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Quarterly Status Report on the

Space Electric Power R and D Program (U)

for the Period Ending January 31, 1969

Part II

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

AEC RESEARCH AND DEVELOPMENT REPORT

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LA-3881-MS (Part I)	LA-3987-MS (Part II, Conf. RD)
LA-3882-MS (Part II, Conf. RD)	LA-4039-MS (Part I)
LA-3941-MS (Part I)	LA-4040-MS (Part II, Conf. RD)
LA-3942-MS (Part II, Conf. RD)	LA-4109-MS (Part I)
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C-93b, ADVANCED CONCEPTS
FOR FUTURE APPLICATION--
CONVERSION DEVICES
M-3679 (60th Ed.)

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THERMIONIC POWER SYSTEMS

ZrH_x-Moderated Reactors with ²³⁵U Fuel

Introduction

The results of scoping studies of space power systems based on the use of ²³⁵U-fueled, ZrH_x-moderated thermionic reactors have been reported previously in LA-3987-MS and LA-4040-MS. These studies encompassed a broad range of reactor sizes, compositions, and electric power levels. Recent, more detailed, studies of selected reactor designs have led to the choice of a single radial configuration of a reactor core which could generate nominally a power varying from ~ 35 to ~ 100 kWe. The output is varied by changing one or more of the following: reactor core length, number of thermionic fuel rods (TFRs), reflector thickness, fuel concentration in the moderator, and/or reactor power-density distributions.

Estimates of system power outputs and component weights reported previously were determined by scaling the results of detailed analyses of a reference design system. Because of differences in reactor power-density distributions, however, this procedure resulted in some inaccuracies. Therefore, the electric power output, the efficiency, and the system component weights for designs of particular interest have been recalculated.

Total system weights are based, to a large extent, on assumed requirements relating to the radiation shielding. The weight estimates for LASL-designed power supplies based on shielding criteria used by various organizations were obtained to illustrate this sensitivity.

Reactor Designs

The reactor core concept utilized in reactor designs of current interest is described in LA-4040-MS; the TFRs, the rods containing ZrH_x-moderator, and the reflector and reactor control systems are also described in this report.

The reactor core configuration for reactor designs with power outputs from ~ 35 to ~ 100 kWe has an o.d. of 12 in. and contains 211 rods, of which either 55 or 73 are TFRs and the remainder

are (U-Zr)H_x rods. The TFRs contain 2-in. long thermionic diodes, 8-12 per rod, and utilize W-coated Mo-UO₂ cermet as thermionic fuel. The U-concentration in the (U-Zr)H_x rods varies from 0 to 10 w/o in the U-Zr alloy. The reflectors are made of Be and contain rotating poison-backed control drums. Reflector thicknesses vary from 3 to 4 in. Cesium reservoirs, fission-product storage areas, power leads, and instrumentation leads are connected to the TFRs which penetrate the core vessel at one end.

These reactor designs are now divided into two groups depending on whether or not they include core-compensation variations for the purpose of flattening the power-density distributions in the TFRs. Initial evaluations were based on reactor cores with uniform core composition; this resulted, in some cases, in very poor power-density distributions. The ZrH_x-moderated reactors being studied are readily amenable to power flattening. (Power flattening is a tedious and time-consuming procedure; the results of initial calculations are discussed in a subsequent section.)

The estimated excess reactivity requirement for 10,000 h of full-power operation for U-fueled ZrH_x-moderated thermionic reactors is from 5 to 7%. The minimum initial excess reactivity of the reactor designs discussed is 9.1%, i.e., $k_{\text{eff}} = 1.10$.

Power Output

The electric-power outputs for the reactors studied were calculated with the LASL code developed for this purpose. The calculations were based on detailed power-density distributions and on current-voltage data from the SIMCON code. Since the thermionic diodes in each TFR have a common Cs supply and because all Cs reservoirs are probably located in an isothermal region, all electric-power outputs were calculated by assuming a constant Cs pressure. Other assumptions made for these calculations included the following: maximum emitter temperature is 1830°C; voltage

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produced by all TFRs in a reactor is the same; and all TFRs in a reactor are of identical design.

Some variation in electric-power output can be obtained from a particular design by selecting an operating point. Unless otherwise noted, the electric-power levels given for the various designs correspond to the maximum-efficiency operating point. Increases in electric-power output can be obtained by operating at the maximum-power operating point.

Waste-Heat Radiators

The reactor designs which appear most attractive for power levels of < 100 kWe are driver-core designs, i. e., designs where part of the fuel is contained in the moderator which, therefore, also produces part of the power. Such designs are attractive because they are relatively small and thus require only relatively small radiation shields. Overall efficiencies are penalized, however, because part of the power is produced in the driver fuel and because relatively large waste-heat radiators are required. The tradeoff between shield weight and radiator weight unquestionably favors the use of moderated driver-core designs; nevertheless, since overall efficiencies are low, i. e., 6 to 8%, there is a considerable incentive to design low-weight waste-heat radiators.

Waste-heat radiators have been designed which are based on the use of heat pipes. The technology for very reliable heat-pipe radiators is available and such designs should be considered for all space applications. Some of the advantages compared to conventional radiator designs are: (1) Heat pipes enclose nearly empty spaces and are inherently light in weight. (2) Heat pipes transport heat at nearly uniform temperatures, and this tends to reduce radiator area requirements. (3) Heat pipes may be operated either in series or in parallel, permitting the design of radiators which would take maximum statistical advantage from system redundancy. (4) No mechanical devices (e. g., pumps) are required in secondary loops.

Two radiators have been designed in considerable detail for possible use in space power

supplies. Both designs incorporate numerous cylindrical heat pipes and contain Li as the working fluid. The pipes of one design are made of armored Be whereas those of the other are made of stainless steel armored with Be. The weights per kWe radiated of the two heat-rejection systems, including primary coolant pumps and coolant loops external to the reactor, are 0.606 and 1.675 lb for Be construction and stainless-steel construction, respectively; their weights for the systems considered are from ~ 30 to ~ 45% of the total weights of the reactor heat-rejection systems. The total system weights given in Table I, include the weight of waste-heat radiators constructed of stainless steel.

Radiation Shields

The weight of radiation shielding has been calculated for two configurations of shadow shields. The first shield, referred to as a SNAP shield, was designed for manned missions with a SNAP-8 reactor and a thermoelectric power conversion unit; it has a 12° cone angle, with a 100-ft separation implied between the reactor and the crew compartment. The shield is divided into two parts of about equal thickness by a 10-in. high gallery section which could be used to enclose a heat exchanger between the activated primary coolant and a heat-pipe radiator. Power-conditioning equipment could be placed adjacent to such a shield. Shield weights for the thermionic reactors were determined by scaling the weight of the SNAP shield on the basis of its volume.

The second shield design considered, referred to as an AEC shield, was suggested by the AEC for use in system studies. It has a 15° cone angle and consists of 2-in. thick W followed by 34 in. of LiH.

About half the weight of shielded systems is contributed by the shield; however, the difference in weight between the two shield designs is ~ 1000 lb for the systems considered. Therefore, weight comparisons between the systems presented here and others should be based on the use of equivalent shields.

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TABLE I. VARIATIONS OF 211-ROD THERMIONIC REACTOR

No. TFR	U-Content in U-Zr, %	Reactor Diam, in.	Reactor Length, in.	Power, kWe	System Weight, lb		
					Non-Shielded	SNAP Shield, 20° Cone, 10-in. Gallery	AEC Shield, 15° Cone, No Gallery
73	5	20	32.6	55.7	2058	4956	4041
55	5	20	24.4	36.3	1620	4154	3351
55	3	20	30.5	36.9	1758	4499	3634
55	5	18	28.5	37.4	1748	4109	3353
73	3	20	42.9	79.5	2481	5839	4919

Weights and Power Levels

The weights and electric-power levels of five space-power system designs are given in Table I. Weights are given for nonshielded systems and for those including the SNAP and the AEC shadow shields. These total weights include, in addition to the items listed above, the weights of control-rod drives, Cs reservoirs, instrumentation, γ shielding for Cs reservoirs and instrumentation, and power-conversion equipment.

The electric-power levels given in Table I are for reactors with nonflattened power-density distributions and have been reduced to account for losses in the power-conditioning equipment and for power required to operate the control-rod drives and the instrumentation. The maximum-efficiency operating point has been assumed for all cases.

Flattening of Power-Density Distribution

The electric-power outputs of the systems described in Table I can be increased, without significant weight penalty, by flattening the power-density distributions. The 18-in. diam reactor was chosen for further analysis to determine the gains in power output that can be obtained in this way.

Several power-flattening procedures could be used, including variations in H concentration, in ^{235}U concentration in the moderator, or both. Preliminary calculations were made to

determine the effectiveness of varying the ^{235}U concentration axially in the 18-in. diam reactor, with the maximum ^{235}U concentration limited to 10 w/o of the Zr. The results of these calculations are plotted in Fig. 1. The axial peak-to-minimum power density distribution was improved from ~ 2.2 for the unflattened reactor to ~ 1.34 for the power-flattened design. Although the upper limit on ^{235}U concentration in the moderator is the same as the nominal SNAP fuel concentration, there is no assurance that upper limit on power density in the moderator, which is related to acceptable H loss rates, has not been exceeded. This aspect of the design requires further analysis. In any case, ZrH_x -moderated thermionic reactors appear to be amenable to power flattening, and it is expected that even better power-density distributions can be obtained.

Electric power outputs were calculated as a function of axial peak-to-minimum power density ratio for both the 18-in. diam reactor with 55 thermionic fuel rods and for the 20-in. diam reactor with 73 thermionic fuel rods (first row in Table I). Nonflattened radial power-density distributions were assumed for these calculations. The results are plotted in Figs. 2 and 3. The cross-hatched regions in these figures indicate possible operating points between maximum-efficiency and maximum-power operating points. For an axial peak-to-minimum power-density ratio of 1.34, the 18-in. diam reactor containing 55 TFRs can be operated between 60 and 77 kWe;

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Fig. 1. Axial power-density distribution in 18-in. diam. reactor.

Fig. 2. Electric-power output vs axial peak/minimum power density ratio. (Reactor diam 20 in.; 73 TFRs; radial peak/min. power density 1.25.)

Fig. 3. Electric-power output vs axial peak/minimum power-density ratio. (Reactor diam 18 in.; 55 TFRs; radial peak/min power density 1.41.)

the corresponding power range for the reactor containing 73 TFRs is from 95 to 120 kWe.

Although this study is not complete, there seems to be no reason why a shielded space-power system with a net output of 50 kWe weighing less than 4000 lb cannot be designed. A shielded system with a net power output of 100 kWe will apparently weigh between 4000 and 5000 lb.

ZrH_x-Moderated Reactors: ²³³U Fueled

Moderated thermionic reactors fueled with ²³³U are being investigated to determine criticality-limited reactor sizes and to evaluate the performance of such designs. Initial results of this investigation are reported here.

The scope of current studies is constrained by the following ground rules: (1) Consider only one set of moderator and thermionic fuel-rod diameters. (2) Maintain a minimum reflector thickness of 3 in. (3) Emitters consisting of W, enriched to 84% of ¹⁸⁴W, are permitted. (4) Consider only fueled-moderator concepts because designs of near minimum size are of interest. (5) Center-line temperatures in moderator rods are limited by design values for SNAP-8 reactors. (6) Estimates of electric power outputs are based on SIMCON current-voltage data.

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TABLE II. MATERIAL COMPOSITIONS AND RADIAL DIMENSIONS OF MODERATOR AND THERMIONIC FUEL RODS

Thermionic Fuel Rods		Moderator Rods	
Material	Outside Radius, in.	Material	Outside Radius, in.
$^{233}\text{UO}_2$, 85% ρ_o	0.230	$(\text{U-Zr})\text{H}_x$, $N_H = 0.63$	0.335
W, 84% ^{184}W	0.270	Gap	0.340
Gap	0.280	Hastelloy N*	0.350
Nb	0.310	--	--
Al_2O_3	0.330	--	--
Nb	0.340	--	--

*Moderator cladding assumed to be coated with an appropriate glass seal.

Material compositions and radial dimensions of moderator and emitter sections of TFRs are given in Table II. The emitters are 1.57 in. long and are spaced axially 0.47 in. apart. The reactor cores consist of hexagonal arrays of moderator and TFRs with a center-to-center spacing of 0.72 in. Parametric calculations indicate that the moderator-to-thermionic rod ratios in minimum-size reactors will be ≥ 3 and that the U concentration in the moderator should be ~ 8 w/o of the metal. Arrays containing relatively few thermionic rods are of particular interest, but approximately cylindrical geometric arrangements of small numbers of rods are not very numerous. The radial reactor configuration chosen for initial evaluation contains a total of 85 rods, 19 which are TFRs. A cross-section of this reactor is shown in Fig. 4.

The overall diameter of the reactor is 13.57 in., including 3-in. thick radial reflectors. This reactor has sufficient excess reactivity for 10,000 h of full-power operation with a U-concentration in the moderator of 7.5 w/o of the U-Zr alloy, the length is 18.2 in., including 4-in. thick end reflectors. Each TFR contains five thermionic diodes. The cold-clean initial excess reactivity is 8.8%.

The electric-power output was calculated on the basis of the constraints enumerated earlier for the ^{235}U -fueled designs; the results are plotted in Fig. 5 as a function of thermionic efficiency.

Fig. 4. Cross section of ^{233}U -fueled, ZrH_x -moderated reactor.

Fig. 5. Calculated electric-power output of ^{233}U -fueled reactor.

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These calculations were based on nonflattened power-density distributions. The peak-to-minimum power-density ratios are 1.27 radially and 1.84 axially. Additional information for the operating points corresponding to maximum efficiency and maximum power are given in Table III.

Table III. Calculated Power of ^{233}U -Fueled, ZrH_x -Moderated Reactor

	Max. Efficiency Operating Point	Max. Power Operating Point
Electric power, kW	8.2	9.9
Thermionic efficiency, %	14.04	12.88
Electric potential/ TFR, V	3.81	2.89
Total reactor power, kW	146.05	192.15
System efficiency, %	5.61	5.15

TABLE IV. WEIGHT SUMMARY AND POWER DATA FOR NON-SHIELDED THERMIONIC REACTOR SYSTEM (9.9 kWe Reference Design)

Component	Weight, lb
Reactor (core, pressure vessel, reflector)	242
γ -heat shield	38
Cs reservoirs	3
Control-rod drives	53
Instrumentation	44
Reactor total	380
Heat-rejection system	110
Overall Total:	490
	Power, kWe
Pump requirement	0.7
Instrumentation requirement	0.5
Net reactor output	8.7
Total Generated:	9.9

A weight summary for the 9.9-kWe design is given in Table IV. The instrumented reactor weighs 380 lb; and the combination of stainless-steel heat-pipe radiator, heat exchanger, and primary coolant pump weighs 110 lb. Total system weight is consequently slightly less than 500 lb. After supplying power for the primary coolant pump and for the instrumentation, the net power output of this reactor is 8.7 kWe.

Weight summaries for shielded and power-conditioned systems are given in Table V. Shield weights were calculated for four different sets of assumptions.

1. Conventional SNAP shield, with a 12° cone angle and a 10-in. high gallery section, weighing 1310 lb. Power conditioning equipment was included at 4.4 lb/kWe and was assumed to be 90% efficient. The net power output was reduced to 7.7 kWe, and total system weight was 1844 lb.
2. Conventional SNAP shield, but with its gallery removed, which reduced shield weight by $\sim 30\%$. This shield was included in the evaluation to indicate the weight penalty associated with shielding the primary coolant. Shielding of the heat exchanger need only be attenuated by a factor of ~ 10 for the power-conditioning equipment so that the gallery could be moved farther away from the reactor than in the conventional SNAP design. This would reduce shield weight to an intermediate value.

3. SNAP shield with a 0° cone angle. If the power-conditioning equipment can be contained within the diameter of the reactor, shield weight can be reduced by a factor of ~ 3 , resulting in a total shielded-system weight of less than 1000 lb. This clearly indicates the importance of the cone angle on shield weight.

4. The AEC shield design described in a previous section.

Because of geometrical considerations, significant reductions in reactor diameter do not appear to be possible. The electric-power output could be increased by power-flattening and by increasing the emitter surface area. The latter could be achieved either by decreasing the diameter of the ZrH_x and TFRs, thereby increasing the total number of rods in a given core diameter, or by increasing the core diameter. By increasing the core diameter from 7.57 to 8.57 in., the number of thermionic rods and the electric-power output could be approximately doubled. This could be done without increasing the overall size of the reactor by decreasing the radial reflector thickness from 3 in. to 2.5 in., which seems perfectly feasible. Such an arrangement would

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TABLE V. WEIGHT SUMMARY FOR SHIELDED REACTOR SYSTEMS*

	SNAP Shield			AEC Shield
	12° Cone Angle 10-in. Gallery	12° Cone Angle No Gallery	0° Cone Angle --	15° Cone Angle No Gallery
Shield weight, lb	1310	1036	420	797
Weight of power- conditioning equipment, lb	44	44	44	44
Total system weight, lb	1844	1570	954	1331

*The electric power output of all four combinations evaluated was reduced to 7.7 kW.

result in a power supply of ~ 15 kWe, shielded and power conditioned, weighing between 1000 and 1500 lb depending on radiation-shield requirements.

40:60 v/o UO₂-Mo fuel could be used in the 55-TFR reactor and that the only adverse effect would be a decrease of ~ 15% in overall reactor efficiency.

Miscellaneous

One of the most serious problems confronting realistic appraisals of the future of thermionic space-power supplies is the uncertain performance of various possible thermionic fuels. This is due, in large part, to the fact that irradiation testing of the several prospective fuels has been insufficient. The fuel which has been tested most thoroughly at this time is W-coated Mo-UO₂ cermet. Irradiation tests, including 10,000 h tests at approximately twice the fuel power density of current reactor designs, have been completed for 40:60 v/o UO₂-Mo. These tests are discussed in detail in a later section of this report. It has to be pointed out, however, that current reactor designs are based on the use of 57:3:40 v/o UO₂·Y₂O₃-Mo. Obviously, fuel testing is not keeping pace with fuel design. However, because the 40:60 v/o UO₂-Mo fuel tests were very encouraging, a calculation was made for the 18-in. diam, 55-TFR reactor including this fuel. It was found that excess reactivity changed from 10.56% in the original design to 9.88%, and that the fraction of power produced in the moderator increased from 58.59 to 64.94%. These results indicate that the

Irradiation Testing of Fuel Specimens

Introduction

Additional burnup, metallographic, and fission-gas escape data have been obtained from the fuel specimens contained in the four test capsules which comprised the second series in the Mo-UO₂ fuel-pin irradiation program (see LA-3987-MS). All the specimens in this series consisted of Mo-40 v/o UO₂ cermet (~ 10% enriched ²³⁵U) clad with a nominal 0.005 in. of CVD W. They were ~ 1 in. long and 0.25 in. in diam. Irradiation time ranged from 2297 to 10,400 h. Data on particle size, density, open porosity, irradiation conditions, and dimensional changes are summarized in Table VI.

Burnup Measurements

The burnup data given in Table VI are the averages of two separate determinations, one based on the reduction of the ²³⁵U/²³⁸U ratio by burnup and the other on the increase in the ²³⁶U/²³⁸U ratio by neutron capture in ²³⁵U. Separate values are listed in Table VII together with those obtained by radiochemical determination of the amount of ¹⁴⁴Ce

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TABLE VI: IRRADIATION TEST DATA FOR Mo 40 v/o UO₂
Cermet Fuel Pins (10% Enriched UO₂)

Test No.	Irradiation Time, h	Fuel Specimen No.	Initial Particle Size, μ		Density, % Theoretical	Open Porosity, %		Diametral Increase, %		Length Increase, %	Burnup, Fissions/cm ³	Mean Power Density, W/cm ³	Overall Mean Temp., °K
			UO ₂	Mo		Initial	Post-Test	Max	Avg				
LA 26-11	2297	5128 R1-10	125	4	87	10	6.3	0.4	0.2*	0	9.22×10^{19}	328	1620**
		5128 R1-12	125	4	87	10	6.9	1.0	0.6*	0.5	9.42×10^{19}	336	1620**
		5117 R2-15	4	4	91	7	12.7	5.2	0.6	6.5	1.025×10^{20}	365	2140
LA 26-13	3245	5116 R2-2	4	4	89	11	10.9	1.6	0.8	-0.7	1.043×10^{20}	263	1980
		5127 R2-1	125	4	91	7	8.2	4.8	2.4	1.9	1.152×10^{20}	290	2030
		5127 R2-4	125	4	91	7	--	4.2	2.2	2.5	1.152×10^{20}	290	2030
		5127 R2-6	125	4	91	7	6.8	4.0	2.4	1.0	1.165×10^{20}	293	2040
LA 26-12	6113	5116 R2-17	4	4	89	11	--	2.4	1.8	1.9	2.09×10^{20}	280	2000
		5116 R2-14	4	4	89	11	9.6	3.1	2.0	1.7	2.09×10^{20}	280	2000
		5128 R2-8	125	4	87	10	--	4.1	2.6	2.4	2.20×10^{20}	294	2030
		5128 R2-9	125	4	87	10	9.3	4.0	2.4	1.7	2.23×10^{20}	298	2040
LA 26-10	10,400	5116 R2-4	4	4	89	11	--	6.5	3.5	0.6	---	---	--
		5116 R2-12	4	4	89	11	--	5.7	3.5	0.5	3.35×10^{20}	264	1980
		5128 R1-13	125	4	87	10	--	10.0	5.3	4.7	3.46×10^{20}	272	1990
		5128 R1-11	125	4	87	10	--	9.1	4.8	4.3	3.60×10^{20}	283	2000

*Water leaked into test chamber during irradiation.

**estimate based on pressure of 3 atm H₂ in test chamber resulting from water leak.

TABLE VII. COMPARISON OF BURNUP DETERMINATION

Test No.	Irradiation Time, h	Fuel Specimen No.	Fissions/cm ³ x 10 ⁻²⁰		
			²³⁵ U/ ²³⁸ U	²³⁶ U/ ²³⁸ U	¹⁴⁴ Ce
LA 26-11	2297	5128 R1-10	0.86	0.98	0.84
		5128 R1-12	0.94	0.94	0.81
		5117 R2-15	1.04	1.02	0.91
LA 26-13	3245	5116 R2-2	1.09	0.99	--
		5127 R2-1	1.21	1.09	--
		5127 R2-4	1.28	1.06	--
		5127 R2-6	1.19	1.11	--
LA 26-12	6113	5116 R2-17	2.04	2.13	--
		5116 R2-14	2.08	2.13	--
		5128 R2-8	2.17	2.22	--
		5128 R2-9	2.22	2.25	--
LA 26-10	10,400	5116 R2-4	--	--	--
		5116 R2-12	3.35	3.36	3.75
					3.73
		5128 R1-13	3.51	3.41	3.90
					3.71
		5128 R1-11	3.59	3.62	--

produced during irradiation. The burnup values based on isotopic ratios are seen to be in close agreement for the longer irradiation times for which the inherent accuracy of the technique is highest. However, the ¹⁴⁴Ce values differ from the isotopic values by more than the expected error of $\pm 5\%$. For the two tests of shorter duration, the ¹⁴⁴Ce values were lower, and the discrepancy was believed to have been caused by diffusive loss of ¹⁴⁴Ce from the fuel. But this

may not be the correct explanation, as attested by recent data from the 10,400-h test for which higher burnup values were obtained by the ¹⁴⁴Ce technique. The cause of the discrepancies has not yet been determined, but the internal consistency of each set of data suggests the possibility of a systematic error.

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TABLE VIII. RESULTS OF COLLECTION OF CHAMBER GASES LA-26-10
(Results are normalized to Bottle 1 for each chamber)

Bottle No.	Relative Activity	Relative Pressure	Ratio by Volumes	Total ^{85}Kr Atoms in Chamber $\times 10^{-17}$
Top Chamber:				
1	1.00	1.00	1.00	6.84
2	0.76	0.77	0.76	
3*	2.42	--	} 2.36	
4*	0	--		
Total Chamber Content	4.18		4.12	
Lower Chamber:				
1	1.00	1.00	1.00	7.43
2	0.80	0.76	0.76	
3*	2.27	--	} 2.36	
4*	0.11	--		
Total Chamber Content	4.18		4.12	

*Bottle contains 23 g of 4A/5A-molecular sieve powder.

Fission Gas Release

General

Measurements have been completed of fission-gas location in the various samples of the Mo-40 v/o UO_2 cermet specimens from Test Unit LA-26-10 (irradiation time, 10,400 h). Analysis of the gases indicates release values ranging from 50 to 70% for the small-grained cermet samples (5116-R2 series in Table VI) and from 50 to 80% for the large-particle UO_2 cermet specimens (5128-R1 series). Improvements in gas-collection techniques have led to good agreement between the amount of ^{85}Kr actually measured (by collecting the gas vented from the fuel specimens during irradiation and the gas given off when the fuel specimens are dissolved) and the amount calculated from burn-up determinations. Confidence in the measurements of fission-gas retention was enhanced by the fact that adjacent sections of the same fuel specimen gave consistent results, even when one of these sections was dissolved in one stage (Mo and UO_2 together) and the other in two stages (one component at a time).

Gas Escape During Irradiation

Gases released during irradiation were collected by puncturing the irradiation chamber and expanding the gas into 228-cm³ collection bottles. Four bottles were used on each of the two chambers of Test Unit LA-26-10. The gases were sequentially expanded into the first two, and measurements of pressure and comparative volumes in the gas-collection system were used to determine the amount of gas collected in each bottle relative to the total amount of gas initially in the irradiation chamber. A charge of molecular sieve was used in the third and fourth bottles to collect the gases remaining in the system after the two expansions. The purpose of this rather elaborate procedure was to provide a multiple check on the accuracy of the measurement procedure and to ensure that all the gas was collected. The relationship between relative pressures, activities, and volume ratios for the two chambers is given in Table VIII. Of particular interest is the agreement between the relative amount of gas collected by the bottles charged with molecular sieve and the amount calculated by volume ratios to be remaining in the collection system after the first two samples were taken.

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Because the first two samples from each chamber contained no molecular sieve, they presented an opportunity to check the accuracy of the γ -counting measurements of the amount of ^{85}Kr released by mass-spectrometric measurements. These tests indicated that values for total ^{85}Kr obtained by counting should be increased by a factor of 1.24 ± 0.03 if they are to be in agreement with the mass-spectrometer results. Because the initial calibration of the counting method involved the use of known sources at numerous point locations to simulate the extended-source configuration of the sample bottles, this calibration was less direct and more subject to error than the calibration based on mass-spectrographic measurements. Therefore, the latter method was used to compare measured ^{85}Kr yields with the values obtained from burnup measurements. However, the difference between the two calibrations is larger than expected and is the subject of investigation.

Gas Retention During Irradiation

The amount of gas retained in the fuel specimens is measured by collecting all gases released either when both the Mo and the UO_2 are dissolved simultaneously, or when the Mo and the W cladding are dissolved electrochemically and the remaining UO_2 is then dissolved separately. The collection procedure has been modified extensively by using 10 g of 13X molecular sieve in the 230-cm³ collection bottles and sequentially pumping with a minimum of two, and usually three, such bottles to ensure collection of all the gases. The amount of molecular sieve used forms a layer ~ 1 cm deep in the bottom of the bottles. The bottles are cooled in LN_2 before counting to ensure that all the gaseous contents are contained in the molecular sieve. They are placed 60 cm from the γ detector so that the uncertainty in the measurements which could result from nonuniform distribution of the ^{85}Kr in the layer of molecular sieve is less than 3%.

In the cases where either Mo- UO_2 or just UO_2 alone was dissolved, all activity was collected into the first bottle. When W and Mo were selectively dissolved from the fuel samples,

two bottles were required to collect all measurable activity. A third bottle used in a number of such dissolvings was always devoid of measurable activity.

When the measured ^{85}Kr amounts are based on the mass-spectrometric calibration and compared with the predicted total obtained from burnup measurements, the agreement is very good as shown below.

	Total ^{85}Kr Content of Test Unit LA-26-10, atoms ($\times 10^{-17}$)	
	Upper Chamber	Lower Chamber
Gamma activity measurements	13.6	13.0
Burnup measurements	13.1	13.0

The fractional release for ^{85}Kr has been obtained at numerous locations along the length of the fuel specimens from Test Unit LA-26-10. The results are given in Fig. 6 where the percentage released is plotted vs distance from the supported end for each of the four fuel specimens. In each case the release value increases monotonically from the supported end to the free end of the specimen. The dashed line shown for Specimen 5116-R2-4 reflects the increase in diameter of this sample during irradiation. Swelling curves are similar for the other fuel specimens and have already been reported (see LA-4040-MS).

Gas release and diametral increase are obviously correlated. To a certain extent this may reflect an increase in open porosity as swelling occurs. However, the main effect is one of temperature distribution in the specimen. The temperature increases $\sim 200^\circ\text{C}$ from the supported end to a broad maximum located at least 70% of the way from this end toward the free end. The diffusion coefficient for ^{85}Kr in UO_2 is thus much larger near the free end, and escape from the UO_2 particles takes place more readily. This can be seen in Fig. 7 where the gas-retention fractions in both UO_2 and Mo are plotted vs axial position along the specimens. Retention is seen to be much higher in the colder regions (toward the supported ends) indicating that the relative inability of the gas to escape from the UO_2 is the limiting factor.

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Fig. 6. Fractional release of ^{85}Kr (and Cs) along fuel specimens from test unit LA-26-10.

Fig. 7. Gas-retention along fuel specimens from test unit LA-26-10.

The fraction of the gas retained in the Mo phase of the cermet is not strongly dependent on position, i.e., it is not temperature dependent. This indicates that the gas observed is that which recoiled into the Mo and is held either in the Mo lattice or in very small bubbles which are not mobile enough to induce a significant release to open channels in the cermet structure. It is calculated that $\sim 12\%$ of the ^{85}Kr fission product is put into the Mo matrix by recoil from the large ($125\text{-}\mu$ diam) spherical UO_2 particles in the 5128-R1 type cermets. This fraction of fission products for the small-particle cermet (5116-R2) is $\sim 39\%$. The average UO_2 particle size and configuration are the factors which determine the difference of retention in the Mo phase for the two types of cermet.

Cesium Retention Measurements

In addition to the burnup results discussed earlier, burnup determinations have also been made based on the amount of ^{137}Cs found in sections of the fuel specimen. For the porous cermets these measurements invariably gave burnup values much lower than those determined by isotopic ratio from the ^{144}Ce data. The difference in the values is a measure of the amount of Cs fission product which escapes from the fuel during irradiation. The Cs escape percentages were always larger than those obtained for the inert gases. Data for the specimens from Test Unit LA-26-10 are included in Fig. 6. The general behavior as a function of specimen temperature was the same as that observed for ^{85}Kr .

It has recently been realized that the relative escape percentages of Cs and Kr or Xe are significant for determining whether a fuel is venting fission gases continuously at high temperature or releasing gas primarily during temperature cycling or during periods when the fuel is cold. In virtually every test reactor, unprogrammed shutdowns occur with sufficient frequency - on the order of every 100 to 200 h - to preclude the possibility of determining whether a fuel is stable under long-term irradiation conditions when thermal cycling does not occur.

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This factor is particularly important in the case of clad UO_2 fuels where fission gases are vented to a central void by a differential fuel-evaporation sweeping mechanism. Escape from this void appears to occur primarily, if not only, when the UO_2 cracks due to thermal cycling.¹

For the metal- UO_2 cermet fuels, gas escapes at high temperatures by a diffusion process to interface regions between the fuel and the matrix and to UO_2 grain boundaries, where bubbles form and cracking eventually occurs.² If sufficient interconnected porosity exists at operating temperature, a significant fraction of the gas escaping from the fuel particles will also escape from the cermet.^{3,4} If interconnected porosity exists at low temperature but closes up as operating temperature is approached because the UO_2 expands more than the metal matrix, then the gas pressure building up during irradiation will be released only when the cermet temperature has dropped sufficiently for interconnected porosity to open up again.

As the cermet temperature is reduced, any Cs trapped in small pockets will eventually begin to condense (at a temperature which depends on the amount of initially trapped Cs) and the Cs pressure will drop much more rapidly than that of the inert gas which is trapped in the same pockets. If reactor shutdown and startup times are short compared to down times, most of the inert gas will escape at near-ambient temperatures for which the vapor pressure of Cs is insignificant compared to the inert-gas pressures which build up in long-term irradiations. As a result, the escape of Cs will generally be inconsequential compared to the escape of inert fission gases.⁵

The significance of the effect is that gas-release measurements which show a large ratio of inert-gas escape to Cs escape indicate that the release has occurred primarily at low cermet temperatures, i.e., during periods of reactor shutdown. Conversely, comparable escape percentages for Cs and inert gases can be explained in the following two ways: (1) Venting occurs continuously during irradiation, and the diffusion coefficient of Cs in UO_2 is about the same as that for the inert gases. (2) Roughly equal concentra-

tions of Cs and inert gases build up in trapping sites of a cermet having no interconnected porosity at operating temperature and are released during reactor cooldown before the fuel-specimen temperature becomes too low for appreciable Cs transport to occur.

The latter mechanism requires a switching effect on open porosity as the temperature decreases, i.e., from essentially zero to values large enough (with pore diameters large enough) for complete venting of Cs in the affected trapping sites to take place at driving pressures much lower than those for the inert gases (since the Cs pressure for large-enough concentrations will be following the vapor-pressure curve for Cs). The mechanism also requires that the solubility of Cs in UO_2 at high temperatures be very low so that the equilibrium pressure of Cs in the trapping sites be of the same order as that of the inert gases.

Although models can be constructed in principle which would give the behavior required in this mechanism, it is very unlikely that in typically complex cermet structures all the trapping sites would open up to the cermet exterior to the same degree as temperature is reduced. The expected behavior is that the access to the exterior would vary over a wide range including zero. For this situation, the inert gases and the Cs would fractionate and the percentage release of Cs would be reduced relative to that of the inert gases.

Because of the foregoing considerations, it appears logical to ascribe comparable escape percentages of Cs and inert gases to Alternative 1, above, and thus to use the ratio of Cs-to-inert gas escape as an indicator of the amount of continuous venting which is taking place during irradiation.

Variations of Specimen Temperature with Irradiation Time

Because the U in the Mo- UO_2 fuel specimens described above was only 10% enriched in ^{235}U , considerable burnup occurred during irradiation amounting to 48% for the specimens in Test Unit LA-26-10, the 10,400-h test.

Compensation for the effect of burnup required several shifts of position of the test capsules in the MTR during irradiation. The

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Fig. 8. Power density for Mo-UO₂ fuel density vs irradiation time.

criterion for shifting the capsules was to maintain the power density in the fuel within a $\pm 5\%$ range about the design test value; this would give a $\pm 1.5\%$ variation in emitter temperature. The criterion was met fairly well for the three tests of shorter duration, but because of limitations of available flux levels and changes in flux caused by variations in adjacent experimental configurations the criterion was not well met for Test Unit LA-26-10.

Figure 8 shows the variation of power density for one of the center fuel specimens in this test as a function of irradiation time. The power-density values are based on burnup measurements and on MTR flux monitoring results. Test positions were chosen to limit the MTR cycle variations to $\pm 5\%$ so that the actual shape is a saw-toothed curve with a period of ~ 400 h superimposed on the curves shown. Variations in the maximum temperature of the fuel specimens averaged for the MTR cycle variation are also shown in Fig. 8. These values are based on a calibration of power density vs temperature obtained in a test of an identical unit in the OWR.

The initial values of power density and temperature were within $\pm 1\%$ of the design test conditions. The change occurring at 3060 h was not a change in reactor position, but rather a change in neutron flux level. The total spread in cycle-averaged temperature is seen to be 150°K .

UO₂ Diffusion Barriers

Two of the fuel specimens from Test Unit LA-26-12 (irradiation time, 6100 h) contained porous plugs of Mo in the center hole, to serve as a diffusion barrier to UO₂ loss while still providing channels through which fission gas could escape. The differential impedance of the porous structure is based on the fact that the force tending to drive UO₂ through the pores is limited (the vapor pressure of UO₂ is $\sim 10^{-4}$ torr at 2000°K), whereas the pressure available to drive fission gases through is in essence unlimited.³

The porous Mo plugs were manufactured, by electric-spark machining to the desired shape, from a Mo-40 v/o UO₂ cermet block and evaporating the UO₂ by heating to high temperature in vacuum. The resulting piece was slip-fitted into the central fission-gas vent hole of the fuel specimen and held by the specimen support stem when the latter was brazed into place.

Photomicrographs of the fuel specimens irradiated for 6113 h at an average temperature of 2020°K showed that little UO₂ had collected in the plug or in the gap between the plug and the cermet, and that very little depletion of UO₂ had occurred from the cermet. In contrast, the photomicrograph of an unplugged fuel specimen irradiated under the same conditions of time and temperature shows a UO₂ depletion zone ~ 8 mils thick. The photomicrographs are shown on p. 16.

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Photomicrograph of irradiated fuel specimen with porous Mo plug (~ 30X).

Photomicrograph of irradiated fuel specimen with porous Mo plug (~ 150X).

Photomicrograph of irradiated fuel specimen without porous plug (~ 150X).

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Fabrication and Materials Development

Fuel Pin Development

The Mo-60 v/o UO_2 fuel pins are made by dry-blending UO_2 and Mo powders in the presence of paraffin dissolved in hexane and then hydrostatically pressing and sintering the resulting mixture; 1.5 w/o Y_2O_3 is added to the mix to stabilize the UO_2 . In some instances, 9 w/o W was added to the blend to strengthen the Mo matrix. Pins with ~15% open porosity and no closed porosity are considered optimum at this time, the open porosity permitting fission gas to escape to the vent thus minimizing internal pressure and resultant pin swelling.

Tests were conducted to determine the effects of sintering time and temperature on the growth of UO_2 particles in the cermet pin; the results are summarized in Fig. 9. Data are also shown on the effect of Y_2O_3 additions to the UO_2 and to the UO_2 -Mo mix.

It is apparent that UO_2 particle growth occurs as a function of sintering time and temperature with the small (4μ) UO_2 showing significantly greater growth than the 20μ UO_2 starting material. The extent of growth is not as great in the 40 v/o UO_2 cermets as in the 60 v/o cermet. It is felt that this is due to the higher Mo content of the former which blocks sintering and growth of the UO_2 . Blocking of sintering by the Y_2O_3 added to the Mo powder further reduced the UO_2 growth.

In practically every case, higher sintering temperatures and longer times at temperature tend to decrease the open porosity significantly, as expected. The closed porosity changes are not consistent in these experiments. A peculiar effect is noted for the 4μ UO_2 powder in both the 40 and 60 v/o UO_2 cermets after the 24-h treatment at 2200°C . In these specimens, the open porosity increased instead of decreasing as anticipated. The cause for this is not known, but it may simply be specimen to specimen variations.

Metallography on the 60 v/o UO_2 cermets made with 4μ UO_2 indicates a UO_2 matrix surrounding the Mo particles. Structurally this would be a very weak fuel pin since the UO_2 would crack when exposed to any appreciable stress. The pins containing 20μ UO_2 exhibited a Mo matrix and should be relatively strong.

The 20μ average UO_2 particles, when used in conjunction with 4μ Mo particles, tended to block sintering, yielding pins with greater than 20% open porosity. Pins were then made with an intermediate UO_2 particle size (8.8μ) which had densities within acceptable limits.

The average particle sizes of the Mo powders evaluated in conjunction with the 8.8μ UO_2 were 1, 2, and 4μ . After final sintering at 2350°C for 1.5 h in Ar, the pins were heated to 1900° for 100 and 500 h. After the 100 and 500-h treatments, the open and closed porosities of the pins were measured. The results are given in Table IX.

The data show that, although there is little sintering after 100 h, significant sintering occurs after 500 h at 1900°C . The sintering is most pronounced in the cermets prepared from 1μ

Fig. 9. Effect of sintering temperature and time on grain growth in Mo- UO_2 cermets.

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Table IX. Mo-60 v/o UO₂ Cermet*

Metal Composition	Average Particle Size, μ				% Porosity					
					As-Sintered		100 h at 1900°C		500 h at 1900°C	
	Mo	W	UO ₂	Y ₂ O ₃	Open	Closed	Open	Closed	Open	Closed
(Mo-9 w/o W) + 60 v/o UO ₂	1.0	0.7	8.8	0.07	12.72	0.19	12.90	0.0	3.95	3.27
	2.0	0.7	8.8	0.07	20.12	0.14	20.07	0.0	16.50	0.0
	4.0	0.7	8.8	0.07	19.21	0.00	16.44	0.0	12.76	0.0
Mo+60 v/o UO ₂	2.0	-	8.8	0.07	21.13	0.65	19.74	0.0	15.24	0.0

*All samples contained 1.5 w/o Y₂O₃ as a UO₂ stabilizer, were hydrostatically pressed at 30,000 psi and sintered at 1700°C for 3 h in H₂ plus 2350°C for 1.5 h in argon.

Mo where the open porosity is reduced to ~4%, considerably below the desired porosity. The 2 μ and 4 μ Mo powders yield pins with open porosities close to 15% after 500 h at 1900°C.

Pins for MTR irradiation tests are being made using both 1 and 4 μ Mo powders. The decision to use the 1 μ Mo powder was made after 100-h tests and prior to completion of the 500-h tests. The 2 μ powder also gave favorable results after 500 h, but this powder is not considered reproducible enough to use for fabrication of MTR pins because of the presence of large particles. The 4 μ Mo powder has been used extensively and is a more reproducible material.

Tacks must be attached to both ends of the fuel pins for support purposes. It is proposed that these TZM (Mo) tacks be diffusion bonded to the Mo-UO₂ pin. To prevent degradation of the joint by the UO₂, the pins are coated with 0.001-0.002 in. W by pyrolytic deposition from W(CO)₆ prior to diffusion bonding. After the tack is bonded to the pin, additional W is deposited by the H₂ reduction of WF₆. The WF₆ reacts with the exposed UO₂, thus dictating the use of the W(CO)₆ process for the initial thin coating.

The latest design consists of a 0.462-in. o.d. x 1.420 in. long Mo-60 v/o UO₂ pin with a 0.012-in. wide cruciform slot Eloxed the length of the pin. The slots are Eloxed to within 0.015-

0.025-in. of the outside surface of the pin. It is necessary to Elox from both ends approximately halfway to prevent longitudinal bowing of the slot. This leaves a small offset (~0.002 in.) at the center. It should be of little consequence as far as swelling is concerned since mating faces have the proper spacing. The thin wall over the slot eliminates the problems involved in masking the slot to permit a continuous W coating to be applied over the entire external surface of the pin.

One attempt was made to diffusion-bond a TZM (Mo) tack to a pin of this design. However, too high a pressure was applied during bonding and the pin split. Additional pins are on hand for similar experiments.

Tri-Layer Development

In the development of metal-sheathed tri-layers, both pure Nb and Nb-1% Zr are being considered as the metallic component. The latter is generally preferred because of its superior mechanical properties and chemical stability at high temperatures.

Earlier results of the various attempts made to produce satisfactory pressure-bonded tri-layers, using both Nb and Nb-1% Zr, were described in detail in LA-4040-MS. In most cases there appeared to be good contact between the metal and the ceramic, but in only one out of 21

autoclaved assemblies was the insulator free of cracks.

Other investigators have demonstrated that Nb/Al₂O₃/Nb tri-layers can be fabricated without cracking the insulator if the radial clearance between the inside sheath and the ceramic tube is kept relatively wide^{1,2} and if bonding temperatures are around 1300°C. Four new assemblies were fabricated to investigate the applicability of this technique to the smaller-diameter LASL tri-layers. The compositions and wall thicknesses were as follows:

Spec. No.	Inner Sheath	Insulator	Outer Sheath
26	Nb (0.030 in.)	Lucalox (0.020 in.)	Nb (0.010 in.)
27	Nb (0.050 in.)	Lucalox (0.020 in.)	Nb (0.010 in.)
28	Nb (0.050 in.)	Lucalox (0.020 in.)	Nb-1% Zr (0.010 in.)
29	Nb-1% Zr (0.050 in.)	Lucalox (0.020 in.)	Nb-1% Zr (0.010 in.)

The radial clearances between the inner sheaths and the Lucalox tubes were increased from 0.001 ± 0.0005 to 0.004 ± 0.001 in., but the radial clearances between the Lucalox tubes and the outer sheaths remained unchanged (0.001 ± 0.0005 in.). These units were autoclaved by an outside vendor at 1300°C for 3 h at a pressure of 10,000 psi. Cooling time was ~ 4 h. The pressure was released when the pressure vessel reached a temperature of ~200°C.

Specimen 28 developed a leak while being autoclaved and was not bonded. The outer sheaths of Specimens 26, 27, and 29 were etched away from the insulators in three $\frac{1}{4}$ -in. wide bands, exposing each end and a center section of the insulators. There were no cracks in Specimen 26. In the remaining two specimens there were as few or fewer cracks than in the best of the specimens from previous runs. Thus, increasing the clearance between the inner sheath and the ceramic tube proved to be beneficial. Furthermore, as has been previously observed, the thinner the wall of the inside sheath, the better the chances of producing a crack-free assembly.

In attempting to cut cylindrical sections from these pieces, however, new cracks appeared in the insulators as soon as the cutoff wheel touched the surface of the ceramic. Cutting was done as carefully as possible with the best equipment using No. 80 metal-bonded diamond wheels. In a final effort to obtain crack-free samples, an abrasive-coated wire was drawn across the ceramic tube, but it still produced cracks. Specimen 26, which had not cracked during autoclaving, was quite obviously the most sensitive to any of the cutting operations. Specimens 27 and 29 seemed to be less sensitive. Two similar tri-layer assemblies from a previous set which had been cooled slowly (18 h) from 1650°C to ambient and which also contained only a few cracks could be sectioned quite easily without producing additional cracks.

Evidently, the current pressure-bonding schedules leave high residual stresses in the insulators. It is often assumed that a mismatch of the coefficients of thermal expansion (CTE) between the metal and the ceramic may be responsible for such stresses; CTE data on AD 995,³ Lucalox,⁴ Coors AD 999,⁵ and Nb⁶ (all from different sources) do show significant differences. When the CTEs of Nb, Nb-1% Zr, and Lucalox were measured at LASL, however, all in the same equipment, the differences were negligibly small over a temperature range from 25° to 1800°C. If, as seems likely, the latter comparative data are more reliable, then a mismatch of CTE is not responsible for the stressed insulators.

A stress analysis was made using a finite-element computer code to determine the stress distributions within a tri-layer assembly caused by relieving the autoclave pressure. The analysis showed that the tensile stresses within the Al₂O₃ layer (away from the ends) were of the order of 10,000 psi, in both the tangential and axial directions, but the stresses peaked very sharply at or near the ends of the Al₂O₃ layer. Because the stress code is a finite-element code, the stresses at the end cannot be calculated; they can only be approximated by extrapolation. When a finer mesh with elements of 0.001 in. thickness

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were used in the code, a maximum stress of ~ 60,000 psi was obtained at a distance of 0.005 in. from the ends and the stress gradient was still increasing.

Creep data for Nb and Nb-1% Zr show a very high creep rate at elevated temperatures. This indicates that the stresses would be relieved by the creep of the metal if the autoclave pressures were relieved slowly at elevated temperature. Because there appears to be no appreciable CTE mismatch, no stresses would be introduced by reducing the temperature.

To test this conclusion, four new tri-layer assemblies, composed as those described above, are now being autoclaved by an outside vendor. After holding the assemblies at 1525°C and 10,000 psi for 3 h, the pressure will be released slowly over a period of 2 h. The assemblies will be held at 1525°C for an additional 3 h after the start of the pressure release. The power will then be shut off and the assemblies will be allowed to cool to ambient in the autoclaving vessel. This cooldown requires ~ 5 h.

An additional eight tri-layer assemblies are being autoclaved by an outside vendor. Two Nb/Lucalox/Nb assemblies similar to specimen 26, will be used in thermal conductivity studies; whereas three Nb/Lucalox/Nb and three Nb-1% Zr/Lucalox/Nb-1% Zr assemblies of a modular design suitable for a single thermionic cell will be used in a study of fabrication techniques for multiple-cell fuel rods. They will be autoclaved in accordance with the proposed new schedule, except that two assemblies of modular design (one Nb/Lucalox/Nb and one Nb-1% Zr/Lucalox/Nb-1% Zr) and the two assemblies similar to Specimen 26 will be autoclaved at 1300° instead of 1525°C.

Thermal Conductance of Tri-Layer Assemblies

Further thermal-conductance measurements have been made on the tri-layer assemblies described in LA-4040-MS, and on one additional assembly. Investigations of the sources and magnitudes of errors in the temperature readings indicate that the information available is insufficient to differentiate between the temperature-drop data taken in vacuum and those taken in inert gas in a water calorimeter (see LA-4040-MS, Figs. 8 and 9). Figure 10 shows the dependence of temperature drop on radial heat flux for tri-layer Assembly TLT-11 when one curve is fitted to the data points obtained both in vacuum and in inert gas (see LA-4040-MS, Fig. 9). Additional data points obtained for TLT-2 are also shown in Fig. 10 as well as the single curve which fits them. Both of these units had a 0.010-in. thick Nb-1% Zr outer sheath, a 0.020-in. Lucalox insulator tube, and a 0.050 in. Nb-1% Zr inner tube. They were autoclaved at 10,000 psi for 3 h at a temperature of 1260°C for TLT-2 and at 1460°C for TLT-11. The accuracy of the data is not sufficient to distinguish a significant difference in thermal conductance between the two samples.

Both of these units had the ceramic insulator tube exposed to the gas in the calorimeter so that gas may have diffused into the tri-layer region and resulted in a thermal conductance higher than one attainable in vacuum, assuming that high enough heat fluxes could be obtained for such measurements. Because of the uncertainty introduced by this possible effect, a third unit was assembled by building a Na heat pipe into an autoclaved sample for which the ceramic tube was not exposed. Data obtained for this unit (TLT-21), which was bonded for 3 h at 1650°C and 10,000 psi, are shown in Fig. 11.

The finite temperature drop observed for zero heat flux is assumed to be a result of a difference in emissivity between the two points on the assembly from which the temperature differences were obtained. In Fig. 12 the data for all three units are corrected for this zero displacement and compared to the curve labelled "ideal" which is expected when no temperature drop occurs at the metal-ceramic interfaces.

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Fig. 10. Temperature drop vs radial heat flux for tri-layer assemblies bonded at 1260 and 1460°C.

Fig. 11. Temperature drop vs radial heat flux for tri-layer assemblies bonded at 1650°C.

Fig. 12. Data in Figs. 10 and 11 corrected to zero temperature at zero flux.

The curves show that near-ideal behavior was obtained for all three units. A test unit autoclaved at 1300°C is currently being assembled with the precaution, again, of not exposing the insulator to gas. Data from this unit should determine whether any significant differences in thermal conductance arise from varying the autoclaving temperature.

Helium Diffusion Through Nb and Nb-1% Zr

It was suggested that He might diffuse at an external pressure of 10,000 psi (~700 atm) during autoclaving of tri-layer assemblies through the thin Nb or Nb-1% Zr walls, which are at 1300-1500°C, into the space between the metal sheaths and the ceramic tube in sufficient quantities to inhibit bonding.

It would have required an elaborate experiment to measure the diffusion of He through the metal during autoclaving. Therefore, a simpler experiment at lower pressures was devised, which, it is hoped, would at least determine the limits of the problem.

Closed cylinders of Nb and Nb-1% Zr (0.680-in. o.d. by 3-in. long with 0.010-in. thick walls) were attached to $\frac{1}{4}$ -in. o.d. tubes of similar metal. Each of the assemblies was sealed into a quartz tube with the small-diameter tube projecting through the seal, and the quartz tube was connected to a He leak detector to indicate the rate of He diffusion. The metal cylinders were heated inductively through the quartz tube; thus, the inside of the metal cylinders could be either evacuated or pressurized with He while being heated to various temperatures.

To summarize the results, the leak detector at a maximum sensitivity of 1.3×10^{-10} atm/cm³/sec could not determine whether there was a vacuum or an atmosphere of He inside the metal cylinder at any temperature up to 1500°C. The quartz tube was shut off for periods up to 30 min to see if He might accumulate in this tube. Although there was some deflection of the gauge when the stopcock was opened, the leak detector

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still could not distinguish between an internal vacuum or a He atmosphere in the metal cylinder. The deflection may have been due to outgassing of the apparatus. These results establish an upper limit on a possible rate of He diffusion through Nb or Nb-1% Zr, but they are inconclusive as to whether the diffusion is of sufficient magnitude to be detrimental to the pressure-bonding process.

EBR-II and OWR Isothermal Irradiation Tests

The EBR-II isothermal irradiation experiment was designed to determine the long-term irradiation effects on Al 995, AD 999, Lucalox, and Y₂O₃ insulators. Since the start of the experiment, the samples have accumulated 1260 h of irradiation time at full power (50 MW) in an estimated neutron flux of 1.8×10^{14} n/cm²/sec ($E_n > 1$ MeV) at a nominal temperature of 750°C.

Samples OWR-I and OWR-II, which are mockups of EBR-II, were removed from the Omega West Reactor. The OWR-I experiment was terminated according to plan, after a total irradiation time of 7779 h. The OWR-II test, which had accumulated 4850 h of irradiation time, was terminated because the thermocouples on the isothermal section of the heat pipe, which measure the temperature of the ceramic test specimens, indicated that the temperature had dropped over the last eight weeks from 750°C to less than 620°C. The reason for this temperature decline has not yet been determined.

Both experiments are now in the hot cell for evaluation.

High-Temperature Metal-Ceramic Bonds

The requirement for high-temperature metal-ceramic bonds in a thermionic reactor resulted in a study over the past several years of high-temperature (1800-1900°C) brazing materials and techniques. Recent design changes in insulators and the acquisition of a new braze material necessitated a reevaluation of the older techniques.

Insulators of Al 995 and Lucalox were bonded to Nb-1% Zr skirts with two types of brazing material: 65:35 w/o V-Nb, 0.004 in. thick by 0.02 in. wide -- the older brazing alloy; and

52:38:10 w/o Nb-V-Ti, 0.010 in. thick by 0.020 in. wide. The brazing areas of the insulators were W-metallized and brazed to the Nb-1% Zr test pieces. The thickness of the metal at the braze joint was 0.010 in. The insulators and the Nb-1% Zr test pieces were cleaned with Freon TF solvent and were outgassed in a vacuum furnace ($< 10^{-4}$ torr) at 1900°C. The braze material was cleaned and also outgassed in vacuum at 1000°C. A W brazing jig was used to maintain alignment of the insulator assembly. This jig, with the assembly in place, was placed inside a W radiation shield inside the vacuum furnace. A 920-g W weight kept pressure on the assembly and, at the time of melt, ensured proper mating of the components. The assembly was then brought to a temperature of 1000°C and allowed to equilibrate before raising to brazing temperature. The 65:35 w/o V-Nb assemblies were then heated to 1900°C (observed by optical pyrometer) and held for 5 min, whereas the 52:38:10 w/o Nb-V-Ti assemblies were brought to 1875°C and held for 4 min. A cooldown period of 3 h was allowed before removing the assemblies from the furnace.

Metallographic examination of the sectioned 65:35 w/o V-Nb assemblies showed that the braze material did not flow the full length of the lips and did not form a fillet at the top. The upper end of the lips was belled out, pulling away from the ceramic, and there were stress cracks at the bottom of the insulator. These results are typical of all the assemblies. The 52:38:10 w/o Nb-V-Ti assemblies, however, showed good flow characteristics with a good fillet at the top and good penetration of the braze material into the ceramic, but the lip at the top was belled and pulled away from the ceramic.

To relieve the stress cracking and belling of the lips, the end of the insulator was tapered and the clearance was opened up (by ~0.002 in.) between the insulator and the Nb-1% Zr lips. In addition, 30% more braze material was used to allow for the increase in clearance. Photomicrographs taken from a 52:38:10 w/o Nb-V-Ti assembly after modification showed only very slight belling, good flow characteristics, and good penetration of the brazing material into the ceramic. One sample showed a slight crack at the bottom of the braze, but this was not general.

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Fig. 13. Electron microprobe (x-ray intensity) data indicating penetration of Nb, Al_2O_3 , V, and W through braze area.

An electron-microprobe examination of the 65:35 w/o V-Nb assembly, made at the metal-ceramic interface, revealed that the V had penetrated the Nb-1% Zr lips to depths of $\sim 100 \mu$. The results are shown in Fig. 13. Trace amounts of V, but not of Nb or W, were detected in the grain boundaries in the Al_2O_3 to a distance of 100μ from the metal-ceramic interface. An electron microprobe examination of the 52:38:10 w/o Nb-V-Ti assembly is being made.

Diffusion Bonding Studies

Samples of bonded assemblies which had been subjected to post-heat treatment at 1100°C for different time intervals (see LA-4040-MS) were evaluated by electron-microprobe analysis to determine the depth of diffusion of Ni into the Nb.

The x-ray intensities of the Ni and Nb were measured at appropriate intervals, proceeding from one piece of Nb through the Ni-bonded area to the second piece of Nb. The intensities, after correcting for detector dead time and background, were divided by the intensities from the respective pure metals to obtain the intensity ratio. The ratio units obtained have standard deviations of $\sim \pm 0.006$ and ± 0.00017 for the Nb and the Ni, respectively. The highest intensity ratio for the Ni was at the bonded-joint area. This ratio for the sample heat-soaked at 1100°C for 80 h averaged 0.0520, whereas that for the sample heat-soaked

for 200 h averaged 0.00215. Because suitable alloys with known Nb and Ni content were not available, the intensity ratios cannot be interpreted as concentrations.

The x-ray intensity ratios indicate diffusion of Ni into the Nb to a depth of 50 to 80μ for the first sample (80 h) and to a depth of $\sim 100 \mu$ for the second sample (200 h). It is estimated that the electron-microprobe detection limit for Ni in Nb ranges between 0.01 and 0.1%.

The depletion of Ni from the joint after the heat soaks did not leave a low-density zone because the x-ray intensity ratio for the Nb in the bonded area was within the deviation limit (± 0.006) of unity. This shows that the density is the same from one piece of Nb through the bonded area to the second piece of Nb.

Miscellaneous

Six heavy-walled Al tubes were loaded with 95% enriched UO_2 powder for "worth" determinations in the SNAPTRAN assembly at Atomics International. Two of the tubes had enriched U disks placed ~ 1.5 in. on either side of the center.

The powder was compacted in the tubes by using a Syntrol vibrator polisher during loading. After loading, the tubes were radiographed to determine the exact location of the ^{235}U disks and if any density variations were present. Tube No. 5 had the ^{235}U disks 4.05 and 7.02 in. from the top and tube No. 6 had the disks 4.17 and 7.30 in. from the top. The UO_2 was uniformly packed. The weights of UO_2 ranged from 291.68 to 302.97 g and the densities from 6.251 to 6.556 g/cm^3 (56.65 to 59.76 % of theoretical).

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HEAT PIPE SYSTEMS

The results obtained in the work on Heat
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in Space Electric Power R and D Program Report,
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