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SOME SCOPING EXPERIMENTS
FOR A SPACE REACTOR

Task 128

BATTELLE
Columbus Laboratories

July 7, 1983

by

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Cc: C. A. Alexander
J. S. Ogden
J. E. Davis/Contracts

August 10, 1983

R. E. Smith Jr., Lt. Col.
Department of Energy
Washington, D. C.

Dear Colonel Smith:

Ref: W-7405-Eng-92-128

Attached is our final report entitled "Some Scoping Experiments For A Space Reactor". If you have any questions of a technical nature please do not hesitate to contact me at (614)-424-5233.

Sincerely,

C. A. Alexander
Manager
Physico-Chemical Systems Section

CAA:ahf

Enc.

EXECUTIVE SUMMARY

Some scoping experiments were performed to evaluate fuel performance in a lithium heat pipe reactor operating at a nominal 1500K heat pipe temperature. Fuel-coolant and fuel-coolant-clad relationships showed that once a failed heat pipe occurs temperatures can rise high enough so that large concentrations of uranium can be transported by the vapor phase. Upon condensation this uranium would be capable of penetrating heat pipes adjacent to the failed pipe. The potential for propagation of failure exists with UO_2 and a lithium heat pipe. Changing the composition of the metal of the heat pipe would have only a second order effect on the kinetics of the failure mechanism.

Uranium carbide and nitride were considered as potential fuels which are nonreactive in a lithium environment. At high temperatures the nitride would be favored because of its better compatibility with potential cladding materials. Compositions of UN with small additions of YN appear to offer very attractive properties for a compact high temperature high power density reactor.

CONTACT CONDUCTANCE

Introduction

Contact conductance is a measure of the effectiveness of heat transfer surfaces. Anytime there is a gap or two dissimilar materials are in contact then there is a temperature discontinuity at this interface. For the SP-100 then the interface between the fuel and refractory metal fin forms such a gap. Even with very good bonding between metal and UO_2 it would be expected that a temperature discontinuity would exist because of the differences in thermal conduction mechanism at the interface. Conduction in UO_2 is overwhelmingly phonon induced, while in the alloy the heat is carried principally by electrons. There must be then a phonon-electron interaction at the interface between the materials. Also there would be photon transport across the interface. These two processes electron-phonon interactions and net photon transport would combine and form the contact conductance across the interface.

Experimental

There are four devices at BCL with which contact conductances can be determined. Three of these utilize a heat meter of some sort. Two of them require that the thermal conductivity of the heat meter be accurately known. A stainless steel - intercompared with ORNL is used in the low temperature heat meter (700 C). A higher temperature model utilizing NBS thermal conductivity tungsten (NBS 730) has been employed to about 1200 C but this is close to its upper limit of applicability. A third device employs a long heat pulse and does not require knowledge of thermal conductivity, only heat capacity of one material. This device is very good for thin films of the order of 0.05 mm (2 mil). It is not applicable where radiant transfer becomes significant, hence was not applicable for the present application. For this application a diffusivity technique was selected. The flash diffusivity technique ⁽¹⁾ was selected. In this method as developed at BCL ⁽²⁾ a small disk (ideally) about the size and thickness of a dime is irradiated by a laser pulse of several joules in a 1.6 millisecond pulse. The diffusivity is computed from

the rise time of the temperature pulse on the side opposite the input laser pulse. It is customary to use $t_{1/2}$, the time from initiation of the pulse to where the temperature reaches one half its maximum value. A correction due to Cowan⁽³⁾ accounts for heat loss from the material during the actual experiment.

The components in the BCL diffusivity device are a Korad 1.2 cm dia neodymium glass laser with a Spectra-Physics Helium-Neon alignment laser coaxial to the Korad laser. The sample is maintained in a diffusion pumped freon trapped vacuum furnace where the heater is a tungsten mesh resistance element. IRTRAN windows at the bottom and top allow the beam and signal passage out to the detector. An IRTRAN lens focuses the output pulse onto the detector. The detector is an InSb infrared detector built into the bottom of a liquid nitrogen dewar. A sapphire window protects the InSb crystal. It is this sapphire window which limits the low temperature operation to about 100 C. Below 100 C the light output has a wavelength of several microns and infrared of this wavelength is not transmitted through sapphire. The output of the InSb detector is routed to a Nicolet 2094 digital storage oscilloscope. The output is stored on a floppy disk and can then be transferred through an RS 232 interface to a mainframe on to the DEC PDP 11-34 for further workup.

Diffusivity then can be calculated from the experimentally determined $t_{1/2}$ by the relation

$$\alpha = \frac{0.139 \ell^2}{t_{1/2}}$$

here $t_{1/2}$ is the experimentally determined half rise time and ℓ is specimen thickness expressed in centimeters. Use of $t_{1/2}$ is convenient but in general

$$\alpha = \frac{k_T \ell^2}{t_T}$$

here k_T is a constant related to the T percent rise. In contact conductance use is made of this more general relationship. The overall diffusivity apparatus is shown in Figure 1.

FIGURE 1. OVERALL VIEW OF DIFFUSIVITY APPARATUS

Results

For contact conductance the two layer diffusivity relationships developed by Larson and Kayama⁽⁴⁾ have been modified at BCL so that a term involving an interfacial resistance can be added to the two known thermal conductivities of the materials. The input data needed for computation are thickness, heat capacity, and diffusivity of each individual layer. Values for $t_{1/4}$, $t_{1/2}$, and $t_{3/4}$ for the composite diffusivity is given to the computer along with an initial estimate for this interfacial resistivity. The computer then performs an iterative repetitive computation where it computes a response curve, determines $t_{1/4}$, $t_{1/2}$, and $t_{3/4}$ from the computed curve and compares these computed values with the observed values.

The first step was to determine diffusivity and thermal conductivity of a prototypical UO_2 . A PWR fuel pin obtained from EG and G Idaho was selected because its density and its processing parameters were considered most nearly comparable to the reference SP-100 fuel. Results obtained are tabularized in Table 1. The molybdenum utilized was cut from a casting of ClimeIt molybdenum from the BCL diffusivity archive. This choice - while not a molybdenum-rhenium alloy was electron-beam melted and subsequently drawn. It was the contention that this structure would more closely approximate the Mo-Re alloy than would a powder-compacted specimen. Values obtained for the molybdenum are shown in Table 2.

Two sets of experiments were performed. In the first, the UO_2 and molybdenum were both surface lapped with 600 grit paper and ethanol. The lapped surfaces were in contact and only gravity maintained contact. The molybdenum was on top so that the heat pulse passed through the UO_2 into the molybdenum as is the case for SP 100. One typical result of the computer interaction is shown in Figure 2. This was for a temperature of 1500 K and gives a value of $0.29 \text{ w/cm}^2\text{sec}$ for contact conductance. A temperature of 1800 K was also run. Here the contact was $0.25 \text{ w/cm}^2 \text{ sec}$.

A second set of experiments were run. In this case the UO_2 and molybdenum was diffusion bonded at about 10 atmospheres pressure (150 psi) at 2100 K for 16 hours. The diffusion couple was removed from the bonding furnace

TABLE 1. THERMAL DIFFUSIVITIES AND CONDUCTIVITY
OF UO_2 OF 95% THEORETICAL DENSITY

Temp ^o C	Thermal Diffusivity Cm^2/Sec	Thermal Conductivity W/Cm^o
840	0.01037	0.0338
970	0.00881	0.0288
1060	0.00816	0.0268
1170	0.00721	0.0239
1260	0.00680	0.0227
1345	0.00663	0.0224
1420	0.00610	0.0209
1520	0.00630	0.0208
1625	0.00578	0.0193
1725	0.00563	0.0183
1825	0.00500	0.0191
1930	0.00477	0.0191
2025	0.00475	0.0200
2075	0.00475	0.0205

TABLE 2. THERMAL DIFFUSIVITIES AND CONDUCTIVITIES OF CLIMELT MOLYBDENUM

Temp ^o C	Thermal Diffusivity cm ² /sec	Thermal Conductivity w/Cm ^o
1200	0.34	1.14
1400	0.33	1.13
1600	0.32	1.11
1800	0.31	1.09

and loaded into the diffusivity apparatus. In general values about 50 percent higher were observed.

Discussion

Under design conditions the Sp-100 produces about 16 watts/cm². Computed values for ΔT between fuel and fin are shown in Table 3.

TABLE 3. VALUES FOR ΔT BETWEEN FUEL AND FIN FOR 16W/CM² GENERATION

Nominal Temperature K	Initial Gravity Gap ^o C	Diffusion Bonded Gap ^o C
1500	55	45
1800	64	47

From the above it can be seen that the fuel will run cooler once some fuel movement has occurred, but there will remain a significant difference and for evaluation of vapor transport, and creep, and swelling characteristics these temperatures should be utilized as the minimum value for fuel temperature.

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R DOUBLE
PULSE TIME? .002
ALPHA(1),L(1),CP(1),D(1)? .34,.24,.343,10.2
ALPHA(2),L(2),CP(2),D(2)? .0072,.063,.315,10.
T(25%),T(50%),T(75%)? .30,.62,1.1
MAXIMUM TERMS? 10
ENTER SGL .5
0.11761778D+01 0.36309932D+01 0.57201519D+01 0.65962442D+01
0.96258075D+01 0.11372626D+02 0.12725575D+02 0.15830974D+02
0.17029577D+02 0.18955448D+02
CONTACT CONDUCTANCE PARAMETER 0.7200D+00
5 TERMS
25.0 0.383321022D+00
50.0 0.723534968D+00
75.0 0.917111439D+00
0.94659639D+00 0.34224213D+01 0.56997445D+01 0.64441427D+01
0.95284374D+01 0.11354128D+02 0.12646207D+02 0.15770648D+02
0.17015081D+02 0.18902618D+02
CONTACT CONDUCTANCE PARAMETER 0.3600D+00
5 TERMS
25.0 0.262497806D+00
50.0 0.561226927D+00
75.0 0.738909600D+00
0.72226220D+00 0.32915798D+01 0.56853963D+01 0.63636239D+01
0.94772691D+01 0.11344231D+02 0.12606256D+02 0.15739563D+02
0.17007973D+02 0.18676092D+02
CONTACT CONDUCTANCE PARAMETER 0.1800D+00
5 TERMS
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50.0 0.378863829D+00
75.0 0.605632614D+00
0.91454946D+00 0.34000474D+01 0.56973671D+01 0.64295940D+01
0.95192451D+01 0.11352356D+02 0.12638946D+02 0.15765038D+02
0.17013782D+02 0.18697798D+02
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75.0 0.776046563D+00
0.88549136D+00 0.33609285D+01 0.56953428D+01 0.64174287D+01
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0.17012701D+02 0.18693780D+02
CONTACT CONDUCTANCE PARAMETER 0.3000D+00
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50.0 0.512887367D+00
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5 TERMS
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75.0 0.732532003D+00
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0.95192451D+01 0.11352356D+02 0.12638946D+02 0.15765038D+02
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CONTACT CONDUCTANCE PARAMETER 0.3273D+00
5 TERMS
25.0 0.246740483D+00
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0.88827559D+00 0.33627140D+01 0.56955349D+01 0.64185546D+01
0.95122588D+01 0.11351007D+02 0.12633454D+02 0.15760783D+02
0.17012801D+02 0.18694152D+02
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25.0 0.234066902D+00
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0.95108440D+01 0.11350734D+02 0.12632344D+02 0.15759922D+02
0.17012603D+02 0.18693415D+02
CONTACT CONDUCTANCE PARAMETER 0.2975D+00
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75.0 0.751724965D+00
0.87999913D+00 0.33774346D+01 0.56949662D+01 0.64152315D+01
0.95101535D+01 0.11350600D+02 0.12631802D+02 0.15759501D+02
0.17012507D+02 0.18693056D+02
CONTACT CONDUCTANCE PARAMETER 0.2951D+00

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FIGURE 2. ITERATIVE COMPUTATION OF CONTACT CONDUCTANCE

ADVANCED FUELS

Introduction

UO_2 is a very forgiving material. It is compatible with stainless steels, refractory metals, and even other ceramics. It is relatively stable, easy to fabricate and, nonhygroscopic (as UO_2). It does possess a complex vapor phase, and has nearly the poorest thermal conductivity of any potential nuclear fuel. It is capable of long time operation at low temperature or short time operation at high temperature. It was early recognized (1973) in the breeder program that doubling rates with $UPuO_2$ would be unsuitably low because of the temperature limitation of oxide base fuels. For the breeder both the carbide and nitride proved greatly superior. A comparison is given in Table 4.

TABLE 4. OPERATING POWER LEVELS FOR THE LMFBR

Fuel Type	Power KW/ft
$UPuO_2$	11
$UPuC$	32
$UPuN$	44

As can be seen there is a gain by a factor of three for carbide over oxide, and by four for nitride over oxide. The carbide is cheaper to make and easier to fabricate so the gain of nitride over carbide is not as great as is shown. There would be two cases where nitride would be superior. Case I. would be when center temperature do not exceed 1350 C. Figure 3 reproduced from BMI 1918 show how well $UPuN$ can retain fission products at these temperatures. Swelling is even less than that predicted for solid fission products, thus indicating that much of the swelling is contained in the porosity of the fuel. The carbide is just not this strong. For Case II. the nitride is superior to the carbide for $T > 1600$ C here, superiority is not based on swelling but on compatibility. Alexander et. al.⁽⁵⁾ showed that compatibility was very limited for all common

FIGURE 3. VOLUME CHANGE OF (U, Pu)N SPECIMENS AS A FUNCTION OF BURNUP

materials with carbides at elevated temperatures. These results are summarized in Figure 4 taken from Reference 5.

Selection of Nitride

The nitride suffers from one drawback and that is a preferential loss of nitrogen with concomitant production of molten uranium. Alexander, Ogden and Pardue⁽⁶⁾ showed this phenomenon occurs at all temperatures above the melting point of uranium (1133 C). The same authors showed that the evaporation coefficient of nitrogen from the nitride was about 0.001 so that the rate at which the molten metal formation occurs is very slow below about 1700 C. In a closed fuel pin it is easy to add a bit of nitrogen or even let the reaction go a bit till the equilibrium nitrogen pressure limits the reaction. Not much nitrogen pressure is needed, as can be seen from Figure 5. At 2000K the nitrogen decomposition pressure is about 10^{-8} . In an open system it would be likely that this pressure would be reached. It is possible to calculate the generation rate of uranium at the surface. For UN the nitrogen volatilizes as N_2 so the molar rate of uranium formed is twice the molar rate of nitrogen removed. The molar rate of nitrogen removed is given by

$$Z = \frac{\alpha 44.3P}{\sqrt{MT}}$$

Here Z is moles nitrogen volatilized per second per square centimeter of surface, α is the evaporation coefficient whose value is 0.001, P is the equilibrium decomposition pressure 10^{-8} atm, M is the molecular weight of nitrogen, 28, and T is the temperature, 2000K. Solving the above gives 1.8×10^{-12} for the molar rate of nitrogen loss the uranium buildup then is twice this, or 3.7×10^{-12} moles/sec cm^2 . Multiplying by 237 (for 20 percent enriched) gives 8.9×10^{-10} g/sec cm^2 . This is a rate of metal formation of about 5×10^{-11} cm/sec or about 2×10^{-8} mil/sec. In a year then a little over a half mil of uranium would form on the surface with a surface temperature 2000 K. At 1800 K it would take about ten years to generate this much molten metal. A complete fix to the above situation is to maintain enough nitrogen around to keep from reaching the phase boundary where the free metal forms. This can be done by simply putting a nitrogen source in the system. Boron Nitride for example could be employed as

FIGURE 4. COMPARISON OF FREE ENERGY OF FORMATION OF U, W, Mo, Ta, Zr AND Ti CARBIDES

FIGURE 5. URANIUM-NITROGEN PHASE DIAGRAM

a burnable poison as in a control rod configuration. Its slow decomposition could supply the nitrogen overpressure to prevent liquid formation.

There is a more direct method however to tailor a fuel for a specific application and that is to form a mixed nitride in such a manner that decomposition gives rise to a dissociation pressure high enough to prevent metal formation. This concept sounds more complex than it really is. It was observed⁽⁷⁾ that UN and PuN form nearly ideal solutions ranging from 20^m/o UN to 80^m/o UN. By forming a solution of two nitride the activities of the individual constituents readjust and by judicious choice of the second nitride it is possible to completely suppress liquid formation. Some results from Reference 7 are given below in Table 5 .

*TABLE 5 . ACTIVITY OF URANIUM AND PLUTONIUM
U_{.8}Pu_{.2}N SOLID SOLUTION

Temperature K	Activity	
	Uranium	Plutonium
1600	.06	.015
1800	.21	.032
2000	.51	.044
2200	.61	.056
2400	.78	.062

* At 2400 K incipient liquid would form.

Experimental

Plutonium is necessary in a breeder but for a space reactor a less potentially toxic metal would be preferred. A number of candidate metals are available and two of the most attractive are titanium and yttrium. Yttrium has been selected for experimentation because of the higher melting point of the metal. For an application involving T2M as a clad titanium would be the obvious choice.

A quantity of yttrium nitride was prepared. Yttrium powder of 99.93 percent purity was obtained from Ventron and was hydrided to YH_3 at 500 C. Nitrogen gas was introduced and the system heated to 900 C. Nitriding began at about 850 C. A limiting composition of $\text{YN}_{.90}$ was reached.

From observable decomposition pressured and utilizing a prediction of ideality the following values would be predicted for a 5 m/o, 10 m/o, and 20 m/o addition to UN.

*TABLE 6. ACTIVITY OF URANIUM IN $\text{UN}-\text{YN}_{-x}$ SOLID SOLUTIONS

Temperature K	Activity of Uranium		
	5 m/o YN_{-x}	10 m/o YN_{-x}	20 m/o YN_{-x}
1800	.045	.022	.01
2000	.089	.043	.019
2200	.137	.068	.032
2400	.234	.113	.053

* An activity of about 0.78 must be reached before a molten phase appears.

No direct evidence exists applicable to $\text{UN}-\text{TiN}$ but the heat of vaporization of the metal, and heat of formation of the nitride would indicate that the titanium nitride addition would as well. There is some loss in thermal conductivity by addition to UN. Tabularized below are thermal conductivity values for UN and for $\text{U}_{.8}\text{Pu}_{.2}\text{N}$. Similar measurements have not been made for UYN but it would be anticipated that the electronic contributions would be about the same for the two alloys, and electrons carry almost all the heat in the nitrides. A comparison of the values in Table 7 for mixed nitride with that of Table 1 for UO_2 shows that even for the mixed nitride the thermal conductivity is a factor of ten higher than for oxide.

TABLE 7. THERMAL CONDUCTIVITY OF UN AND $U_{.8}Pu_{.2}N$

Temperature K	Thermal Conductivity w/cm deg	
	UN 95% TD	$U_{.8}Pu_{.2}N$ 93% TD
1000	0.205	0.155
1250	0.238	0.167
1500	0.255	0.175
1750	0.263	0.188
2000	0.265	0.198
2250	0.267	0.205
2500	0.271	0.213

Discussion

Forming solid solutions is an attractive way to counteract the formation of a molten phase for UN. The more additive, the higher the temperatures of the fix. Operation for extended times at >2200 K is conceivable so use of nitride for very high temperature direct conversion devices is feasible.

To recapitulate, the nitride would be the choice for steady state high power density operation with surface temperatures of 1000 C or so. No need for stabilizers would arise in this temperature regime. For a few hundred degrees cooler, say 700 C the carbide would be the choice. Its good conductivity and the low diffusion rates at 700 C would allow high power operation with acceptable compatibility with clad materials. Between 1000 C and 1200 C UO_2 might be the choice. This 1200 C might be extended some by forming a cermet but now the cost is approaching the nitride but the power density still nowhere near. For high power in a compact reactor as Table 4 attests either the carbide or nitride are the choice. It has been demonstrated here that indeed the stabilized nitride is a viable choice even under open core configurations such as the SP-100.

LITHIUM FUEL INTERACTIONS

Introduction

Consideration of the condensed state reaction

Such a reaction will have a very high temperature dependence and at high temperatures this reaction will proceed to the left. At high temperature (above the normal boiling point of lithium) lithium will not reduce UO_2 to free metal in the condensed state. The real situation is a bit more complicated - especially over extended times. The gas phase over UO_2 at any time is comprised of O, O_2 , U, UO , UO_2 , and UO_3 . At any temperature only one composition is the congruent subliming composition. That is, at only one composition of solid does the gas and solid have the same composition. In 1966 a high temperature transpiration study⁽⁸⁾ established a relationship between composition and congruent sublimation. From the transpiration experiments it was possible to determine when the composition of the gas phase and condensed phase were exactly the same. Some values from Reference 8 are tabularized below.

TABLE 4. URANIA PRESSURE VERSUS COMPOSITION AT 2000 K

O/U Ratio		Pressure, atm					
Solid	Gas	Total	O_2	O	UO_2	UO	UO_3
2.03	2.61	1.2×10^{-7}	1×10^{-10}	7×10^{-9}	5×10^{-8}	2×10^{-10}	6×10^{-8}
2.02	2.32	7.3×10^{-8}	7×10^{-12}	2×10^{-9}	5×10^{-8}	9×10^{-10}	2×10^{-8}
2.01	2.07	5.9×10^{-8}	1×10^{-12}	7×10^{-10}	5×10^{-8}	2×10^{-9}	6×10^{-9}
2.00	1.93	5.8×10^{-8}	2×10^{-13}	3×10^{-10}	5×10^{-8}	6×10^{-9}	2×10^{-9}
1.99	1.86	6.1×10^{-8}	3×10^{-14}	1×10^{-10}	5×10^{-8}	1×10^{-8}	1×10^{-9}
1.98	1.63	8.1×10^{-8}	6×10^{-15}	5×10^{-11}	5×10^{-8}	3×10^{-8}	5×10^{-10}
1.97	1.42	1.2×10^{-7}	1×10^{-15}	2×10^{-11}	5×10^{-8}	7×10^{-8}	2×10^{-10}
1.96	1.20	2.5×10^{-7}	2×10^{-16}	9×10^{-12}	5×10^{-8}	2×10^{-7}	8×10^{-11}

The implications from Table 8 are clear. Generally the composition of the gas phase differs from that of the condensed phase, and while lithium cannot reduce the uranium oxide to free metal it can influence the gas phase and concomitantly the condensed phase. The reaction envisioned as most significant would be

Here we might expect a slight dependence on temperature with the equilibrium shifting slightly to the right with temperature.

Experimental

Initial experiments were performed with the nuclide 12-90 HT mass spectrometer. A 'double boiler' assembly was fabricated from molybdenum for these experiments. A drawing of the boiler assembly and the mass spectrometer housing is shown in Figure 6. It was possible to independently vary the temperature of each part of the double boiler over a limited range, enough to get a variation of a factor of two in lithium pressure with a constant UO_2 temperature.

The temperature of the top was maintained at 1700 K for the bulk of the experiments, but a temperature of 2100 K was also run to simulate temperatures prevalent under failed heat pipe conditions⁽⁹⁾. Experimentally a fuel pellet obtained from Westinghouse was maintained in the molybdenum cell. Lithium gas passed up the molybdenum tube from the reservoir and contacted the UO_2 . Vapor products were monitored. Lithium, uranium monoxide, uranium dioxide and dilithium oxide were the gaseous species observed and their intensities were in the order named. The UO/UO_2 ratio observed with 10^{-4} atm lithium was 4 to 1 at 1700 K and 3 to 1 at 2100 K. Of course the absolute intensities were much greater at 2100 K. The overall reaction which was monitored was

NUCLIDE 12-90
DOUBLE OVEN ASSEMBLY

At 2100 K under failed pipe conditions with a lithium pressure of 10^{-4} atm the partial pressure of UO reaches 10^{-6} atm. Some previous experiments⁽¹⁰⁾ on deposition of UO on polycrystalline tungsten and irridium indicated a stiking factor of 0.4. That is, forty percent of all molecules striking the surface remained. Experiments showed that the grain boundaries were the preferential sites and that migration occurred to these sites. If a value of 0.4 be utilized and a pressure of 10^{-7} atm UO be employed as conservative for 2100 K then about 7×10^{-7} grams UO per second per square centimeter would be deposited on the molybdenum surfaces. Although UO is very stable in the gas phase it is unstable at all temperatures with respect to U and UO_2 . The reaction then upon condensation would be

For every gram of UO which strikes a surface 0.47 gram of molten uranium would be deposited. Molybdenum can retain about 5 a/o uranium before forming a liquid. For a molybdenum heat pipe 30 mils thick (0.075 cm) the mass of a square centimeter would be about 0.8 gms. When a deposit of .01 grams of liquid uranium from UO had deposited then there would be sufficient uranium to cause massive failure of the pipe. Since only 0.47 grams out of every gram of UO is free uranium it would take 0.021 grams UO deposited on the molybdenum before failure could occur. Since the deposition rate is 7×10^{-7} g/sec cm^2 a time of about 10^4 seconds, or three hours would be required before enough uranium could deposit to cause failure. In the real case the time could be considerably longer because diffusion or permeation through the UO_2 could be rate controlling. Figure 7 shows Robinson-backscattered electron images of the molybdenum cell utilized in this investigation.

Discussion

The view is along the top surface of the cell and into the cell cavity. As can be seen there are significant voids in these cavities to a depth of about $40\mu\text{m}$ (1.5 mil). The cell was used for many hours at 1700 and 1800 K, but only briefly about one and one-half hours at 2100 K. If the damage observed occurred

FIGURE 7. INNER SURFACE AND WALL CROSS SECTION
IN CONTACT WITH LITHIUM- UO_2 VAPOR PRODUCTS

at 2100 K and is diffusion controlled when the penetration/reaction rate is dependent on $t^{1/2}$ then, $kt^{1/2} = \ell$. for ℓ of 40 μm and t of 5400 seconds then k would be 0.54. For a molybdenum wall thickness of 30 mil, or 760 μm , this rate constant would predict a time period of 500 to 600 hours before complete penetration would occur. This value is of course predicated on the assumption that the rate is $t^{1/2}$ dependent and that this same rate would hold over extended periods. This may prove to be conservative. It must be noted however that the uranium transport to the heat pipe occurs over a relatively short time period, a few hours, and then further migration can occur with the result that molten uranium can reach the heat pipe over this extended period. It would not be necessary for lithium to remain as a gas within the reactor more than a few hours for the sequence of events to start. Actually a scenario involving gaseous uranium transport could occur almost instantaneously. As Table 8 indicates these reduced species of uranium can exist in the gas phase at 1200 K and 2100 K with lithium present at 10^{-4} atm our measurements indicated the following values.

TABLE 9. URANIUM SPECIES OVER UO_{2-x} IN CONTACT WITH LITHIUM GAS

Temperature	Pressure, atm		
	UO_2	UO	U
1800K	2×10^{-9}	8×10^{-9}	5×10^{-10}
2100K	4×10^{-7}	1×10^{-6}	1×10^{-7}

The values of uranium would indicate a thermodynamic activity of uranium of about 0.2 in the UO_{2-x} . An optical micrograph of the specimen supports this activity value. As can be seen in Figure 8 there is definite evidence of free uranium in the grain boundaries and pores of the charge.

250X

FIGURE 8. LIGHT MICROGRAPH OF UO_2 -WITH
FREE METAL IN GRAIN BOUNDARIES

The information obtained to date was with an essentially isothermal system. The reactor is far from that. It is difficult to predict what would happen in a nonisothermal system with high reliability. It must be considered however that measurements to date would cause one to question whether the heat pipe concept at these temperatures and with UO_2 really form a redundant system.

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