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MATERIALS TECHNOLOGY FOR AN ADVANCED SPACE POWER NUCLEAR REACTOR CONCEPT - PROGRAM SUMMARY

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MATERIALS TECHNOLOGY FOR AN ADVANCED SPACE POWER NUCLEAR REACTOR CONCEPT - PROGRAM SUMMARY by Richard E. Gluyas and Gordon K. Watson Lewis Research Center

SUMMARY

A concept for a compact, lithium-cooled, nuclear reactor to operate for about 50 000 hours with a coolant outlet temperature of about 950⁰ C was studied for space power applications. This report summarizes the materials technology parts of this study with major emphasis on the development of fabrication methods and the testing of chemical compatibility, mechanical properties, and irradiation resistance of the fuel pin materials. Also, more limited work is reported on materials for control systems, reflectors, reactor vessel and piping, and other reactor structural elements. The major conclusions of this effort are summarized in the following paragraphs.

The most promising materials for fuel pins were found to be 85 percent dense uranium mononitride (UN) fuel clad with tungsten-lined T-111 (Ta-8W-2Hf). But further work is required to avoid hydrogen embrittlement of the T-111 cladding of fuel pins during irradiation testing at a cladding temperature of about 990° C and to determine the effects of fast neutrons on the behavior of fuel pins.

The effects of thermal aging, hydrogen and nitrogen impurities, and straintemperature cycling on T-111 have been studied. More information is needed on the effects of irradiation on the properties of UN and T-111 for design and modeling purposes.

No significant chemical compatibility problems were found for the fucl pin and reflector materials in static or flowing lithium. Based on compatibility tests, a promising bearing material (HfC-10TaC-10W) was found for a rotating fueled-drum control concept, and promising poison control materials (especially HfB_2 and ZrB_2) were found for a poison rod or drum control system concept.

Fabrication methods for fuel pins are well developed. More work is required to find a satisfactory joining method for the fuel pin support assembly and the thick section T-111 and to inspect welds.

INTRODUCTION

A compact, liquid-metal-cooled, fast-spectrum nuclear reactor concept for space power applications was investigated at the NASA Lewis Research Center. The objectives of this investigation were to establish feasibility of the reactor design concept, to develop necessary long lead-time technology, and to test critical components (see refs. 1 to 3). The materials technology program conducted in support of this investigation was concentrated primarily on the reactor design concept depicted in figure 1. Associated with this concept is a reactor power level of about 2 megawatts (thermal) and an assumed coolant outlet temperature of about 950⁰ C with the capability of temperature growth to about 1230⁰ C. (Capability of growth means either that the materials selected would be acceptable for operation at the higher temperature or that a rather straightforward substitution with other more suitable materials could be made.) Because of heat-transfer considerations, the maximum fuel element cladding temperatures $(\sim 990^{\circ} \text{ and } \sim 1270^{\circ} \text{ C}, \text{ respectively})$ would be slightly higher than the coolant outlet temperature. Also, an operating life of at least 50 000 hours was selected with a goal of 1 percent or less of diametral creep strain on the fuel pins. This life goal implies no harmful reaction between materials or degradation of properties which would lead to failure.

The selection of materials for the proposed reactor concept made use of information developed in previous, closely related, nuclear space power reactor programs such as the work at Pratt & Whitney - CANEL (e.g., ref. 4), Oak Ridge National Laboratory (e.g., refs. 5 and 6), and Lawrence Radiation Laboratory (e.g., ref. 7). The materials technology program concentrated primarily on the fuel element materials and included fabrication and processing studies, mechanical property studies, and chemical compatibility studies. In addition, in-reactor tests were conducted to investigate the effects of selected materials variables on the irradiation behavior of the fuel pins. Limited work was done on the control system and structural materials. This research work was conducted with both in-house and contracted programs. Major contractors included Oak Ridge National Laboratory (ORNL), the General Electric Company -Nuclear Systems Programs (GE-NSP), Westinghouse Astronuclear Laboratory (WANL), and TRW Materials Technology Laboratory. The materials technology program was not completed because of the cancellation of the nuclear space power reactor work at the Lewis Research Center. Significant progress was made, however, in a number of areas.

The main objective of this report is to summarize the results and status of the materials technology program for the proposed reactor concept. The major outstanding problem areas are discussed in the report along with suggested methods for solving these problems. The majority of the references in this report are to reports generated

out of the NASA advanced space power nuclear reactor program; however, some pertinent references from other sources are also included.

SELECTION, FABRICATION, AND EVALUATION OF MATERIALS

The grounds for selection of candidate materials and the status of a technology program for fabricating, testing, and evaluating these materials to determine if they meet the goals for the reactor are discussed in this section. Also, some materials investigations based on variations of the basic reactor concept (ref. 8) are presented - particularly variations of the control system, the 1 percent maximum fuel pin diametral creep strain goal, and the operating temperatures.

FUEL PIN MATERIALS

Selection of Materials

A drawing of the reference fuel pin configuration and a listing of the nominal operating requirements and the prime materials of construction are shown in figure 2. The selection of the fuel was based on the following considerations: density of fissionable atoms, thermal properties, stresses transmitted to the cladding because of fuel growth, chemical compatibility with the cladding and coolant, and fabricability. The thermal properties include heat capacity, thermal conductivity, thermal expansion, melting point, and thermal stability. Fuel growth can occur because of fission-product-induced swelling or, as observed in some cases with cermet fuels, as a result of thermal cycling. Ceramic fuels, in particular uranium dioxide (UO_2) , uranium monocarbide (UC), and uranium mononitride (UN), were selected as the candidate fuels to be considered for the reactor. Ceramic fuels were selected over metallic fuels primarily because of their high melting points and probable lower swelling on irradiation. Cermets were not considered because of their low fuel densities and the possibility of an excessive fuel growth problem from thermal expansion differences (thermal ratcheting). Plutonium compounds were not considered because of added difficulty and costs in development due to the reactivity and toxicity of plutonium. Other ceramic compounds, such as uranium monophosphide (UP) and uranium monosulfide (US), were not considered because less is known about them than about UO₂, UC, and UN, and because there are no obvious potential advantages of using these fuels.

The properties of the candidate ceramic fuels are shown in table I. The UN was selected as the prime ceramic fuel candidate. This selection was based primarily on

the following advantages: more successful irradiation studies have been made on UN in the temperature range of interest, UN has the highest fuel density, UN has good thermal conductivity, and UN was expected to be the most compatible with the lithium coolant. The UN undergoes thermal dissociation at high temperatures, but this is not regarded as a serious problem in the temperature range of interest (ref. 2). The UO_2 has been developed extensively for lower temperature applications (e.g., in commercial and naval reactors which operate at approximately 650° C), but little experience has been obtained at higher temperatures. Because of the low thermal conductivity of UO2, high local temperatures could lead to the release of a large fraction of the fission gas generated which might require a complex venting system to avoid stressing the cladding by high fission gas pressures. Also, UO, has a lower uranium density and is not compatible with lithium. The UC has high fuel density and thermal conductivity, but it has been reported to swell more than UN on irradiation (ref. 9). Also, UC is thermochemically less stable than UN. Thus, UC would be more prone to chemical reactions during fabrication and with the cladding and coolant materials during reactor operation. Although UN was selected as the prime fuel materials, UO_2 and UC are regarded as potential alternate fuel materials. For example, UO_2 might be used at lower temperatures with sodium as the coolant.

The severe environmental conditions in the proposed reactor concept and the limited information available on candidate materials made the selection of a cladding material more difficult. Because of the irradiation-induced swelling of nuclear fuels, it is necessary that the cladding material possess high creep strength to minimize the diametral growth of the fuel pins. The reference design concept goal limits the diametral creep strain of the fuel pins to less than 1 percent in 50 000 hours in the 990° to 1270° C cladding temperature range with a fuel burnup of ~3 atom percent of the uranium atoms initially present. In addition, the cladding material must have sufficient ductility to withstand the diametral growth without cracking. The purpose of the low diametral strain (\leq 1 percent) goal is to allow for an unknown amount of radiation-induced loss of ductility.

Almost any high-temperature metal or alloy could meet the strength requirements by using an adequate cladding thickness. But, because minimum practical reactor volume and total powerplant weight are desired for space use, consideration was limited to the refractory metal alloys. The metals considered included the following (alloy compositions included in this report are expressed in weight percent unless specifically noted otherwise):

- (1) Tungsten alloys: tungsten-rhenium (W-25Re) and tungsten-rhenium-molybdenum
 (W 25 at. % Re 30 at. % Mo)
- (2) Molybdenum alloys: TZM (Mo-0.5Ti-0.08Zr-0.03C),

TZC (Mo-1.2Ti-0.25Zr-0.15C), and molybdenum-rhenium (Mo-50Re)

- (3) Tantalum alloys: T-111 (Ta-8W-2Hf), T-222 (Ta-9.5W-2.5Hf-0.01C), and ASTAR-811C (Ta-8W-1Re-0.7Hf-0.35C)
- (4) Columbium alloys: columbium-zirconium (Cb-1Zr), PWC-11 (Cb-1Zr-0.1C), D-43 (Cb-10W-1Zr-0.1C), and FS-85 (Cb-28Ta-10W-1Zr)

The criteria for selection from these included previous experience, availability, fabricability (including weldability), mechanical properties (e.g., tensile and creep), compatibility with fuel and coolant, thermal stability, stability under irradiation, and neutronic properties. Initial selection from among these candidates was made on the bases of high-temperature creep strength, room temperature ductility, previous experience, and fabricability. Also, in the case of alloys of tantalum and columbium, only hafnium or zirconium "gettered" alloys were considered because of the corrosion characteristics of alkali metals; for example, Ta-10W was not included. Some estimated plots of stress to 1 percent creep in 10 000 hours are shown in figure 3. These are based on extrapolation of previously published data and some unpublished data obtained at NASA. The purpose here is to convey an idea of the relative creep strength of some of the refractory metals considered. The tungsten-rhenium alloy appears to be the strongest followed by the tantalum and molybdenum alloys. The columbium alloys are the weakest. Most of the tungsten and molybdenum alloys have poor room temperature ductility - if not in the base material, in the welds. The tungsten alloy W-25Re-30Mo was developed for improved ductility but, unfortunately, has considerably less strength than, for example, the tungsten-rhenium or tantalum alloys (ref. 10). The molybdenum alloys TZM and TZC show poor room temperature ductility. The carbon-strengthened TZC shows good creep strength, but this alloy has poor welding characteristics. The Mo-50Re alloy reportedly has good ductility and high creep strength, but there is little experience with this alloy and it is very expensive because of the rhenium content. (A curve for Mo-50Re is not included in fig. 3 because of insufficient data.) On the basis of creep strength, ductility, and fabricability, the more attractive alloys include the tantalum alloys T-111 and ASTAR-811C and the molybdenum alloy Mo-50Re. The columbium alloys could be considered if the 1 percent diametral creep strain, temperature, or reactor size goals could be relaxed. The D-43 alloy was not considered further because it has exhibited an overaging problem. Also, there has been some concern that alloys strengthened by means of carbide precipitates (e.g., TZC, T-222, ASTAR-811C, PWC-11, and D-43) may be subject to the transport of the carbon in hightemperature liquid-metal loops. Although the amount of experimental data on carbon transport is somewhat limited, carbon transport does not appear to be a problem with ASTAR-811C in a lithium loop operated at 1040°C for 7500 hours (ref. 11). No data are available for higher temperatures. The low ductility materials were not ruled out absolutely, but because of the greater difficulty in fabrication and handling of these materials, a more ductile material is much preferred if its other properties are acceptable.

The prime tantalum alloys considered are T-111 and ASTAR-811C. (T-222 shows some strength advantage over T-111, but it is not nearly as well developed or as available an alloy as T-111.) ASTAR-811C also is not as readily available or as well characterized as T-111, but it shows a significantly better high-temperature creep strength than T-111. Calculations on fuel pin swelling (ref. 12) indicate that 0.147-centimeter-(58-mil-) thick T-111 should meet the 990° C cladding temperature, 50 000-hour, 1 percent maximum creep strain goal but that ASTAR-811C would be needed to meet the higher (1270° C) cladding temperature goal. Because the main emphasis of this program was on the 990° C temperature condition and because of greater existing experience, T-111 was selected as the prime cladding material. In addition, because a chemical compatibility problem was expected between T-111 and UN at these high temperatures from thermochemical considerations, a thin tungsten barrier layer was used to separate the T-111 and UN. For the design application a 0.013-centimeter-(0.005-in.-) thick layer (liner) was considered adequate based on estimates of the interdiffusion rates of tantalum and tungsten.

Fabrication

Fabrication studies on fuel pin materials included developing methods of fabricating UN fuel, T-111 cladding, and tungsten-lined T-111 and of assembling and inspecting completed fuel pins. The objectives were to fabricate a large number (\sim 100) of fuel pins for a variety of tests and to establish feasible methods of producing fuel pins for the reactor design concept.

<u>UN fuel</u>. - The synthesis and fabrication of UN is complicated by the fact that the UN powders are easily contaminated by oxygen and water vapor; thus, the powders must be handled in an inert atmosphere. However, once the nitride powder has been consolidated into solid fuel cylinders, the cylinders are relatively stable in air and may be machined by conventional methods. At the start of this program no technique was available to rouinely produce high quality UN fuel forms to rigid specifications with respect to purity, density, dimensions, and integrity.

The problem of UN fabrication was investigated both in-house (ref. 13) and at ORNL (ref. 14) under NASA sponsorship. High purity (less than 100 ppm oxygen) UN powders were produced from uranium metal using a carefully controlled hydride-dehydridenitride synthesis process. Also, an isostatic pressing and sintering technique was developed for fabricating high density (~95 percent of theoretical) UN cylinders with large length to diameter ratios. These cylinders contained less than 300 ppm oxygen. After sintering the cylinders could be successfully machined to close dimensional tolerances by centerless grinding, and they were free from cracks or chips. The specimens also could be machined using electrodischarge machining (EDM) techniques.

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Examples of the size range of the UN cylinders produced are shown in figure 4 The larger cylinder is of the size required for the reference design concept fuel element. It has a 3.8-centimeter (1.5-in.) length, a 1.6-centimeter (0.62-in.) diameter, and a 0.5-centimeter- (0.2-in.-) diameter axial hole. Ten of these fuel cylinders would be stacked together to make the 38-centimeter (15-in.) fuel column. The smaller cylinder, shown next to a standard paper clip, has a 0.64-centimeter (0.25-in.) length and 0.13centimeter (0.05-in.) diameter with a 0.08-centimeter- (0.03-in.-) diameter axial hole. These small cylinders were used for fission gas release studies (ref. 15). Typical microstructures of ~95 percent dense UN are shown in figure 5.

Two other studies at ORNL were sponsored by NASA to investigate some other aspects of the UN fuel fabrication. One study (ref. 16) showed that low density (~ 85 percent) UN cylinders with thermally stable interconnecting porosity could be produced by modifying the existing fabrication technique. The sintering characteristics of the UN powder could be inhibited either by heat treating the powder prior to pressing and sintering or by using a high nitrogen content powder. In the second case the nitrogen content was reduced to the desired level during the sintering operation. The low density UN compacts produced in this study did not densify after 100 hours at 1400^o C.

In the other study (ref. 17) a preliminary investigation was conducted on using stabilizers to minimize the dissociation of UN and the loss of nitrogen at high temperatures (for possible future applications at temperatures above the range considered in this report). Samples of UN containing 10 mole percent zirconium nitride (ZrN) lost considerably less weight after 16 hours at temperatures as high as 1700° C than samples of unalloyed UN. A metallographic examination of the samples after testing showed much less free uranium in the stabilized UN; this indicated that the nitrogen loss was reduced by the addition of ZrN.

Problems in obtaining good agreement of UN chemical analytical results among different laboratories were encountered during the fabrication studies on UN. Two analytical roundrobins were conducted in an attempt to improve the chemical analytical techniques for UN. Identical samples of UN were analyzed for uranium, nitrogen, carbon, and oxygen by several laboratories. The results of the first roundrobin (ref. 14) showed that the poorest agreement was obtained on the major elements, uranium and nitrogen. In the second analytical roundrobin (ref. 18) standard procedures were developed so that each laboratory was using the same analytical methods. These procedures were as follows: uranium - oxidation, reduction, oxidation; nitrogen - Kjeldahl; oxygen - inert gas fusion, chromatographic finish; and carbon - combustion, chromatographic finish. When these methods were used improved interlaboratory agreement was obtained on the uranium, nitrogen, carbon, and oxegen analyses. Both the uranium and nitrogen analyses were reproducible to 0.04 percent compared to previous values of ± 0.15 and ± 0.08 per -

cent, respectively. For oxygen the reproducibility was ± 15 ppm at the 170 ppm level. And for carbon the reproducibility was ± 46 ppm at the 320 ppm level. The spread in the carbon results appears to be due to an unresolved bias among the laboratories. The goal was a ± 24 ppm or less spread in the carbon results.

<u>T-111 cladding</u>. - The T-111 alloy was developed in 1962 and is produced commercially in a variety of forms including tubing, sheet, plate, and rod. The processing development of this alloy has been well documented in the literature. The procurement specifications for the processing, composition, and inspection of T-111 used in this program were similar to those reported in reference 19. After the T-111 was processed, it normally was recrystallized 'or 1 hour at 1650° C in vacuum. A typical microstructure of T-111 is shown in figure 6.

The T-111 tubes purchased for the fuel claddings appeared to be of uniform high quality and were within the specifications. The dimensional tolerances specified for the drawn tubing, however, could not be as restrictive as the dimensional tolerances $(\pm 0.003 \text{ cm}, \pm 0.001 \text{ in.})$ specified in the reactor design and both the inside and outside diameters of the tubes had to be machined to the proper dimensions.

In general, little difficulty was encountered in machining T-111, and a large number of complex T-111 parts were produced successfully both for this program and for various corrosion-loop test programs. Typical machining conditions are given in reference 20. Some cracking problems were observed, however, when T-111 components were cut using a water-cooled alumina cutoff wheel (ref. 21). One possible explanation for this cracking was hydrogen embrittlement resulting from a breakdown of the coolant or the abrasive binder.

<u>Tungsten-lined cladding</u>. - Various methods have been investigated for lining the T-111 cladding with a thin layer of tungsten. Initial attempts to line the tubing by chemical vapor deposition indicated that contamination of the T-111 and accurate control of the liner thickness were major problems with this method. Other methods were then investigated in which the tungsten liner was produced using multiple wraps of 0.0025-centimeter- (0.001-in.-) thick tungsten foil. All together, four methods were developed for lining T-111 tubing using a foil wrap. But only two of these methods (hot isostatic pressing of a free-standing tungsten tube and a differential thermal expansion method) were used extensively in the fabrication of fuel elements. The third liner method (ref. 22) used the creep of an internally pressurized thin-walled tube to press the tungsten foil against the inside of the T-111 cladding. The fourth method, used primarily in the fabrication of certain test specimens (ref. 23), was the hot isostatic pressing of the tungsten foil directly onto the T-111.

The majority of the short- (up to about 11.5 cm) fuel pins fabricated for the UN irradiation studies for this program were lined using free-standing tungsten tubes. These tubes were produced by hot isostatically pressing multiple wraps of tungsten foil around an accurately machined, cylindrical molybdenum mandrel. Pressing conditions were

2. 1×10^8 newtons per square meter (3×10^4 psi) and 1650° C for 3 hours which resulted in solid-state welding between the foil wraps. After pressing, the tungsten-wrapped mandrels were centerless ground for a slip-fit into the T-111 cladding. Finally, the molybdenum mandrel was removed by dissolution in nitric acid leaving a thin-wall tungsten tube having very accurate dimensions. The tube then could be inserted into the cladding during final assembly. Some problems, however, were anticipated in using this liner method for full length, 42-centimeter (17-in.) fuel elements because the thin-wall tungsten sten liner might be broken easily during insertion.

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The method eventually used for lining full length cladding was the differential thermal expansion method described in reference 24. Tungsten foil was wrapped around an alumina-coated steel mandrel and inserted into the T-111 tube as shown in figure 7. During high-tempe cature exposure, the steel mandrel expanded more than the T-111 and forced the liner against the T-111. If the time, temperature, and pressure conditions were sufficient, solid-state welding occurred between foil wraps. As the assembly was cooled to room temperature, the mandrel contracted more than the lined tubing and was removed easily. An example of a full length Γ -111 fuel element cladding lined by the differential thern.al expansion process is shown in figure 8.

Assembly and inspection. - Assembly procedures for the T-111 clad UN fuel elements are well within the state-of-the-art technology. The major concern during assembly was to avoid contamination of the fuel element components. For example, the T-111 components can be contaminated easily and embrittled by interstitials (such as nitrogen, oxygen, hydrogen, and carbon) and by certain metals (such as copper and nickel) during welding and heat treating (ref. 21). Thus, these operations must be done in vacuum or in a high purity inert atmosphere and contact with copper and nickel must be avoided. Prior to assembly, the T-111 parts were thoroughly cleaned, acid etched, and then heated to 1090° C in vacuum (2.7×10⁻³ N/m² (2×10⁻⁵ torr) or better) for 1 hour to remove any volatile adsorbed impurities. Welding the T-111 end caps to the T-111 cladding was done by electron beam welding in a vacuum chamber evacuated to 6. 7×10^{-3} newton per square meter $(5 \times 10^{-5} \text{ torr})$ or better. In instances where the fuel element was to be backfilled with helium, the end cap to cladding welds still were made by electron beam welding. Then, a fill-hole in one of the end caps was closed by gas tungsten arc welding in a chamber containing purified helium. After welding, the T-111 normally was annealed for 1 hour at 1315° C in a vacuum of 1.7×10^{-3} newton per square meter $(2 \times 10^{-5} \text{ torr})$ or better. The purpose of this anneal was to prevent liquid metal attack of the welds by allowing the hafnium in the T-111 to getter any grain boundary oxygen picked up during welding. Recommended specifications for cleaning, welding, and heat treating of T-111 are given in reference 25.

The high density UN fuel cylinders were relatively stable and were not contaminated by exposure to air at room temperature. On the other hand, low density UN (~85 percent dense) having a large amount of open porosity could play up oxygen at room temper-

ature and therefore hac to be handled only in an inert atr:osphere. In addition, the overheating of the UN fuel in the presence of T-111 (for example, in welding operations) had to be avoided because the UN could have decomposed and contaminated the T-111.

Nondestructive inspection procedures used for the fuel elements included visual examination, dimensional measurements, dye penetrant testing, ultrasonic testing, helium leak testing, and X-ray and neutron radiography. Most of these methods were developed adequately for the required inspections. Some problems still exist, however. in examining the end cap to cladding welds and in detecting inhomogeneities in the T-111 cladding. The problem of obtaining accurate measurements from radiographs of the fuel elements was investigated in some detail by using an electronic image analyzer (ref. 26). Destructive examination included chemical analyses of control specimens processed with each lot of specimens and metallography of typical end cap to cladding welds.

About 100 UN fuel element specimens were fabricated to help evaluate the feasibility of the proposed reactor concept. The majority of these specimens were used in the various irradiation studies. The specimens ranged in size from relatively small fuel pins for accelerated burnup to ts to full-size prototype fuel elements. Details of the fuel element assembly, inspection, and quality control procedures are presented in references 20 and 27.

Properties

Most of the physical and mechanical properties of the fuel element materials were documented previously in the literature and were sufficient for conceptual design needs and for preliminary modeling studies of fuel swelling. Therefore, very little additional work was done in this program on these basic properties. Instead, the main emphasis in properties studies was to determine the effect of impurities and reactor environment on the mechanical properties of the T-111 cladding.

<u>UN fuel.</u> - The available physical and mechanical propertie of UN are well summarized in a 1972 review of nitride fuels by Bauer (ref. 28). This review, which also summarizes work on fabrication, compatibility, and irradiation of UN, includes most of the available references on the properties of UN. But some additional work probably is needed in the future to provide more refined mechanical property data on UN for use in advanced swelling models. This work on UN should include the creep strength as a function of stoichiometry, grain size, density, and purity, the measurement of Poisson's ratio, and the determination of yield strength as a function of temperature. In addition, work is needed on the effects of irradiation on the properties of UN.

<u>T-111 cladding</u>. - The various physical and mechanical properties of the cladding material T-111 have been well documented in the literature. A survey (ref. 29) pub-

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lished in 1970 is a good summary of the available information on T-111. The survey shows that T-111 has good high temperature strength, good low temperature ductility, excellent welding characteristics, and good resistance to alkali-metal corrosion. Additional creep data on T-111 (and on several other refractory metal alloys) are given in reference 30. The good creep strength of T-111 can be seen in the Larson-Miller plot presented in figure 9, which shows 1 percent creep life data. Elevated temperature tensile tests and some of the creep tests have shown that the T-111 alloy is sensitive to strain aging particularly in the 500° to 1100° C temperature range. A complex atmosphere-dislocation interaction involving solid solution oxygen has been suggested (ref. 31) as the mechanism responsible for this behavior.

Although T-111 appears to have relatively good properties for the proposed application, recent test results have identified some possible problems with T-111. As will be discussed in the fact section of this report, long-time aging of T-111 can result in decreased strength and increased sensitivity to hydrogen embrittlement. In addition, the application of low-amplitude, high-frequency fatigue vibrations during creep testing of T-111 significantly reduced the creep strength (ref. 32). It was concluded in this study that the creep rate acceleration resulted from a negative strain-rate sensitivity associated with the strain aging phenomenon in T-111.

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Effect of thermal aging on cladding. - Several studies have been conducted on the response of T-111 to thermal aging. One study (ref. 33) was conducted on the thermal stability of T-111 and other fabricable tantalum-base and columbium-base alloys. Both weld stability and base metal stability were investigated. The alloys were heated in ultra-high vacuum furnaces for up to 10 000 hours at temperatures from 815° to 1315° C. The effects of thermal aging were evaluated by bend testing, tensile testing, and metallography. Although some instabilities were noted in all the alloys, only the response of T-111 is discussed here.

Long-time aging at 980° and 1150° C increased the ductile-brittle bend transition temperature of weld zones in T-111, whereas aging at 815° and 1315° C had no effect (see fig. 10). The aging effects were less pronounced on electron-beam welds than on gas-tungsten-arc welds. The aging conditions appeared to have no effect, however, on the transition temperature of the base metal.

In a more recent investigation (ref. 34) on gas-tungsten-arc welds in T-111, aging $(1000 \text{ hr at } 1150^{\circ} \text{ C})$ appeared to have much less effect on the ductile-brittle bend transition tomperature than observed in the previous study. The reason suggested for the increased ductility was a slight improvement in the purity of the T-111 material. Other results obtained on notched and unnotched tensile test specimens showed no notch sensitivity at temperatures as low as -196° C for T-111 sheet specimens in the as-welded, post-weld annealed, and welded and aged condition.

The effects of long-term, elevated temperature exposure (aging) in vacuum and in

lithium on both tensile and creep properties of T-111 also were investigated as described in reference 35. Sheet specimens were exposed for up to 5000 hours in either vacuum or lithium at temperatures from 980° to 1315° C. The exposure conditions caused significant decreases in the tensile strength and creep life of T-111 in the test temperature range $(500^{\circ}$ to 1100° C) where strain aging contributes an appreciable amount to the strength of T-111. (Similar effects of aging on tensile strength c., welds and base metal were reported in refs. 33 and 34.) The explanation given for this strength decrease was that the exposure conditions depleted oxygen from solid solution in the T-111, thereby reducing the effectiveness of the strain-aging mechanism. The lithium exposures caused the greatest oxygen depletions. Some grain growth in the T-111 occurred in the long-time exposures at 1315° C which resulted in increased creep life in the test temperature range (above 1100° C) where grain boundary sliding occurred.

Although most of the studies on T-111 showed that aging had only a small effect on ductility, brittle intergranular cracking was observed in some cases when aged T-111 was stressed at room temperature following aging. Ring-shaped samples, cut from T-111 tubing from various parts of a lithium corrosion-test loop that had operated for 10 000 hours, showed a large variation in sample ductility (ref. 36). Rings cut from the 1040° C part of the loop were very brittle and fractured intergranularly with very little deformation during room temperature flattening tests. In contrast, rings from the 1200° C part of the loop were ductile and could undergo considerable deformation without cracking. Brittle behavior also was noted for rings cut from the T-111 cladding of simulated fuel element specimens which had been tested for 2500 hours in another lith-jum loop that operated at 1040° C in the test section (ref. 37, also see fig. 11).

The apparent brittleness observed in T-111 aged at 1040° C was investigated in detail and reported in reference 38. Samples of T-111 sheet and tubing were aged for up to about 3000 hours at 1040° C in vacuum and in lithium. Aging the T-111 at 1040° C was found to increase greatly the sensitivity of T-111 to hydrogen embrittlement. Trace amounts of hydrogen, with the resulting embrittlement, were picked up during exposure of the aged T-111 to water during cutting or sanding operations after aging. Testing of some aged T-111 specimens in a moist atmosphere also resulted in embrittlement. This embrittlement problem can be avoided, however, by preventing exposure of the aged T-111 to a source of hydrogen during post-aging processing or testing. For example, a T-111 ring cut from the cladding of the fuel element specimen exposed for 7500 hours in the 1040° C lithium loop test could be flattened completely when cut and tested in a dry argon atmosphere (ref. 39).

A metallographic study was included in reference 38 in an effort to determine why T-111 aged at about 1040° C is more sensitive to hydrogen embrittlement than T-111 aged at higher temperatures (1200° C or above). The major difference seen in the various T-111 samples was in the distribution of precipitate particles in the T-111. The

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T-111 aged at 980° to 1040° C contained numerous hafnium-rich oxide particles (presumed to be HfO₂) located primarily at the grain boundaries, whereas the starting material and the specimens aged at 1200° to 1315° C were essentially free of precipitates. One mechanism proposed for the sensitivity to hydrogen embrittlement was that the presence of free hafnium or hafnium-rich oxide particles could concentrate hydrogen at the grain boundaries which could lead to grain boundary failure.

Unpublished data obtained at the Lewis Research Center has shown that the sensitivity of T-111 aged at 1040° C to hydrogen embrittlement can be eliminated by heating the T-111 to 1315° C for 1 hour after aging at 1040° C. Apparently this heat treatment alters the microstructure produced by aging at 1040° C so that the T-111 is no longer sensitive to hydrogen embrittlement. Additional long-time aging at 1040° C after the 1315° C heat treatment, however, probably would resensitize the T-111 to hydrogen embrittlement.

Samples of T-111 following long-time exposures in the temperature range of 980° to 1315° C were examined using scanning and transmission electron microscopy and Auger electron emission spectroscopy in an attempt to determine the metallurgical factor influencing ductility (ref. 40). No classical aging response could be detected in the T-111 samples over the temperature range studied. Significant concentrations of silicon, potassium, and fluorine were found, however, at the fusion-zone grain boundaries of aged, gas tungsten arc welded samples displaying a tendency toward low temperature intergranular fracture. Other results of this study confirmed that aging at 1040° C increased the sensitivity of T-111 to post-aging hydrogen embrittlement.

The sensitivity to thermal aging and to hydrogen embrittlement following aging was investigated for eight tantalum-base alloys, including T-111, and two columbium-base alloys (ref. 41). The results again showed that T-111 was susceptible to aging embrittlement over a narrow temperature range near 1040° C. Of the alloys tested, only T-111 and a similar alloy (Ta-8W-3Hf) were embrittled by thermal aging for 1000 hours at 1040[°] C. However, all of the aged alloys tested, except for Ta-2Hf and Cb-1Zr, were sensitive to hydrogen embrittlement to some extent following aging. In the case of the tantalum alloys containing tungsten and hafnium, this appeared to be associated with precipitate particles in the grain boundaries. No such grain boundary particles were observed in the two alloys that were not sensitive to hydrogen embrittlement. Therefore, this study suggests that the binary alloys Ta-2Hf and Cb-1Zr are attractive alternate cladding alloys because they have good ductility after aging and because they do not appear to be sensitive to hydrogen embrittlement after aging. Their relatively low strengths at 1040[°] C, however, may limit their use. For example, the tensile strength of Ta-2Hf at 1040⁰ C is about one-half that of T-111 at the same temperature. Another possible alternative is the use of higher strength tantalum alloys that were shown to be less sensitive to hydrogen embrittlement than T-111. These alloys include the inter-

mediate strength, modified composition T-111 type alloys (Ta-8W-0.5Hf; Ta-8W-1Hf; and Ta-4W-2Hf) and the high strength ASTAR 811C alloy (Ta-8W-1Re-0.7Hf-0.35C).

Effect of impurities on cladding. - The problems associated with the contamination of tantalum- and columbium-base alloys by interstitials (carbon, hydrogen, nitrogen, and oxygen) are well known. The amount of interstitials in the alloy can have significant influence on the strength, ductility, and corrosion resistance. Although contamination of T-111 by all of the interstitials was of concern, only the effects of nitrogen and hydrogen were investigated as part of the materials studies for the proposed reactor. The effects of oxygen on the properties of T-111 welds and base metal are reported in reference 42. This study showed that the T-111 alloy could tolerate at least 200 ppm oxygen without any ductility impairment. At higher oxygen levels, the thermal history of the T-111 had a significant effect on the amount of oxygen that could be tolerated. For example, the bend transition temperature of T-111 containing 400 ppn. oxygen and aged 1000 hours at 980° C was below -196° C, whereas the transition temperature of the same material aged at 815° C was about 180° C.

The effect of nitrogen in T-111 is of concern because of possible contamination of the T-111 cladding by thermal decomposition of the UN fuel or by nitrogen released from the UN because of fissioning of uranium. Although insignificant contamination of the T-111 would occur from thermal decomposition at the proposed reactor operating temperature conditions, localized hot spots or temperature excursions could result in some nitrogen pickup by the T-111. Therefore, a study was conducted to determine the effect of nitrogen on the strength and ductility of T-111 tubing (ref. 43). Samples of T-111 tubing were doped with 80 to 1125 ppm nitrogen by heating the samples at about 1430⁰ C in a partial pressure of nitrogen. As expected, the tensile strength increased and the ductility decreased with increasing nitrogen content (see fig. 12). Additions of about 450 ppm or less of nitrogen, however, did not seriously affect either the tensile elongation values or the ductility of rings cut from the tubing and flattened at room temperature. Increasingly severe embrittlement occurred in both tests at nitrogen levels above 500 ppm. Thus, based on nitrogen transport calculations (ref. 2), the amount of nitrogen picked up by the T-111 as a result of thermal decomposition of UN at sustained UN fuel temperatures as high as about 1250° C would not have any adverse effects on the tensile strength and ductility of the T-111 cladding. No work was done, however, to determine the effects of long-time aging on the nitrogen contaminated T-111. Possibly, the thermal history of the T-111 could affect the amount of nitrogen that could be tolerated by the T-111 in a manner similar to that seen in oxygen contaminated T-111.

The influence of hydrogen on the ductility of T-111 was investigated because of unexpected room temperature embrittlement observed in some T-111 samples. As described in the preceding section, long-time aging at about 1040° C greatly increased the sensitivity of T-111 to hydrogen embrittlement. In the sensitized condition the T-111 could be embrittled by only trace amounts of hydrogen. The actual amount of hydrogen necessary for embrittlement could not be determined, however, because a small amount of hydrogen at the grain boundaries would not be detected using conventional analytical techniques. The bulk hydrogen analyses of aged and embrittled T-111 samples was, in many cases, less than one part per million. Annealing these samples in vacuum at temperatures as low as 870° C restored room temperature ductility.

Hydrogen embrittlement also was observed in unaged T-111 following hot sostatic pressing. During pressing, sufficient hydrogen was picked up from trace amounts of moisture in the high pressure helium to embrittle unaged T-111. The problem of hydrogen contamination during pressing and the effect of vacuum annealing on the hydrogen content of the T-111 were investigated as part of a study on tungsten-lined T-111 tubing (ref. 23). The T-111 samples contained more than 30 ppm hydrogen after hot isostatic pressing and were brittle at room temperature. Vacuum annealing was effective in removing hydrogen. As the annealing temperature was increased, the hydrogen content of the T-111 decreased and the ductility increased. These results showed that although T-111 is easily contaminated by hydrogen, a simple vacuum outgassing is sufficient to remove the hydrogen and to restore ductility.

Effect of tungsten liner. - The combination of high temperature during irradiation and pressure from fuel swelling could cause some solid state welding to occur between the tungsten liner and the T-111 cladding at some time in the life of the reactor. This solid state welding could occur regardless of the method used to apply the tungsten liner. A study (ref. 23) was conducted to determine what effect, if any, the tungsten liner would have on the properties of the T-111. Samples of T-111 tubing were lined with tungsten by hot isostatic pressing (1650[°] C and 2. 1×10^8 N/m² (3×10⁴ psi)) to assure welding between the tungsten and the T-111. The lined tubing was evaluated by tensile testing at temperatures up to 1315[°] C and by room temperature tube flattening tests. The liner had essentially no effect on the strength or ductility of the T-111. The effect of tensile test temperature on the behavior of the tungsten liner can be seen in figure 13. Note that at low temperatures the tungsten liner cracked during deformation, but the cracks did not propagate through the T-111. Several of the lined specimens were annealed for 3 hours at 1650° C to increase the width of the interdiffusion zone between the tungsten liner and the T-111. The resulting interdiffusion was about 0.007 centimeter (0.003 in.), which is somewhat greater than that expected in the fuel pins after 50 000 hours at the reactor design temperature. The results of ductility tests and tensile tests on these annealed specimens were similar to the results obtained on the aslined specimens. Thus, it does not appear that the tungsten liner will have any detrimental effects on the T-111 cladding.

<u>Effect of irradiation on refractory metal alloys</u>. - In the past few years, irradiation experiments at elevated temperatures in a fast neutron spectrum have shown g neration

and growth of voids and changes in mechanical properties in many different types of metals, including the refractory metals (e.g., refs. 44 and 45). These voids become visible under transmission microscope examination after exposure of the metal to fluences of about 10^{21} reutrons per square centimeter (E > 0.1 MeV). The size and denity of these voids are influenced strongly by the neutron flux, fluence, and energy spectrum and 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 alloy melting temperature are generally required to generate voids. Voids tend to increase in size and concentration with increasing neutron fluence at constant temperature. But voids tend to increase in size and decrease in concentration with increasing irradiation temperature at constant fluence. These observations indicate that voids arise from vacancy coalescence. The vacancies themselves are generated by the collision cascade resulting from fast-neutron interactions with metal ions.

Void generation and growth result 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. Therefore, performance limits cannot be defined. Because these effects are large for other materials, irradiation damage is considered to be a serious potential problem for the advanced space power reactor. Therefore, the design concept was based on the relatively small strain limit of 1 percent even though the tensile elongation capability of unirradiated T-111 is about 40 percent.

To determine the extent of irradiation effects, an irradiation experiment was initiated for refractory metals of potential interest for this reactor concept. The principal objective of this experiment was to determine the effect of fast neutrons at fluences comparable to those expected in the reference design concept (about 10^{22} neutrons, cm² ($E \ge 0.82$ MeV)) on the ductility and sv lling of selected refractory metal alloys. This experiment was not completed becaue of the cancellation of the program. However, the completed capsules with their Design and Hazard Analysis Manual and Operating Procedures are stored at the Plum Brook Reactor Facility for possible use in future studies of this type.

Compatibility and Corrosion

The various materials recommended for use in the proposed reactor concept must be chemically compatible with each other for times up to 50 000 hours. To evaluate the compatibility of these materials, out-of-pile tests were conducted at about 1040° C for times up to 7500 hours. This test temperature is somewhat higher than the coolant outlet temperature (950° C) of the reference reactor concept to allow for possible hot spots and to provide some acceleration of the compatibility testing. The preferred testing method was in a pumped lithium loop because it best simulated the reactor concept with respect to temperature gradients and lithium flow rates. But, because of the high cost of this type of test, the majority of the compatibility testing was done using isothermal capsules. The results of some of these tests are summarized in reference 2 along with a more detailed discussion of the compatibility problems. In addition to the 1040° C tests, a series of tests were conducted at very high temperatures (up to the melting point of UN) to determine what might happen during brief high temperature excursions.

Capsule tests. - Isothermal capsule tests were conducted as screening tests of the compatibility of the fuel element materials of prime interest. These tests were conducted for about 2800 hours at 1040° C in an ultra-high vacuum furnace (ref. 46). These tests showed that, even with very high purity UN, some reactions occurred when UN and T-111 were in direct contact and thus confirmed that a tungsten liner is required between the fuel and the cladding. As can be seen in figure 14, the areas of contact between a high density UN cylinder containing about 100 parts per million oxygen and polished T-111 surfaces are clearly defined. Metallographic examination of the contact region in T-111 showed a second phase precipitate thought to be hafnium nitride and/or tantalum nitride. This fuel-clad reaction could lead to embrittlement of the T-111 cladding. Other capsule tests in this study showed, however, that no reactions were observed which were due to vapor transport and that physical separation of the UN and the T-111 was sufficient to prevent reaction. Thus, cracks in the tungsten liner should not cause any compatibility problems providing the UN does not contact the T-111. Although tests have not been made to 50 000 hours, insignificant reaction is expected based on the known temperature dependence of the dissociation pressure of UN and simple kinetic theory considerations.

The isothermal capsule tests also were used to evaluate the effect of oxygen content in the UN on lithium compatibility. Samples of UN containing different amounts of oxygen (approx. 800, 1490, and 2300 ppm) were exposed to high purity lithium for about 2800 hours at 1040° C. No changes were detected in the microstructures or the surfaces of the UN samples. In addition, no attack of the T-111 capsule walls was observed. The only observed change was a large increase in the oxygen content of the lithium. Since most of this oxygen had to come from the UO₂ in the UN, the possible problem of free uranium production cannot be ignored. Thus, the best choice for fuel would be UN with the lowest oxygen content possible. Because UN containing less than 300 parts per million oxygen now can be produced routinely, this purity of fuel should be used.

The effect of vacuum level during testing on the contamination and compatibuity of lithium-filled T-111 capsules was reported in reference 47. In this study, the capsules

were heated for about 100 hours at 980° and 1260° C at vacuum levels ranging from 4×10^{-2} to 1.3×10^{-6} newton per square meter $(3 \times 10^{-4} \text{ to } 1 \times 10^{-8} \text{ torr})$. No lithium corrosion was observed in any of the capsules even though the bulk analysis of the oxygen concentration in the T-111 after testing was as high as 3600 parts per million. This is in contrast to previous studies of others (ref. 48) where lithium corrosion occurred in T-111 containing only 1500 parts per million oxygen (analysis before testing). In the more recent study, the oxygen was picked up slowly from the test atmosphere at the atmosphere/T-111 interface, and the lithium was able to getter the oxygen at the T-111/lithium interface at a rate such that oxygen content of the T-111 at the interface was quite low, corrosion did not occur. Although the increased oxygen content of the T-111 did not appear to influence its corrosion behavior, the oxygen is assumed to be detrimental to the mechanical properties of the T-111. Therefore, it was recommended in this study that for long-time tests the vacuum should be 2.6×10⁻⁵ newton per square meter (2×10⁻⁷ torr) or less.

<u>Loop tests</u>. - Although the isothermal capsule tests were useful for materials screening studies, they did not simulate the temperature gradients and lithium flow rates found in the reactor. Therefore, additional compatibility tests of the fuel pin materials were conducted under the more severe conditions present in a pumped-lithium loop.

Tungsten-lined T-111 clad UN fuel pin specimens were exposed in a 1040° C pumped-lithium loop for up to 7500 hours (ref. 11). A schematic drawing of the loop is shown in figure 15. Lithium was circulated by an electromagnetic pump through the loop as shown. In the specimen test section, the temperature was about 1040° C and the lithium flow velocity was about 1.5 meters per second (5 ft/sec). Two fuel pin specimens were tested for 2500 hours, two for 5000 hours, and one for 7500 hours. A cladding crack, which allowed the lithium to contact the UN, was simulated in one of the two specimens exposed for 5000 hours by an axial slot through the cladding and liner. Preliminary results from the specimens tested for 2500 hours are presented in reference 37. The final evaluation of results from all of the specimens is given in reference 39.

All of the fuel pin specimens after removal from the loop are shown in figure 16. No evidence of any corrosion of the T-111 cladding by the lithium could be detected by visual examination or by weight change measurements. Examination of the T-111 cladding microstructures also showed no evidence of any corrosion and no contamination of the T-111 by the UN fuel. Metallographic comparison of the UN before and after testing showed no apparent effects on density, grain size, or overall appearance.

Both the T-111 and the UN were analyzed before and after testing. The only significant change in the chemical composition of the claddings was a decrease in oxygen content which is typical of refractory metals exposed to lithium. No detectable changes

were noted in the composition of the UN fuel.

Examination of the UN fuel cylinder from the specimen containing the simulated cladding crack showed some loss of UN in the immediate area of the defect (fig. 17). The total amount of loss, however, was quite small, amounting to only about 0.03 percent of the weight of the fuel cylinder. The UN microstructure appeared eroded in this area, but there was very little evidence of any compatibility problems between the UN and the flowing lithium. Thus, no compatibility problems are expected in the proposed reactor because of fuel element cladding cracks.

<u>Off-design temperature tests.</u> - Tests were conducted on tungsten-lined, T-111 clad UN fuel specimens to determine the behavior of the fuel element materials at very high temperatures (ref. 49). These tests were run to help ascertain the maximum permissible fuel element temperature and to help establish requirements for reactor safety and emergency core cooling. A total of eight specimens was tested in vacuum over a temperature range of 1800° to 2700° C. Test times ranged from 1 minute to 8 hours.

Tests above 2500° C resulted in damage to the fuel element materia... with the amount of damage increasing with increasing time and temperature. A cross section of the test specimen heated to 2600° C for 10 minutes is shown in figure 18. At these high temperatures nitrogen is lost from the UN fuel leaving free uranium. The molten uranium can then migrate and attack both the liner and the cladding. No compatibility problems, however, were observed in the specimens tested at temperatures as high as 2400° C for times up to 8 hours. No free uranium was detected in the fuel, lining, or cladding. For these specimens, the cladding was ductile after the high-temperature exposure.

Based on these results, the fuel elements can probably tolerate brief temperature excursions to 2400° C without damage. But more work is needed to quantify these results.

Fuel Pin Irradiation Performance

As mentioned earlier, an important goal for the design of fuel pins for the reference reactor was to limit cladding diametral strain to 1 percent during an operating life of 50 000 hours to a fuel burnup of about 3 atom percent. This low strain limit was imposed to allow for a potentially large decrease in ductility of the cladding without rupturing the cladding and to limit the restriction of coolant flow around the fuel pins. Therefore, the fuel pin irradiation program included experiments to assess the effects of irradiation on fission-induced fuel swelling, fission gas release, fuel pin integrity, dimensional stability, and materials compatibility. The specific experiments carried out were the following:

(1) Sweep gas experiments to investigate the mechanisms of fission gas release

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- (2) Highly accelerated burnup rate tests on miniature fuel pins to obtain early information on the effects of irradiation on compatibility of materials, fission gas release, and fuel swelling
- (3) Fuel pin design proof tests to determine if the preliminary design and prime materials selected (i.e., tungsten-lined T-111 cladding and dense UN fuel) showed promise of meeting performance goals and to provide data for fuel pin modeling efforts
- (4) Evaluation of the effect of selected materials variables on the irradiation performance of fuel pins under comparable conditions

The results from these experiments are summarized and discussed briefly in the following sections.

<u>Fission gas release rates</u>. - Fission gas release rate measurements were made in a sweep gas facility (ref. 50) on small vented pins containing UN fuel. Fuel temperatures ranged from 330° to 1510° C and burnups ranged up to 8 atom percent uranium at high fission rate densities. Gas release rates were found to decrease slightly at burnups of up to about 1 atom percent and to increase linearly at burnups from about 1.5 to about 8 atom percent (refs. 15 and 51). The measured release rates were correlated with a preliminary analytical model. In reference 52 the model is refined further and correlated with available measured total gas release data obtained from the literature over a range of fuel temperatures from 1000° to 1390° C and fuel burnups up to 4.6 atom percent.

<u>Accelerated burnup rate tests</u>. - The capsule design and in-pile operation of the accelerated burnup rate tests are described in reference 53. Small fuel pins (~0.46 cm (0.18 in.) o.d.) consisting of UN clad with T-111 were irradiated for 1500 hours to a maximum burnup of 0.7 atom percent uranium and for about 3000 hours to a maximum burnup of about 1.0 atom percent uranium (ref. 54). The average cladding temperature for all fuel pins was about 930° C. Post-irradiation examination showed the following: (1) no cladding failures or measurable fuel swelling occurred, (2) less than 1 percent of the fission gas escaped from the fuel, and (3) the cladding of the pins irradiated for 1500 hours was ductile. The latter performance was attributed to improved fabrication procedures and reduced contamination of the second group of pins.

Unrestrained fuel swelling information was obtained to check fuel swelling models (e.g., ref. 12); these measurements were obtained as described in reference 55. Fuel pins of 304L stainless steel clad UN were irradiated for about 4000 hours to burnups ranging from 1 to 1.8 atom percent uranium. The average cladding temperature was about 830° C. At this temperature the stainless steel offers very little restraint to irradiation swelling of UN fuel. Under these conditions, the unrestrained swelling of all

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of the fuel pellets examined was less than 3 percent and appeared to be isotropic.

<u>Fuel pin design proof tests</u>. - These tests were made on T-111 clad dense UN fuel pins at a cladding temperature of about 990⁰ C. Reference 56 gives the results of postirradiation examination of three fuel pins after 8070 hours of irradiation to a maximum burnup of 0.9 atom percent uranium. Two of these pins were 0.914 centimeter (0.360 in.) in diameter and one was 1.822 centimeters (0.717 in.) in diameter. The fuel length was 5.72 centimeters (2.25 in.) in all cases. The larger diameter corresponds to that of the reference fuel pin. Under these conditions, the fuel swelled about 0.5 percent which was less than the fuel-cladding assembly clearances. Fission gas release was less than 0.05 percent. The cladding, however, was embrittled. This is believed to be due to sensitization of the T-111 to hydrogen embrittlement and subsequent embrittlement by hydrogen produced by the { $14N(n, p)^{14}C$ } reaction.

Reference 57 presents the results of examination of 27 pins similar to the previous ones which were irradiated for up to 13 000 hours to a maximum burnup of 2.34 atom percent. Although there was no evidence of incompatibilities between fuel, liner, and cladding, this cladding also was embrittled during irradiation. This embrittlement (together with the swelling fuel) resulted in cladding cracks and fission gas leaks in about one-half of the pins. Again the embrittlement was attributed to aging sensitization and hydrogen. In most cases, the UN fuel pellets remained intact and free from cracks. The amount of fission gas released from the fuel was low, generally less than 0.5 percent.

Alternate materials and porous UN. - The objectives of this experiment were to compare the irradiation performance of (1) T-111 clad fuel pins containing 95 percent dense UN fuel with pins containing porous 85 percent dense UN fuel and (2) T-111 clad UN, T-111 clad UO₂, and Cb-1Zr clad UO₂ fuel pins. The irradiations were carried out in the Oak Ridge Reactor at ORNL (fabrication and irradiation testing performed at ORNL under NASA-AEC Interagency Agreement 40-184-69) on 9 pins at nominal cladding temperatures of 990^O C, for times up to 10 450 hours, and to burnups up to 3.11 atom percent. The evaluation of these fuel pins and associated unirradiated thermal control pins is described in reference 58. The fuel pin description and results for four representative irradiated pins are summarized in figure 19 and table II. Also, some photographs of selected pins before and after irradiation are shown in figure 20.

The major results from this experiment are as follows:

(1) Post-irradiation examination of fuel pins of dense UN fuel clad with T-111 showed cladding cracking. For example, one pin irradiated for 10 037 hours to a burnup of 3.11 atom percent showed a large axial crack in the cladding. Based on fission gas monitoring during irradiation, it was estimated that the cracking of this pin first occurred at about 8500 hours (at about 2.63 atom percent burnup). Also, based on measurements of the crack width and of the axial profile of the cladding opposite the

crack, the cladding diametral strain at the time of cracking was estimated as about 1.5 percent. On the other hand, similarly irradiated fuel pins of 85 percent dense UN clad with T-111 showed cladding diametral strains of only about 0.3 percent and no cracking of the cladding. (The 95 percent dense UN compacts had an axial hole whereas the 85 percent dense UN compacts did not. All of the compacts contained the same amount of fuel per unit length.)

(2) Fuel pins of 95 percent dense UO_2 clad with T-111 or with Cb-1Zr showed no cracking of the cladding during irradiation for 8333 hours to a burnup of about 2.28 atom percent. The diametral strain on the T-111 cladding was about 0.5 percent and on the Cb-1Zr cladding was from about 0.5 to 1.0 percent.

(3) In all cases the T-111 was brittle as-irradiated. This is believed to be due to aging sensitization and hydrogen embrittlement. Except for the T-111 cladding on UO_2 , ductility could be restored by a 1-hour 1040° C vacuum outgassing. (There was some microstructural evidence of attack of the T-111 cladding by the UO_2 fuel.) The Cb-1Zr was ductile as-irradiated and was not sensitized to hydrogen embrittlement.

(4) The fission gas release from the UO_2 fuel was about 30 percent. But the release from 85 percent dense UN fuel was only about 4 percent. Values of fission gas release could not be measured accurately for 95 percent dense UN fuel because the fuel pin cladding cracked during irradiation. Calculations indicate that the diametral increase of Cb-1Zr clad UO_2 because of 30 percent fission gas release would be about 5 percent in 50 000 hours based on the unirradiated creep properties of Cb-1Zr and the conditions in the reference reactor. The shorter test time (8333 hr) and the external NaK pressure used in the test capsules limited the strain to 1.0 percent or less in this experiment (see point (2)).

(5) Fuel cracking occurred for the irradiated fuels but not for the thermal controls, possibly because of temperature gradients. The cracking problem was the most severe for UO_2 . Fuel cracking is undesirable because it may lead to redistribution of fuel.

<u>Discussion</u>. - Probably the major problem with the UN/T-111 fuel-cladding combination was the sensitization of T-111 to hydrogen embrittlement by long-term aging at about 990^o C coupled with the production of hydrogen by the (n, p) reaction with nitrogen 14. (The exception to this, the 3000-hr test discussed under "Accelerated Burnup-Rate Tests," may be due to the low (1 percent) burnup, the small size of the fuel pin relative to the capsule, and the fabrication of the T-111 fuel pin cladding from bar stock rather than drawn tubing.) This problem resulted in embrittled T-11. cladding on irradiated fuel pins. This probably should not be a problem in an actual reactor because the hydrogen should be lost by permeation through the hot containment vessel. But, for irradiation testing of fuel pins, special provision will have to be made to remove the hydrogen as it is generated because the irradiation test capsule walls are usually water-cooled and not permeable to hydrogen. It might be possible to accomplish this with a

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getter for hydrogen incorporated inside the capsule; for example, zirconium wire or turnings maintained at about 600° to 700° C. Furthermore, it has been found from out-of-reactor tests that T-111 is not sensitized to hydrogen embrittlement at 1315° C or perhaps as low as 1200° C. Further work would be required in this area.

Uranium mononitride appears to be a more promising fuel choice than uranium dioxide for this lithium-cooled reactor concept. In addition to its higher uranium density and compatibility with lithium, UN would release less fission gas during normal operation as well as during brief excursions to higher temperatures and it appears less prone to cracking. Porous UN fuel (85 percent dense with interconnected porosity) is preferred over dense UN fuel (95 percent dense with enclosed porosity) because it causes less cladding strain, probably because of a lower compression \oplus creep strength. Both T-111 and Cb-1Zr should be considered as potential candidate cladding materials for UN. The Cb-1Zr appears to be less sensitive to embrittlement than T-111, but it has a lower creep strength and therefore less temperature enowth capability. Both alloys require further intensive study of aging and hydrogen embrittlement effects. The combination of Cb-1Zr cladding with 85 percent dense UN has not been tested in this program. This might be a promising combination for applications at 990° C and lower and should be tested further if a similar program is resumed in the future.

These fuel pin irradiation tests were run in a thermal flux, and effects on cladding properties (ductility and creep strength) would not be expected to be as great as in a fast flux environment such as expected in the reactor concept. Also, the pins were not tested in lithium which is known to deplete T-111 of oxygen and alter its creep properties. Thus, further tests would be necessary to obtain quantitative measurements of cladding creep strain.

Coolant

The selection of the coolant was based on considerations of vapor pressure, pumping power requirements, pressure drop through the reactor core, convective film coefficient, compatibility with fuel, cladding, and other reactor materials, neutronic characteristics, and previous experience. Only liquid metals were considered primarily because of their superior heat-transfer characteristics and relatively low vapor pressures. And, of the liquid metals, the main consideration was given to the alkali metals lithium (Li), sodium (Na), sodium-potassium eutectic (NaK), and potassium (K) because of fairly extensive experience in this cour ry with these materials in flowing coolant systems.

Lithium was selected as the prime coolant candidate for use in the 950° to 1230° C coolant outlet temperature range The lithium isotope, lithium-7, must be used because

of an undesirable neutron absorption resonance of the less common lithium-6. The choice of lithium was made primarily because of its low vapor pressure over this temperature range, superior engineering properties (i.e., heat-transfer and fluid flow characteristics), and low density (ref. 59). The melting point of lithium (180° C) is well above room temperature: therefore, provision must be made to melt the lithium before eactor startup (ref. 60).

Sodium was selected as an alternate coolant candidate. But it is not recommended for temperatures above about 950° C because of the high vapor pressure which would require a heavier pressure vessel. The main advantage of sodium is the more extensive experience existing for this coolant. Sodium has been extensively studied throughout the world as a coolant for fast breeder reactors at temperatures of about 650° C. Also, sodium has been studied extensively for thermal reactors (e.g., the Seawolf). Therefore, much technology is available for sodium.

The principal areas of work on the lithium coolant were to further develop methods for purification, removal from test specimens, and analysis for impurities - especially oxygen. Commercial-purity lithium was purified further for use in the various loop tests and isothermal capsule tests associated with the advanced space power reactor program by a combination of hot trapping and vacuum distillation. Hot trapping, or gettering, of the lithium was performed in vacuum at about 800° C using either titanium or zirconium chips. The vacuum hot trapping resulted in substantial decreases in the oxyger, and nitrogen content of the lithium. Additional purification was obtained by vacuum distillation at about 700° C in either a titanium-lined or Cb-1Zr lined system.

Several methods were investigated for removing the lithium from the test specimens after the desired lithium exposures were completed. One unpublished study at General Electric - Nuclear Systems Programs on welded and unwelded T-111 test specimens showed that the lithium could be dissolved rapidly with liquid ammonia. The use of the ammonia caused little or no change in the impurity concentrations or physical properties of the test specimens. Removing the lithium with water resulted in brittle test specimens having much higher hydrogen concentrations. In another study on removal techniques (ref. 61) both liquid ammonia and vacuum distillation were proved satisfactory for removing lithium from T-111 aged at 1040° C without embrittlement or contamination of the T-111 (see <u>Effect of thermal aging on cladding</u> section, p. 11). Some T-111 specimens having traces of lithium on the surfaces, however, were embrittled by exposure to moist air.

A vacuum distillation technique also has been used to determine the oxygen content in the lithium (ref. 62). The nonvolatile impurities, assumed to be primarily lithium monoxide (Li_2O) , were separated from the lithium by distillation of the lithium. The residue was dissolved in water and the solution analyzed for alkalinity. This alkalinity was then used to calculate the oxygen concentration.

Control System

The prime control system studied for this reactor concept consisted of rotatable fueled drums inside the reactor vessel (ref. 1). But the use of poison control rods or drums, or of movable reflectors external to the reactor vessel, was also considered. Materials for movable reflector control systems are not discussed here because movable reflectors offer less reactivity control than fueled drums or poison control devices.

The control system selected must be capable of reliable operation throughout the reactor lifetime of 50 000 hours. Because of the high operating temperatures, potential materials compatibility problems and degradation of mechanical properties are of concern.

<u>Fueled rotating drums</u>. - The materials requirements for rotating fueled drums include material for a flexible bellows for a pressure vessel penetration device. structural materials for the drum and drum shaft, and a bearing material for reliable operation in lithium at high temperatures. The material selected for the bellows was T-111 because of its ductility, fabricability, and compatibility with lithium. The material selected for the drums was TZM because it is a good high temperature moderator. Both TZM and T-111 were considered for the drum shafts. Because of the lack of past experience in the application of bearing materials in a lithium environment at the temperatures of interest, several candidates were selected for evaluation (composition in wt. %): (1) HfC + 10W, (2) HfC + 10TaC + 10W, (3) HfC + 2CbC + 8Mo, (4) HfN + 10W, (5) HfN + 10TaN + 10W, and (6) ZrC + 17W. These materials were selected on the basis of their predicted thermochemical stability in lithium, their hardness, their possible toughness, their high melting points, and, in some cases, by analogy with tungsten-carbide-base alloys which have been studied at lower temperatures (ref. 63).

The control system employing rotating fueled drums operating inside the reactor vessel and immersed in the lithium coolant received the most attention for the advanced power reactor concept. This system is described in reference 1. The main potential materials problems in this system are associated with (1) the T-111 bellows in the reactor vessel penetration device to allow movement of drums through a seal by actuators external to this vessel, and (2) the bearing materials which must operate in hightemperature lithium - preferably at the temperatures prevailing in the reactor vessel to avoid complications involved in cooling the bearings.

It was found that the bellows may be susceptible to low cycle fatigue failure, particularly to cycling involving tensile deformation at high temperature followed by a compressive deformation at low temperature (ref. 64). Failure of the bellows must be avoided because it could lead to loss of the coolant.

The bearing materials must be compatible with lithium and other materials they contact, they must not diffusion bond and seize at mating surfaces, and they must main-

tain mechanical integrity. The bearing design considerations are given in reference 65. Isothermal compatibility capsule studies of six bearing material candidates with lithium (ref. 66) for times to 4000 hours and at temperatures to 1090° C show that a HfC-10TaC-10W cermet offers the best promise for meeting the 50 000-hour life requirement at about 950° C. The fabrication of the bearing material specimens is described in reference 67. Compatibility of bearing materials was not tested in pumped loops with a temperature gradient or under dynamic conditions (i.e., ruobing surfaces under load). At the interfaces the bearings will be under thrust loads up to about 1.38×10^{7} newtons per square meter (~2000 psi) and may have long dwell times during which no motion occurs (2000 hr was assumed as the maximum dwell time). Under these conditions the possibility of diffusion bonding is of concern.

Tests of three cermet materials (HfC + 10TaC + 10W, HfC + 2CbC + 8Mo, and HfN + 10W) in various combinations indicated that these materials do not diffusion bond to each other at temperatures up to 1200° C under a load of $1...6\times10^{7}$ newtons per square meter (~2000 psi) for up to 2000 hours (ref. 66). Similar tests of these materials with the potential drum and shaft materials, TZM and T-111, showed no bonding after 2000 hours at 980° C. However, at 1200° C the HfC + 10TaC + 10W was the only material that did not bond to T-111. And, in contrast to the other two cermet materials tested, it showed only slight evidence of bonding with TZM. In addition, for design purposes, the thermal expansion coefficients were measured for these three materials. The HfC-10TaC-10W composition appears to be the most promising bearing material based on the compatibility and diffusion bonding tests. Dynamic testing in a pumped lithium loop is needed to evaluate further the usefulness of this bearing material.

<u>Poison control rods and drums</u>. - Because of concern about the possibility of loss of coolant through failure of the penetration device for the fueled-drum control concept, the feasibility of using passively cooled, metal-clad, poison control rods moving in wells in the reactor vessel or of metal-clad rods in rotatable drums arrayed around the reactor vessel was examined. It was estimated that such rods or drums, if passively cooled by thermal radiation, would operate at about 1400° C for the reactor coolant outlet temperature of 950° C. Active cooling to achieve lower operating temperature (e.g., with an inert gas) leads to increased complexity and the possibility of failure of the control device because of loss of this gaseous coolant. Since no past work had been done on poison control materials at these temperatures, an evaluation of potential control rod materials was required. The materials selected for evaluation included boron carbide (B₄C) and the refractory metal diborides (e.g., HfB₂, ZrB₂, TaB₂) as poison materials and tungsten, T-111, TZM, and Cb-1Zr as candidate cladding materials. The poison control material probably would be in the form of cylindrical rods clad with a refractory metal somewhat analogous to a fuel pin.

Out-of-reactor studies were made on the compatibility of some candidate boron con-

trol materials with potential cladding materials (ref. 68). Specimens of B_4C , the most desirable control material because of its high boron content, were heated out-of-pile in contact with tungsten, TZM, T-111, and Cb-1Zr for times to 1000 hours. Unfortunately, all combinations were found to be incompatible at both the 1200^o and 1400^o C test temperatures. The compatibility of the diborides of tantalum, columbium, hafnium, and zirconium was also checked against the refractory metals. The best results were obtained with HfB₂ and ZrB₂ against tungsten. These combinations were compatible at temperatures up to 1650^o C for 1000 hours. For example, a comparison of the reaction zone in tungsten which was in contact with B₄C to the reaction zone in tungsten which was in contact with ZrB₂ for 1000 hours at 1400^o C is shown in figure 21. A preliminary check was made of the effectiveness of a thin layer of ZrB₂ between B₄C and tungsten as a compatibility barrier. This method shows some promise; however, further tests are required to see if a continuous barrier is needed (i. e., without cracks or discontinuities) to avoid vapor transport.

<u>Discussion</u>. - The materials problems associated with the fueled drum concept need considerably more study before materials could be selected for application in the advanced power reactor. The bellows section of the penetration device, especially, needs to be tested under conditions that simulate those of the application to evaluate reliability and failure mechanisms. There appears to be no serious compatibility or diffusion bonding problems with the bearing materials (especially for HfC-10TaC-10W); however, proof testing in pumped lithium loops and dynamic testing of bearing design concepts remains to be done. Also, irradiation effects in the bearing material candidates have not been evaluated.

Preliminary out-of-reactor studies indicate that HfB_2 and ZrB_2 clad with tungstenlined T-111 (or, perhaps, Cb-1Zr) show promise for poison control rods or drums for long-term operation at 1400° C. Also if higher boron density is required, HfB_2 and ZrB_2 show some promise as barrier materials to prevent reaction between W and B_4C . Further out-of-reactor work is required to confirm and develop these findings. Longer term compatibility testing is necessary. Fabrication development is needed for porous poison materials to facilitate venting of helium generated from the (n, α) reaction with B^{10} during irradiation to minimize swelling. Mechanical properties information will be needed for design purposes. Barrier fabrication methods may have to be developed, and the effectiveness of these barriers must be tested. Also, the performance of poison controls rods must be tested in a reactor in the 1400° C temperature range. The effects of irradiation on compatibility, poison material swelling, and cladding integrity need to be determined.

Some promising materials candidates for control systems and associated problems have been identified for further investigation. But, development of materials for control system concepts still requires a major effort.

Other Reactor Components

The unique core support structure and fabrication studies (particularly welding) for the core support structure are described in references 1 and 69. The prime material selected for the core support structure was T-111. This alloy was also selected for the reactor vessel and piping. Again the reasons for this selection are the good hightemperature strength, ductility, fabricability, and lithium corrosion resistance of T-111. Also, because T-111 was selected as the fuel pin cladding material, it was desired to avoid unnecessarily mixing materials to minimize chances for bimetal corrosion. The reactor vessel and piping are expected to have wall thicknesses of about 0.64 centimeter (0.25 in.). The molybdenum alloy TZM was selected for the internal reflector material because of its desirable neutronic properties. The Cb-1Zr was selected as a potential alternate material for the pressure vessel and piping in case (1) external reflectors or poison drums are used for reactor control (because of the lower neutron croos section of Cb-1Zr compared to T-111) and (2) the joining of T-111 in thick sections proved to be a serious problem.

Reflector

The main concern with using the molybdenum alloy TZM in the reflector was the possibility of bimetallic corrosion in circulating lithium in the T-111 primary reactor loop. If TZM were subject to lithium corrosion, it would be necessary to either protect it by a cladding with T-111 or choose a different reflector material. As described earlier in this report (Loop Tests of Compatibility and Corrosion of Fuel Pin Materials) and in reference 11, specimens of TZM were tested in circulating lithium at 1040° C for up to 7500 hours. The weight of the specimens increased slightly (<0.1 percent) as a result of the lithium exposure. This weight gain was no greater after 7500 hours than after 5000 hours so it may be self-limiting. Thus TZM appears to have acceptable compatibility in a lithium-cooled T-111 containment structure.

Core Support Assembly

The core support structure concept is described in reference 1, and a representative portion is shown in figure 22. It consists of a bundle of T-111 tubes each having a 2.16-centimeter (0.85-in.) diameter, a 0.025-centimeter (0.010-in.) wall thickness, and about a 45-centimeter (18-in.) length. Each tube has internal standoff projections on reinforcing inserts to position and restrain movement of a fuel pin. The tubes are joined together and to a header plate of T-111 which, in turn, is joined to the T-111

pressure vessel. Fabrication studies were carried out under a NASA contract (ref. 69) and in-house. Unfortunately, none of these fabrication attempts were successful because of distortion problems. Thus, it was concluded that fusion-type welds (electron beam and gas tungsten arc) do not appear suited for joining the tubes axially and then to the header plate because of the excessive localized expansion and deformation. Other joining methods such as brazing (perhaps using Cb-1Zr) or the use of fasteners need to be investigated.

Pressure Vessel and Piping

Fabrication and joining of heavy section T-111 plate and large diameter tubing were studied under NASA-supported contract programs. Tubing having a 10.8-centimeter (4.25-in.) outside diameter by a 0.318-centimeter (0.125-in.) wall thickness and a 7.6-centimeter (3-in.) outside diameter by a 0.20-centimeter (0.08-in.) wall thickness with a maximum single continuous length of 3.4 meters (11 ft) was produced (ref. 70). Another study indicated that heavy section T-111 is subject to underbead cracking in multipass welds (ref. 34). Modified joint designs showed minimal improvements. The authors conjectured that this problem was probably due to the relatively high hafnium content in T-111 because ASTAR-811C, with a lower hafnium content, showed no tendency to underbead cracking. They suggest that underbead cracking in T-111 welds might be alleviated by using a modified filler wire composition (i.e., lower hafnium content). More work is required to develop a successful process for joining thick section T-111.

CONCLUSIONS

The results and status of a materials technology program for an advanced nuclear space power reactor concept have been reviewed. General areas of work included fabrication studies, irradiation behavior, compatibility testing, and property measurement. The following major conclusions were drawn from the results of this program:

1. The most promising combination of fuel pin materials for the proposed reactor concept is porous UN fuel and a cladding of tungsten-lined T-111. However, the sensitization of T-111 to hydrogen embrittlement during long-time aging and the subsequent embrittlement by hydrogen from the (n, p) reaction with N¹⁴ in the UN fuel during irradiation presents a major problem with the use of T-111 cladding in fuel element test programs. This problem must be solved, possibly by removing hydrogen during irradiation or by modifying the composition of the T-111, before the capabilities of the

T-111 cladding can be fully realized. Also, the effects of a fast neutron environment on fuel pin performance must be evaluated.

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2. Fabrication, assembly, and welding processes for UN fuel pins with tungstenlined T-111 cladding are well developed. But some additional work is needed on the nondestructive evaluation of welds to assure freedom from cracks or flaws which could cause leaks during reactor operation. Also, more effort is required to solve joining problems in construction of the T-111 fuel pin support assembly and the T-111 reactor vessel.

3. No significant chemical compatibility problems are expected for the fuel pin materials (UN, tungsten, and T-111) and the reflector material (TZM) in cont. ct with lithium.

4. Adequate data on the physical and mechanical properties of unirradiated UN and T-111 for preliminary design considerations are available in the literature. More information is needed, however, on the effects of irradiation on the properties of UN and T-111 for more refined modeling studies. Also, further study is needed on the effects of thermal aging, interstitials, and thermal and strain cycling on the properties of T-111.

5. A cermet of HfC + 10TaC + 10W offered the most promise for use as a bearing material in a rotating fueled drum control concept. But additional tests are needed with this material; for example, pumped lithium loop tests to determine if carbon transport is a problem. Tests of boron containing compounds for use in poison reactor control concepts showed that HfB_2 and ZrB_2 are promising materials for long-time operation at 1400^o C in contact with a tungsten cladding. Under the same conditions, B_4C shows gross compatibility problems with any refractory metal.

Lewis Research Center,

National Aeronautics and Space Administration, Cleveland, Ohio, November 22, 1974, 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. Mayo, Wendell; Klann, Paul G.; and Whitmarsh, Charles L., Jr.: Nuclear Design and Experiments for a Space Power Reactor. NASA TM X-67857, 1971.

- DeCresente, M. A.; Freed, M. S.; and Caplow, S. D.: Uranium Nitride Fuel Development SNAP-50. PWAC 488, Pratt & Whitney, 1965.
- Weaver, S. C.; Scott, J. L.; Senn, R. L.; and Montgomery, B. H.: Effects of Irradiation on Uranium Nitride under Space-Reactor Conditions. ORNL-4461. Oak Ridge National Lab., 1969.
- Cuneo, D. R.; Long, E. L., Jr.; Jostons, A.; and Washburn, T. N.: Examination of Irradiated Uranium Nitride Fuel Clad with Tungsten-Rhenium or T-111 Alloy. ORNL-TM-3895, Oak Ridge National Lab., 1972.
- 7. Albrecht, E. D.; Rothman, A. J.; Lee, J. D.; Johnson, J. M.; and Hayes.
 W. N., Jr.: A High-Temperature Irradiation and Post-Irradiation Analysis of Uranium Nitride Fuel. UCRL-50727, Lawrence Radiation Lab., 1969.
- Davison, H. W.; Kirchgessner, T.; Springborn, R. H.; and Yacobucci, H. G.: Advanced Power Reactor Design Concepts and Performance Characteristics. NASA TM X-2957, 1974.
- Chubb, W.; Hilbert, R. F.; Storhok, V. W.; and Keller, D. L.: Fission Gas Swelling of Refractory Nuclear Fuels. Mater. Sci. Eng., vol. 9, 1972, pp. 293-300.
- 10. Buzzard, Robert J.: Effect of Grain Size on Creep Properties of a Tungsten 25-Atomic-Percent-Rhenium 30-Atomic-Percent-Molybdenum Alloy From 180C⁰ To 4000⁰ F (982⁰ To 2204⁰ C). NASA TN D-6285, 1971.
- Brandenburg, 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.
- Davison, Harry W.; and Fiero, Ivan B.: Calculation of Radiation Induced Swelling of Uranium Mononitride Using the Digital Computer Program CYGRO-2. NASA TM X-2224, 1971.
- Metroka, Robert R.: Fabrication of Uranium Mononitride Compacts. NASA TN D-5876, 1970.
- Tennery, V. J.; Godfrey, T. G.; and Potter, R. A.: Synthesis, Characterization, and Fabrication of UN. ORNL-4608, Oak Ridge National Lab. (NASA CR-72764), 1970.
- Weinstein, Michael B.; Kirchgessner, Thomas A.; and Tambling, Thomas N.: Fission-Gas Release from Uranium Nitride at High Fission Rate Density. NASA TN D-7171, 1973.

- Potter, R. A.; Tennery, V. J.; and Scott, J. L.: Sintering Studies and Fabrication of Uranium Nitride. ORNL-TM-4395, Oak Ridge National Lab. (NASA CR-121121), 1973.
- Fotter, R. A.; and Scott, J. L.: Continuation of Development of Nitrides for Space Nuclear Reactors. ORNL-TM-4496, Oak Ridge National Lab. (NASA CR-134498), 1974.
- Merkle, Emery J.; Davis, Warren F.; Halloran, John T.; and Grabb, Judson W.: Interlaboratory Comparison of Chemical Analysis of Uranium Mononitride. NASA TN D-7536, 1974.
- Frank, R. G.; Miketta, D. N.; Kearns, W. H.; Young, W. R.; and Hand, R. B.: Material and Process Specifications for Refractory Alloy and Alkali Metals. R66SD3007, General Electric Co. (NASA CR-88711), 1965.
- Keeton, A. R.; Stemann, L. G.; and Lessmann, G. G.: Capsule Assembly Fabrication. WANL-TNR-227, Westinghouse Aeronautical Lab. (NASA CR-72905), 1970, p. 38.
- Ekvall, R. A.; Frank, R. G.; and Young, W. R.: T-111 Alloy Cracking Problems During Processing and Fabrication. Recent Advances in Refractory Alloys for Space Power Systems. NASA SP-245, 1970, pp. 221-260.
- Maag, William L.; and Mattson, William F.: Lining Metal Tubes by Creep Forming. NASA TM X-2347, 1971.
- Buzzard, R. J.; and Metroka, R. R.: Tensile Properties from Room Temperature to 1315^o C of Tungsten-Lined Tantalum-Alloy (T-111) Tubing Fabricated by Hot Isostatic Pressing. NASA TM X-2964, 1973.
- 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.
- 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.
- Vary, Alex; and Bowles, Kenneth J.: Application of an Electronic Image Analyzer to Dimensional Measurements from Neutron Radiographs. NASA TM X-68200. 1973.
- Keeton, A. R.; and Stemann, L. G.: Fabrication of Fuel Pin Assemblies, Phase 3. WANL-M-FR-72-004, Westinghouse Electric Corp. (NASA CR-120956), 1972.
- Bauer, Arthur R.: Nitride Fuels: Properties and Potentials. Reactor Tech., vol. 15, no. 2, Summer 1972, pp. 87-104.

,

29. Moorhead, Paul E.; and Stone, Phillip L.: Survey of Properties of T-111 (Tantalum -8 Tungsten-2 Hafnium). NASA TN D-5873, 1970.

١

30. Sheffler, K. D.: Generation of Long Time Creep Data on Refractory Alloys at Elevated Temperatures. TRW Materials. Technology Lab. (NASA CR-72632), 1969.

1

- 31. Sheffler, K. D.; Sawyer, J. C.; and Steigerwald, E. A.: Mechanical Behavior of Tantalum-Base T-111 Alloy at Elevated Temperature. NASA CR-1436, 1969.
- 32. Sheffler, K. D.: Interactions Between Creep, Fatigue and Strain Aging in Two Refractory Alloys. TRW-ER-7451, TRW Inc. (NASA CR-120891), 1972.
- Lessmann, G. G.; and Gold, R. E.: Determination of the Weldability and Elevated Temperature Stability of Refractory Metal Alloys.
 Long-Time Elevated Temperature Stability of Refractory Metal Alloys.
 NASA CR-1608, 1970.
- 34. Gold, R. E.; and Lessmann, G. G.: Influence of Restraint and Thermal Exposure on Welds in T-111 and ASTAR-811C. WANL-PR(VVV)-001, Westinghouse Electric Co. (NASA CR-72858), 1971.
- 35. Buzzard, Robert J.: and Sheffler, Keith D.: Effect of Long-Time, Elevated-Temperature Exposures to Vacuum and Lithium on the Properties of a Tantalum Alloy, T-111. NASA TN D-7548, 1974.
- 36. Harrison, R. W.; and Smith, J. P.: Advanced Refractory Alloy Corrosion Loop Program. GESP-562, General Electric Co. (NASA CR-72818), 1970.
- 37. 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.
- 38. Watson, Gordon K.; and Stephens, Joseph R.: Effect of Aging at 1040^o C (1900^o F) on the Ductility and Structure of a Tantalum Alloy, T-111. NASA TN D-6988. 1972.
- Watson, Gordon K.: Evaluation of Tantalum Alloy Clad Uranium Nitride Fuel Specimens from 7500-Hour, 1040⁰ C Pumped Lithium Loop Test. NASA TN D-7619, 1974.
- 40. Gold, R. E.: Metallurgical Evaluation of Factors Influencing the Ductility of Aged T-111. WANL-M-FR-70-012, Westinghouse Astronuclear Lab. (NASA CR-120865), 1972.
- 41. Stephens, Joseph R.: Role of Hf and Zr in the Hydrogen Embrittlement of Ta and Cb Alloys. NASA TM X-68293, 1973.
- 42. Stoner, D. R.: Determination of the Weldability and Elevated Temperature Stability of Refractory Metal Alloys.
 3: Effect of Contamination Level on the Weldability of Refractory Metal Alloys NASA CR-1609, 1970.

Buzzard, Robert J.; and Metroka, Robert R.: Effect of Nitrogen on Tensile Properties and Structures of T-111 (Tantalum - 8-Percent-Tungsten - 2-Percent Hafnium) Tubing. NASA TN D-6999, 1973.

ļ

- Moteff, J.: Radiation Damage in Body-Centered Cubic Metals and Alloys. Metallurgical Society Conferences, vol. 37, Sec. F, Mechanical Properties, Gordon and Breach Science Publishers, Inc., 1967, pp. 727-808.
- 45. Wiffen, F. W.: Radiation Damage to Refractory Metals as Related to Thermionic Applications. ORNL-TM-3629, Oak Ridge National Lab., 1972.
- 46. Sinclair, John H.: Compatibility Tests of Materials for a Lithium-Cooled Space Power Reactor Concept. NASA TN D-7259, 1973.
- Gahn, Randall, F.: Effects of Low-Pressure Air on Oxygen Contamination and Lithium Corrosion of a Tantalum Alloy, T-111, at 980^o and 1260^o C. NASA TN D-7588, 1974.
- 48. Sessions, C. E.; and DeVan, J. H.: Effect of Oxygen, Heat Treatment, and Test Temperature on the Compatibility of Several Advanced Refractory Alloys with Lithium. ORNL-4430, Oak Ridge National Lab., 1971.
- 49. Bowles, Kenneth J.: Off-Design Temperature Effects on Nuclear Fuel Pins for an Advanced Space-Power-Reactor Concept. NASA TM X-2981, 1973.
- 50. Kirchgessner, Thomas A.; Weinstein, Michael B.; Tambling. Thomas N.: A Sweep Gas Facility for Fission Gas Release Studies at the NASA Plum Brook Reactor. NASA TM X-2267, 1971.
- 51. Weinstein, Michael B.; Kirchgessner, Thomas A.; and Tambling, Thomas N.: Fission-Gas-Release Rates from Irradiated Uranium Nitride Specimens, NASA TM X-2890, 1973.
- 52. Weinstein, Michael B.; and Davison, Harry W.: A Fission Gas Release Correlation for Uranium Nitride Fuel Pins. NA3A TN D-7401, 1973.
- 53. Rohal, Robert G.: The Design and In-Pile Operation of Instrumented Hydraulic Capsules. NASA TM X-52643, 1969.
- 54. Rohal, Robert G.; Tambling, Thomas N.; and Smith, Roger L.: Testing of Uranium Nitride Fuel in T-111 Cladding at 1200 K Cladding Temperature. NASA TM X-2732, 1973.
- 55. Rohal, Robert G.; and Tambling, Thomas N.: Unrestrained Swelling of Uranium-Nitride Fuel Irradiated at Temperatures Ranging from 1100 to 1400 K (1980 to 2520 R). NASA TM X-2907, 1972

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- 56. Slaby, Jack G.; Siegel, Byron L.; Gedeon, Louis; and Galbo, Robert J.: Irradiation of Three T-111 Clad Uranium Nitride Fuel Pins for 8070 Hours at 990^o C (1815^o F). NASA TM X-2878, 1973.
- 57. Slaby, Jack G.; Siegel, Byron L.; Bowles, Kenneth J.; and Galbo, Robert J.:
 Examination of T-111 Clad Uranium Nitride Fuel Pins Irradiated Up to 13 000
 Hours at a Clad Temperature of 990⁰ C. NASA TM X-2950, 1973.
- 58. Bowles, K. J.; and Gluyas, R. E.: Evaluation of Refractory Metal Clad UN and UO₂ Fuel Pins After Irradiation for Times up to 10 450 Hours at 990⁰ C. NASA TN D-7891, 1975.
- 59. Davison, Harry W.: Compilation of Thermophysical Properties of Liquid Lithium. NASA TN D-4650, 1968.
- 60. Lustig, Paul H.: PreStartup Thawing of the Lithium Cooling in a Nuclear Reactor for a Space Power Plant. NASA TM X-2177, 1971.
- 61. Gahn, Randall F.: Techniques for Lithium Removal from 1040⁰ C Aged Tantalum Alloy, T-111. NASA TM X-2775, 1973.
- 62. Gahn, Randall F.: Determination of Oxygen in Lithium by the Vacuum Distillation Method. NASA TN D-5197, 1969.
- 63. Wallace, M. J.: Potential Liquid Metal Bearing Materials for SNAP-50/SPUR Pumps. PWAC-468, Pratt & Whitney Aircraft Corp., 1965.
- 64. Sheffler, K. D.; and Doble, G. S.: Influence of Creep Damage on the Low Cycle Thermal-Mechanical Fatigue Behavior of Two Tantalum Base Alloys. TRW-ER-7592, TRW Equipment Labs. (NASA CR-121001), 1972.
- 65. Yacobucci, H. G.; Waldron, W. D.; and Walowit, J. A.: The Preliminary Design of Bearings for the Control System of a High-Temperature Lithium-Cooled Nuclear Reactor. NASA TM X-68218, 1973.
- 66. Sinclair, John H.; and Hendrixson, William H.: Evaluation of Cermet Materials Suitable for Lithium-Lubricated Thrust Bearings for High-Temperature Operation. NASA TN D-7655, 1974.
- Heestand, R. L.; Jones, R. A.; Wright, T. R.; and Kizer, D. E.: Preparation of Refractory Cermet Structures for Lithium Compatibility Testing. BMI-762-11, Battelle Memorial Institute (NASA CR-121130), 1973.
- 68. Sinclair, John H.: Compatibility of Refractory Materials for Nuclear Reactor Poison Control Systems. NASA TM X-2963, 1973.

69. Thompson, S. R.; and Young, W. R.: Fabrication Studies for T-111 Honeycomb Structure. GESP-570, General Electric Co. (NASA CR-72851), 1971.

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 70. Stoner, D. R.; and Buchman, R. W.: Development of Large Diameter T-111 (Ta-8-W-2Hf) Tubing. WANL-PR-222-001, Westinghouse Electric Corp. (NASA CR-72869), 1970.

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		Fuel	
	UN	U02	UC
Uranium density, g of uranium/cm ³ fuel Thermal conductivity, $W/(cm)(^{O}C)$ Melting point. ^{O}C	13.5 0.24 2850	9.67 0.025 2800	12.9 0.23 2375

TABLE I. - COMPARISON OF FUELS

TABLE II. - SUMMARY OF POST-IRRADIATION RESULTS ON SELECTED FUEL PINS

Fuel	Pre-irradiation fuel density, percent theo- retical density	Cladding (0. 069 cm thick)	Irradiation time, hr	Average measured cladding temperature, ^O C	Burnup, atom percent	Maximum clad diameter increase, ΔD/D - 100	Post-irradiation clad due, percont	Fost-irradiation clad duct after 1 hr in vacuum at 10:0 ⁰ C, percent	Fission gas release, percent
UN	84.8	T-111	10 450	976	2.76	0.3	1. 0	6.0	4.3
UN	94.3	T-111	10 037	991	3.11	^a 3.7	Brittle	Ductile	
UO ₂	95.2	T-111	8 333	^b \$42	2 28	.5	1. 0	Brittle	34.9
UO ₂	96.1	Cb-1Zr	8 333	^c 978	1.95	1.0	7. 0	8.0	26.4

^aCladding cracked.

^bIrradiated for 1500 hr at an average measured temperature of 845^o C. Value given is for subsequent 6833 hr.

Irradiated for 1500 hr at 887° C. Value given is for subsequent 6833 hr.

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Figure 1. - Compact fast reactor reference design.

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Figure 3. - Creep strength of some refractory metals and alloys for 1 percent creep in 10 000 hours.



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Figure 4. - Machined 95 percent dense UN fuel forms showing range of sizes fabricated.

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Figure 6. - Typical microstructure of T-111 cladding. Etchant: 30 grams ammonium bifluoride, 50 milli/iters nitric acid, 20 milliliters water. ×100.



Figure 7. - Schematic drawing of differential thermal expansion liner technique





Figure 8. - T-111 tube lined with tungsten by the differential thermal expansion lining technique for use as cladding for full-length fuel element.

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Figure 11. - Cross section of ring ductility test on T-111 cladding from fuel element specimen after 2500 hours at 1040° C showing intergranular cracking. Etchant - 30 grams ammchium bifluoride, 50 millititers nitric acid, 20 millititers water, =100.



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Figure 13. - Tungsten-tined T-111 tensile specimens tested in this study at various temperatures.



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Figure 15. - Schematic drawing of 1040⁰ C pumped lithium loop and fuel element test section,





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LT-5 (5000 hr) LT-6



LT-2 (7500 hr)

Figure 16. - T-111 clad UN fuel element specimens after exposure in 1040⁰ C pumped lithium loop.







Figure 18. Cross section of firel pin after 10 minutes at 2600° C Contrat portion of firel not shown, X20.





Figure 19. - Typical fuel pin for capsules UN-4, 5, and 6 and thermal aging tests. All dimensions given in centimeters.

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