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MATERIALS TECHNOLOGY PROGRAM FOR A COMPACT FAST REACTOR FOR SPACE POWER

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TABLE OF CONTENTS

E-6415

ABSTRACT

The suitability of T-111 (Ta-8W-2Hf) clad uranium mononitride (UN) fuel for a lithium-cooled space power reactor concept is examined. Methods for fabrication of fuel pins are described. Out-of-pile compatibility testing indicates that at 1040 °C a 0.013 cm (0.005 inch) thick tungsten barrier is required between the fuel and cladding; UN with $<$ 300 ppm of oxygen is compatible with lithium; no corrosion of T-111 was observed after 7500 hours in a pumped-lithium loop. An irradiation program on fuel pins and cladding materials is also described.

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SUMMARY

A compact, liquid metal cooled, fast reactor concept is being investigated at the NASA-Lewis Research Center. This paper describes selected parts of the materials technology program for this concept. The emphasis is on uranium mononitride (UN) as the nuclear fuel, T-111 (Ta-8W-2Hf) alloy for fuel pin cladding and reactor structural components, and lithium as the coolant. Also, under study are alternate fuel and cladding materials, reflector materials, and refractory carbides and nitrides for bearing materials.

Fabrication methods have been developed for routine preparation of high purity UN fuel forms, for applying tungsten liners to the inside wall of T-111 tubing used for fuel pin cladding, and for assembly of fuel pins. Inspection methods are under development.

Compatibility and corrosion studies performed at 1040°C with isothermal capsules and with a pumped-lithium loop show: reaction of UN in direct contact with T-111 but no reaction when these materials are separated by a 0.013 cm tungsten barrier; compatibility of UN with lithium provided the oxygen impurity content of UN is low (less than 300 ppm); and no significant corrosion of T-111 or of TZM in a pumped-lithium loop for times up to 7500 hours.

A fuel pin irradiation program to investigate fission gas release, materials compatibility under irradiation, and fuel pin dimensional stability are briefly described. Most of this program is on UN clad with tungsten-lined T-111, but some work is being done with UO_2 and Cb-1Zr. So far, for UN pins at low burnup levels (about 1 a/o), it appears that fission gas release is very low ($\leq$ 1%), and that there are no irradiation-induced compatibility problems.

One of the greatest unknowns is the effect of fast neutron fluences to about 1.2×10^{22} neutrons/cm^2 (E > 0.82 Mev) on the ductility and swelling of potential structural materials. Capsules incorporating specimens of various refractory alloys immersed in lithium are being fabricated for irradiation tests of this effect.

INTRODUCTION

A compact, liquid-metal cooled, fast spectrum reactor concept for space power applications is being investigated at the NASA-Lewis Research Center. The investigation is concentrated on a reference reactor design concept with a reactor power level of 2.2 MW (thermal), a coolant outlet temperature of 950°C, and an operating life of 50,000 hours. The study is intended to establish feasibility of a reactor design concept and includes the development of necessary long lead-time technology and testing of critical components. This report is one of a group of three reports describing the overall effort. The conceptual reactor design is described in Reference 1, and Reference 2 discusses the nuclear design and associated critical experiments. The materials technology and fuel pin test programs are discussed in this report.

The materials which have been selected for intensive study are: uranium mononitride (UN) as the nuclear fuel, T-111 (Ta-8W-2Hf) alloy for fuel pin cladding and reactor structural components, and lithium as the coolant. Uranium mononitride (fully enriched with the isotope U-235) was selected because of its high uranium density, high thermal conductivity, the possibility of low fission-induced swelling (ref. 3), and compatibility with lithium. The T-111 alloy is ductile, readily fabricated, and has good high temperature creep strength for application as fuel pin cladding. Lithium is attractive as the coolant because of its relatively low vapor pressure, good heat transfer and fluid flow characteristics, and low density. The lithium isotope, lithium-7, must be used because of an undesirable neutron absorption resonance of the less common lithium-6. Also under study are: TZM (Mo-0.5Ti-0.08Zr-0.02C) for the reflector and control drums and refractory carbides or nitrides as the bearing material for a rotating drum control concept. Some materials for alternate reactor concepts are being studied to a lesser extent. These include: uranium dioxide (UO_2) as the nuclear fuel; other refractory metal alloys such as Cb-1Zr, ASTAR-811C (Ta-8W-1Re-0.7 Hf-0.035C), and W-25Re for fuel pin cladding; sodium as the coolant; and B_4C and TaB_2 as potential control rod materials. The control rod materials and sodium will not be discussed in this report.

The high temperature and long operating life goals put exacting demands on the materials of construction. The most serious potential problems probably are associated with the fuel pins. The fuel pin for the reference reactor design concept (fig. 1) is 1.9 cms (0.75 inch) in outside diameter and approximately 43 cms (17 inches) long with a fuel column length of approximately 38 cms (15 inches). The T-111 cladding is 0.15 cm (0.058 inch) thick. The fuel pin contains helium to promote heat transfer across the gap between the fuel and cladding. This 0.005 cm (0.002 inch) radial gap is necessary to allow sufficient clearances for assembly of the fuel pin. The tungsten liner between the UN and T-111 is 0.013 cm (0.005 inch) thick. This liner is needed to prevent chemical reaction of the UN and T-111 as will be discussed later in this report. Fuel burnups up to about four atom percent uranium are expected and a one percent diametral creep strain limit has been imposed on the cladding. A fluence of about 1.2×10^{22} neutrons/cm^2 ($E > 0.82$ Mev) is expected in the cladding.

Initially, it was considered that major potential problems with this type of fuel pin may arise in development of suitable fabrication methods from chemical reactions of fuel and cladding, from liquid metal corrosion, from fission-induced fuel swelling, and from deterioration of the properties of the cladding through irradiation damage. Therefore, work has been concentrated on determining the extent and effect of these problem areas. The major purpose of this report is to review the present status of knowledge concerning these potential problems and to identify the current major unknowns.

FABRICATION PROCESS DEVELOPMENT

To fabricate fuel pins it is necessary to select or develop suitable methods for preparation of the UN fuel forms, for fabrication of the T-111 cladding and fuel pin end caps, for application of tungsten liners to the inside diameter of the T-111, for assembly of the fuel pin, and finally, for inspection of the complete fuel pin.

Uranium Mononitride Fuel

Uranium mononitride is a relatively new fuel material compared to uranium dioxide. At the outset of this program, it was not clear how to routinely make UN fuel forms of high purity, single phase, close dimensional tolerances, and free from cracks and chips. Since that time, UN fuel forms have been prepared by two different methods at the Oak Ridge National Laboratory (working under NASA sponsorship). Both of these methods utilized powder hyperstoichiometric in nitrogen prepared by a hydriding-dehydriding-nitriding sequence on uranium metal. One method involved die pressing of the powder with a binder and subsequent vacuum outgassing and sintering. The other method consisted of cold isostatic pressing of the powder without a binder followed by vacuum out-gassing and sintering. The latter method is preferred because it consistently yields UN fuel forms of high purity (approximately 300 ppm oxygen compared to approximately 2000 ppm for the die-pressed material). Also, the fuel forms can be made with a greater length-to-diameter ratio and are free of cracks and chips. (The fabrication methods are described in detail in References 4 and 5.)

Examples of some of the specimens are shown in Figure 2. The larger of these is 3.8 cms (1.5 inches) long and 1.6 cms (0.62 inch) in diameter with a 0.5 cm (0.2 inch) diameter axial hole. This is the size required for the reference design concept fuel pin. Ten of these fuel cylinders are stacked to make the 38 cms (15 inches) fuel column. The smaller specimen, compared here to a standard paper clip, is 0.64 cm (0.25 inch) long and 0.13 cm (0.05 inch) in diameter with a 0.08 cm (0.03 inch) diameter axial hole. These and several other intermediate sizes are being used in the NASA experimental program.

At this point, it is felt that the fabrication technology of UN is adequately developed and does not present any problem to future development of a reactor of this type.

T-111 Cladding and Tungsten Liner

The T-111 tubing, sheet, and rod required for this program have been obtained from commercial vendors. The specifications are similar to those in Reference 6. (A useful survey of properties of T-111 is given in Reference 7.) Normally all T-111 material delivered receives a one hour, $1650^{\circ}C$, vacuum heat treatment to recrystallize the worked structure. The T-111 can be readily machined to close tolerances. After machining and prior to welding or assembly, T-111 parts are cleaned (with a mixture of two parts of 48 percent HF, four parts of concentrated HNO_3, one part of concentrated H_2SO_4, and two parts H_2O) then rinsed and air dried. The parts are then heated to $1090^{\circ}C$ for one hour in a vacuum of 2.6×10^{-3} newtons/meter^2 (2×10^{-5} mm) or lower to remove any volatile impurities. After welding, usually by electron beam but occasionally by gas tungsten arc welding, the T-111 assemblies are given a one-hour anneal at $1310^{\circ}C$ in vacuum. The purpose of this anneal is to combine any excess oxygen with the hafnium to prevent grain boundary corrosion of welds in liquid alkali metals. This is done as a standard post-weld operation.

Several methods have been investigated for applying the thin (0.013 cm, 0.005 inch) tungsten liners to the inside surface of T-111 tubing for fuel pin cladding. But only two of these have been extensively used. These are insertion of a loose, free-standing tungsten tube or utilization of a differential thermal expansion method to press a liner to the cladding. To produce the free-standing tube, multiple wraps of 0.0025 cm (0.001 inch) thick tungsten foil are pressed around a molybdenum mandrel in a high pressure autoclave at $1650^{\circ}C$ and a pressure of 2.1×10^{-8} newtons/meter^2 (30,000 psi). The outer surface of the tungsten can then be ground to a precise diameter. The finished tungsten-liner in the form of a thin-walled, free-standing tube is obtained by dissolving the mandrel in nitric acid. This tungsten tube of precise dimensions then can be slipped into the T-111 tube to be lined. The differential expansion method (see fig. 3) utilizes a cylindrical mandrel of AISI 1020 steel which is coated with alumina and ground to close tolerances. This mandrel is wrapped with 0.0025 cm thick foil to the desired total thickness, inserted in the T-111 tube to be lined, and heated in vacuum for one hour at $1200^{\circ}C$. Because the mandrel has a higher coefficient of thermal expansion than T-111, it presses the liner to the inside wall of the T-111 tube producing a mechanical bond. This method avoids the undesirable gap between the tungsten liner and the T-111 necessary for insertion of a free standing liner. Both of these methods are superior to chemical vapor deposition or direct hot isostatic pressure bonding of a tungsten liner on to T-111 because they are less prone to contaminate the T-111.

Fuel Pin Assembly and Inspection

Fuel pin assembly procedures are fairly straightforward. Some discussion of these procedures is given in Reference 8. It is important to avoid contamination of the parts at all steps in the assembly by interstitials such as nitrogen, oxygen, hydrogen, and carbon) and by metallics (such as copper and nickel). All heat treatments must be done in vacuum. A vacuum of 2×10^{-3} newtons/meter^2 (2×10^{-5} mm) is adequate for several heat treatments each of about one hour duration at 1310°C. Overheating of the UN fuel in the presence of T-111 (for example, in welding operations) must be avoided because the UN will decompose and contaminate the T-111. Welding of the end caps to the T-111 cladding is ordinarily done by electron beam welding. In instances where the pin is backfilled with helium, a fill-hole in one of the end caps is closed by gas tungsten arc welding in a chamber containing purified helium. For the most part, assembly procedures have been well worked out and present no major problems.

Inspection procedures currently being used include visual examination (for example, for cracks and chips in the UN pellets), dimensional measurements, ultrasonic testing (for example, of the T-111 tubing), dye penetrant testing, helium leak testing, and X-ray and neutron radiography. These methods are adequately developed for most of the inspections required. However, some problems still exist in detecting weld imperfections and inhomogeneties in the T-111 cladding. So more work is needed to improve inspection methods for these areas.

MATERIALS COMPATIBILITY STUDIES

Several combinations of reactor materials are being tested out-of-pile to determine their chemical compatibility with each other. Most of these tests are conducted at about 1040°C for times ranging up to 7500 hours. This test temperature is higher than the reference reactor coolant outlet temperature to allow for possible higher temperatures in the reactor core and to accelerate the effects of any chemical reactions. The preferred testing method is in a pumped-lithium loop because it best simulates the actual system concept as to temperature gradients and lithium flow rate. But, because of the high cost of this type of test, screening tests have been done using thermal convection loops and isothermal capsules, particularly in cases where reactions may occur which could cause damage to a pumped loop. Some combinations of materials being studied are: (1) T-111 and UN in direct contact, (2) T-111 and UN separated by tungsten, (3) UN in contact with lithium, (4) T-111 in contact with lithium, (5) TZM and T-111 in lithium, and (6) Bearing materials and T-111 with lithium.

Fuel-Cladding Compatibility

Because tantalum and hafnium form nitrides of greater stability than uranium nitride, T-111 and UN would be expected to react if they are in direct contact at the 1040°C test temperature. In view of this, the fuel pin is designed with a tungsten barrier between the UN and T-111. However, it is desirable to know how extensive the reaction is between UN and T-111 in direct contact and how effective a tungsten liner is in preventing this reaction.

The extent of the UN/T-111 reaction was tested in an evacuated isothermal capsule which consisted of a 1.3 cms (0.5 inch) diameter by 5 cms (2 inches) long T-111 capsule containing a cylindrical specimen of UN sandwiched between two T-111 cylinders (see fig. 4). The UN contained less than 100 ppm of oxygen impurity. Capsules of this type have been tested for 1000 and 2860 hours at 1040°C. The overall extent of reaction is shown in Figure 4. The microstructure (fig. 5) of the T-111 cylinder at the interface with UN after the 1000-hour test shows a second phase at the grain boundaries and in some of the grains. This occurs only at locations where the UN directly contacts the T-111 suggesting a solid-state reaction. The second phase is probably nitrides of tantalum and hafnium. The microstructure of a 2860-hour test specimen is similar and qualitatively shows about the same amount of reaction. These tests confirm that for long-term compatibility, UN should not be in direct contact with T-111 at temperatures of interest for this reactor concept.

To prevent this reaction, tungsten has been selected as the liner because UN is more stable than the nitrides of tungsten. Also, diffusion-rate calculations indicate that a 0.013 cm (0.005 inch) thickness of tungsten whould be sufficient to prevent significant diffusion of tantalum or hafnium from the T-111 to the tungsten-UN interface for 50,000 hours at temperatures to 1260°C. However, it is impractical to assure a hermetic envelope of tungsten around the UN. Because tungsten is a brittle material, cracks may occur. Assuming that cracks and discontinuities are present in the liner but that a gap is always maintained between the UN and T-111, any reaction will have to occur through the gas phase. Assuming that molecular flow of nitrogen is rate controlling, the calculated maximum increase of nitrogen in the 0.15 cm (0.058 inch) thick T-111 of the reference fuel pin at a crack site is less than one part per million by weight after 50,000 hours at 1040°C (see Appendix). This should cause no harm to the properties of the T-111. But the calculated nitrogen transport increases rapidly with temperature: 5 ppm at 1140°C, 200 ppm at 1240°C, and 5000 ppm at 1340°C. The higher levels of nitrogen could affect the T-111 properties. So the maximum fuel temperature should be limited to less than about 1300°C. The reader is reminded that in an actual fuel pin the fuel-cladding gap will contain about one atmosphere pressure of helium at standard temperature and pressure to augment heat transfer. The diffusion transport of nitrogen across this helium-filled gap will be slower than the molecular flow assumed in the calculation.

These results indicate that thermal dissociation of the fuel should not be a problem at expected reactor operating conditions and simple physical separation of UN and T-111 will avoid compatibility problems between these materials. This conclusion is supported by the results of tests on sealed T-111 capsules containing UN cylindrical specimens. Direct contact of the UN and T-111 was prevented by means of a tungsten wire coil around the UN specimens. These capsules were tested at 1040°C for about 3000 hours in a vacuum of 1.3 x 10^{-8} newtons/meter^2 (10^{-10} mm). Metallographic examination indicated no apparent effects of the test on either the

UN pellets or the T-111 capsule. A typical analysis of the T-111 from these capsules compared to T-111 from an empty capsule tested under the same conditions shows no significant change in the nitrogen content (see Table I). The same result has been obtained from pumped-lithium loop tests discussed later in this report.

Fuel-Coolant Compatibility

Normally, UN and lithium are not expected to be in contact with each other in the reactor because they are separated by the tungsten-lined T-111 cladding. But the consequence of a crack or hole in the cladding must be considered. If a reaction occurs between the UN and lithium, early failure might result. Pure UN and lithium are expected to be compatible; this is one of the main reasons for preferring UN over UO_2 for this type of reactor. However, to confirm this expected compatibility and to find the effect of oxygen impurity, tests have been made with isothermal capsules and with simulated fuel pins in a pumped lithium loop. The results of the loop test will be summarized in the next section.

The T-111 capsules used for the isothermal tests are of a cylindrical configuration ((1.3 cm (0.5 inch) diameter by 5 cm (2 inches) long)). These are filled with lithium which is purified by hot gettering and vacuum distillation to a typical impurity level of about 80 ppm oxygen, 40 ppm carbon, and 5 ppm of nitrogen. The UN fuel pellets immersed in the lithium are kept from contacting the T-111 with a coil of tungsten wire. Factors which may affect the compatibility of UN with lithium are test time, temperature, and purity of the UN. Oxygen is an impurity normally found in UN and is of concern because of the stability of lithium oxide.

An extreme example of the effect of oxygen impurity is shown in Figure 6 where UN specimens containing 6000 ppm of oxygen and showing UO_2 as a second phase were exposed to lithium at 1400°C for 20 hours. (The high test temperature was used to accelerate the rate of reaction.) Apparently, under these conditions, the lithium has removed the UO_2 from the UN. Whereas in the past, as-fabricated UN compacts ordinarily contained from 1000 to 2500 ppm of oxygen, UN is now available with less than 300 ppm of oxygen.

Unlike the 6000 ppm material, uranium nitride containing up to about 2500 ppm of oxygen usually does not show second phase UO_2 in the microstructure at 500X magnification. Specimens of UN containing about 250 ppm of oxygen and tested for about 1000 hours at 1040°C in T-111 capsules filled with lithium show no significant change in weight of the UN, in the chemical analyses of the UN or T-111, or in the microstructure near the surface of the UN (fig. 7). However, the microstructure of UN containing 2300 ppm of oxygen after testing for 1000 hours at 1040°C shows a thin layer at the surface of the UN (fig. 7). Similar tests on UN pellets containing 800 ppm and 1400 ppm of oxygen also resulted in this surface layer. Tests to identify the composition of this surface layer have not yet been successful. So more specimens tested for 3000 hours are being evaluated to check the composition of the surface layer.

It is concluded from the tests completed so far that UN containing less than about 300 ppm of oxygen should be used to avoid problems from reaction with lithium. This presents no difficulty from the standpoint of fuel fabrication.

Cladding-Coolant Compatibility

Generally, refractory metals gettered with Hf or Zr are compatible with alkali metal coolants providing that proper processing methods are used to prevent oxygen contamination. Thus, no major corrosion problems are expected with the use of T-111 and lithium. However, more subtle effects of long-term exposure of fuel elements to lithium might be encountered in this type of reactor system. For example, the combined effects of long-term aging plus the oxide reducing effects of lithium could affect the properties of the T-111. Therefore, long-term corrosion tests are being conducted.

The most meaningful tests of corrosion effects and effects on properties of materials exposed to liquid metals are made in pumped loops. A pumped loop test has the advantage of most closely simulating the conditions that would occur in an actual system--aside from the nuclear environment, of course. Temperature gradients and lithium flow rates can be adjusted to the values anticipated in an actual system. A schematic diagram of a T-111 loop built and operated by the General Electric Co., Nuclear Systems Programs, under NASA contract is shown in Figure 8. (For further details concerning this loop and testing procedures, see Reference 9.) Lithium is circulated by an electromagnetic pump through a coil where it is heated resistively, through a specimen test section at a velocity of about 1.5 meters per second at about 1040 °C, through a radiator section where the lithium is cooled to about 990 °C, and then back to the pump. The loop is operated in an ultra-high vacuum chamber at about 1.3×10^{-8} newtons/meter^2 (10^{-10} mm). The specimen test section contains three UN fuel specimens clad with tungsten-lined T-111 and also two ring-shaped specimens of TZM. All of the prime reactor materials except for bearing materials are included in the test section.

Seven simulated fuel pins (designated LT-1 through LT-7) were prepared. Five of these pins were tested in the 1040 °C pumped loop. Two of these pins (LT-1 and LT-3) were tested for 2500 hours, two (LT-5 and LT-6) for 5000 hours, one (LT-2) for 7500 hours, and two (LT-4 and LT-7) were left untested as controls. The design of the pins is essentially as shown in Figure 9 except that one of the 5000-hour test pins (LT-6) has a 0.6 cm x 0.008 cm (.25 inch x 0.003 inch) axial slot purposely cut through the cladding and liner to allow contact of UN and lithium during the test. The tungsten washers and the 0.013 cm (0.005 in) thick tungsten lining the cladding shown in the figure prevent contact of the UN and T-111. The dished washers provide an axial gap that prevents over-heating of the UN during electron beam welding of the end caps.

Evaluation has been completed on LT-1 and LT-3 after 2500 hours of testing and also on LT-4, the untested control. This evaluation is described in detail in Reference 10. Evaluation of LT-2, LT-5, and LT-6 is underway and some results are available. All of the specimens are shown in Figure 10 after removal from the loop. No corrosion of the T-111 cladding by the lithium is apparent from visual inspection or from weight change measurements. The results of chemical analysis of the T-111 from LT-1, LT-3, and LT-4 are listed in Table II. The cladding and end caps were analyzed separately because they were made from different T-111 stock. The only significant change in composition as a result of testing is a small decrease in oxygen content. The nitrogen contents have not changed within the precision of the analysis. This supports the expectation stated earlier that reaction of UN with T-111 is not a problem at 1040 °C when they are not in direct contact. The microstructure of the fuel and cladding from these 2500-hour test specimens are also unchanged by testing.

In the case of the slotted fuel pin (LT-6) which allowed contact of UN and lithium, no evidence of attack of the T-111 or UN is apparent from weight measurements or visual examination. The microstructure in the vicinity of the slot still has to be examined. It appears that a defected fuel pin will cause no significant compatibility problem.

Other Reactor Materials

Normally, the materials exposed to the lithium coolant in the reactor design concept will be T-111 in the cladding and structural components, TZM in the reflector and control drums, and a refractory carbide or nitride as the bearing surface for control drums. In the following two sections, the compatibility of the dissimilar metals TZM and T-111 in lithium will be examined and work on potential problems with bearing materials will be discussed.

Control Drum and Reflector Structural Material

In the T-111 pumped-lithium loop described earlier, specimens of the potential control drum and reflector material, TZM, have been tested for 2500, 5000, and 7500 hours. Preliminary evaluation of these specimens has been done. All of the specimens increased slightly in weight. The 2500-hour specimens gained about 4 milligrams, and the 5000 and 7500-hour specimens each gained about 7 milligrams. The original weight of each specimen is about 9 grams. Chemical analyses of the TZM specimens showed very small changes in interstitial content: carbon (217 ppm untested versus 213 ppm tested), oxygen (22 ppm untested versus 17 ppm tested), and nitrogen (2 ppm untested versus 12 ppm tested). In all cases, the hydrogen content was less than one ppm by weight. These tests show a high degree of compatibility of T-111 and TZM in flowing lithium at 1040 °C.

Bearing Materials

Several compositions are being considered as potential candidates for control drum shaft bearings in the space power reactor concept under consideration. Because these bearings have to operate in the lithium coolant, it is necessary that they also are compatible with the lithium at operating temperature. Also, it is necessary to know if bearing gliding surfaces will stick together after hold times of about 500 hours under load. Studies of these potential problems are underway at General Electric, Nuclear Systems Programs, under NASA contract. The bearing compositions under study are:

1. HfN + 10 w/o W

2. HfC + 10 w/o TaC + 10 w/o W

3. HfC + 10 w/o W

4. ZrC + 17 w/o W

5. HfC + 2 w/o CbC + 8 w/o Mo

6. HfN + 10 w/o TaN + 10 w/o W

These materials were selected for their potential hardness, toughness, high melting points, and thermochemical stability in lithium. Specimens of these materials were prepared by hot pressing mixed powder into compacts, sintering, and grinding into 5 cm x 0.62 cm x 0.62 (2 in x 0.25 in. x 0.25 in) pieces. These specimens were than tested in lithium-filled T-111 capsules for 500 hours at 980°C. All of the compositions appear to be compatible with lithium according to this test. Testing is being continued for 4000 hours at 870°, 980°, and 1090°C. Based on other factors as fabricability, density, metal distribution, and phase stability, two of these materials (90 HfN-10W and 90 HfC-2CbC-8Mo) have been selected for simulated out-of-pile service testing with full-scale control drums and actuators.

Because the control drums must always be free to move, the potential problem of diffusion bonding of bearing surfaces at their sliding interface must be investigated. Three bearing compositions (90 HfN+10W, 80HfC+10TaC+10W, and 90HfC+2CbC+8Mo) are to be tested for 500 hours in lithium in contact with each other and with TZM and T-111. The test temperature will be about 980°C and the load will be 9.7×10^{6} newtons/meter^2 (1400 psi).

So far, it appears from these isothermal capsule tests that all of the bearing materials are compatible with lithium in the presence of T-111 at 980°C for 500 hours. It remains to be seen if longer term (4000 hours) testing will show otherwise. Also, as yet none of these materials have been tested in flowing lithium with a temperature gradient.

IRRADIATION EFFECTS TESTS

An important basis for the design of fuel pins for the reference reactor concept is to limit cladding strain to one percent during an operating life of 50,000 hours to a fuel burnup of 4 atom percent. This low strain limit is imposed to allow for a large decrease in ductility of the cladding without rupturing the cladding and to limit the restriction of coolant flow around the fuel pins. Four experiments are underway to assess the effects of irradiation on: (1) fission-induced fuel swelling and fission gas release; (2) fuel pin integrity, dimensional stability, and materials compatibility; and (3) fuel pin cladding and structural materials properties. Each of these experiments will be described briefly, the objective stated, and the status outlined. Except where noted, the irradiations are being carried out in the NASA-Plum Brook Reactor Facility (PBRF).

Fuel Pin Irradiations

Three experiments are being done in this area: (1) accelerated burnup-rate tests of fuel-cladding materials on miniature fuel pins to obtain early information on the effect of irradiation on compatibility of materials, fission gas release, and fuel swelling; (2) fuel pin design proof tests on fuel pins to determine if the preliminary design shows promise of meeting the goal of 1 percent maximum diametral fuel pin strain; and (3) irradiation of fuel pins to find the effect of UN fuel density on swelling and fission gas release and to test Cb-1Zr cladding and UO_2 fuel under comparable temperature and burnup conditions.

Accelerated Burnup - Rate Tests

The capsule design and in-pile operation for the accelerated burnup-rate tests are described in Reference 11. The objectives of the tests, the materials, and the test conditions are summarized in Table III. Eight miniature T-111 clad UN fuel pins 0.46 cm OD x 2.5 cms long (.180" OD x 1" long) have been irradiated for 1500 hours to one atom percent (1 a/o) burnup (a burnup rate of about eight times that of the reference concept; i.e., 6×10^{13} fissions/cm^3 sec). Six of these were irradiated at 830°C and two at 990°C. Irradiation of four additional pins is being conducted at 990°C with a goal of 3 a/o burnup in 4500 hours. Preliminary results of the 1500-hour tests are: (1) no materials compatibility problems between the UN and cladding; (2) no detrimental effects of fission products; (3) less than one percent fission gas release from the UN; and (4) no measurable dimensional changes of the fuel or cladding.

Further tests are being prepared to obtain unrestrained UN fuel swelling data using a relatively weak cladding (like 304 stainless steel) at a 990°C irradiation temperature to a fuel burnup of about 1.3 a/o in 3000 hours. Also, it is planned to obtain partially-restrained UN fuel swelling data using a stronger cladding (such as T-111 or a tungsten alloy) at 1370°C and a test time of 4000 hours to a burnup of about 3 a/o. It is expected that measurable fuel cladding strain should be obtained in these small diameter fuel pins under these conditions. These data will be used to check analytical models for fuel swelling and fuel-cladding interactions (for example, see References 3 and 12).

Also, alternate materials, UO_2 and Cb-1Zr, are included in this program primarily to check the fuel-cladding compatibility of these materials under irradiation.

Fuel Pin Design Proof Tests

The objective of the fuel pin design proof tests is to find if the T-111 clad UN fuel pin design concept will meet the design goal of one percent maximum diametral fuel pin cladding strain in 50,000 hours at 990°C. Because it is impractical to test for 50,000 hours, irradiations are being carried out over the time range of 5000 to 20000 hours. But increased burnup rates and thinner claddings to accelerate tests are being included in the program to improve ability to extrapolate fuel pin performance to the 50,000-hour life goal.

To achieve higher potential cladding strain at the reference reactor design burnup rate (about 8×10^{12} fission/cm^3 sec) in 20,000 hours, some of the full-size pins (1.9 cms diameter) will have a T-111 cladding thinner than the design value (i.e., 0.102 cm compared to 0.146 cm). To achieve the design burnup in 20,000 hours or less and without an increased radial temperature drop across the fuel, smaller diameter pins (0.95 cm) are being used to permit testing at accelerated burnup rates of 1 to 4 times the design concept (i.e., 8 to 32×10^{12} fission/cm^3 sec). The cladding of these pins is about one-half as thick as that of the 1.9 cm diameter pins (i.e., 0.069 and 0.051 cms).

Three full-length (38 cm long fuel column) 1.9-cm diameter pins are to be irradiated. Data from these will be used mainly to check for axial fuel-cladding interaction.

Thirteen pins from this 28-pin program are now in-pile. Three of these pins have been irradiated for about 5400 hours. The maximum burnup at this time is about 0.5 atom percent. Irradiation is continuing. Neutron radiography after 5300 hours shows no diametral swelling (within precision of measurement of 0.005 cm) or any other problems.

Fuel Pins of Alternate Materials and Porous UN

One objective of this experiment (being done under an AEC-NASA Interagency Agreement by ORNL) is to compare the irradiation performance of fuel pins containing cored, high-density (about 96% dense) UN fuel to other pins with uncored, porous, low-density (about 85% dense) UN fuel. Another objective of the experiment is to test UO_2 fuel pins clad with tungsten-lined T-111 or with tungsten-lined Cb-1Zr. This should afford a comparison of the swelling behavior of UO_2 and UN under similar irradiation test conditions.

The entire experiment consists of three capsules each containing three pins. The capsule design is essentially as described in Reference 13. The fuel pins are 0.95 cm outside diameter x 11.3 cms long with a cladding thickness of 0.071 cm and a 0.008 cm thick tungsten liner. The fuel column in each pin is 7.5 cms long, and the amount of fuel is the same for the cored high density and the porous low density fuel. Two capsules are essentially duplicates and contain the T-111 clad UN pins. The third capsule contains three pins with UO_2 fuel: two are clad with Cb-1Zr and the other is clad in T-111. The pins are immersed in pressurized NaK in a Cb-1Zr container and are being irradiated in the Oak Ridge Reactor. The nominal cladding temperature of all pins during irradiation is 1000°C. The goal of the experiment is about 3 a/o burnup in 10,000 hours--an acceleration factor of about four times the reference reactor design concept. The experiment is designed such that the centerline temperature of the UO_2 will not exceed 1600°C. Because of the high thermal conductivity of uranium nitride, its centerline temperature will not exceed 1100°C.

At present, the two capsules containing UN pins are in-pile. One capsule has been in-pile for 2000 hours; the other for 1500 hours. The capsule containing the UO_2 pins is expected to go in-pile in July 1971. It is planned to neutron radiograph the capsules periodically and, after completion of the irradiations, to evaluate the pins for dimensional changes, fuel swelling, fission gas release, burnup, microstructural changes, chemical analysis of cladding, and ductility of cladding. Unirradiated thermal controls will also be tested and evaluated.

Cladding and Structural Materials Irradiation

In the last few years, irradiation experiments at elevated temperatures in a fast neutron spectrum have shown generation and growth of voids and changes in mechanical properties in many different types of metals, including the refractory metals (e.g., refs. 14, 15, and 16). These voids become visible under transmission electron microscope examination at fluences of about 10^{21} neutrons/cm^2 ($E > 0.1$ Mev). The shape and density of these voids is strongly influenced by the irradiation temperature, the thermomechanical history of the metal, and the alloy composition. Irradiation temperatures in the range of 0.3 to 0.5 of the melting temperature are generally required to generate voids. With increasing neutron fluence at constant temperature, voids tend to increase in size and concentration. But, with increasing irradiation temperature, voids tend to increase in size and decrease in concentration. These observations indicate that voids arise from vacancy coalescence. The vacancies are generated by the collision cascade resulting from fast-neutron interactions with metal ions.

Void generation and growth results in swelling, embrittlement (i.e., reduced tensile ductility and creep-rupture strain and increased ductile-to-brittle transition temperature), and microstructural changes such as precipitation. The magnitudes of these effects are not well known for the materials and conditions being considered here. Because the effects are large for other materials, this is considered to be the most serious potential problem for this reactor design concept.

Thus, an unfueled irradiation experiment is being conducted with refractory metals of interest for this reactor concept. The principal objective of this experiment is to determine the effect of fast neutrons at fluences comparable to those expected in the reference design concept (about 1.2×10^{22} neutrons/cm^2, ($E > 0.82$ Mev) on the ductility and swelling of the refractory metal alloys.

The experiment consists of two irradiation capsules designed and constructed by GE-NSP. Specimens are to be irradiated at fluences of 1 and 5×10^{21} n/cm^2 ($E > 0.82$ Mev) and temperatures of 1000° and 1270°C. The irradiations are to be carried out in the Plum Brook Reactor in an in-core shim rod position to take advantage of the relatively high fast flux (2.8×10^{14} neutrons/cm^2 sec ($E > 0.82$ Mev) at this position. The neutron spectrum is also tailored by use of cadmium and boron-10 filters to reduce the thermal and epithermal components. Heating of the specimens is to be achieved through γ heat. The temperatures are to be controlled by a flowing binary gas (argon and helium) mixture and measured by thermocouples in wells in the specimen containers. The specimen containers are constructed of TZM, and the specimens are immersed in lithium to achieve temperature uniformity. About 100 specimens of each material will be irradiated under each fluence-temperature condition. These include tensile, bend, and weld evaluation specimens of T-111 (Ta-8W-2Hf), ASTAR-811C (Ta-8W-1Hf-0.7Re-0.035C), TZM (Mo-0.5Ti-0.08Zr-0.02C) W-25Re, W-25Re-30Mo, and diffusion couples of T-111/TZM and T-111/W. The tantalum alloy, ASTAR-811C, and the tungsten alloys are included as potentially higher strength or more compatible materials for use at higher temperatures. A set of out-of-pile controls also will be run following approximately the same temperature and cycling history as the specimens in the reactor. The irradiated specimens and controls are to be evaluated for changes in tensile properties (especially the ductility) for ductile-brittle transition temperature, density, chemistry, and microstructure.

At present, the capsule design has been completed, the specimens are prepared, and capsule fabrication is underway. Irradiation testing should be initiated later this year.

SUMMARY OF RESULTS

Three major areas of a materials technology program underway at the NASA-Lewis Research Center for a liquid metal-cooled space power reactor concept have been discussed in this report. The areas cover fabrication process development, out-of-pile materials compatibility studies, and irradiation effects tests. Results and comments from these areas are briefly summarized in the following paragraphs.

Fabrication Process Development

A program for development of fabrication methods for fuel pins has given the following major results:

1. Satisfactory methods have been developed for routine fabrication of uranium mononitride fuel forms of about 95 percent theoretical density, free of cracks and chips, and with oxygen impurity contents less than 300 ppm.

2. Methods have been developed to apply a thin (0.013 cm) tungsten liner to the inner wall of T-111 tubing to be used for fuel pin cladding. The most suitable of these methods is a differential thermal expansion method which results in mechanical bonding of the liner to the cladding.

3. Fabrication of T-111 components and assembly and welding of fuel pins presents no serious problems. Nondestructive evaluation of finished fuel pins needs further development particularly in the area of detection of weld imperfections.

Out-of-Pile Compatibility Studies

Some results of out-of-pile compatibility tests at 1040°C for times up to 7500 hours are summarized below:

1. A 0.013 cm (0.005 in) tungsten barrier is effective in preventing reaction between T-111 and UN even if the tungsten is cracked. This conclusion is based on calculations from the known dissociation pressure over UN and simple kinetic theory considerations and has been confirmed with capsule tests.

2. Tests on simulated UN fuel pins clad with tungsten-lined T-111 in a pumped lithium loop show:

   a. No corrosion of the T-111 cladding.

   b. No contamination of the T-111 by nitrogen from the fuel.

   c. No significant changes in the microstructure of the fuel or cladding.

   d. No apparent effect of an artificial defect through the fuel pin cladding which allows contact between UN and lithium on the UN or corrosion of the cladding.

3. There is no evidence of reaction between UN and lithium in direct contact provided that the oxygen content of the UN is 300 ppm by weight or less.

4. TZM gains slightly in weight (less than 0.1%) when tested with T-111 in a pumped lithium loop. The weight gain was no greater after 7500 hours than at 5000 hours, so it may be self limiting.

5. Tests of bearing material candidates (in this case for 500 hours at $980^{\circ}C$) in T-111 capsules containing lithium show no compatibility problems.

Irradiation Effects Tests

Several irradiation tests are underway to determine the effects of irradiation on the fuel, cladding, and structural materials. These tests are in the beginning stages and only limited results are currently available. For fuel burnups to about one percent in a time of 1500 hours at $990^{\circ}C$, less than one percent fission gas release occurred from UN fuel. Also, no irradiation-induced compatibility problems with UN fuel in tungsten-lined T-111 cladding were observed under these conditions.

The area of greatest uncertainty is the effect of fast neutrons at fluences to about 10^{22} neutrons/cm^2 ($E > 0.82$ Mev) on the ductility and swelling of refractory metals at high temperatures. Tests of these effects will be initiated soon.

APPENDIX

THE CALCULATED MAXIMUM NITROGEN CONTAMINATION OF T-111 BY THERMAL DISSOCIATION OF UN IN 50,000 HOURS AS A FUNCTION OF TEMPERATURE

The expression:

$$G = 5.833 \cdot 10^{-2}\ P\ (M/T)^{\frac{1}{2}} \qquad \text{(see ref. 17)}$$

gives G the mass of gas incident on a unit area per unit time (in $gm \cdot cm^{-2} sec^{-1}$), where

P = pressure of the gas (in millimeters)

M = molecular weight of the gas

T = absolute temperature of the gas

The molecular weight of nitrogen is 28 and the pressure at the temperature of interest is obtained from extrapolation of the curve given by Inouye and Leitnaker (see ref. 18).

$T(^{o}C)$	P(mm)	$G(g.cm^{-2}sec^{-1})$	$G'(g.cm^{-2}/$ 50,000 hours)	Nitrogen (ppm in T-111)
1040	$1.7x10^{-12}$	$1.5x10^{-14}$	$2.7x10^{-6}$	1
1140	$9.9x10^{-11}$	$8.4x10^{-13}$	$1.3x10^{-4}$	50
1240	$3.8x10^{-9}$	$3.1x10^{-11}$	$4.6x10^{-3}$	2000
1340	$8.4x10^{-8}$	$6.5x10^{-10}$	10^{-1}	50000

The amount of nitrogen in the T-111 is calculated on the basis of a 0.15 cm thick cladding. The weight of one square centimeter of this cladding is about 2.5 grams (the density of T-111 is 16.7 grams/cm^3).

Inouye (ref. 19) finds a sticking coefficient of about 0.08 for nitrogen on Cb-1Zr. It is reasonable to assume that the sticking coefficient for nitrogen on T-111 is not any greater than about 0.1. Therefore, the amount of nitrogen in the T-111 will be one-tenth of the amounts given in the last column of the table after 50,000 hours of exposure.

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TABLE I

CAPSULE TESTS OF NITROGEN TRANSPORT FROM UN TO T-111 (3000 HOURS AT 1040°C)

Capsule	N(ppm)	O(ppm)	C(ppm)	H(ppm)
Empty Control Capsule	18	46	42	1.1
Test Capsule With UN Pellets	16	45	70	0.6

TABLE II

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 (LT-4)	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 (LT-4)	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

TABLE III

ACCELERATED BURNUP-RATE TESTS OF FUEL CLADDING MATERIALS ON MINIATURE FUEL PINS

Objectives:
1. Early information on effect of irradiation on compatibility of materials
2. Fission gas release data
3. Swelling data

Materials: UN/T-111; UN/Cb-1Zr; UN/W-25Re-30Mo; UN/304SS; UO_2/Cb-1Zr

Pin Size: 0.46 cm diameter x 2.5 cms long pins (0.38 cm outside diameter x 0.12 cm inside diameter cored fuel pellets)

Test Temperature: 830° to 1370°C

Burnup: 1 to 3 atom percent in times of 1500 to 4000 hours

Burnup Rate: 5 to 10 times reference design concept (i.e., 4 to 8 x 10^{13} fissions/cm^3 sec.)

E-6415

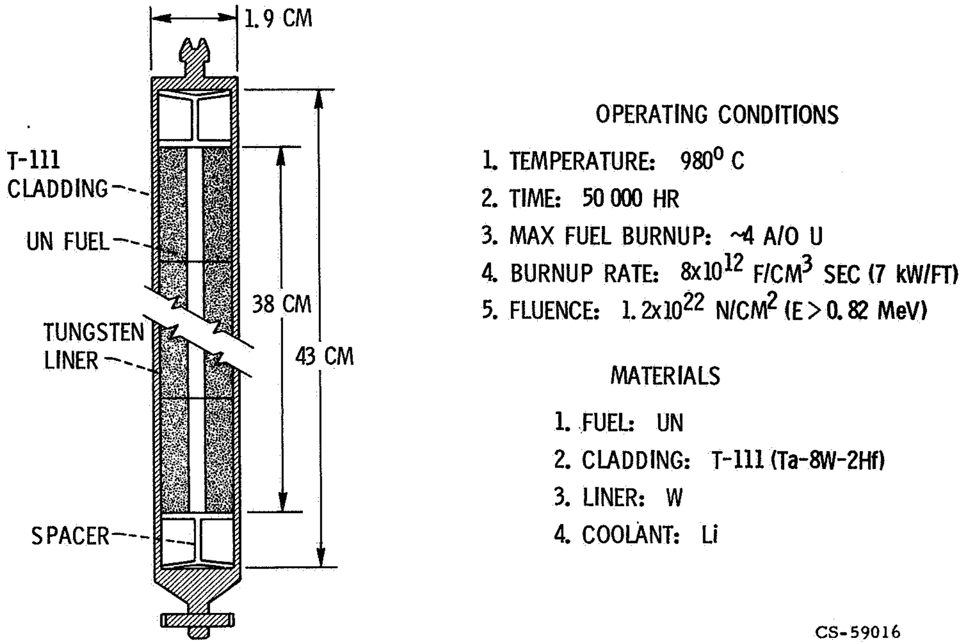


Figure 1. - Space power reactor fuel pin.

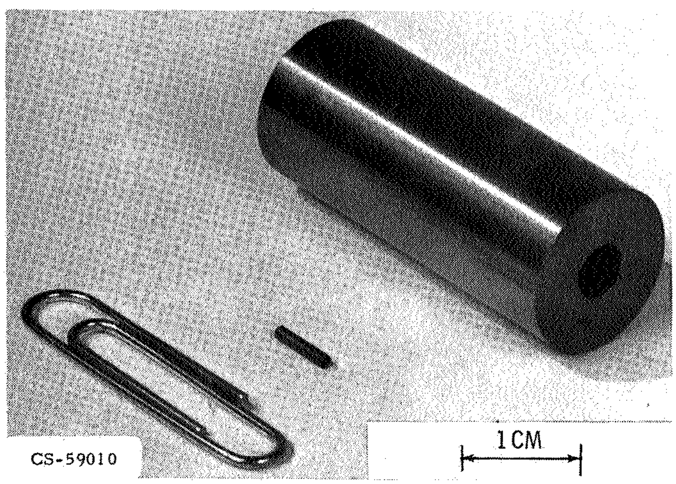


Figure 2. - UN fuel forms.

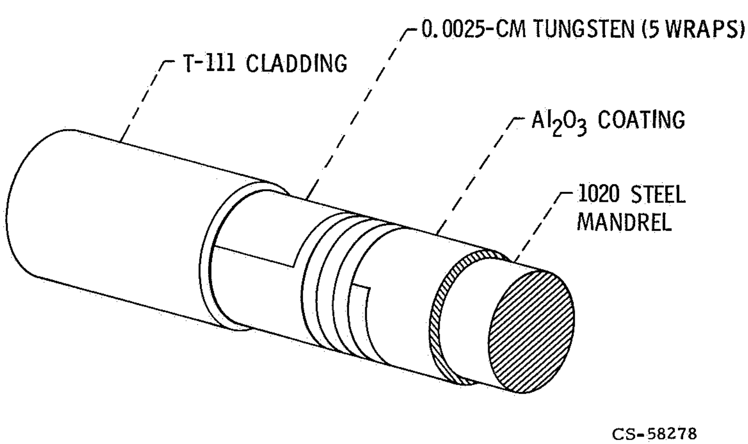
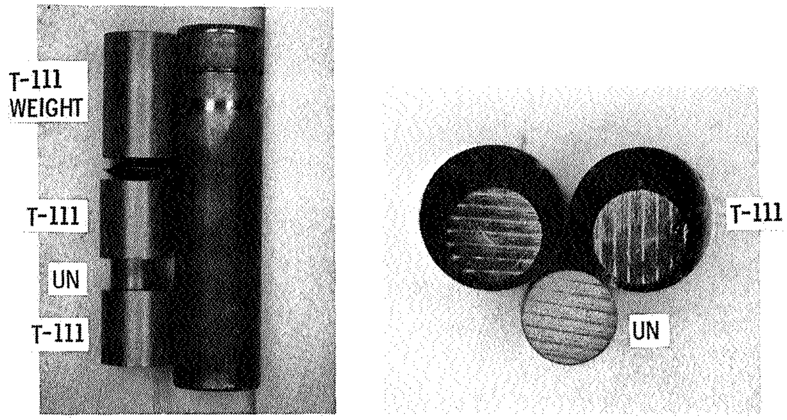


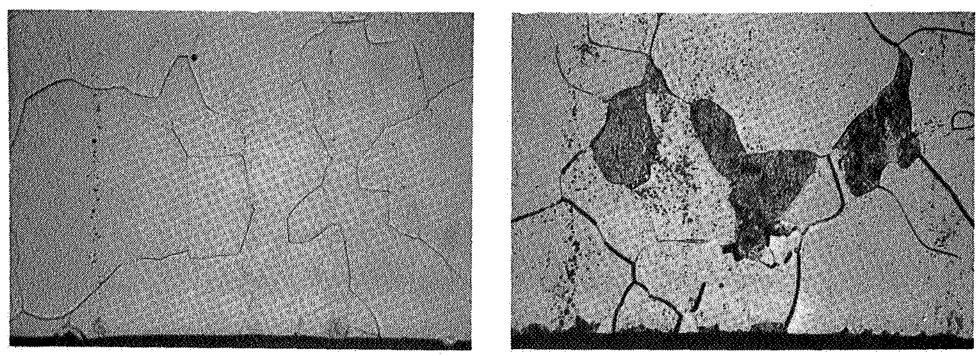
Figure 3. - Thermal expansion method for applying tungsten liners to T-111 cladding.



CAPSULE ASSEMBLY REACTION SURFACES

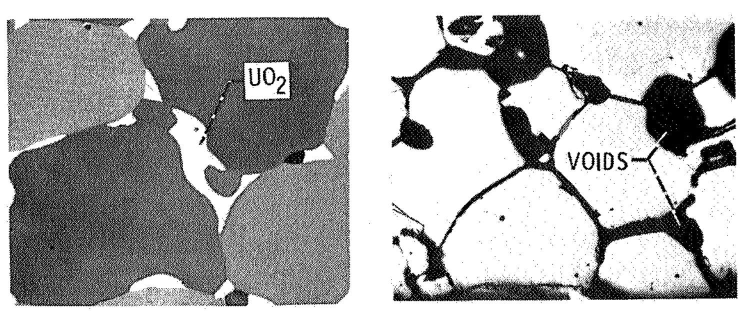
CS-58279

Figure 4. - Effects of T-111 in direct contact with UN 1000 Hr at 1040^{0}C.



T-111 NOT IN CONTACT WITH UN T-111 IN CONTACT WITH UN

Figure 5. - Microstructures of T-111 and UN in direct contact 1000 Hr at 1040^{0}C.



AS-SINTERED EXPOSED TO Li FOR 20 HR AT 1400^{0} C

CS-46307

Figure 6. - Effects of lithium exposure on UN specimens with 0.6 W/O O_2.

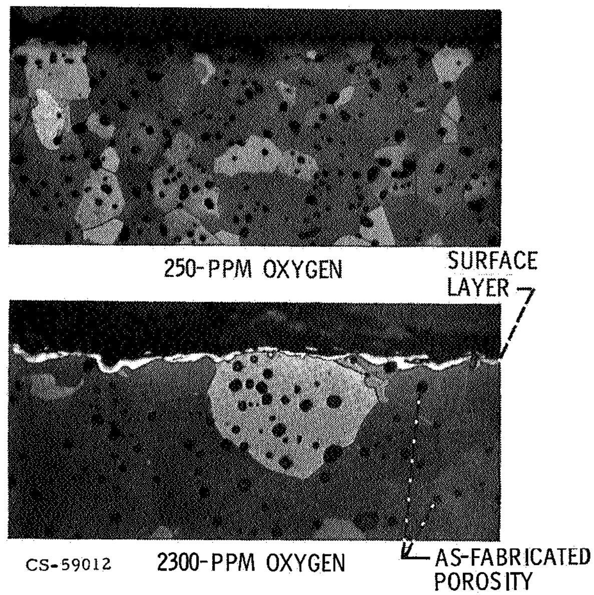


Figure 7. - Effect of lithium exposure on UN fuels with different initial oxygen contents. Lithium exposure at 1040° C for 1000 hrs. X500.

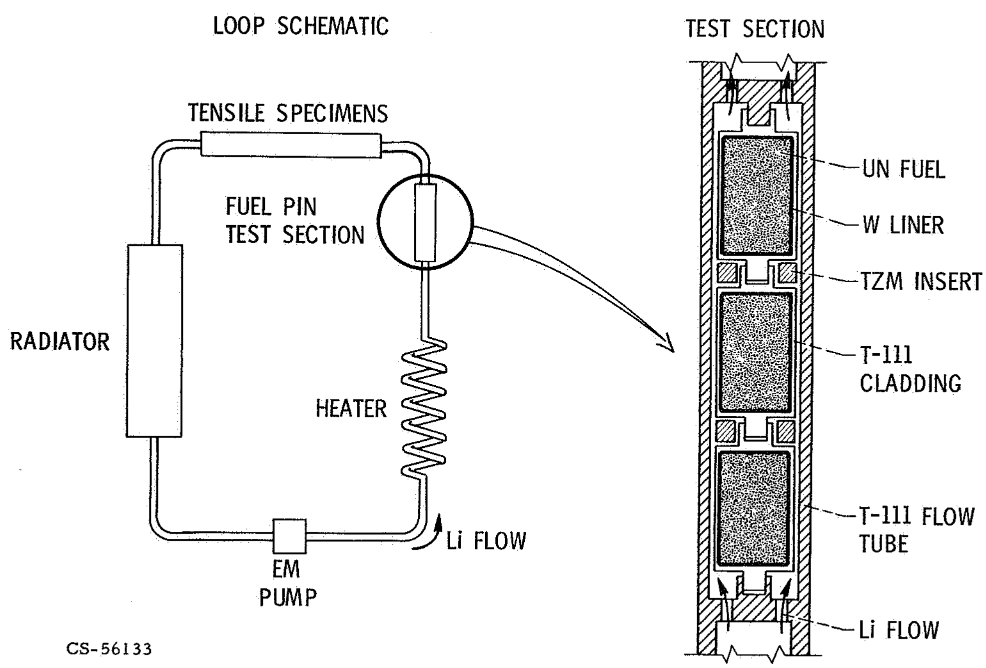


Figure 8. - 1040° C pumped lithium loop test.

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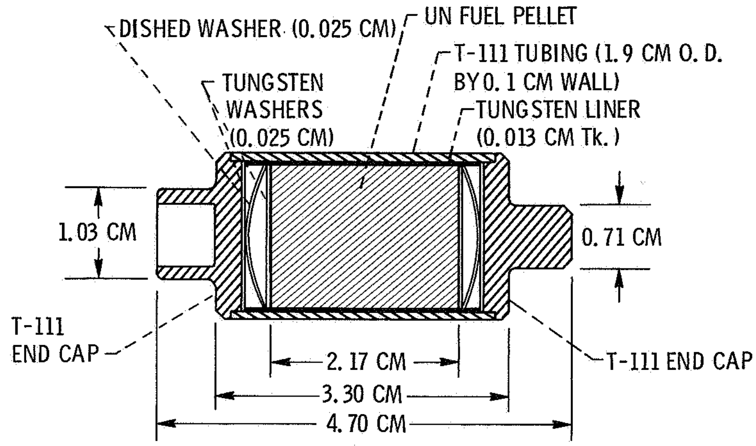

Figure 9. - Design of fuel pin specimen used in 1040° C pumped lithium loop test.

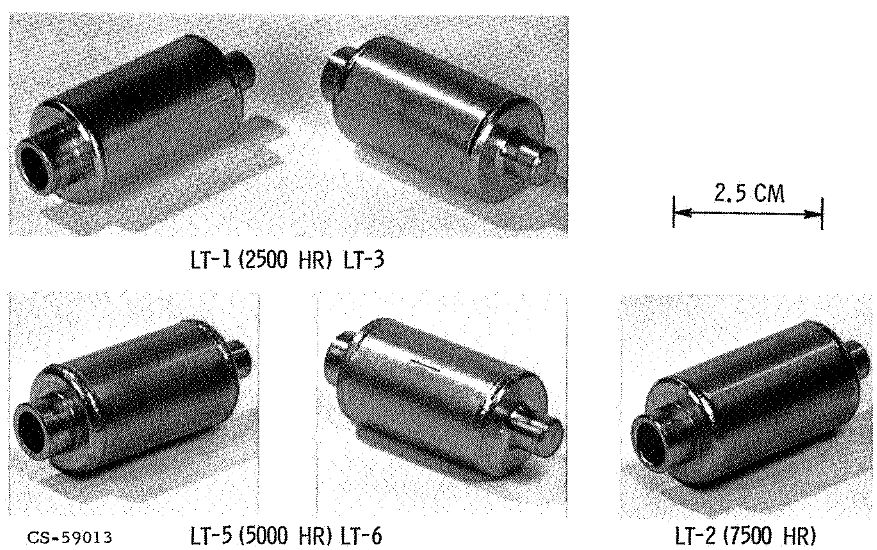

Figure 10. - T-111 clad UN specimens from 1040°C lithium loop.