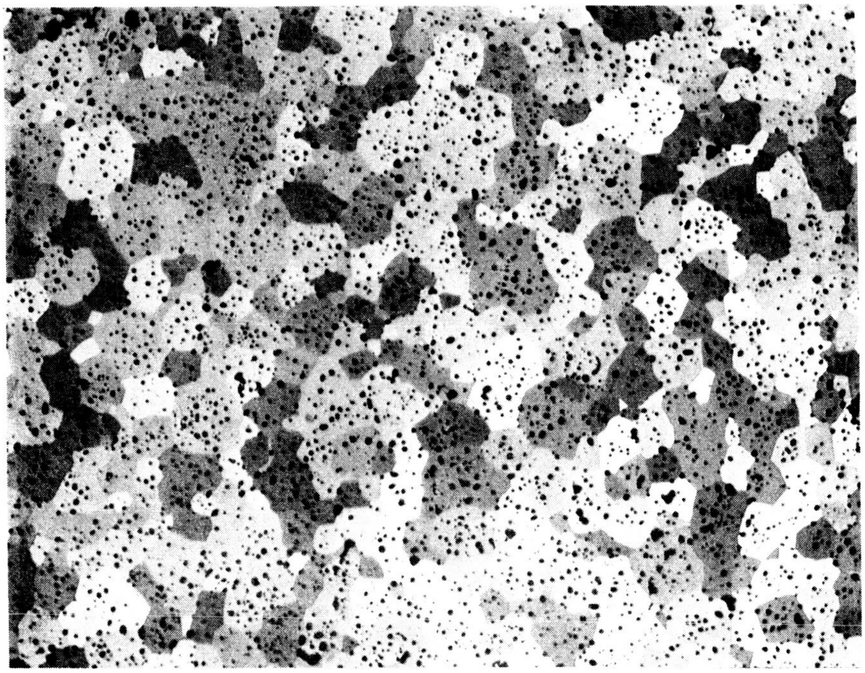
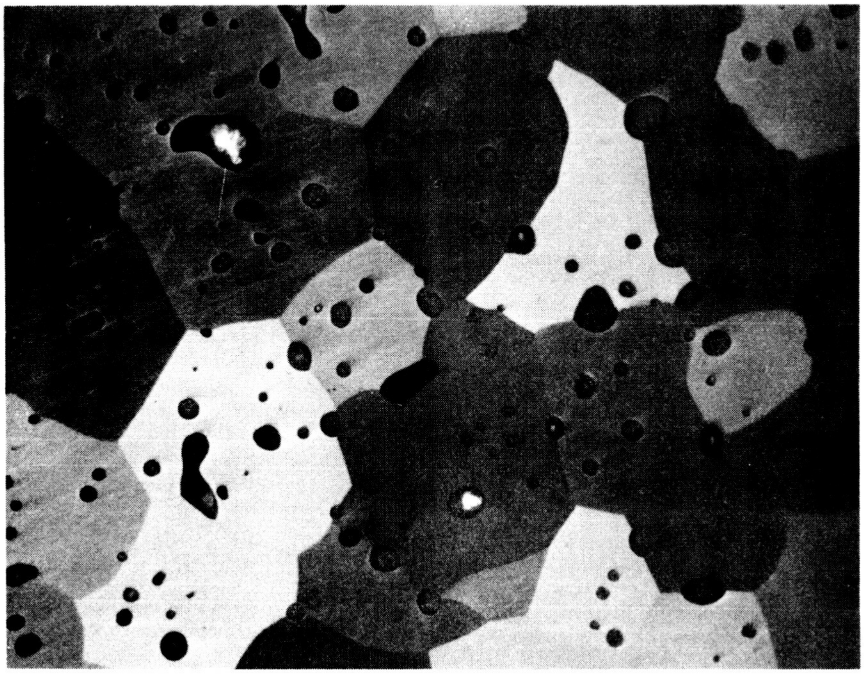


(a) Untested UN. X100.

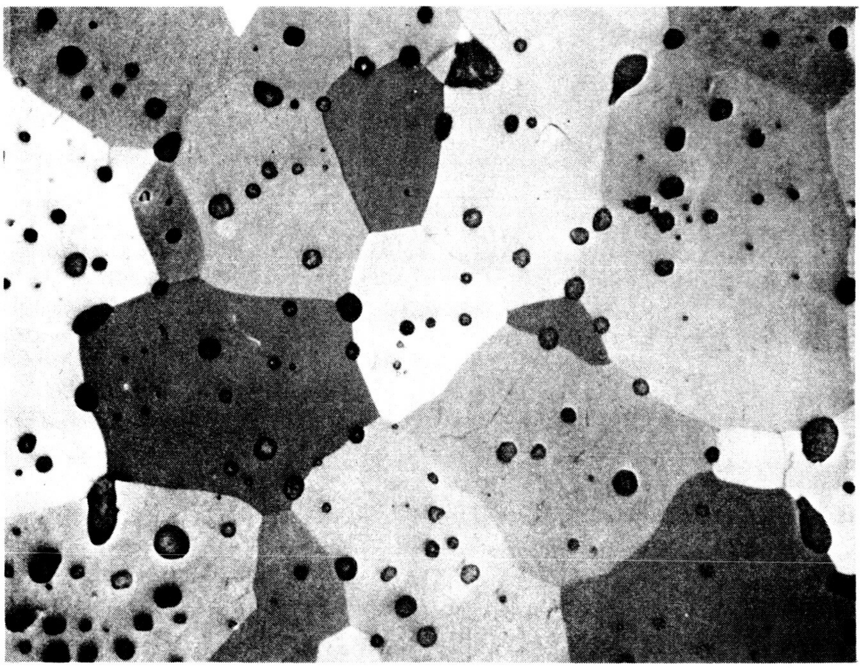


(b) UN from specimen LT-1 after 2500 hours in lithium loop. X100.

Figure 8. - Comparison of UN fuel in the untested condition and after 2500 hours in 1040° C (1900° F) lithium loop. Etchant - 60 ml lactic acid, 24 ml nitric acid, 2 ml hydrofluoric acid.



(c) Untested UN. X500.



(d) UN from specimen LT-1 after 2500 hours in lithium loop. X500.

Figure 8. - Concluded.

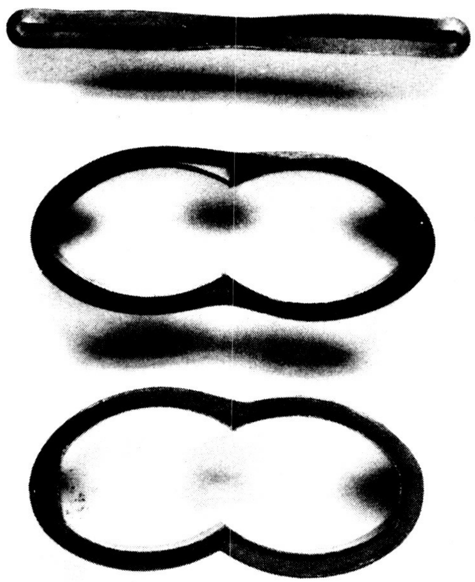


Figure 9. - Results of ring ductility tests on fuel element specimens in both the untested condition and after 2500 hours in 1040° C (1900° F) lithium loop. Magnification X2.

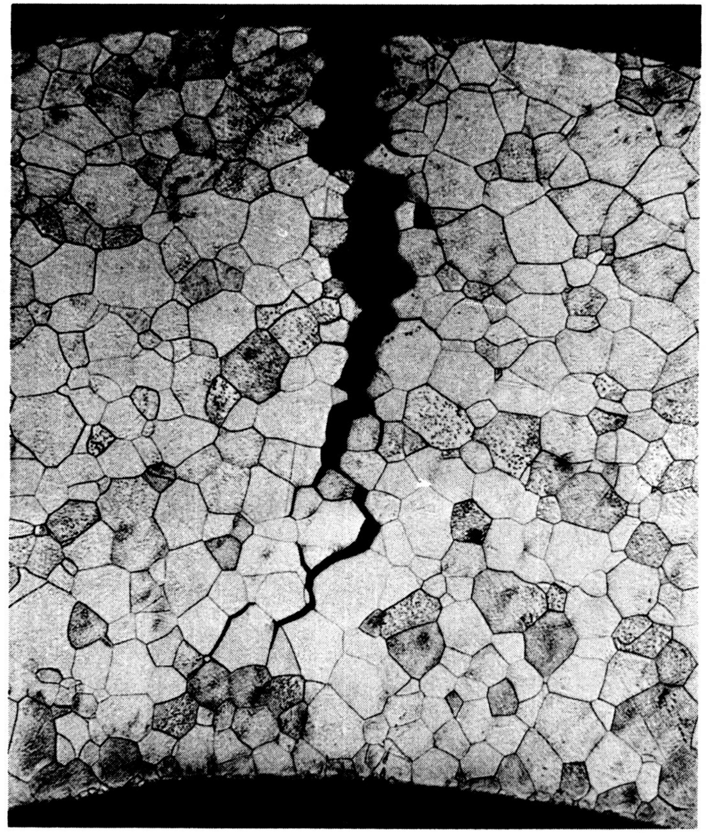


Figure 10. - Cross section of ring ductility test on T-111 cladding from fuel element specimen LT-3 after 2500 hours in 1040° C (1900° F) lithium loop. Magnification, X100. Etchant - 30 grams ammonium bifluoride, 50 ml nitric acid, 20 ml water.

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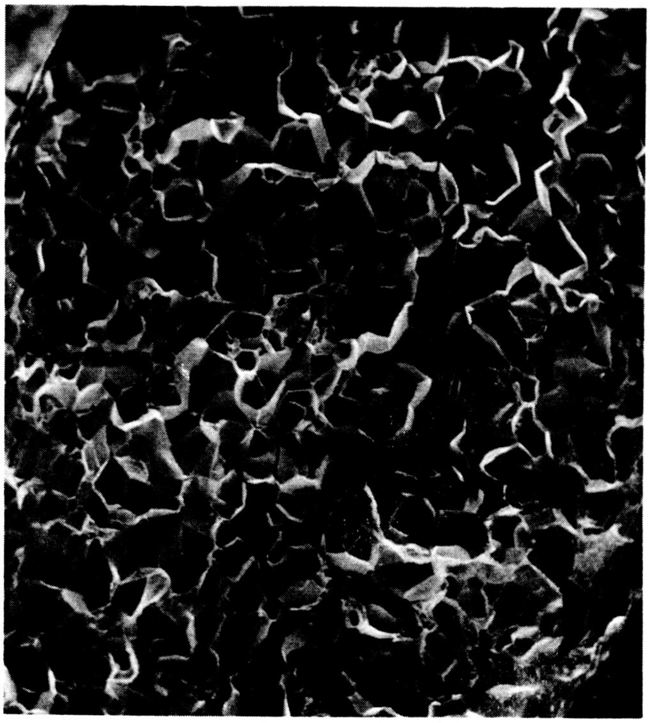
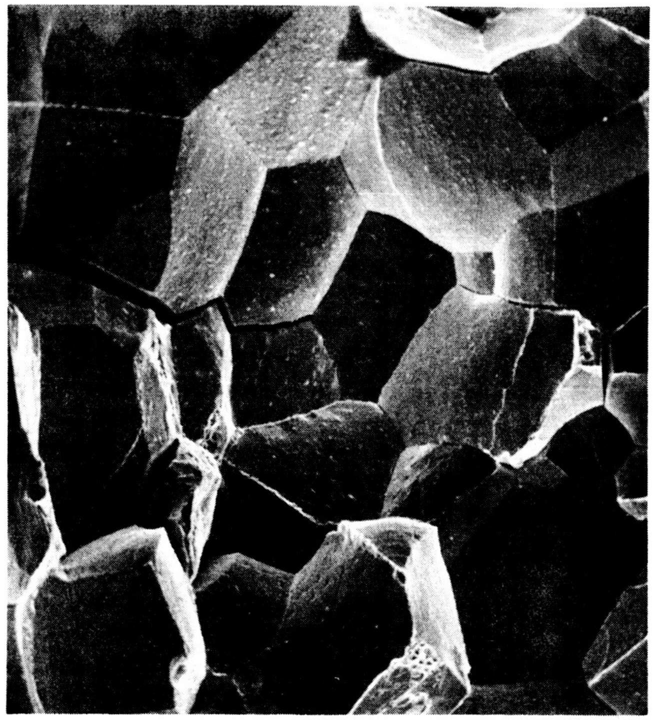
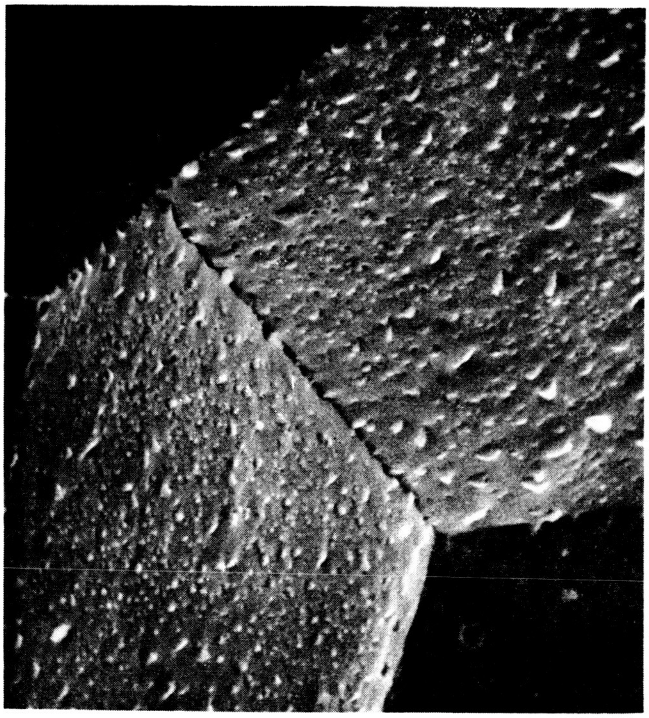


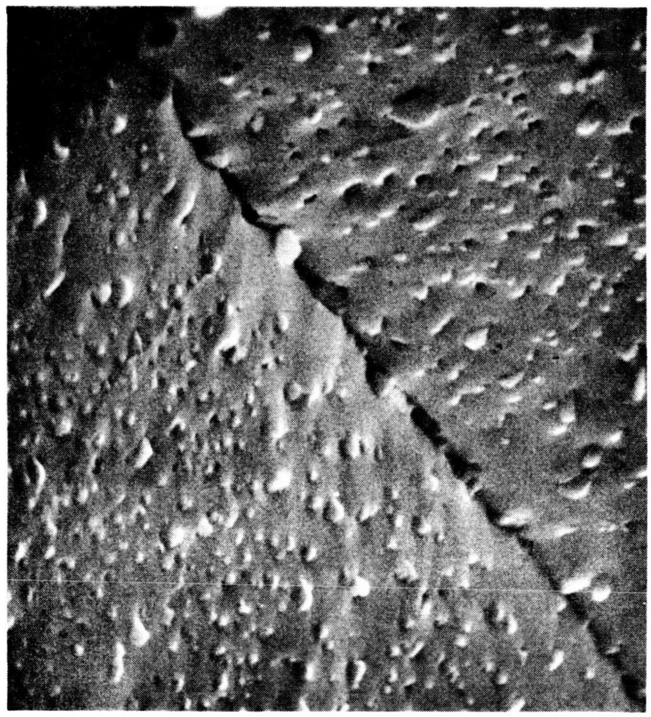
(a) X100.



(b) X500.



(c) X2500.



(d) X5000.

Figure 11. - Scanning electron micrographs of fractured surface of ring ductility test on T-111 cladding from fuel element specimen LT-1.