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EVALUATION OF TANTALUM-ALLOY-CLAD URANIUM MONONITRIDE FUEL SPECIMENS FROM 7500-HOUR, 1040° C PUMPED-LITHIUM-LOOP TEST

by Gordon K. Watson

Lewis Research Center

Cleveland, Ohio 44135

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16. Abstract

Simulated-nuclear-fuel-element specimens, consisting of uranium mononitride (UN) fuel cylinders clad with tungsten-lined T-111, were exposed for up to 7500 hr at 1040° C (1900° F) in a pumped-lithium loop. The lithium flow velocity was 1.5 m/sec (5 ft/sec) in the specimen test section. No evidence of any compatibility problems between the specimens and the flowing lithium was found based on appearance, weight change, chemistry, and metallography. Direct exposure of the UN to the lithium through a simulated cladding crack resulted in some erosion of the UN in the area of the defect. The T-111 cladding was ductile after lithium exposure, but it was sensitive to hydrogen embrittlement during post-test handling.

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SUMMARY

Simulated-nuclear-fuel-element specimens, consisting of uranium mononitride (UN) fuel cylinders clad with tungsten-lined T-111 (a tantalum alloy containing 8 percent tungsten and 2 percent hafnium), were exposed in a pumped-lithium loop operating at 1040° C (1900° F). The lithium flow velocity was 1.5 meters per second (5 ft/sec) in the specimen test section. Two fuel-element specimens were exposed for 2500 hours, two specimens were exposed for 5000 hours, and one specimen was exposed for 7500 hours. A cladding crack was simulated in one of the specimens exposed for 5000 hours by an axial slot machined through both the cladding and the tungsten liner. After exposure, the specimens were evaluated on the basis of weight and dimensional changes, chemistry, metallography, and cladding ductility.

All the fuel-element specimens appeared to be in excellent condition after the test. No evidence of any chemical compatibility problems between the specimens and the flowing lithium was found. Except for a slight reduction in the oxygen content of the T-111, very little change in chemistry was observed in the T-111 or the UN. No microstructural changes were noted in the UN, but bands of fine precipitates were seen in the T-111 after the lithium exposures. These precipitates were thought to be the result of thermal aging and not the result of the lithium exposure.

Direct exposure of the UN to the lithium through the simulated cladding crack resulted in some erosion of the UN and in some nitrogen contamination of the T-111 cladding in the area of the defect.

The T-111 cladding on the fuel-element specimens was ductile after the long-time lithium exposure. The thermal aging at 1040° C (1900° F), however, resulted in the T-111 becoming sensitive to hydrogen embrittlement during post-test handling and testing.

INTRODUCTION

A lithium-cooled, fast-spectrum, nuclear reactor was investigated at the NASA Lewis Research Center for use in advanced space power applications (ref. 1). The reference reactor was designed to operate at a power level of 2.2 megawatts (thermal) for up to 50 000 hours with a fuel-element cladding temperature of about 980° C (1800° F). There is a possibility, however, that local cladding hotspots could reach temperatures of about 1040° C (1900° F). The reactor concept is based upon the use of uranium mononitride (UN) as the fuel and a tantalum alloy, T-111 (tantalum - 8 wt. % tungsten - 2 wt. % hafnium), as the fuel cladding. The T-111 cladding would be lined with a thin layer of tungsten (ref. 2) to prevent contact and possible reactions between the fuel and cladding.

The chemical compatibility of the various proposed reactor materials has been verified primarily by out-of-pile, isothermal capsule tests (ref. 3). No compatibility problems were observed in these tests between T-111 and UN as long as the two materials were physically separated with a layer of tungsten. The tests also showed that lithium and UN were compatible providing the oxygen content of the UN was less than about 800 parts per million by weight. Although isothermal capsule tests are useful for materials screening studies, they do not simulate the temperature gradients and lithium flow rates found in the reactor. Thus, additional compatibility tests of the fuel-element materials are necessary under the more severe conditions present in a pumped-lithium loop.

The purpose of the work presented in this report was to test T-111 clad UN fuel-element specimens in a 1040° C (1900° F) pumped-lithium loop for up to 7500 hours. The loop was operated successfully for a total of 7500 hours with one scheduled shutdown after 2500 hours of operation. Two fuel-element specimens were exposed for 2500 hours, two specimens were exposed for 5000 hours, and one specimen was exposed for the entire 7500 hours. After exposure in the loop, the specimens were evaluated on the basis of weight and dimensional changes, chemistry, and metallography to determine the compatibility and corrosion behavior of the fuel-element specimens in flowing lithium. Preliminary evaluation of the two specimens exposed for 2500 hours is presented in reference 4. The results of the detailed evaluation of all the fuel-element specimens are summarized in this report. Fabrication and evaluation of the fuel-element specimens were done at the NASA Lewis Research Center. Exposure of the specimens in a 1040° C (1900° F) lithium loop was performed under contract.

LITHIUM-LOOP TEST DESCRIPTION

The 1040° C (1900° F) pumped-lithium loop used in this study was designed, fabri-

cated, and operated by the General Electric Company under NASA contract NAS 3-6474. A detailed description of the loop and the complete operating history for the entire 7500 hours are given in references 5 to 12. These reports are summarized in the final report on the corrosion loop program (ref. 13).

A schematic drawing of the lithium loop and a cross section of the fuel-element test section are shown in figure 1. Also included in the loop was a tensile-test-specimen assembly (containing advanced tantalum and tungsten alloys) and a corrosion-specimen assembly (containing T-111 and advanced tantalum alloys). The entire loop, with the exception of a columbium - 1-percent-zirconium surge tank (not shown in fig. 1) was fabricated from T-111. Lithium, at the rate of about 1.8 kilograms per minute (4.0 lb/min), was circulated through the loop by an electromagnetic (EM) pump. As the lithium passed through a resistance-heated coil of T-111 tubing, it was heated to 1040° C (1900° F). From there, the lithium flowed through the fuel-element test section at a flow velocity of 1.5 meters per second (5 ft/sec), through the heat-rejection part of the loop, where the lithium was cooled to about 1000° C (1830° F), and finally back to the pump. The loop was contained in a 61-centimeter- (24-in.-) diameter vacuum chamber that maintained a vacuum during loop operation of 1.3×10^{-6} newton per square meter (10^{-8} torr) or better.

The T-111 clad UN specimens were arranged in series in the fuel-element test section as shown in figure 1. Specimen position was maintained by two TZM spacers (molybdenum-0.5titanium-0.08zirconium-0.03carbon) and by the interlocking design of the specimens. The TZM spacers were included in the test to provide some information on the compatibility of TZM in a flowing lithium - T-111 system because some of the components in the proposed reactor were to be fabricated from TZM. Post-test evaluation of the TZM spacers is presented in reference 13. The lithium flow in this section of the loop was in the annulus between the fuel specimens and the containment tube, as can be seen in figure 1.

Three fuel-element specimens were exposed to the flowing lithium for the first 2500 hours of operation. Then, during a planned shutdown, two of these specimens were replaced with two new fuel specimens, and the loop was run for an additional 5000 hours. A cladding crack was simulated in one of the specimens exposed for 5000 hours. After corrosion testing, the fuel-element specimens were evaluated at the NASA Lewis Research Center.

FUEL-ELEMENT SPECIMEN FABRICATION

A total of seven fuel-element specimens (designated LT-1 to LT-7) were fabricated for this study. Two of the specimens (LT-4 and LT-7) were left untested as control

specimens. The other five specimens were tested in the loop: LT-1 and LT-3 were tested for 2500 hours, LT-5 and LT-6 were tested for 5000 hours, and LT-2 was tested for 7500 hours. A cladding crack was simulated in specimens LT-6 and LT-7 by an axial slot electrodischarge-machined through both the T-111 cladding and the tungsten liner.

Although the design of all the fuel-element specimens is essentially as shown in figure 2, they were fabricated in two different lots. Specimens LT-1 to LT-4 were fabricated at one time, and specimens LT-5 to LT-7 at another. The two lots of specimens were similar except for the quality of the UN, the method used to line the T-111, and the material for the dished washers.

Uranium Mononitride Fuel Cylinders

All the UN cylinders were about 94 percent of theoretical density and were produced by the Oak Ridge National Laboratory as described in detail in reference 14. The UN in the first set of specimens was produced by a die pressing and sintering technique. These fuel cylinders were relatively short because of the length-diameter-ratio limits associated with die pressing. Thus, two fuel cylinders were used in each of the first four fuel-element specimens. The oxygen content of this UN was about 1000 to 1500 parts per million by weight, which could cause compatibility problems if directly exposed to lithium. No problems were anticipated, however, because the UN was completely encapsulated in T-111 and was not directly exposed to the lithium. But for the fuel-element specimen with an intentional cladding defect through which lithium could contact the UN (LT-6), a much lower oxygen content was desired.

For the second set of fuel-element specimens (LT-5 to LT-7), the UN cylinders were fabricated by a subsequently developed fabrication process, which used an isostatic pressing and sintering technique. As indicated in reference 14, this improved fabrication method is superior to die pressing because the length of the UN cylinders is not limited and thus only one UN cylinder was needed for each fuel-element specimen. In addition, because binders or die lubricants are not needed in isostatic pressing, the oxygen content of these UN cylinders was quite low ($\sim$200 ppm by wt.).

The UN cylinders for both sets of specimens were thoroughly cleaned to remove any machining residue prior to final assembly. The cylinders were ultrasonically cleaned, first in acetone and then in methyl alcohol. After drying, any remaining volatile impurities were removed by heating the UN cylinders for 30 minutes at 540° C (1000° F) in a vacuum of 2.7×10^{-3} newton per square meter (2×10^{-5} torr) or better.

T-111 Cladding and End Caps

The claddings for the fuel-element specimens were fabricated from commercially produced T-111 tubing (1.9-cm o.d., 0.1-cm wall (0.750-in. o.d., 0.040-in. wall)). The tubing was inspected thoroughly by nondestructive methods before being cut into the lengths required for the fuel-element specimens. Any part of the tubing having defects deeper than 3 percent of the tube wall was rejected. The T-111 rod for the end caps was also obtained from a commercial vendor. Following nondestructive inspection, the end caps were machined from the T-111 rod. All the T-111 components were cleaned before specimen assembly (as described in ref. 15) and then heated to 1090° C (2000° F) for 1 hour in vacuum (2.7×10^{-3} N/m^2 (2×10^{-5} torr) or better) to remove any volatile adsorbed impurities.

Tungsten Liners

Contact between the fuel-element cladding and the UN fuel was prevented by lining the T-111 tubing with a thin (0.013-cm (0.005-in.)) layer of tungsten. The tungsten liners for the first set of specimens were in the form of thin-wall, free-standing tungsten tubes. These tubes were produced by isostatically hot-pressing multiple wraps of 0.0025-centimeter- (0.001-in.-) thick tungsten foil around an accurately machined cylindrical molybdenum mandrel. Pressing conditions were 2.1×10^{8} newton per square meter (3×10^{4} psi) and 1650° C (3000° F) for 3 hours. After pressing, the tungsten-wrapped mandrels were shaped on a centerless grinder for a slip-fit into the T-111 cladding and then cut to the proper length. Finally, the molybdenum mandrel was removed by dissolution in nitric acid, and a thin-wall, tungsten tube having very accurate dimensions was left. The tungsten tube was inserted into the T-111 cladding as a liner during final capsule assembly.

The free-standing tungsten liner method was not used for the second set of specimens, which included specimens having simulated defects, because the alinement of the defect slot between the cladding and the loose liner would be difficult to maintain. Instead, a differential-thermal-expansion liner method (ref. 16) was used. The tungsten liner, in the form of multiple layers of thin (0.0025-cm- (0.001-in.-) thick) foil, was wrapped around an alumina-coated steel mandrel and inserted into the T-111 tube. During high-temperature exposure in vacuum (1.3×10^{-3} N/m^2 (1×10^{-5} torr)), the steel mandrel expands more than the T-111 and forces the liner against the T-111. Under the conditions used in this study (1 hour at 1315° C (2400° F) and a 0.008-cm (0.003-in.) initial gap size), enough solid-state welding occurred between the liner and the T-111 to prevent any movement of the liner during subsequent operations. The tungsten liner

was recessed 0.23 centimeter (0.090 in.) from each end of the T-111 cladding to provide clearance for the end caps. A cladding crack was simulated in specimens LT-6 and LT-7 by electrodischarge-machining a small axial slot (0.008 cm wide and 0.64 cm long (0.003 in. wide and 0.25 in. long)) through both the T-111 cladding and the tungsten liner. After machining, the specimens were ultrasonically cleaned in acetone and methyl alcohol and then vacuum (1.3×10^{-3} N/m^2 (1×10^{-5} torr)) degassed at 1090° C (2000° F).

Specimen Assembly

Great care was taken to prevent contamination of the fuel-element specimens during final assembly. The components were handled only with nylon gloves after cleaning. Assembly of both sets of specimens was similar. The female end cap and the cladding of each specimen were electron-beam welded, and then the assemblies were annealed at 1315° C (2400° F) for 1 hour in vacuum (2.7×10^{-3} N/m^2 (2×10^{-5} torr) or better). After this anneal, the loose tungsten liners for the first set of specimens were inserted. For the second set of specimens, no separate liner operation was needed during assembly because the bonded liners were already in place.

As shown in figure 2, the UN fuel cylinders were positioned axially by dished washers sandwiched between flat, 0.025-centimeter- (0.010-in.-) thick tungsten disks. The main purpose of the flat tungsten disks on each side of the dished washers was to prevent contact of the UN fuel with the T-111 end caps. The gap between the UN fuel and the end caps served to minimize heat input to the UN during welding of the male end cap to the cladding. The gap also minimized the effects of thermal expansion differences between the fuel and the cladding. Tungsten sheet (0.025 cm (0.010 in.) thick) was used for the dished washers in the first set of specimens. Some of these relatively brittle washers cracked during shipment of the specimens to the contractor for testing. Although it was felt that the cracked washers would not adversely affect the experiment, the dished washers for the second set of specimens were fabricated from T-111, a more ductile material, and no further cracking problems were encountered. The flat tungsten disks served an additional function in the second set of specimens by also preventing contact of the UN fuel and the dished T-111 washers. The components for a typical fuel-element specimen (LT-6), prior to assembly, are shown in figure 3.

After all components were in place, the male end caps were electron-beam welded to the claddings. These final closure welds were made in a vacuum of 1.3×10^{-2} newton per square meter (1×10^{-4} torr) or better; thus, the vacuum level inside all the fuel-element specimens, except the two having intentional defects, should have been approximately 10^{-2} newton per square meter (10^{-4} torr). After welding, the fuel-element

specimens again were annealed at 1315° C (2400° F) for 1 hour in a vacuum of 2.7×10^{-3} newton per square meter (2×10^{-5} torr) or better (ref. 15). The purpose of this anneal was to allow the hafnium in the T-111 to getter any grain boundary oxygen picked up during welding and thus prevent subsequent liquid metal attack of the welds.

The integrity of the fuel-element specimens after annealing was verified by visual inspection and by the use of a helium mass spectrometer leak detector (except for the specimens having intentional defects, LT-6 and LT-7). Then, the specimens were X-ray radiographed to verify the location of the various components. Finally, the specimens were weighed, measured, and shipped to the contractor for testing in the loop.

POST-TEST EVALUATION RESULTS

General Observations

All five of the fuel-element specimens tested in the lithium loop appeared to be in excellent condition after removal from the loop, as can be seen in figure 4. The specimens were clean and bright and showed no evidence of any corrosion of the T-111 claddings. Excessive weld heat input during the fabrication of specimens LT-5 and LT-6 resulted in rather large weld beads, which were filed down to minimize possible flow restrictions in the fuel-element test section. Thus, these welds (see fig. 4(b)) look different from the welds on the other specimens. Except for the simulated cladding crack in specimen LT-6, no leaks or cracks could be found in the fuel-element specimens with a helium mass spectrometer leak detector. No dimensional changes as a result of the lithium exposure were observed.

The weights of the fuel-element specimens before and after testing and the resulting weight changes are listed in table I. The weights listed are based on measurement made by NASA and the contractor. The data in table I show that exposure in the lithium loop resulted in only minor weight changes for all the fuel-element specimens except LT-6. The fact that some samples had a slight weight gain and other samples had a slight weight loss is not considered significant because of the very small weight changes involved. As will be shown in the section Specimen Disassembly, the weight loss from specimen LT-6 can be attributed to erosion of the UN fuel cylinder by the flowing lithium.

Specimen LT-2 was weighed after 2500 hours in the loop and at the completion of the test after a total of 7500 hours in the loop. About 80 percent of the total weight loss from this specimen occurred during the first 2500 hours of testing. The remaining weight loss took place during the final 5000 hours of testing. Based on these results, testing for even longer times in the loop would probably not have resulted in any significant increase in the total weight loss of the defect-free fuel-element specimens.

Specimen Disassembly

The two fuel-element specimens removed from the loop after 2500 hours (LT-1 and LT-3) and the two control specimens (LT-4 and LT-7) were disassembled in air. Because of cladding embrittlement problems observed with specimens LT-1 and LT-3 (see the section Ductility Tests), the other three specimens exposed in the loop were disassembled in an argon atmosphere to minimize reaction of the specimens with the atmosphere during disassembly. The general procedures used in disassembly, however, were the same for all the specimens. The male end cap was carefully cut from each fuel-element specimen with a hacksaw (13 teeth/cm (32 teeth/in.)). Then the flat tungsten disks, dished washers, and UN fuel cylinders were removed and examined.

No evidence of any reactions between the UN fuel and the tungsten components was found. In all cases, the UN fuel cylinders were removed easily from the tungsten-lined T-111 claddings. Removal of the tungsten liners, however, was more difficult. Only the free-standing liner from control specimen LT-4 could be removed. Similar liners could not be removed from specimens LT-1, LT-2, and LT-3, even though these liners were loose prior to testing in the loop. A small amount of solid-state welding probably occurred between the tungsten and the T-111. The tungsten liners from specimens LT-5, LT-6, and LT-7 also could not be removed because the differential-thermal-expansion liner method used for these specimens resulted in some solid-state welding between the liner and the cladding prior to testing.

All the specimen components were clean and bright except for the UN fuel cylinder from LT-6, which showed some erosion and discoloration at the simulated cladding defect. Typical components from a specimen with no defect (LT-1) are shown after disassembly in figure 5. Only one dished tungsten washer is shown because the other washer was broken during shipment. Note that one of the tungsten disks is stuck to the dished washer. The other disks are not shown. The components from a specimen with a defect (LT-6) are shown in figure 6 after 5000 hours in flowing lithium. (Prior to assembly, one of the dished T-111 washers was dimpled to provide additional axial clearance for the UN fuel cylinder.) The marks on the outer surface of the T-111 cladding were caused by slippage in the vise during removal of the end cap. The discolored area on the UN cylinder coincided exactly with the intentional defect in the fuel-element cladding. A matching discolored area was observed on the inside surface of the tungsten liner also. The eroded area of the UN fuel cylinder from specimen LT-6 is shown in more detail in figure 7. Note how the discolored area fans out from the defect slot in the direction of the lithium flow. In this side-lighted photograph, the erosion depth is very apparent. The weight loss from the 61-gram UN cylinder, however, was only 0.0162 gram (about 0.03 percent of the total UN weight), which is quite low considering the large gap width and the long exposure time (5000 hr). The weight loss from the UN

cylinder was somewhat larger than the weight loss from the entire fuel-element specimen (0.0133 g). This difference might have been the result of some reaction between the UN and the cladding in the defect area (see the section Metallography) or of some trace amounts of lithium remaining in the fuel-element specimen having a defect when it was weighed.

Chemical Analyses

Samples of the T-111 end caps and the T-111 claddings from the fuel-element specimens exposed in the lithium loop were analyzed for interstitials (carbon, oxygen, nitrogen, and hydrogen) and major constituents. The results are listed in table II along with the results obtained for the unexposed control specimens. Very little change was observed in the interstitial contents of the samples as a result of the lithium exposures, except for a slight reduction in oxygen content. Both the claddings and end caps showed similar decreases in oxygen content, which is typical of metals subjected to long-time lithium exposures. The observed variations in the tungsten content and some of the variation in hafnium content were probably caused by inhomogeneities in the T-111 starting material. The hafnium analyses, however, showed an apparent trend toward increasing hafnium content with increasing loop exposure. The reason for this increase is not readily apparent. Although an analytical round robin on T-111 (ref. 17) indicated that no serious problems were encountered in the determination of alloying elements in T-111, it is possible that the observed increase in hafnium content is the result of some undefined problems in chemical analyses. Since the three specimens with the highest hafnium contents (LT-5, LT-6, and LT-7) were analyzed about a year later than the other specimens, slight changes in the analytical procedures could account for the apparent increase in hafnium content. The hafnium values also are questionable based on the before and after exposure weights of the specimens (table I) because the weight changes are not large enough to account for the apparent increases in hafnium content.

The UN fuel cylinders from the loop test specimens and from the control specimens were analyzed for uranium, nitrogen, oxygen, and carbon by the analytical methods described in reference 18. These results are presented in table III. As can be seen in the table, two different lots of UN cylinders were used for the fuel-element specimens. The UN in the first set of specimens contained much more oxygen than the UN in the second set of specimens. Comparing the analyses of the unexposed control specimen to the analyses of the tested specimens shows that testing in the lithium loop had essentially no effect on the analyses of any of the UN samples. Even specimen LT-6, exposed directly to lithium, showed no significant change in composition. The amount of scatter in the analyses is within the range normally seen in as-received UN.

Metallography

All the fuel-element specimens were examined metallographically to evaluate the effects of the lithium exposure on the microstructures of the T-111 cladding, the tungsten liners, and the UN fuel cylinders.

T-111 cladding. - Welds between the end cap and the cladding from specimens typical of each exposure condition are shown in figure 8. The difference in joint penetration during welding between the first set of specimens fabricated (LT-1 to LT-4) and the second set of specimens fabricated (LT-5 to LT-7) can be seen clearly in this figure. The weld heat input for the second set of specimens, typified by specimen LT-6 (fig. 8(c)), was much greater than that for the first set of specimens and resulted in a larger weld fusion zone and greater joint penetration. The larger welds, however, did not cause any problems during fabrication or during lithium exposure. Comparison of the microstructure of the untested specimen with the microstructures of the lithium-exposed specimens shows that the T-111 end caps and welds were in excellent condition and there was no evidence of any corrosion or chemical compatibility problems. The only noticeable change in microstructure was the formation of dark bands of precipitates in the T-111. There was no apparent trend for the amount of precipitates to increase with increasing exposure time. These precipitates have not been identified, but they are probably the result of localized areas of inhomogeneities in the T-111. At a higher magnification (see fig. 9), grain-boundary precipitates also can be seen in the T-111 microstructures of the tested specimens. It was shown in the work of reference 19 that precipitates of this type are commonly formed during long-term thermal aging of T-111 at about 1040° C (1900° F) regardless of aging environment (lithium or vacuum). Thus, it is reasonable to assume that the precipitates observed in this present study are also the result of thermal aging and not the result of the lithium exposure.

Examination of the T-111 claddings from all the lithium-exposed specimens showed no evidence of any lithium corrosion. No apparent contamination of the T-111 by the UN was observed except for the specimen having a defect (LT-6). The cladding microstructure from specimen LT-2 exposed for 7500 hours in the loop is shown in figure 10. As can be seen, the cladding is in excellent condition. A thin layer (about 1×10^{-3} cm (4×10^{-4} in.) thick) of a second phase, however, is visible at high magnification on the inside surface of the T-111 cladding (fig. 10(b)). This layer is thought to be a tungsten-rich layer formed as a result of diffusion from the tungsten liner. A diffusion layer of this type has essentially no effect on the strength or ductility of the T-111 cladding, as shown in reference 20.

The T-111 microstructure around the defect slot in specimen LT-6 is shown in figure 11 along with the similar defect region in the untested control specimen LT-7. In the unetched condition (figs. 11(a) and (c)), both samples appear to be uncontaminated.

In the etched condition, however, a large number of precipitates can be seen in the lithium-exposed sample (fig. 11(d)), while no precipitates can be seen in the untested control sample (fig. 11(b)). Thus, the observed precipitates are considered to be the result of the UN fuel being directly exposed to the flowing lithium.

A small sample of the T-111 cladding surrounding the defect slot in specimen LT-6 was analyzed for nitrogen in an attempt to identify the precipitate around the defect. Whereas the average nitrogen content of the cladding was about 25 parts per million by weight, the nitrogen content of the defect area sample was about 400 parts per million by weight. Right at the defect the nitrogen content of the cladding was probably much higher, because the nitrogen content at the defect would be diluted by the uncontaminated T-111 around the reaction zone.

The nitrogen pickup by the T-111 at the defect region was probably the result of the erosion of the UN fuel cylinder by the flowing lithium. The exact mechanism for the nitrogen pickup in the T-111, however, is not known. One possible explanation is that small particles of UN eroded from the fuel cylinder contacted the T-111 and reacted. The resulting nitrogen could have diffused into the T-111, and the free uranium could have been carried away by the lithium flow. (The reactions between UN and T-111 in direct contact and the resulting contamination of the T-111 were demonstrated in capsule tests in the study of ref. 3.)

Tungsten liners. - None of the tungsten liners showed any evidence of any chemical compatibility problems with the UN fuel. As can be seen in figure 12, the only noticeable change in the microstructures of the tungsten liners as a result of the long thermal exposure was some grain growth and the elimination of the visible bond lines between the layers of tungsten foil.

Uranium mononitride fuel cylinders. - A comparison of the UN microstructures before and after testing showed no change in general appearance as a result of the exposure in the pumped-lithium loop. The UN used in the first set of specimens is shown in figure 13 in the untested condition and after 7500 hours in the loop. The UN from the second set of specimens is shown in figure 14 in the untested condition and after 5000 hours in the loop. As can be seen in both figures, long-time exposure in the loop at 1040° C (1900° F) had no apparent effect on the density, grain size, and pore structure of the UN fuel cylinders. The white spots that appear in the pores in two of the photomicrographs (i.e., figs. 13(b) and 14(d)) are probably the result of reflected light from the bottom of the pores. The densities and microstructures of both lots of UN were quite similar except that the grain size (ASTM No. 6) of the UN used in the first set of specimens (fig. 13) was slightly larger than the grain size (ASTM No. 7) of the UN used in the second set of specimens (fig. 14). It was shown in the work of reference 14 that die-pressed UN would have to be sintered at a higher temperature than isostatically pressed UN in order to achieve similar densities. Thus, it is assumed that the die-

pressed UN used in specimens LT-1, LT-2, LT-3, and LT-4 was sintered at a higher temperature than the isostatically pressed UN used in specimens LT-5, LT-6, and LT-7. (The actual sintering temperatures used for the two lots of UN were not supplied by the contractor.) Such a difference in sintering temperature could account for the observed difference in grain size.

The UN fuel cylinder from specimen LT-6, with an intentional defect, was sectioned through the eroded area with a water-cooled silicon carbide abrasive saw. The cut section then was examined metallographically to determine what effect direct exposure to flowing lithium had on the UN. The cross section of the UN cylinder adjacent to the simulated cladding defect is shown in figure 15 in both the unetched and etched conditions. The erosion of UN by the lithium is very apparent in this figure. There is no evidence, however, of any chemical compatibility problem between the UN and the flowing lithium. It appears in figure 15 that the UN lost from the surface of the fuel cylinder was the result of simple erosion. It is possible that some surface reactions occurred between the UN and the lithium, but it is not apparent from the metallography. The crack in the UN fuel cylinder probably occurred during sectioning of the UN cylinder for examination and not during the lithium exposure. The low-density region to the left of the eroded area was probably present in the UN cylinder prior to testing. A similar low-density area can be seen in the upper left corner of the photomicrograph of the untested UN cylinder from the same lot of material (fig. 14(a)).

Ductility Tests

The ductility of the T-111 cladding from the lithium-exposed specimens and from the untested control specimens was determined by a ring-flattening test at room temperature. Rings, about 0.3 centimeter (1/8 in.) wide, were cut from the specimens and flattened either in a hydraulic compression machine or in a hand-operated vise. In both cases, the flattening rate (ram speed) was about 2.5 centimeters per minute (1 in./min).

As discussed in reference 4, initial flattening tests on the T-111 cladding were conducted on rings cut from the cladding of the two fuel-element specimens exposed in the lithium loop for 2500 hours (LT-1 and LT-3). All the ring samples for these initial tests were cut from the specimens with a water-cooled silicon carbide abrasive saw. Then the cut surfaces were wet-sanded with 400-grit silicon carbide paper. When the rings from the lithium-exposed specimens were flattened, they fractured in a brittle, intergranular manner with very little deformation. The ring samples from the untested control specimens were very ductile and could be flattened completely with no evidence of any cracking. Hydrogen embrittlement is suggested in reference 4 as a possible explanation for the brittle behavior.

Later studies conducted on the embrittlement of T-111 (ref. 19) showed that thermal aging of T-111 at 1040° C (1900° F) for long periods of time greatly increased the sensitivity of T-111 to hydrogen embrittlement. Embrittlement resulted from exposure of the T-111 to water during cutting and sanding after aging. Testing of the tubing samples in a moist atmosphere also resulted in embrittlement. In view of these results, the T-111 rings from the fuel-element specimens exposed for 5000 and 7500 hours in the lithium loop were prepared and tested in an argon atmosphere without any exposure to a source of moisture. Rings were cut from the T-111 claddings on specimens LT-2 and LT-5 with a hand-operated hacksaw (13 teeth/cm (32 teeth/in.)), filed to remove worked metal, and finally dry-sanded with 400-grit silicon carbide paper. The rings then were flattened in a vise at room temperature in argon. The rings from both specimens were very ductile and could be flattened completely, as shown in figure 16. There was no evidence of any cracking or embrittlement of the T-111. The tungsten liners cracked and spalled during deformation, but this was expected.

A ring sample from specimen LT-2 also was prepared by the same techniques that had been used on ring samples from specimens LT-1 and LT-3 (i.e., wet-cutting and wet-sanding). The ring sample from LT-2 fractured during flattening in air, which again showed the sensitivity of the aged T-111 to hydrogen embrittlement.

DISCUSSION

The evaluation of the fuel-element specimens exposed in the lithium loop for up to 7500 hours showed that there are no apparent chemical compatibility or corrosion problems between defect-free fuel-element specimens and flowing lithium. The appearance, integrity, weight, dimensions, composition, and metallography of the defect-free specimens were essentially unchanged as a result of the lithium exposure. Based on these results and the assumption that no other reactions would occur with longer exposure times, the fuel-element specimens should be compatible with the flowing lithium at 1040° C (1900° F) for the desired reactor lifetime goal of 50 000 hours.

A small amount of UN was lost from the fuel cylinder directly exposed through a simulated cladding defect to flowing lithium for 5000 hours. The UN loss appeared to be the result of simple erosion. Metallographic examination of the UN cylinder showed no evidence of any chemical compatibility reactions between the lithium and the UN. The T-111 cladding near the defect slot, however, was contaminated with nitrogen from the UN fuel and contained large amounts of precipitates. The fuel loss and the contamination of the T-111 would probably increase with increasing exposure time. Thus, additional tests are needed for longer times to investigate the amount of fuel loss and to determine if the contamination of the T-111 at a cladding defect could have adverse effects

on the ductility and compatibility of the T-111.

Another area that has not been investigated is the effect of irradiation on the compatibility of the fuel-element materials with flowing lithium. Therefore, some additional compatibility tests in an irradiation environment are needed to evaluate fully the compatibility of the fuel-element materials.

CONCLUSIONS

A total of five simulated nuclear fuel-element specimens (consisting of uranium mononitride (UN) fuel cylinders clad with tungsten-lined T-111) were exposed in a pumped-lithium loop at 1040° C (1900° F) for up to 7500 hours. Lithium flow velocity through the specimen test section was 1.5 meters per second (5 ft/sec). After exposure, the specimens were thoroughly evaluated and compared to unexposed control specimens. The results of this evaluation lead to the following major conclusions:

1. The UN fuel cylinders clad with tungsten-lined T-111 were compatible with flowing lithium at 1040° C (1900° F) for at least 7500 hours. Based upon the compatibility results after 7500 hours in the lithium loop, no compatibility problems are expected between fuel elements without defects and flowing lithium at 1040° C (1900° F) for the reactor life-time goal of 50 000 hours.

2. Direct exposure of the UN fuel to flowing lithium through a simulated crack in the T-111 cladding for 5000 hours resulted in only minor fuel loss and reaction with the T-111 cladding. Longer tests are needed, however, to determine if the rate of fuel loss and the cladding reaction would increase with increasing exposure time.

3. The T-111 fuel-element cladding was ductile at room temperature after exposure in flowing lithium for up to 7500 hours at 1040° C (1900° F). The effects of thermal aging at this temperature, however, made the T-111 sensitive to hydrogen embrittlement during post-test handling, as had been demonstrated in previous studies.

4. The long-time lithium exposure at 1040° C (1900° F) had no significant effects on the microstructures of the UN fuel and the T-111 cladding.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, November 28, 1973,
502-21.

REFERENCES

1. Krasner, Morton H.; Davison, Harry W.; and Diaguila, Anthony J.: Conceptual Design of a Compact Fast Reactor for Space Power. NASA TM X-67859, 1971.

2. Gluyas, R. E.; and Lietzke, A. F.: Materials Technology Program for a Compact Fast Reactor for Space Power. NASA TM X-67869, 1971.

3. Sinclair, John H.: Compatibility Tests of Materials for Lithium-Cooled Space Power Reactor Concept. NASA TN D-7259, 1973.

4. Watson, Gordon K.: Preliminary Evaluation of T-111 Clad UN Fuel Specimens from 2500-Hour 1040° C (1900° F) Lithium Loop Test. NASA TM X-52998, 1971.

5. Harrison, R. W.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-303, General Electric Co. (NASA CR-72592), July 30, 1969.

6. Harrison, R. W.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-376, General Electric Co. (NASA CR-72620), Oct. 23, 1969.

7. Harrison, R. W.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-410, General Electric Co. (NASA CR-72662), Feb. 4, 1970.

8. Harrison, R. W.; and Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-491, General Electric Co. (NASA CR-72739), May 11, 1970.

9. Harrison, R. W.; and Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-546, General Electric Co. (NASA CR-72782), Aug. 12, 1970.

10. Harrison, R. W.; and Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-562, General Electric Co. (NASA CR-72818), Nov. 12, 1970.

11. Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-606, General Electric Co. (NASA CR-72853), Feb. 1, 1971.

12. Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. Rep. GESP-652, General Electric Co. (NASA CR-72985), May 13, 1971.

13. Brandenberg, G. P.; Hoffman, E. E.; and Smith, J. P.: Pumped Lithium Loop Test to Evaluate Advanced Refractory Metal Alloys and Simulated Nuclear Fuel Elements. General Electric Co. (NASA CR-134527), 1974.

14. Tennery, V. J.; Godfrey, T. G.; and Potter, R. A.: Synthesis, Characterization, and Fabrication of UN. Rep. ORNL-4608, Oak Ridge National Lab. (NASA CR-72764), Dec. 1970.

15. Moore, T. J.; Moorhead, P. E.; and Bowles, K. J.: Specifications for Cleaning, Fusion Welding, and Postheating Tantalum and Columbium Alloys. NASA TM X-67879, 1971.

16. Watson, Gordon K.; Whittenberger, John D.; and Mattson, William F.: Thermal-Expansion Method for Lining Tantalum Alloy Tubing with Tungsten. NASA TN D-7426, 1973.

17. Chase, D. L.: Comparison of Chemical Analyses of Refractory Alloys. DMIC Rep. 220, Battelle Memorial Inst., 1965.

18. Merkle, Emery J.; Davis, Warren F.; Halloran, John T.; Graab, Judson W.: An Interlaboratory Comparison of the Chemical Analysis of Uranium Mononitride. NASA TN D-7536, 1974.

19. Watson, Gordon K.; and Stephens, Joseph R.: Effect of Aging at 1040° C (1900° F) on the Ductility and Structure of a Tantalum Alloy, T-111. NASA TN D-6988, 1972.

20. Buzzard, Robert J.; and Metroka, Robert R.: Tensile Properties From Room Temperature to 1315° C of Tungsten-Lined Tantalum Alloy (T-111) Tubing Fabricated by Hot Isostatic Pressing. NASA TM X-2964, 1974.

TABLE III. - CHEMICAL ANALYSES OF URANIUM MONONITRIDE FUEL CYLINDERS FROM FUEL-ELEMENT SPECIMENS BEFORE AND AFTER EXPOSURE IN 1040° C (1900° F) LITHIUM LOOP

Specimen	Exposure time, hr	Element concentration			
		Uranium, wt. %	Nitrogen, wt. %	Oxygen, ppm by wt.	Carbon, ppm by wt.
LT-4	0	94.56	5.31	982	208
LT-1	2500	94.47	5.35	1122	192
LT-3	2500	94.40	5.33	983	189
LT-2	7500	94.38	5.38	1100	180
LT-7	0	94.42	5.52	115	265
LT-5	5000	94.41	5.52	95	255
LT-6	5000	94.40	5.52	105	250

TABLE II. - CHEMICAL ANALYSES OF FUEL-ELEMENT CLADDINGS AND END CAPS BEFORE AND AFTER EXPOSURE IN 1040° C (1900° F) LITHIUM LOOP

Specimen	Exposure time, hr	Element concentration					
		Carbon, ppm by wt.	Oxygen, [a] ppm by wt.	Nitrogen, [a] ppm by wt.	Hydrogen, [a] ppm by wt.	Tungsten, wt. %	Hafnium, wt. %
T-111 Cladding							
[b]LT-4	0	61	72	22	1	7.70	2.22
[c]LT-7	0	65	40	17	<.5	7.91	2.06
LT-1	2500	69	35	20	↓	7.92	2.09
LT-3	2500	71	27	15	↓	7.63	2.20
LT-5	5000	64	25	27	↓	7.75	2.63
LT-6	5000	57	13	25	↓	7.70	2.52
LT-2	7500	57	21	40	↓	7.64	2.72
T-111 end caps[d]							
[b]LT-4	0	24	68	12	1	7.51	2.19
[c]LT-7	0	50	47	10	<.5	7.54	1.92
LT-1	2500	53	47	10	↓	7.89	2.14
LT-3	2500	43	45	10	↓	7.55	2.02
LT-5	5000	32	35	16	↓	7.71	2.23
LT-6	5000	44	28	11	↓	7.86	2.34
LT-2	7500	52	40	16	↓	7.86	2.37

[a]Average of duplicate analyses.

[b]Control specimen for LT-1, LT-2, and LT-3.

[c]Control specimen for LT-5 and LT-6.

[d]Combined average analyses of male and female end caps.

TABLE I. - WEIGHTS OF FUEL-ELEMENT SPECIMENS

BEFORE AND AFTER EXPOSURE IN

1040^{o} C (1900^{o} F) LITHIUM LOOP

Specimen	Exposure time, hr	Weight, g		Weight change, g
		Before test	After test	
LT-4	0	135.7926	--------	-------
LT-7	0	133.6238	--------	-------
LT-1	2500	135.9871	135.9901	+0.0030
LT-3	2500	135.5784	135.5817	+.0033
LT-5	5000	133.5430	133.5417	-.0013
LT-6	5000	134.3446	134.3313	-.0133
[a]LT-2	2500	135.2545	135.2524	-.0021
LT-2	7500	135.2545	135.2519	-.0026

[a]Specimen removed from loop and weighed after 2500 hr.

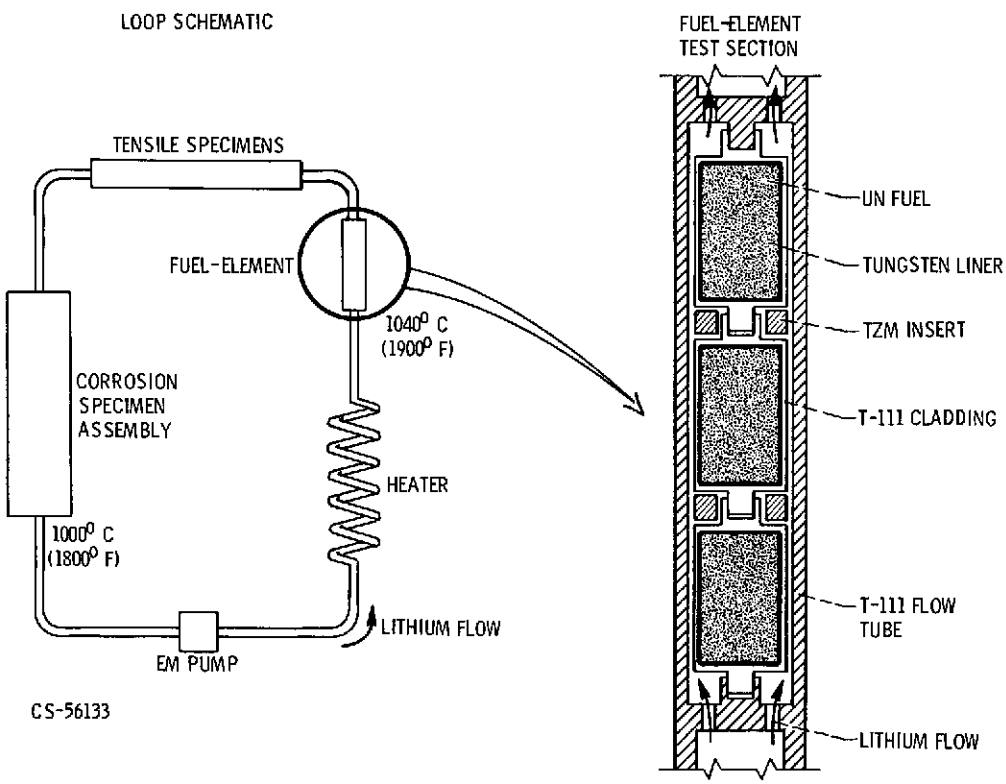


Figure 1. - Schematic drawing of 1040^{o} C (1900^{o} F) lithium loop and fuel-element test section.

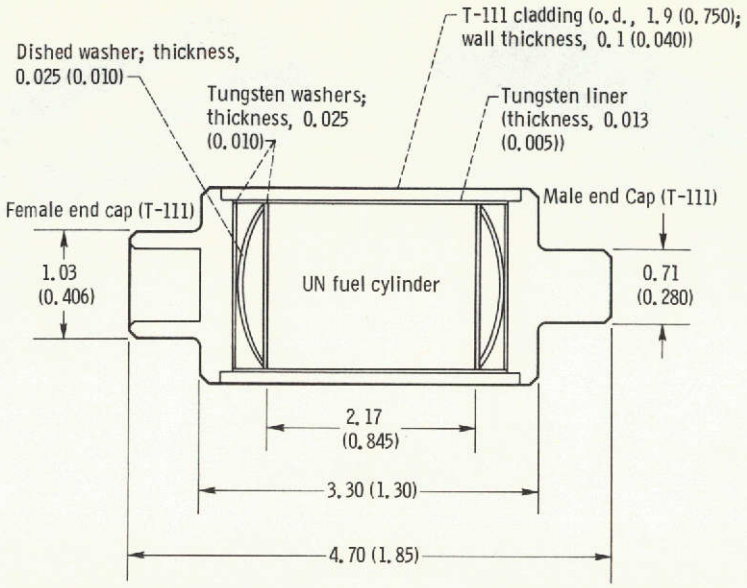


Figure 2. - Design of fuel-element specimens used in 1040° C (1900° F) lithium-loop test. (Dimensions in centimeters (in.).)

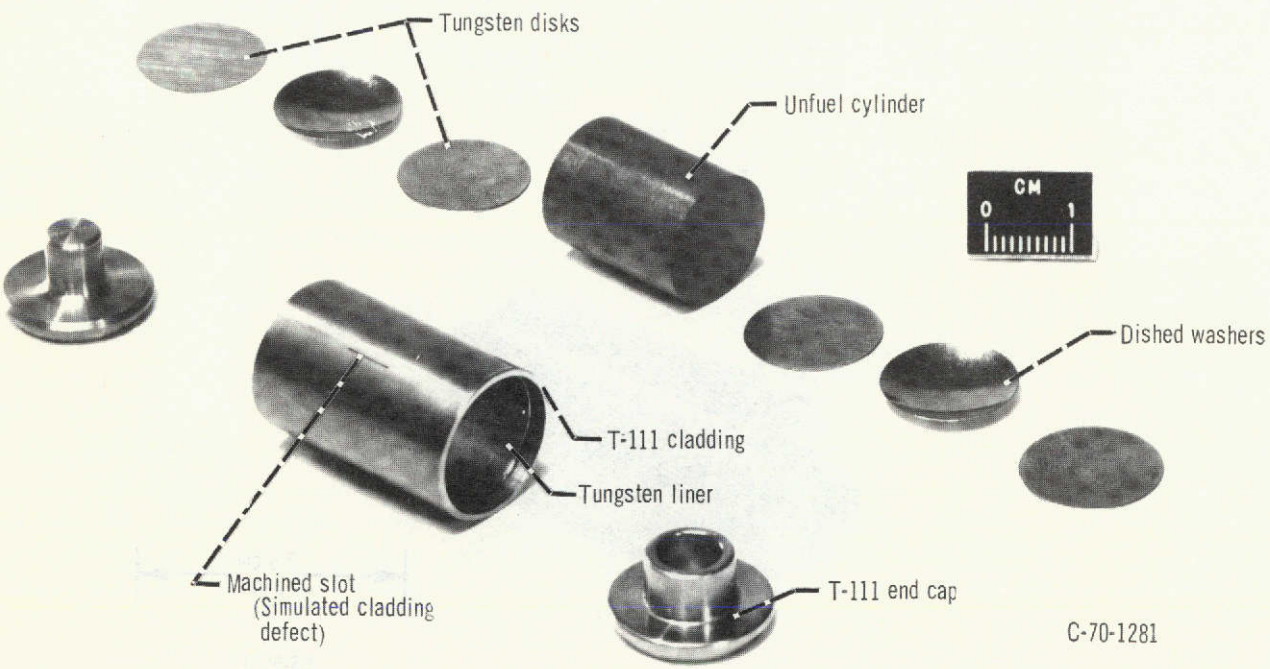
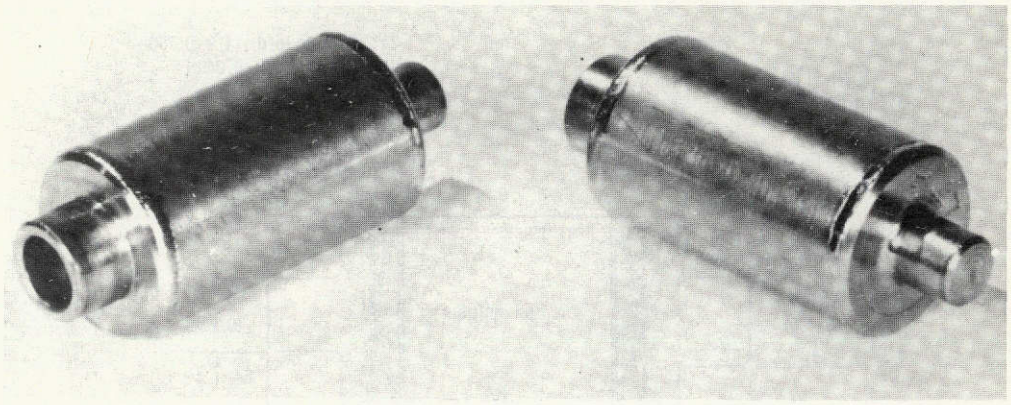
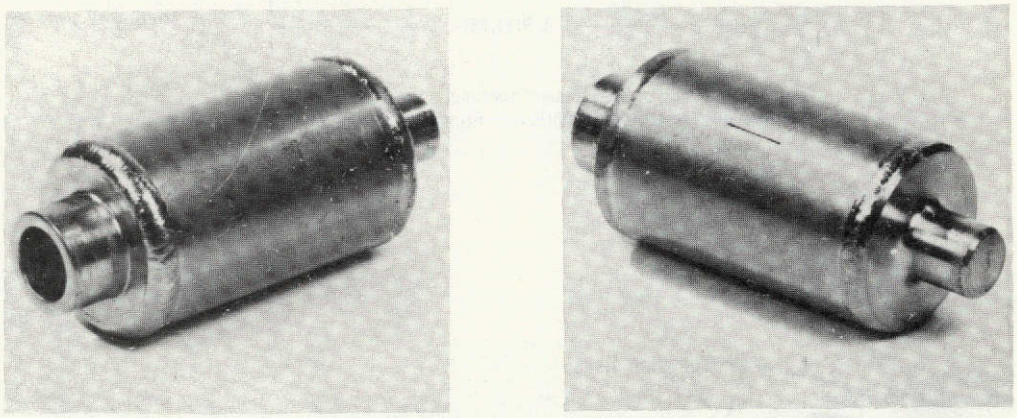


Figure 3. - Components for typical fuel-element specimen (LT-6) prior to assembly.



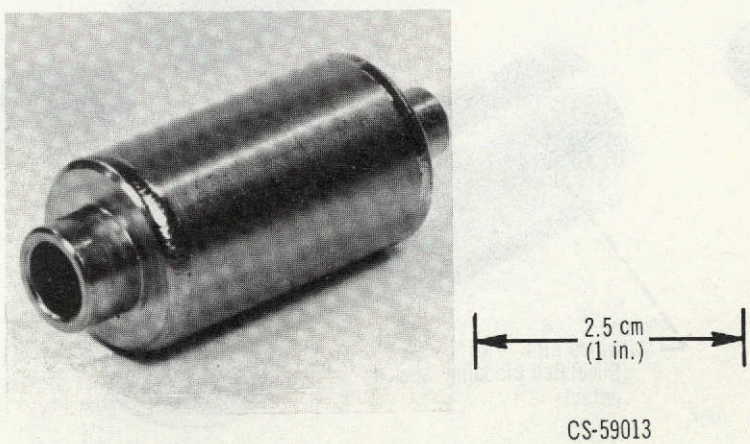
LT-1 LT-3

(a) Exposed 2500 hours.



LT-5 LT-6

(b) Exposed 5000 hours.



(c) Exposed 7500 hours; LT-2.

Figure 4. - Fuel-element specimens after exposure in 1040° C. (1900° F) lithium loop.

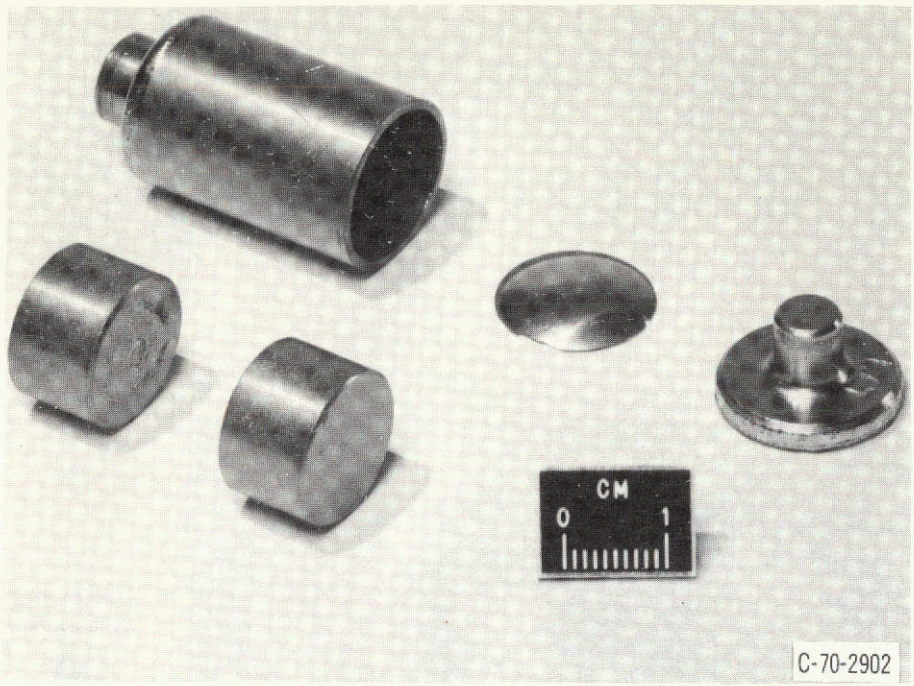


Figure 5. - Fuel-element specimen LT-1 disassembled after 2500 hours in 1040° C (1900° F) lithium loop.

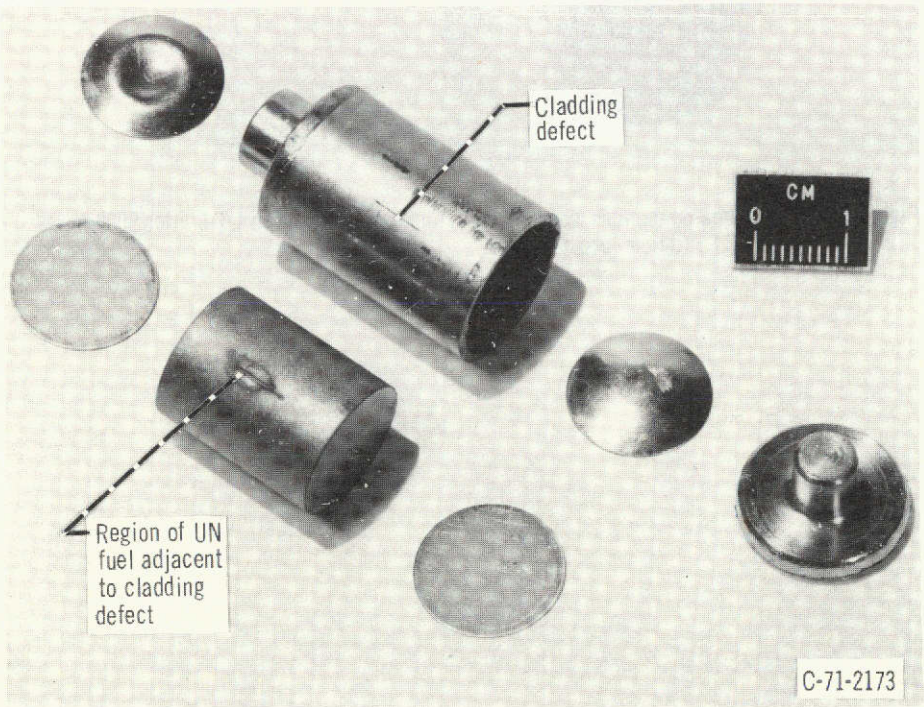


Figure 6. - Fuel-element specimen LT-6 disassembled after 5000 hours in 1040° C (1900° F) lithium loop. Note cladding defect and discolored area on uranium mononitride fuel cylinder.

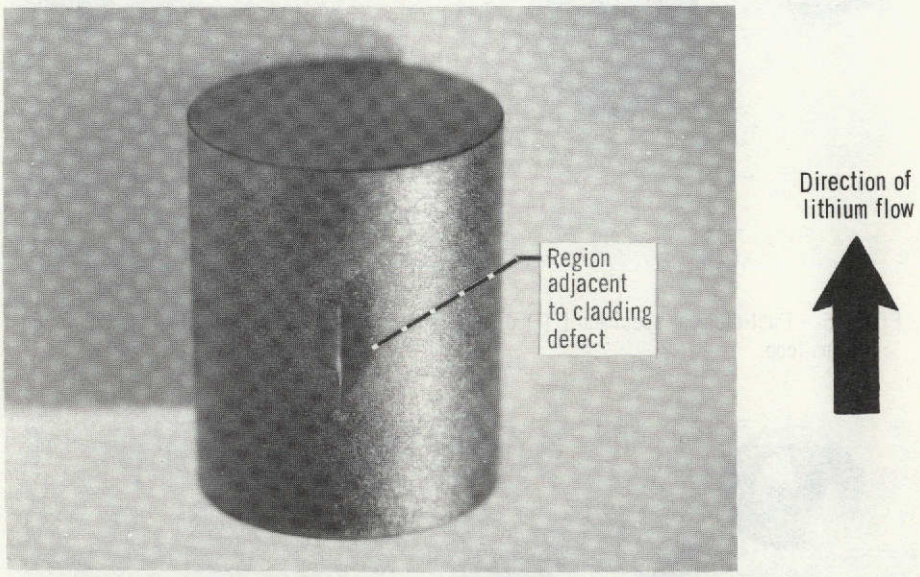
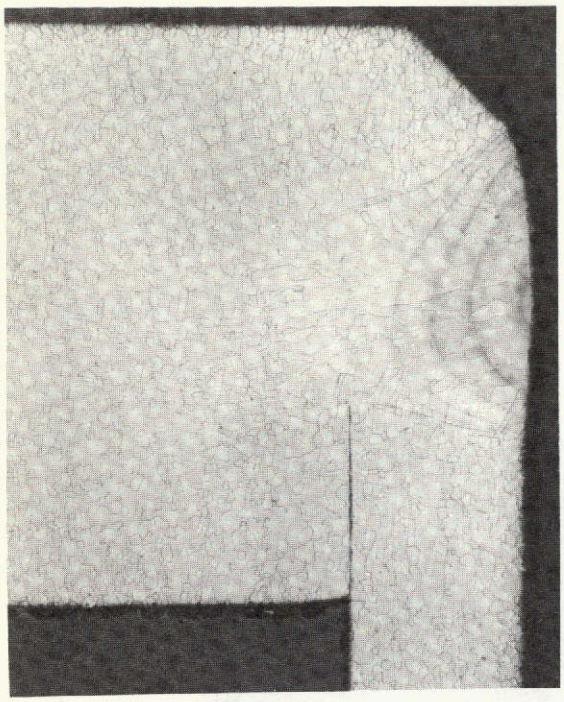
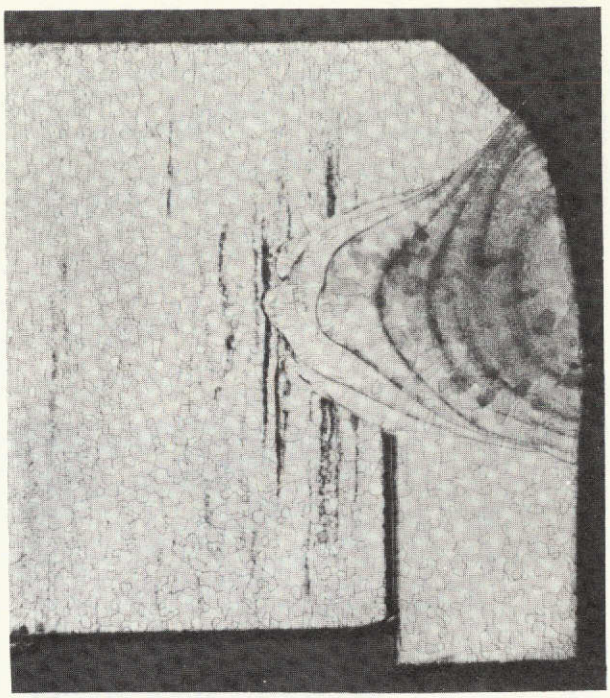


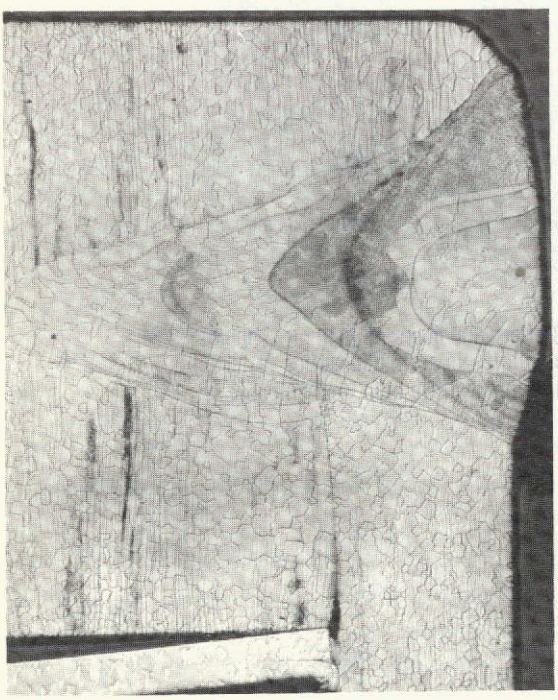
Figure 7. - Eroded area on uranium mononitride cylinder from specimen LT-6 after 5000 hours in 1040° C (1900° F) lithium loop.



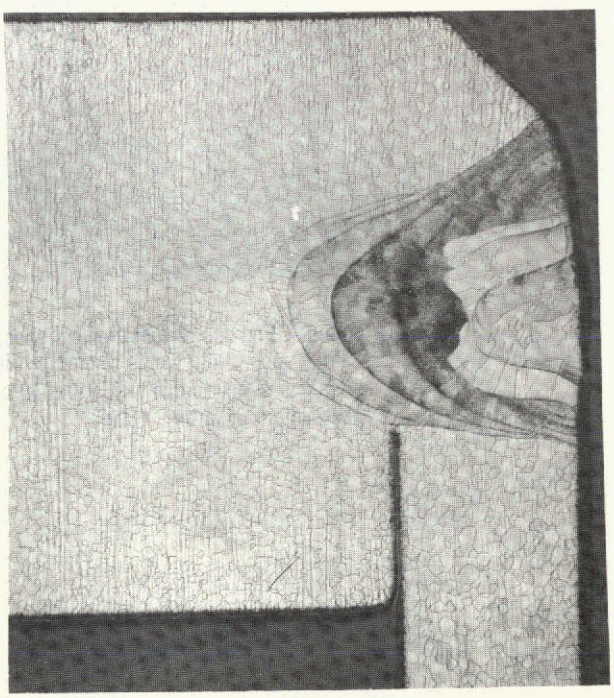
(a) LT-4; untested.



(b) LT-1; exposed 2500 hours.



(c) LT-6; exposed 5000 hours.



(d) LT-2; exposed 500 hours.

Figure 8. - End-cap weld area of T-111 fuel-element specimens in untested condition and after various exposures in 1040° C (1900° F) lithium loop. Etchant: 30 grams of ammonium bifluoride, 50 cubic centimeters of nitric acid, and 20 cubic centimeters of water. X25.

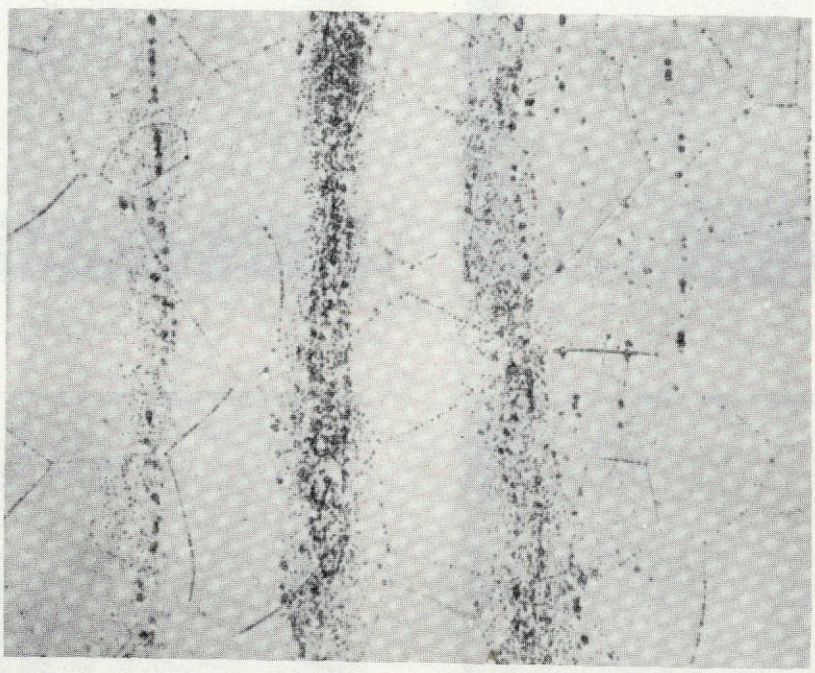
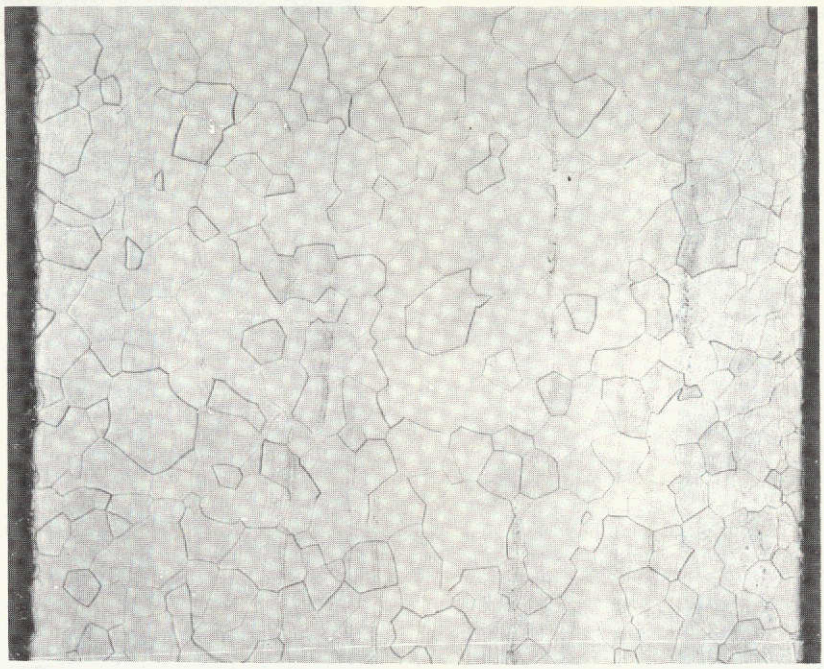


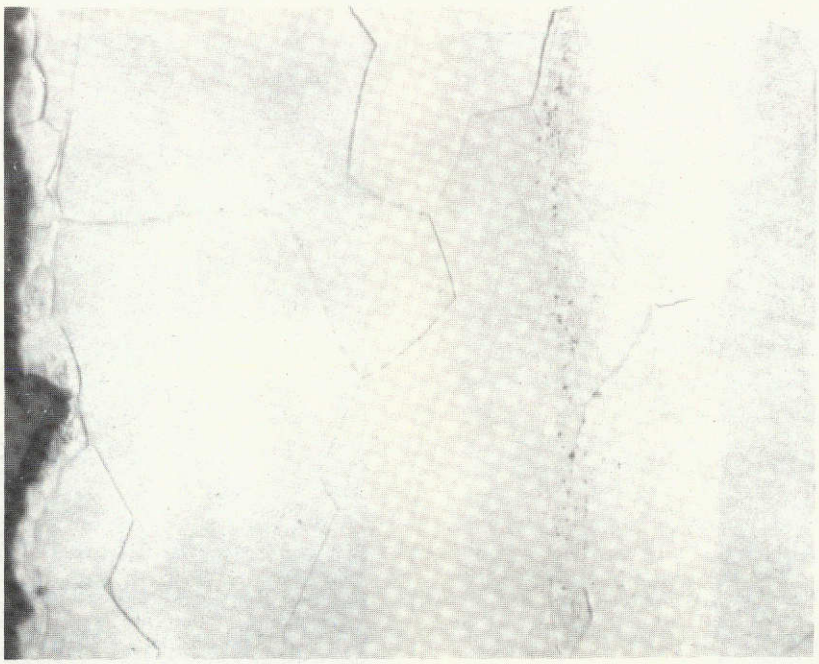
Figure 9. - End cap of fuel-element specimen LT-3 after 2500 hours in 1040° C (1900° F) lithium loop. Note bands of precipitates and precipitates in grain boundaries. Etchant: 30 grams of ammonium bifluoride, 50 cubic centimeters of nitric acid, and 20 cubic centimeters of water. X500.



Inside surface

(a) X100.

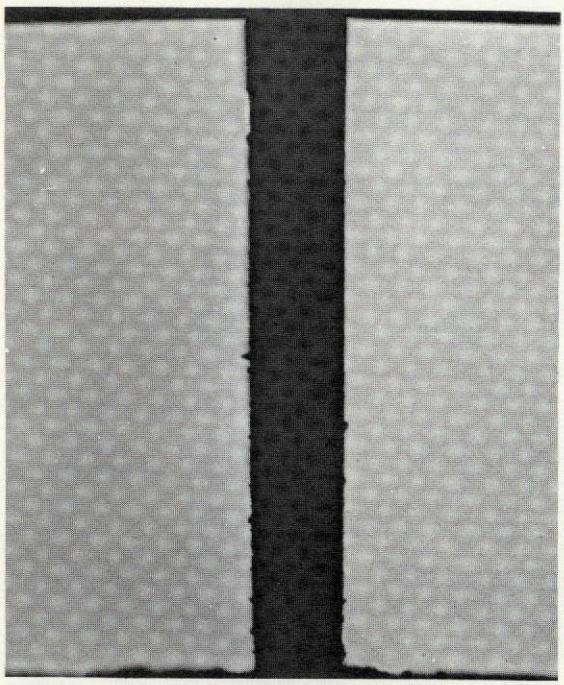
Outside surface



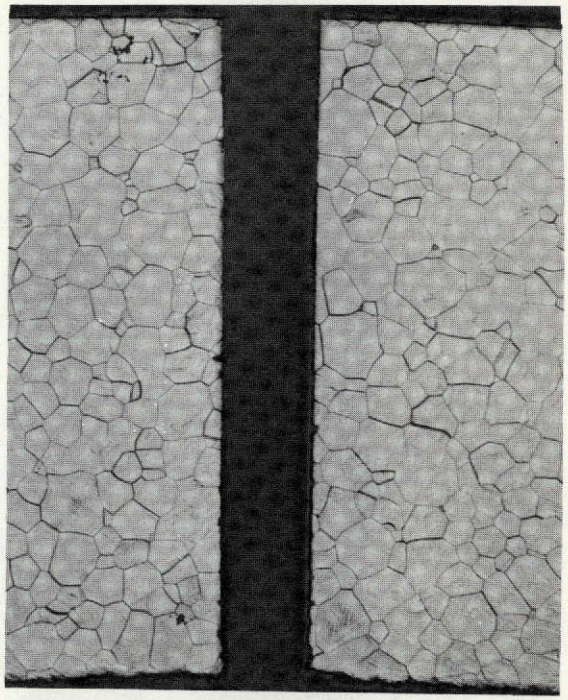
Inside surface

(b) X500.

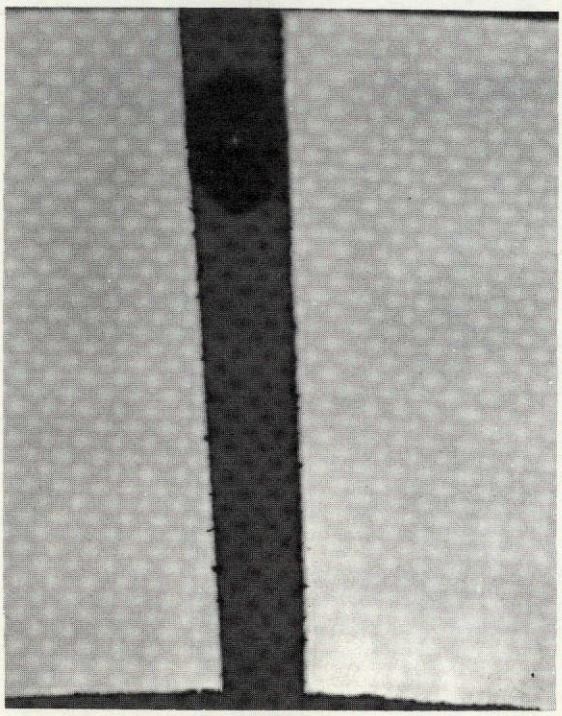
Figure 10. - Longitudinal section of T-111 cladding from fuel-element specimen LT-2 after 7500 hours in 1040° C (1900° F) lithium loop. Note tungsten-rich layer on inside surface of cladding. Etchant: 30 grams of ammonium bifluoride, 50 cubic centimeters of nitric acid, and 20 cubic centimeters of water.



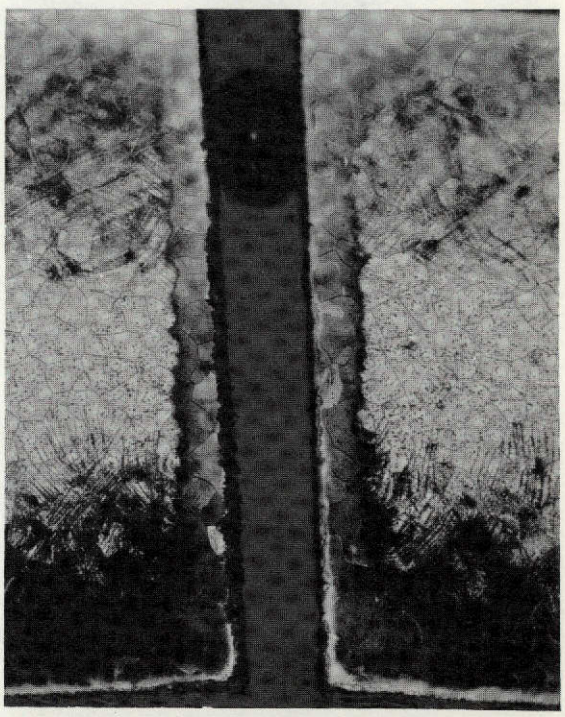
(a) LT-7; untested; unetched.



(b) LT-7; untested; etched.

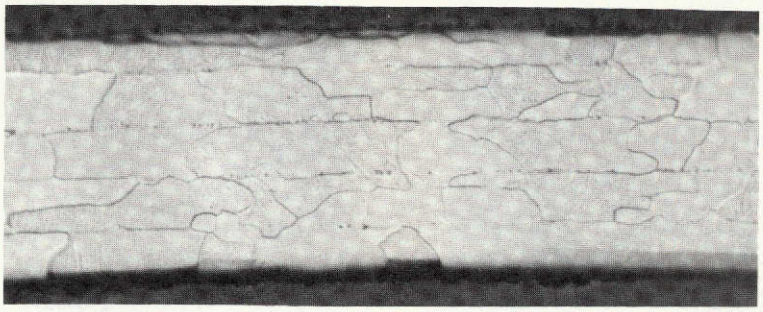


(c) LT-6; exposed 5000 hours; unetched.

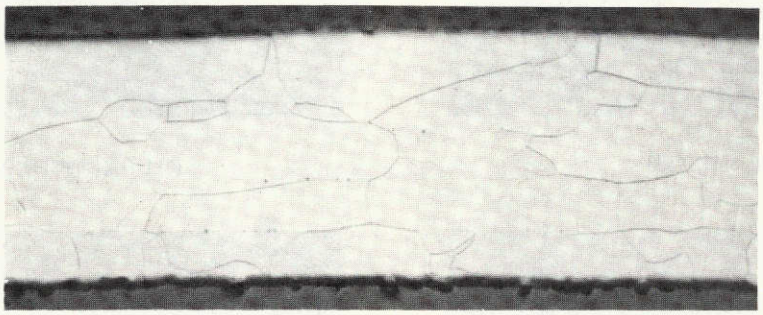


(d) LT-6; exposed 5000 hours; etched.

Figure 11. - Microstructures of defect region in T-111 cladding of fuel-element specimens LT-7, untested, and LT-6, exposed 5000 hours in 1040° C (1900° F) lithium loop. Etchant: 30 grams of ammonium bifluoride, 50 cubic centimeters of nitric acid, and 20 cubic centimeters of water. X100.

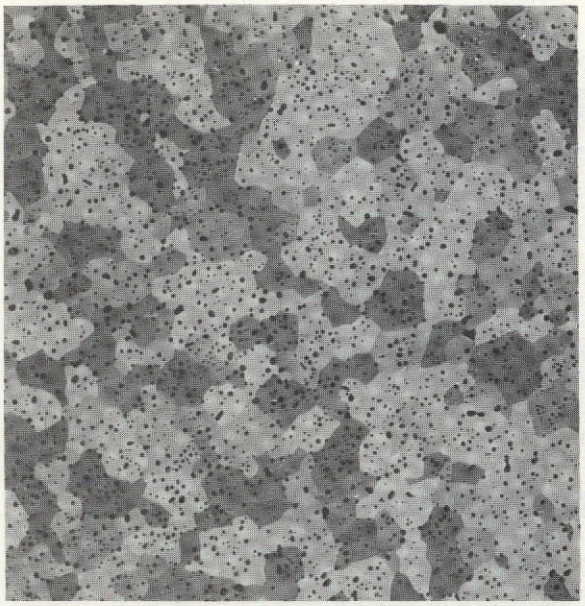


(a) LT-4; untested.

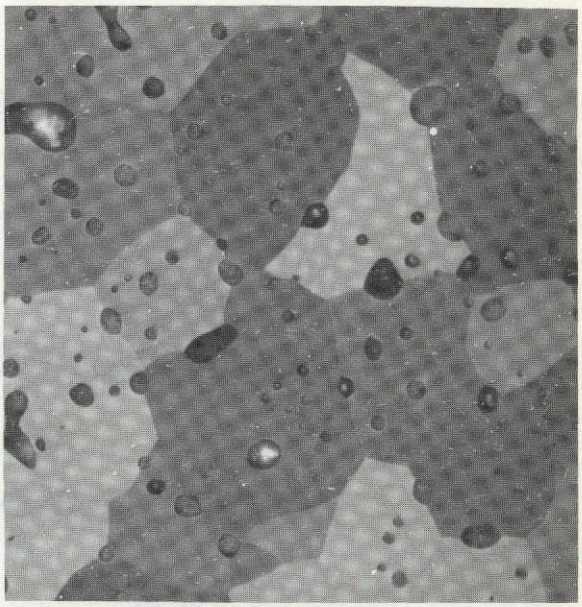


(b) LT-2; exposed 7500 hours.

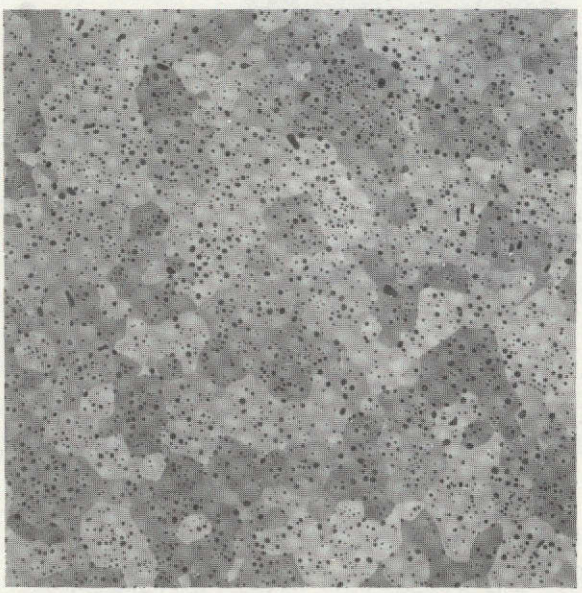
Figure 12. - Comparison of cross section of tungsten liners in untested condition and after 7500 hours in 1040° C (1900° F) lithium loop. Etchant: 3 parts lactic acid, 2 parts nitric acid, and 1 part hydrofluoric acid. X250.



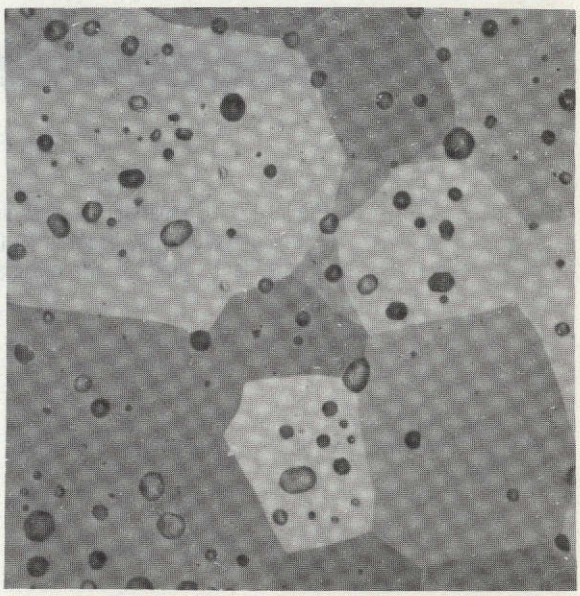
(a) LT-4; untested; X100.



(b) LT-4; untested; X500.

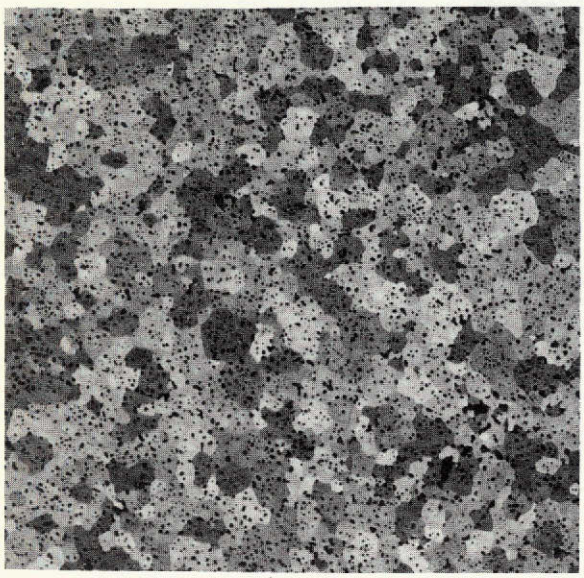


(c) LT-2; exposed 7500 hours; X100.

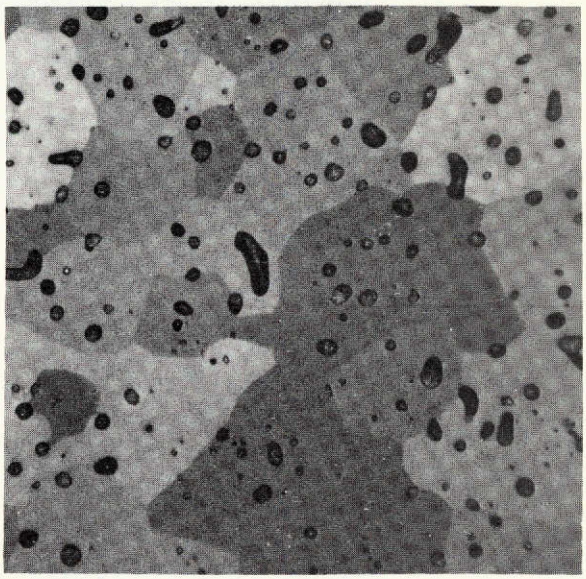


(d) LT-2; exposed 7500 hours; X500.

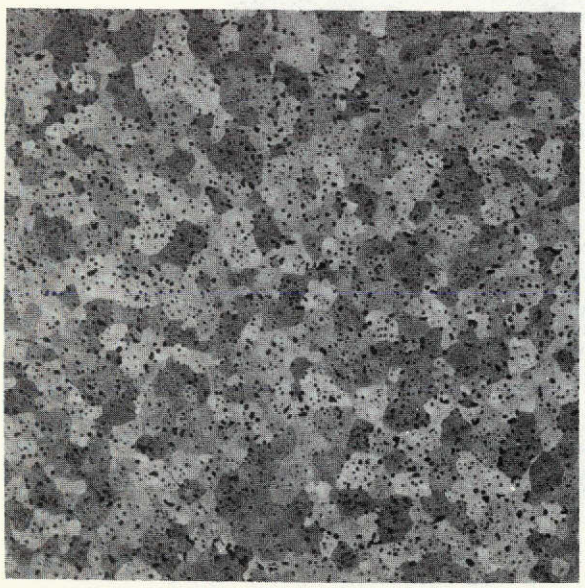
Figure 13. - Comparison of uranium mononitride fuel microstructures in untested condition and after 7500 hours in 1040° C (1900° F) lithium loop. Etchant: 60 cubic centimeters of acid, 24 cubic centimeters of nitric acid, and 2 cubic centimeters of hydrofluoric acid.



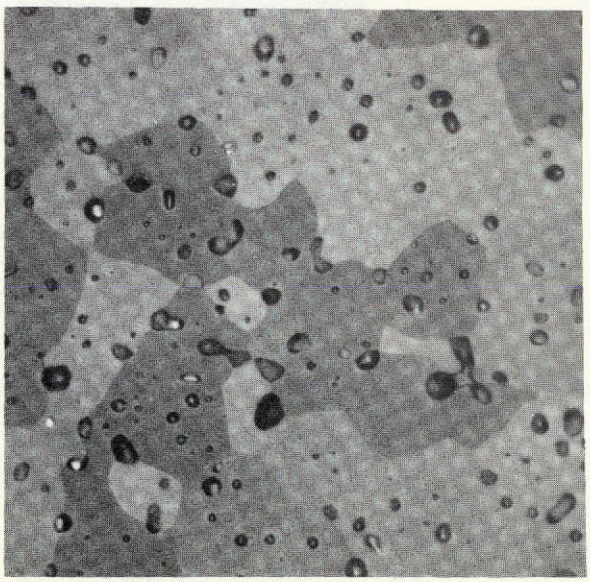
(a) LT-7; untested; X100.



(b) LT-7; untested; X500.

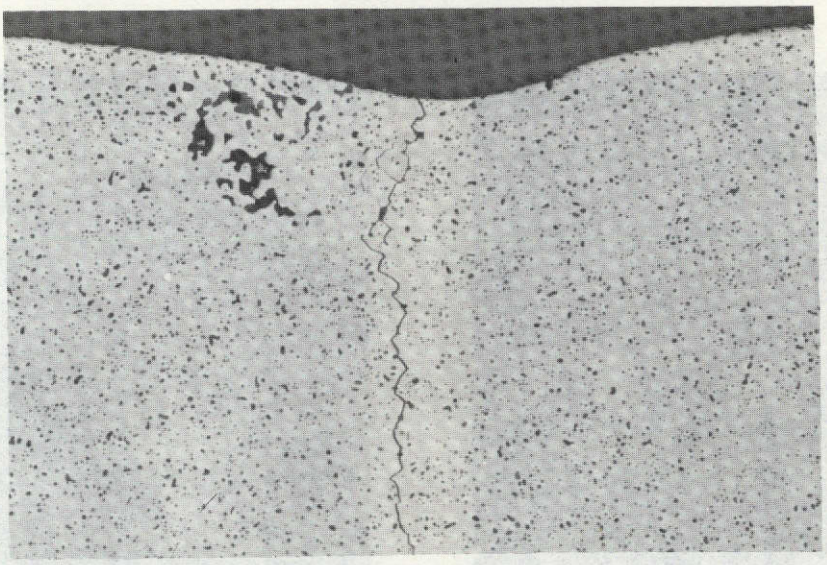


(c) LT-6; exposed 5000 hours; X100.

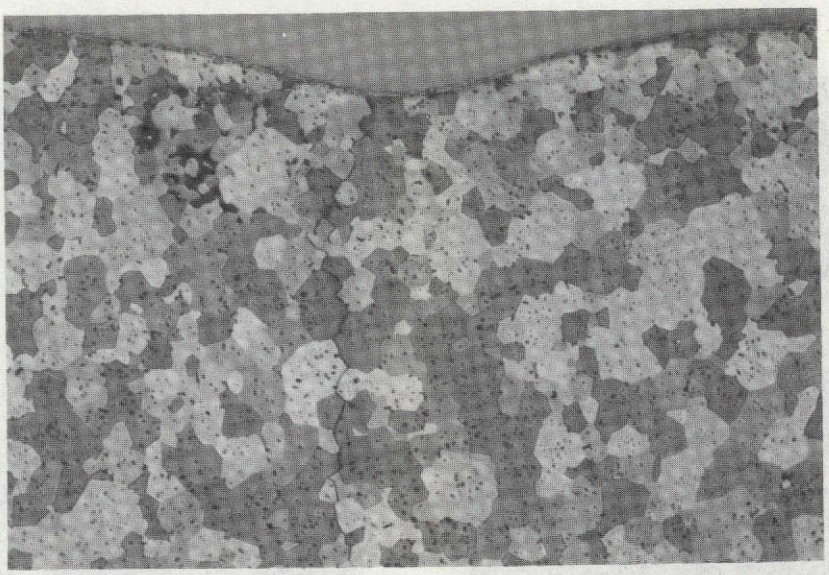


(d) LT-6; exposed 5000 hours; X500.

Figure 14. - Comparison of uranium mononitride fuel microstructures from untested fuel-element specimen LT-7 and from fuel-element specimen LT-6, with intentional defect, exposed for 5000 hours in 1040° C (1900° F) lithium loop. Etchant: 60 cubic centimeters of lactic acid, 24 cubic centimeters of nitric acid, and 2 cubic centimeters of hydrofluoric acid.



(a) Unetched.



(b) Etched.

Figure 15. - Uranium mononitride fuel microstructures in immediate area of cladding defect in fuel-element specimen LT-6 after 5000 hours in 1040° C (1900° F) lithium loop. Etchant: 60 cubic centimeters of lactic acid, 24 cubic centimeters of nitric acid, and 2 cubic centimeters of hydrofluoric acid. X100.

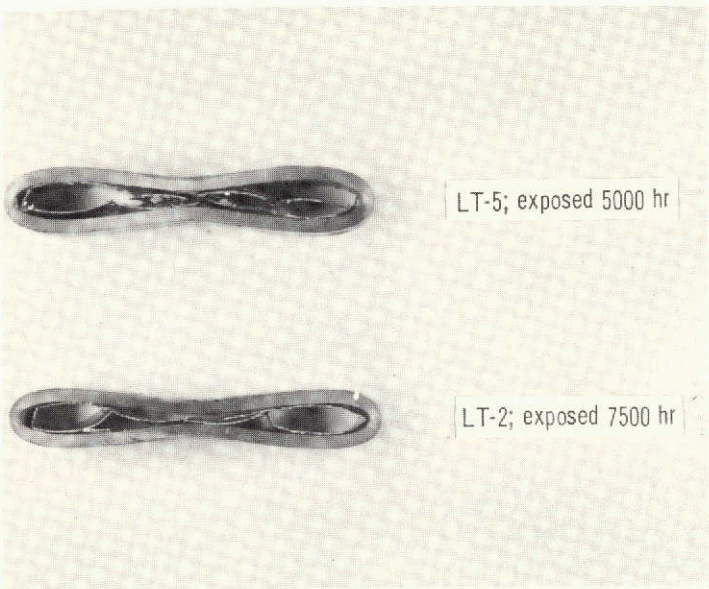


Figure 16. - Results of tube flattening tests conducted on T-111 claddings from fuel-element specimens LT-5 and LT-2. Test rings prepared and tested in argon atmosphere; both rings flattened completely without cracking.