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Quarterly Status Report on the

Space Electric Power R and D Program (U)

for the Period Ending July 31, 1971

Part II

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Los Alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

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ISSUED: August 1971

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QUARTERLY STATUS REPORT ON THE
SPACE ELECTRIC POWER R AND D PROGRAM (U)
For the Period Ending July 31, 1971

PART II

SUMMARY

Irradiation Capsule K-2 was disassembled after having been irradiated in the EBR-II (Experimental Breeder Reactor) to a maximum fluence of 4.6×10^{21} nvt ($E_n > 0.1$ MeV). The neutron-induced dimensional changes of the Y_2O_3 and Al_2O_3 specimens, and of the Al_2O_3 Nb-ceramic-to-metal seals have been determined. The results confirm earlier data: Y_2O_3 again showed no dimensional increase and AD-999 alumina swelled less than Lucalox. Further diagnostic measurements are in progress.

Irradiation Capsules K-3 and K-4 have been assembled and are scheduled for insertion into the EBR-II blanket in mid-August 1971. These units contain a total of 86 ceramic and trilayer specimens to be irradiated at 450°, 600°, and 750°C.

Thermal-diffusivity measurements on irradiated Y_2O_3 indicate a decrease in thermal conductivity with fast-neutron fluence in a manner similar to that of Al_2O_3 . At high irradiation levels and 750°C the thermal conductivity of Y_2O_3 is about two-thirds that of high-grade Al_2O_3 .

A 1.3-in.-diam trilayer specimen was autoclave-bonded without cracking the Al_2O_3 sheath or distorting the circumference of the specimen. A pentalayer test piece was fabricated by a combination of plasma-spraying and autoclave-bonding techniques.

Metallographic examination of the emitters from General Electric Co.'s in-pile thermionic-converter Test Units SCIP-518, TFE-619, and TFE-621 is under way. Initial results show marked interaction between arc-cast tungsten and UO_2 .

Four cermet-fuel irradiation capsules have been assembled for an ETR (Engineering Test Reactor) irradiation series in which the input power densities and irradiation temperatures will range from 100 to 275 W/cm³ and from 1550° to 1750°C, respectively.

Experiments to determine whether liquid lead will prevent excessive loss of hydrogen from $UZrH_x$ moderator rods have shown that lead is a very effective barrier.

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EBR-II ISOTHERMAL IRRADIATION TESTS

Insulator Irradiation Capsule K-2

Irradiation Capsule K-2 was removed from Position 7B3 in the EBR-II reactor and has been returned to LASL. Neutron fluences for this capsule ranged from 2.0 to 4.6×10^{21} nvt ($E_n > 0.1$ MeV). The capsule contained the following test specimens, all of cylindrical shell configuration:

Quantity	Type of ceramic or seal
5	Coors AD-999a Al_2O_3
7	Coors AD-999b Al_2O_3
3	GE Lucalox (fine grain)
1	Feldmühle E-37 Al_2O_3
5	Y_2O_3
2	Nb/Al-995 seal brazed with Nb-V eutectic alloy
2	Nb-1%Zr/AD-999 seal, hot-gas pressure-bonded
1	Nb-1%Zr/E-37 seal, hot-gas pressure bonded
1	Ni/E-37 seal, brazed with Cu-2% Ni alloy.

The difference between the AD-999a and AD-999b specimens was a slightly smaller and somewhat more uniform grain size ($\sim 4 \mu m$) in the latter. The Nb/ Al_2O_3 and Nb-1%Zr/ Al_2O_3 seals consisted basically of thin-walled metal tubes slipped over, and bonded to, each end of a hollow Al_2O_3 cylinder. The Ni/E-37 seal was a butt-joint construction similar to that to be used in the German Incore Thermionic Reactor Program; E-37 is the type of alumina selected for this program.

Hot-cell disassembly of the capsule has been completed and postirradiation diagnostic procedures have begun. Visual inspection shows the usual blackening of the Al_2O_3 specimens and mottling of some Y_2O_3 specimens. All appear to be in good condition with no signs of cracking. The neutron-fluence distribution along the capsules and the dimensional changes due to irradiation have been determined. Vacuum integrity, strength, thermal diffusivity, and electrical resistivity remain to be measured.

The neutron fluences ($E_n > 1.0$ MeV) were determined by measuring the ^{54}Mn γ -ray activity from the $^{54}Fe(n,p)$ reaction in the stainless-steel wall of the heat pipe¹; samples were obtained by slicing very thin rings from the heat-pipe tube and cutting

three small sections, spaced 120° apart, from each of the rings. It was thus possible to measure the average fluence at each chosen axial position as well as the change in fluence across the tube diameter. The average value for the latter was 14%; it occurred over a distance of 1.2 cm, indicating the rapidity with which the neutron fluence decreases with distance from the core in the blanket region of the reactor. The axial dependence of the neutron fluence ($E_n > 1.0$ MeV) is shown in Fig. 1. The curve represents the calculated dependence for the radial position at which the test unit was located² and has been normalized to the maximum experimental flux value. The measured fluence at the core mid-plane was 85% of the calculated value.

Neutron fluences of $E_n > 1.0$ MeV were converted to $E_n > 0.1$ MeV fluences by means of the data computed by General Electric Co.²

Volume increases for the three fine-grained (6 to 10 μm) Lucalox samples are plotted in Fig. 2, together with all the data previously obtained for this material both at LASL and at ORNL (Oak Ridge National Laboratory).^{3,4}

Fig. 1. Comparison of experimental and calculated dependence of neutron fluence ($E_n > 1.0$ MeV) on axial position for Irradiation Capsule K-2. Calculated curve normalized to peak experimental fluence.

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Fig. 2. Volume expansion of fine-grained Lucalox (GE Al_2O_3) as a function of neutron fluence. LASL data were obtained at an irradiation temperature of 700° to 750°C. ORNL irradiation temperatures cover a range of 400° to 1070°C. Solid line is a least-squares straight-line fit to all data.

The error bars for the present data represent a conservative estimate of the measurement error, hence the spread of the data points is more than can be accounted for by this source. Because the values of relative fluence are reasonably accurate (although the absolute values are considerably less so), it is presumed that variations in material properties from one specimen to another may cause much of the observed dispersion in the Lucalox data. The average of the present data is within 5% of the least-squares straight-line fit to all the data.

Volume expansions of the AD-999 samples are plotted in Fig. 3 as a function of neutron fluence ($E_n > 0.1$ MeV) and are fit by the method of least-squares with a straight line through the origin. Data obtained earlier from EBR-II Test Capsule K-1 are included in the fit. Again, there is somewhat wider data-point dispersion than can be accounted for by measurement error. The earlier data appear to indicate swelling saturation with increasing fluence, but the present data cast doubt on this interpretation.

The least-squares fit to the Lucalox data, Fig. 3, shows somewhat higher resistance to swelling for the AD-999. The two heavy black dots are data points for the Feldmühle E-37 alumina. Note that they lie on the Lucalox curve.

Fig. 3. Volume expansion of Coors AD-999 and Feldmühle E-37 Al_2O_3 as a function of neutron fluence for an irradiation temperature of 750°C.

The first EBR-II ceramic irradiation test (Capsule K-1) revealed the excellent resistance of three Y_2O_3 specimens to neutron-induced swelling. Five additional specimens of this material were included in Irradiation Capsule K-2, and again there was essentially no dimensional increase (Fig. 4).

The single data point indicating a shrinkage of 0.4% can not be explained by measurement error; its significance, however, is doubtful despite the fact that these specimens had a density between 90 and 95% of theoretical. A hydrogen firing process has since been developed which produces Y_2O_3 specimens with a density 98% of theoretical. Several such specimens are included in Irradiation Capsules K-3 and K-4, described below.

The diametral increases of the ceramic-to-metal seals, measured at the location where the thin-walled niobium or Nb-1%Zr tubes overlay the ceramic cylinders, are shown in Fig. 5 and are compared with the diametral increases expected for unrestrained Lucalox and AD-999. The Nb-1% Zr tubes had no apparent restraining effect on the AD-999 and E-37, but the niobium tubes may have had such an effect on the Al-995 ceramic bodies. It is possible that the specimen with the least dimensional change was irradiated at a lower temperature than the others because it was located adjacent to the heat dump section of the gas-filled heat pipe used for setting the specimen temperature.

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Fig. 4. Volume expansion of Y_2O_3 as a function of neutron fluence for an irradiation temperature of $750^\circ C$.

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1. "Quarterly Status Report on the Space Electric Power R and D Program for the Period ending July 31, 1970, (U)," LASL Report LA-4497-MS, August, 1970.
2. W. Edwards, W. Henderson, and N. Baumgardt, "Analysis of EBR-II Neutron Spectra by Monte Carlo and Discrete Ordinate Methods," GEMP-742, Dec. 1969.
3. G. Keilholtz, R. Moore, and H. Robertson, "Effects of Fast Neutrons on Polycrystalline Alumina and Other Electrical Insulators at Temperatures from 60° to $1230^\circ C$," ORNL-4678, May 1971.
4. G. Keilholtz and R. Moore, "Irradiation Damage to Alumina exposed to 5×10^{21} Fast Neutrons/cm²," Nucl. Appl. 3, 686 (1967).

Insulator Irradiation Capsules K-3 and K-4

Irradiation Capsules K-3 and K-4 were shipped to Idaho Falls for insertion in the EBR-II reactor. These capsules and a tungsten-specimen capsule from ORNL will be combined in a single hexagonal sub-assembly for irradiation. The two capsules contain four irradiation units: two will run at $750^\circ C$, one at $600^\circ C$, and one at the EBR-II coolant temperature ($\sim 450^\circ C$). Temperatures are established by a sodium-neon isothermal irradiator for the first two units, by a potassium-neon isothermal irradiator for the $600^\circ C$ unit, and by helium coupling to the capsule closure tube for the $450^\circ C$ unit. Samples include AD-999, fine-grained Lucalox, Avco alumina, CeO_2 , both MgO- and Y_2O_3 -stabilized ZrO_2 , Lindsey and MCA yttria, yttralox (GE), and autoclave-bonded trilayer specimens consisting of various combinations of niobium, Nb-1%Zr, AD-999, Lucalox, Avco alumina, and yttria. Eleven GEES (Gulf Energy and Environmental

Fig. 5. Diametral increase of Nb- Al_2O_3 ceramic-to-metal seals irradiated at $750^\circ C$. Seal configuration tends to constrain diametral increase of Al_2O_3 by a 0.010 to 0.014-in.-thick niobium or Nb-1%Zr cylinder.

Systems, Inc., formerly Gulf General Atomic) specimens, one TECO (Thermo Electron Corp.) trilayer, one TECO Al_2O_3 -Nb cermet, and three TECO sapphire specimens are also included. The planned neutron fluence ($E_n > 0.1$ MeV) covers the range of from 1.5 to 6×10^{21} nvt. Insertion of these capsules is scheduled for mid-August 1971. The specimens in these capsules are listed, together with their irradiation temperatures and target fluence ranges, in Table I.

Thermal Diffusivity of Irradiated Y_2O_3

Specimens of Y_2O_3 , fabricated from Lindsey Co. powder at 95% of theoretical density, were irradiated in the initial EBR-II isothermal irradiator at a temperature of $750^\circ C$ to fluences as high as 9.8×10^{21} nvt ($E_n > 0.1$ MeV). These samples were metallized with tungsten bands to facilitate resistivity measurements (see Quarterly Status Report LA-4497-MS). The bands have been removed by chemical dissolution to permit thermal-diffusivity measurements by the laser-pulse technique. Because these samples had been postirradiation-heated to temperatures as high as $800^\circ C$ during the resistivity measurements, some uncertainty existed as to whether annealing effects might or might not have modified the thermal-diffusivity values. However, there was no evidence of annealing in the resistivity measurements, and, because the measuring temperature exceeded the irradiation temperature for only brief periods of time, no appreciable effect was expected.

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TABLE I

TRILAYER AND CERAMIC SPECIMENS IN EBR-II IRRADIATION ASSEMBLIES K-3 AND K-4

	Irradiation Temperature								
	750°C			600°C			450°C		
	H	M	L	H	M	L	H	M	L
<u>Trilayers</u>									
LASL Lucalox Nb-1%Zr	x	x	-	-	x	x	-	x	x
LASL Lucalox Nb collector	x	-	x	-	x	x	-	x	x
LASL AD-999b Nb-1%Zr	-	x	x	-	-	-	-	-	-
LASL AD-999b Nb collector	-	-	-	-	x	-	-	x	-
LASL Avco Al ₂ O ₃ Nb-1%Zr	-	x	x	-	-	x	-	-	-
LASL AD-999x Nb-1%Zr (seal)	x	x	-	-	-	-	-	-	-
LASL Y ₂ O ₃ Nb-1%Zr	x	-	-	-	x	x	-	-	-
GGA Al ₂ O ₃ -Nb graded	-	x	x	-	-	x	-	-	-
GGA Y ₂ O ₃ -Nb graded	x	x	-	-	x	-	-	-	-
GGA Al ₂ O ₃ -3% W cermet-Nb	-	-	x	-	-	-	-	-	-
GGA Al ₂ O ₃ -1% Y ₂ O ₃ -Nb	-	-	x	-	-	-	-	-	-
GGA Lindsey A-Al ₂ O ₃ -Nb	-	-	x	-	-	-	-	-	-
GGA Mellor-Al ₂ O ₃ -Nb	-	-	x	-	-	-	-	-	-
GGA Plasmadyne 301E Al ₂ O ₃ -Nb	-	x	-	-	-	-	-	-	-
Teco Al ₂ O ₃ -Nb	-	x	-	-	-	-	-	-	-
<u>Ceramics</u>									
LASL Y ₂ O ₃ Lindsey	-	-	-	m	-	m	-	-	-
LASL Y ₂ O ₃ Lindsey thin wall	m	m	m	-	-	-	-	-	-
LASL Y ₂ O ₃ Moly Corp.	x	m	-	m	x	-	xx	-	x
LASL Yttralox	x	-	-	-	x	-	x	-	x
LASL AD-999x	mx	mx	m	x	-	-	m	m	-
LASL AD-999b thin wall	-	-	-	-	-	-	-	-	x
LASL Avco thin wall	mx	mx	m	x	-	-	m	x	-
LASL Lucalox thin wall	-	m	-	m	-	-	-	m	m
LASL ZrO ₂ -Y ₂ O ₃ stabilized	-	-	m	x	x	-	x	x	x
LASL ZrO ₂ -CaO stabilized	-	-	-	-	-	-	x	-	-
LASL CeO ₂	-	-	-	-	-	-	-	x	-
Teco cermet	x	-	-	-	-	-	-	-	-
Teco Sapphire	x	-	-	x	-	-	x	-	-
<u>Code</u> <u>Target fluence (E_n > 0.1 MeV)</u>									
H	3.6 to 6 x 10 ²¹ nvt								
M	2.4 to 3.6 x 10 ²¹ nvt								
L	1.5 to 2.4 x 10 ²¹ nvt								

x = unmetallized ceramic or trilayer specimen
m = ceramic specimens metallized for resistivity measurement

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The thermal-diffusivity data are shown in Fig. 6, with the solid line representing data obtained on several unirradiated specimens. Note that fast-neutron damage decreases the thermal diffusivity but apparently does not markedly alter its dependence on specimen temperature. The effect of neutron fluence is shown more clearly in Fig. 7 where thermal diffusivity is plotted as a function of neutron fluence for 25° and for 600°C. The thermal-diffusivity values have been converted to values of thermal conductivity given by the scale on the right-hand side of Fig. 7. It was assumed that the specific heat of Y_2O_3 (0.11 cal/g-°C at 25°C) is neither affected by temperature nor by irradiation. This assumption, is, however, not entirely correct because a slight increase in specific heat is expected as a function of temperature. The effect of irradiation on specific heat is unknown. Measurements of specific heat have been made on irradiated Al_2O_3 samples and no change from values for unirradiated material could be detected. The irradiated Y_2O_3 specimens were too small for accurate specific-heat measurements.

Errors in measurement and, possibly, variations from one sample to another produce a considerable uncertainty, as indicated by the Y_2O_3 data at 25°C (Fig. 7). However, the general trend is similar to that observed for Al_2O_3 specimens (see Quarterly Status Report IA-4497-MS) where the thermal conductivity decreased significantly after exposure to low neutron fluences and became constant at higher

Fig. 6. Measured thermal diffusivity of irradiated Y_2O_3 .

fluences. This trend is shown in Fig. 7 by the curve labelled "Lucalox".

The most significant result of the investigation is the fact that the thermal conductivity of highly irradiated yttria, when extrapolated to 750°C, is 70% of that of highly irradiated Lucalox.

SHEATHED INSULATOR DEVELOPMENT

Fabrication of Pressure-Bonded Tri- and Pentallayers

Two trilayers (TL-113 and TL-120) and one pentallayer (PL-121) were pressure-bonded in accordance with the following conditions:

Time Interval, h	Temperature, °C	Pressure, psig
8.0	20 to 1538	50 (250)*
3.5	1538	50 (250)* to 10,000
3.0	1538	10,000
11.0	1538	10,000 to 250
8.0	1538 to 20	250

*TL-113 was at 250 psig.

Fig. 7. Thermal diffusivity and thermal conductivity of Y_2O_3 and Lucalox vs neutron fluence (specific heat of Y_2O_3 assumed to be constant at 0.11 cal/g-°C).

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In this pressure-bonding program the pressure is reduced more slowly at maximum temperature than in previous programs. The fabrication of crack-free insulators may require slow pressure reductions at high temperature to permit the relief (by creep of the components) of stresses induced by pressure changes. Optimum pressure-bonding schedules have not been determined, but will be investigated in a future parametric study.

The cylindrical collector and the outer sheath of TL-113 were fabricated from Nb-1%Zr sheet that had been rolled, seam-welded, and machined to dimension. The ceramic was Al-995 formed at LASL. The outer-diameter extremes after pressure-bonding were 1.271 and 1.286 in. Microscopic examination of the edges of three cylinders cut from a 2-in. length at the center of the trilayer revealed no evidence of cracks in the ceramic.

The collector and the outer sheath of TL-120 were machined from heavy-walled Nb-1%Zr tubing. Lands, equal in height to the wall thickness of the ceramic, were left on one end of the outside diameter of the collector and on the opposite end of the inside diameter of the outer sheath. This configuration permitted the insulator to be assembled with essentially zero clearance between the ends of the ceramic and the metal lands, thus reducing the strain imposed on the ceramic ends during pressure buildup. The ceramic has WESGO Al-995.

There was no change in the outer dimensions of TL-120 as a result of pressure-bonding, but the inside diameter increased from 1.122 to 1.130 in. (+0.008 in.). A band of metal from the outer sheath was removed by etching to expose one end of the ceramic. No cracks were evident.

The dimensional changes in TL-120 indicate that the ceramic cylinder was in tension during the pressure cycle; it is therefore particularly notable that cracking of the ceramic did not occur. In certain tri- and pentalayer designs currently under consideration, putting the ceramic in tension and even displacing it radially outward without cracking may be necessary.

The collector and the outer sheath of Pentlayer-121 were machined from Nb-1%Zr tubing. The collector was successively plasma-sprayed with

layers of Al_2O_3 (0.010 in.), Nb-1%Zr (0.005 in.), and Al_2O_3 (0.010 in.). The outer layer was ground to dimension (0.643-in. diam).

The outer diameter of Pentlayer-121 decreased from 0.672 to 0.664 in. (-0.008 in.) and the inside diameter increased from 0.5225 to 0.5280 in. (+0.0025 in.) during pressure-bonding. There was no detectable distortion. A circular band of metal ~ 0.375 in. wide near the end of the ceramic, and a longitudinal strip ~ 0.125 in. wide by 1.750 in. long were removed by etching to expose the outer layer of the ceramic. Only one very faint circular crack was visible near the end of the ceramic. No obvious reason for the crack was found. Evaluation of these insulators is continuing.

Voltage-at-Temperature Irradiation Tests of Sheathed Insulators

The in-pile voltage-at-temperature test of pressure-bonded sheathed-alumina insulators (see Quarterly Status Report LA-4625-MS) was removed from the OWR (Omega West Reactor) core on June 25, 1971. The test had accumulated 3150 h of irradiation at full reactor power, equivalent to a neutron fluence of 5.3×10^{20} nvt ($E_n > 0.1$ MeV).

The heat-pipe temperature-control system performed very well throughout the test: trilayer temperatures were maintained at $750 \pm 20^\circ\text{C}$ when the reactor was at power.

The measured resistivity fluctuated erratically during the last few months of the test, sometimes by a factor of ten during a week. However, the values became fairly constant during the last three weeks of the test, i.e., ~ 1×10^6 and ~ 0.6×10^5 $\Omega\text{-cm}$ for Lucalox and Al 995 samples, respectively.

Disassembly in the hot cell will follow shortly, to determine the causes of the erratic behavior before a second unit for testing yttria and AD-999 samples is constructed.

The corresponding out-of-pile test was restarted after repair of the heater. The measured resistivity of both Lucalox and Al-995 in this test is ~ 3×10^{10} $\Omega\text{-cm}$ after 2500 h at 750°C .

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FUELED-EMITTER DEVELOPMENT

Disassembly of Thermionic Converters Tested by
General Electric Co. (GE)

Introduction

The disassembly and post-test diagnostic evaluations of the GE thermionic-converter Test Units SCIP-518, TFE-619, and TFE-621 are continuing. Dimensional measurements of the emitters and collectors from all three assemblies have been completed. The typical hour-glass distortion observed in earlier tests was not found for the emitter of SCIP-518 or the emitters of TFE-619, but was detected for the upper and middle emitters of TFE-621 (see Quarterly Status Report LA-4690-MS). All three emitters in TFE-621 had stress-relief grooves in the end caps.

All emitters have been potted in epoxy to permit sectioning for diagnostic measurements and metallographic examinations. The potting compound was initially introduced at low pressure to prevent regions of fragile UO_2 structure from being swept away by the inflowing potting material.

The emitters are being sectioned as indicated in Fig. 8. The three circular sections taken from the axial center of the emitter will be used for metallographic examination, for isotopic-ratio (burnup) measurements, and for fission-gas retention measurements. The two longitudinal end pieces will be used for metallographic examinations only. The fueled regions in the end pieces are being exposed at successive depths by grinding until the

Fig. 8. Sketch illustrating cutting and grinding procedure used in preparing irradiated emitters for diagnostic measurements.

planes through the center axes are reached. During this process, the fueled regions are polished and photographed to permit study of the fuel structure.

This method of grinding to expose successive layers (in a radial direction) has the disadvantage that relatively coarse diamond compound must be used if the task is to be accomplished within a reasonable length of time. As a result, pullout of tungsten grains during grinding was fairly common, particularly for the arc-cast tungsten components of the emitter body. To overcome this difficulty, polishing periods have been extended to up to 12 h. Photographs of highly polished specimens are not yet available, but some important observations have been made on less-than-optimally polished specimens and are discussed below.

SCIP-518

The emitter of this unit was made by alternately stacking 0.080-in.-thick wafers of UO_2 and 0.010-in.-thick disks of tungsten in a CVD tungsten cladding of 0.464 in. o.d. SCIP-518 was irradiated for 10,200 h at a surface temperature of $\sim 1600^\circ C$.

One unexpected phenomenon which was revealed by postirradiation examination was the extensive interaction between the arc-cast tungsten disks and the UO_2 fuel. This interaction is indicated on a previously published radiograph (see Quarterly Status Report LA-4690-MS, Fig. 2) by areas of apparent low density in the middle of the tungsten disks near the center of the emitter, i.e., in the hottest temperature region. A photomicrograph of two disks adjacent to the tungsten cladding, Fig. 9, shows voids in the region of relatively low temperature. (The notch-like voids in the cladding are clearly tungsten pullout.) A photomicrograph at higher magnification ($\sim 100\times$) of the same area, Fig. 10, reveals extensive penetration of the tungsten disks by UO_2 as well as migration of tungsten into the UO_2 . It appears that extensive migration of tungsten into the UO_2 has occurred at the interface of cladding and fuel (upper right-hand corner of the photomicrograph). However, penetration of the CVD tungsten cladding by the UO_2 is negligible. The interaction between the disk and the fuel near the center of the emitter is shown in Fig. 11. Here the penetration of UO_2 into the tungsten is so pronounced that the structure has the appearance of a cermet.

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Fig. 9. Photomicrograph of tungsten-disk-clad junction of SCIP-518 emitter (20X).

Fig. 11. Interaction between UO_2 and tungsten disk near center of SCIP-518 emitter (200X).

Fig. 10. Tungsten-disk-clad junction of SCIP-518 emitter (~ 100X).

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The notable difference in compatibility of UO_2 with arc-cast and CVD tungsten (as indicated by Fig. 10 where the tungsten disk and the tungsten cladding are at very nearly the same temperature) is as yet unexplained.

TFE-619

The bottom emitter of TFE-619 was also a UO_2 -wafer/tungsten-disk configuration, similar to SCIP-518. The bottom section of this emitter is shown in Fig. 12 after rough grinding to expose the fuel-disk configuration (but before final grinding and polishing). The tungsten disks are seen to be relatively undisturbed in the center of the emitter but taper off and disappear towards the edge of the emitter. The edge of one of the disks is shown in Fig. 13 at high magnification and illustrates the disintegration of the tungsten at that point. The condition of the tungsten disk nearer the center of the emitter is shown in Fig. 14. Although the photograph is marred by the obvious pullout of tungsten grains, it clearly shows a definite lack of interaction between the tungsten and the UO_2 at the upper interface. This lesser interaction in a region of higher temperature (the reverse of the reaction in SCIP-518) is not understood. One contributing factor might have been the radial variation of chemical properties of the UO_2 fuel, brought about by the radial variation of fission density. This latter variation is more pronounced for the

emitters whose diameter is large compared to the diameter of the SCIP diode series. In any case, CVD tungsten appears to better resist interaction with UO_2 than arc-cast tungsten (see SCIP-518 results) and would therefore be the obvious choice for future UO_2 -wafer/tungsten-disk experiments.

TFE-621

The emitters in this test unit operated at open circuit for ~ 100 h with estimated emitter temperatures as high as $2100^\circ C$. As a result of these high temperatures, the columnar grain structure of the UO_2 extends radially, with relative uniformity, from the cladding to the interior of the fuel. Many photomicrographs published in the literature show the columnar structure of the grains in views along the axes of the grains, but few, if any, show this structure in views taken end-on. Such a view of the columnar UO_2 structure in the center emitter of TFE-621 at a location about 0.020 in. radially inward from the inside surface of the cladding is shown in Fig. 15. At this location there is little uniformity in grain cross section and a surprising amount of void space between grains.

Cermet Fuel Irradiation Program

Assembly of the four cermet-fuel irradiation capsules which are to be tested in the Engineering Test Reactor (ETR) has been completed. A schematic drawing of these test capsules is shown in Fig. 16.

Fig. 12. Bottom section of bottom emitter from TFE-619 (before final grinding and polishing)(2.5X).

Fig. 13. Outer edge of tungsten disk from bottom emitter of TFE-619 (200X).

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Fig. 14. Tungsten disk near center of bottom emitter from TFE-619 (200X).

Fig. 15. View of UO_2 perpendicular to axes of columnar grains in center emitter from TFE-621 (200X).

Fig. 16. ETR irradiation capsule for testing tungsten-clad Mo-60 v/o UO_2 cermet fuel specimens containing a 9-w/o addition of tungsten in the molybdenum. Gap between specimen and capsule wall is connected to the rare-gas-filled chamber by a central hole in the fuel specimen.

Each capsule contains two fueled-emitter specimens in separate vacuum-tight chambers. Rare gas is used to fill the chambers, and heat from the emitters is transmitted to the capsule wall by a combination of radiation and gas conduction. In the reference unit (emitter temperature, $1650^\circ C$; input power density, $190 W/cm^2$) each of the two mechanisms accounts for half of the heat transport, and the temperature in

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the range of interest varies approximately as the square root of input power. For other temperatures and power densities the fill gas and the gap between emitter and capsule wall are varied. A fill-gas pressure of 3 atm was chosen to keep the change in gas conductivity (caused by xenon and krypton fission gas escaping from the fuel and entering the gas chamber) to $< 10\%$ in 10^4 h of irradiation.

The emitter support was calculated to represent an optimized design for an emitter operating in a thermionic diode with 12% efficiency at 1650°C and 190 W/cm², i.e., for a condition where ~ 7% of the total heat generated is removed by conduction through the emitter support. The irradiation tests require higher thermal resistances for the emitter supports than are needed for an operating diode because the heat-dump temperature for the irradiation tests is 650°C lower. This accounts for the reentrant design of the emitter support shown in Fig. 16.

Emitter temperature determinations will be based on burnup measurements and on a calibration test of burnup rate vs emitter temperature performed in the OWR in January 1971 (see Quarterly Status Report LA-4625-MS). Thermocouple instrumentation for each test unit, which would be preferable, was not included because of cost considerations. However, calculated and measured performances for the calibration unit agreed to within 4%, so that considerable confidence exists that temperature determinations will be reasonably accurate.

The fuel being tested in this irradiation series is Mo-60 v/o UO₂ cermet, in which 9 w/o W has been added to the molybdenum matrix as a strengthener and 3 w/o Y₂O₃ has been added to the UO₂ as a stabilizer. The cermet has 15% interconnected porosity to allow fission-gas escape and is clad with 0.010 in. of CVD columnar-oriented tungsten.

The variations in specimen temperature and input power density for this test series, together with gap widths and fill gases are listed in Table II. Irradiation will begin at the end of August 1971.

Hydrogen Barrier Experiments

An experiment, conducted to determine the diffusion properties of hydrogen through pure lead, has yielded very promising data. The experiment was

made as part of the continuing low-level investigation of the use of lead as a hydrogen barrier for ZrH_x fuel and neutron moderating rods in reactor cores (see Quarterly Status Report LA-4447-MS).

The experimental assembly is shown schematically in Fig. 17. It consists of a section of SNAP (Space Nuclear Auxiliary Power) fuel (ZrH_{1.715} loaded with 10 w/o uranium) enclosed in a niobium can. The SNAP fuel is centered and supported by alumina cones which fit closely into the can and extend a short distance into a central hole in the fuel. A tungsten spring is used to hold the alumina and fuel pieces firmly against the bottom of the can. After assembly of these parts, lead was melted into the can in vacuum. The end cap was then welded-in, the space above the lead was back-filled with 1 atm of argon, and the assembly was sealed by pinching-off the tube shown in the end cap.

The diameter of the fuel was 0.529 in., and the lead thickness surrounding the fuel was 0.0165 in. at 600°C.

The test apparatus had been used previously on earlier assemblies and consists of a vacuum system of known volume, a vacuum pump, a McCloud pressure gauge, an rf induction coil, and a continuously-recording infrared pyrometer. The assembly was tested in an upright position.

Experimental data are presented in Table III. Leak rates at temperatures $< 550^\circ\text{C}$ were so low that they were indistinguishable from background leakage. The 145.1-h test at 700°C was run after the assembly had been heated at 750°C for 5 h to outgas reasonably well the metal parts in the assembly. The measured pressure increase at 750°C may have included some contribution from additional outgassing.

These data are compared with diffusion rates through tungsten in Fig. 18. (The tungsten data were obtained from an Atomics International Report.)¹ The properties of tungsten were used as a basis for comparison because tungsten has the lowest diffusion coefficient for hydrogen of any refractory metal. In the interesting temperature range (600° to 700°C), a tungsten membrane ~ 0.020 in. thick is equivalent as a barrier (i.e., has the same hydrogen leak rate per unit area) to the ceramic hydrogen-diffusion barrier which has been developed for SNAP fuel rods.

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TABLE II
IRRADIATION CONDITIONS FOR CERMET-FUEL SPECIMENS IN THE ETR

Specimen No.	Surface Temperature, °C	Input Power Density, W/cm ²	Gap Width Between Emitter and Capsule Wall, in.	Fill Gas (3 atm)
1a	1650	190	0.030	Ne
1b	1750	190	0.044	Ne
2a	1635	100	0.150	Xe
2b	1550	100	0.061	Ar
3a	1650	275	0.049	He
3b	1750	275	0.062	He
4a	1650	190	0.030	Ne
4b	1550	190	0.078	He

Fig. 17. Test assembly for hydrogen-retention experiments.

Fig. 18. Hydrogen diffusion rate vs temperature.

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TABLE III

HYDROGEN LEAK RATES FROM NIOBIUM-CLAD LEAD-BARRIER TEST ASSEMBLY

Assembly Temp., °C	Length of Test, h	Pressure Increase, Torr/h	Leak Rates	
			H_2/cm^2-h	$\frac{cm^3(STP)-mm}{cm^2-h-atm^{1/2}}$
550	5	1.5×10^{-5}	1.78×10^{13}	2.73×10^{-6}
600	15.1	7.4×10^{-5}	8.77×10^{13}	6.81×10^{-6}
650	5.1	7.0×10^{-4}	8.28×10^{14}	3.44×10^{-5}
700	145.1	2.26×10^{-3}	2.68×10^{15}	6.50×10^{-5}
750	21.0	15.88×10^{-3}	1.81×10^{16}	2.70×10^{-4}

Data from previous tests are compared with those for tungsten in Fig. 19. Note that the hydrogen retention properties of the current experiment are considerably improved over those of earlier experiments. In fact, the current data indicate hydrogen diffusion rates that are about one order of magnitude lower in lead than in tungsten. The reasons for the dramatic improvement over previous experiments are not entirely understood. The absence of a wire screen in the lead is undoubtedly an important factor due both to the effect of the metal screen on the diffusion coefficient of hydrogen in lead and to the difficulties encountered in attempting to fill the metal wicks with lead by using previous techniques. Another reassuring aspect of the current experiment is the fact that molten lead apparently does not dissolve uranium from the SNAP fuel, at least not in sufficient quantities to markedly increase the diffusion rate of hydrogen.

The difficulties encountered in previous experiments, due in part at least to the presence of the wire screens, should not discourage further attempts to develop lead barriers. A careful comparison of data from the current experiment and from an earlier experiment which contained wire screens (labeled Nb-Nb-W-1) shows that the diffusion rate in the interesting temperature range was less for the current experiment by only a factor of from 2 to 3. Both are superior to tungsten and would therefore be very useful. It should also be pointed out that most LASL reactor designs which incorporate lead barriers do not include wire screens in the barrier regions.

An additional encouraging fact is the relative unimportance of concentricity in experiments such as this. For example, a simple analysis shows that for an extreme case in which the barrier thickness is decreased to one-half its nominal value on one side of the assembly due to nonconcentricity, the hydrogen diffusion rate is increased by only 16% over that of a perfectly concentric case.

A materials compatibility test for lead hydrogen-barrier systems was also completed. The test capsule was maintained at 750°C for 1000 h. The capsule consisted of a section of a SNAP fuel rod that was surrounded by lead and was contained in a Nb-1%Zr can. After the test, the capsule was sectioned and polished for electron-microprobe examination, and part of the lead was melted out for spectrochemical analysis.

Electron-microprobe examination indicated no significant variations in niobium and zirconium x-ray intensities through the niobium can relative to the normal alloy, and uranium and lead were not detected in the alloy. Trace amounts of niobium, estimated to be less than 0.5%, were detected in the lead near the niobium-zirconium container; and trace amounts of uranium and zirconium, estimated to be less than 1.0%, were detected in the lead near the SNAP fuel. No lead or niobium was detected in the SNAP fuel. Detection limits were estimated to be in the range of between 0.05 and 0.5%.

Spectrochemical analysis of the lead indicated the presence of zirconium at a concentration of 400 ppm. No other elements were detected. The limit of detection was ~ 100 ppm.

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Fig. 19. Hydrogen diffusion rate vs temperature for all tests conducted to date.

Additional metallurgical examinations are in progress to investigate the possibility of a reaction zone at the lead-niobium interface.

Reference:

1. R. W. Webb, "Permeation of Hydrogen through Metals," Atomics International Report NAA-SR-10462, July 1965.

HEAT PIPE SYSTEMS

The results obtained in the work on heat pipe systems are unclassified and are described in Space Electric Power R and D Program Report, Part I, LA-4746-MS.

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