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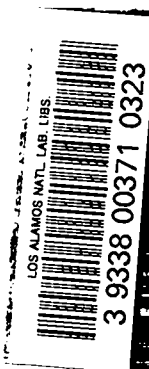
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OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS · NEW MEXICO**

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QUARTERLY STATUS REPORT OF THE LASL  
PLASMA THERMOCOUPLE DEVELOPMENT PROGRAM  
FOR PERIOD ENDING MARCH 20, 1961



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Printed in USA. Price \$ .50. Available from the  
Office of Technical Services  
U. S. Department of Commerce  
Washington 25, D. C.

LAMS-2544  
REACTOR TECHNOLOGY  
(TID-4500, 16th Ed.)

**LOS ALAMOS SCIENTIFIC LABORATORY**  
**OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO**

REPORT COMPILED: April 1961

REPORT DISTRIBUTED: April 28, 1961

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Prepared from material supplied by  
CMB, CMF, GMX, J, N, P, and T Divisions



Contract W-7405-ENG. 36 with the U. S. Atomic Energy Commission

All LAMS reports are informal documents, usually prepared for a special purpose. This LAMS report has been prepared, as the title indicates, to present the status of the LASL Plasma Thermocouple Development Program. It has not been reviewed or verified for accuracy in the interest of prompt distribution. All LAMS reports express the views of the authors as of the time they were written and do not necessarily reflect the opinions of the Los Alamos Scientific Laboratory or the final opinion of the authors on the subject.

## "IN-PILE" TEST PROGRAM

### New Test Cells

In a new design of the "in-pile" plasma thermocouple cell (Fig. 1) the fuel pin-collector assembly has been inverted. This facilitates the use of thermocouples for measuring the fuel pin temperatures during cell operation, and simplifies the cell assembly, particularly when the collector-outer envelope spacing is small. A larger volume gas storage tube has been incorporated in an effort to reduce the pressure effects of pin outgassing, fission gas buildup, etc. The new design also permits the incorporation of a pressure transducer to measure the gas pressure in the cell.

In previous test cells, a packed bed of alpha alumina and helium made up the heat transfer path between the collector and outer envelope. "Gray investment", a dental grade calcium sulphate (Plaster of Paris), is now being used as the heat transfer medium in place of the packed bed. The amount of copper exposed to the reactor flux has been reduced considerably with a subsequent reduction in the gamma heating of these parts. The purpose of the "P" trap assembly is to remove gases.

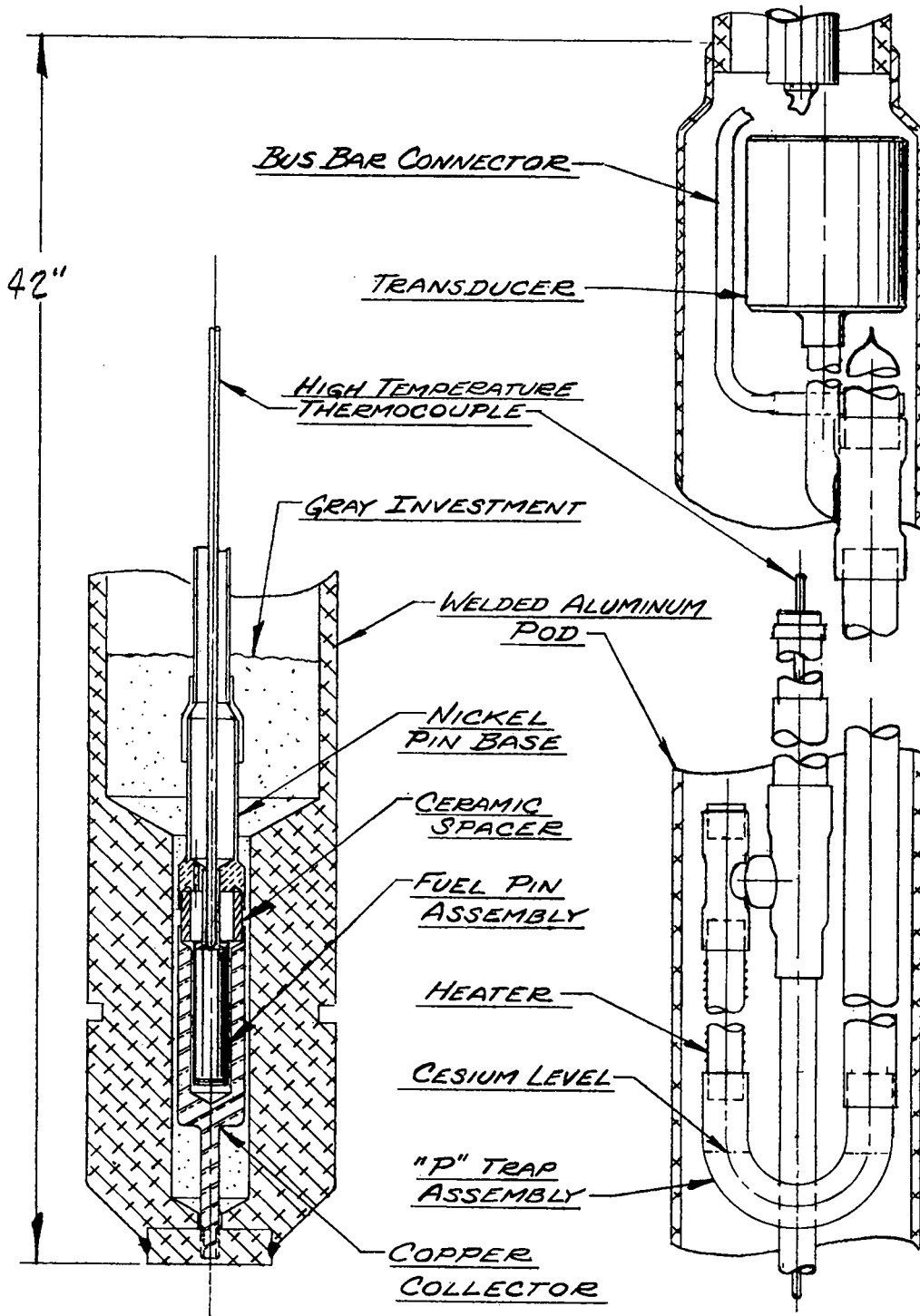


Fig. 1. New "In-Pile" Plasma Thermocouple Cell

In the first successful test (RT-MP-4) of the modified cell (called a "trombone" type), the fuel pin consisted of  $(\text{UO}_2)_{0.5}(\text{Mo})_{0.5}$ , with the uranium-235 content about one fifth that of the pins previously used. The pin was enclosed in a niobium capsule. The collector temperature, with the Omega West Reactor operating at a power of 5 Mw, was  $350^\circ\text{C}$ .

With a collector-emitter gap of 0.015 in., the maximum open-circuit voltage and short-circuit current was 2.2 volts and 34 amp; however, the output was quite erratic. During the course of the run, the pressure transducer indicated that little or no buildup of gas occurred in the cell. The maximum pin temperatures recorded were relatively low (about  $1000^\circ\text{C}$ ). After about 10 hr of running time in the OWR, the cell output dropped to zero. Subsequent x-ray tests showed the fuel pin had warped and shorted out to the collector and that the niobium casing head ruptured and ballooned.

In a second "in-pile" test of the "trombone" cell (RT-122), the fuel pin was of the standard  $(\text{UC})_{0.3}(\text{ZrC})_{0.7}$  form which has been used in many previous tests. There was no high-temperature thermocouple but the pressure transducer was included. This assembly was intended as a life test and, as such, ran in the Omega West Reactor for approximately 230 hr (2 weeks of reactor operating time) at a power of 5.0 Mw. Apart from week-end shut-down effect, the short-circuit current, which was used as a monitor of cell performance during the course of the experiment, ranged from 30 to 55 amp due to deliberate changes in the

cesium pressure. The failure pattern in the current at the end of the test resembled that of previous tests. There was no indication of gas pressure buildup given by the pressure transducer; however, the ability of these transducers to operate in a high flux over an extended time is questionable. It may be noted that no radiation was detected on the reactor top which would indicate a leaking cell.

In cells RT-132 and RT-133 high-temperature thermocouples were incorporated into standard  $(\text{UC})_{0.3}(\text{ZrC})_{0.7}$  fuel pins in order to measure the temperatures of the pins. These cells failed after a short time in the reactor. X-ray examination of cell RT-132 shows that the fuel pin had broken.

#### Flux in Omega West Reactor

In the "in-pile" experiments, the test cell is placed in the center hole of a dummy fuel element. The flux in the pin is monitored by a thermopile, in a nearby hole, and positioned vertically opposite the pin in the test device.

In new dummy fuel elements which have been incorporated into the Omega West Reactor, the axis of the center sample hole and the thermopile hole are not parallel but diverge by  $1^\circ$ . This raised the question as to whether the ratio of flux at the thermopile to that at the "in-pile" test cell remains constant in a traverse up and down inside the dummy fuel element. The axial flux distribution in and around the dummy fuel, with and without a test cell, has been measured. Analysis of the data indicates that the ratio of the flux at the thermopile and test cell remains constant.

### Corrosion of Insulators by Cesium

In trying to establish lifetimes for "in-pile" tests, experiments are being made on the resistance of ceramic insulators to corrosion by cesium, by placing a piece of ceramic in a liquid cesium bath at 420°C for 22 hr. In this test the Frenchtown alumina disintegrated completely. Alite alumina, Morganite, Lucalox (GE) alumina, sapphire, and spinel ( $MgO$ ;  $Al_2O_3$ ) all exhibited defects after the test. Thus far, the only samples of ceramic which have withstood this test are a piece of high purity (99.9%) alumina made at LASL and a piece of BeO from Frenchtown.

### Fuel Element Tests

A vacuum and gas-handling system has been constructed to permit loading fuel test cells and to aid measurement and identification of the gases evolved from fuel samples while the irradiation is in progress. The amounts of krypton and xenon produced provide a measure of the fuel burnup. It has been found that a Phillips ion gage can be used to measure the gas pressure in the expected range, and that the discharge generates enough light to permit spectroscopic identification of the gaseous species.

In preparation for various spectral measurements, the dark current characteristics for RCA type 6199 and type 7102 photomultipliers have been investigated and a mount constructed for use on the Bausch and Lomb Monochromator on order. Dark currents for the 7102 (infra-red) multipliers have been reduced by a factor of approximately  $10^4$  to a few times  $10^{-11}$  amp with maximum permissible voltage (1500 volts). For the 6199 multipliers the dark currents were about the same, and were about  $10^3$  below the values



at room temperature. For both types the residual current is mostly leakage across insulators within the tubes.

#### RADIATION SHIELDS

Limited tests have indicated that a suitable radiation shield between cathode and collector in a plasma thermocouple may cut down the heat loss considerably, with little adverse effect on the electrical output. However, since the spacing between cathode and collector must be maintained as small as practicable (generally less than 0.040 in.) for best operating efficiency, construction of a shield to fit in such a small space without electrically shorting the cell poses somewhat of a problem.

A promising cylindrical shield has been built, of 1/4 in. diameter, with five layers of metal within an effective wall thickness of less than 0.040 in. Molybdenum foil, 0.0005 in. thick, was used for the layers. The foil was first dimpled, and perforated in places by pressing it with emery cloth. A semi-rigid outside cylinder was first rolled up to the desired size and spot-welded along the seam. The outer surface of this cylinder was then coated with a proprietary high-temperature cement (Astro-Ceram Type A) to provide electrical insulation. The inner layers were added by tightly rolling up a suitable length of the dimpled foil like a watch spring, then letting this unwind slightly to form snug-fitting layers inside the outer cylinder.

Preliminary tests of radiation shields constructed in this manner indicate that they cut down radiant heat transfer very effectively and that the Astro-Ceram provides reliable electrical insulation up to

temperatures of at least 1700°C. However, neither compatibility with cesium vapor nor the exact composition of the baked cement have yet been determined.

#### ION DENSITIES AND ELECTRON TEMPERATURES IN PTC CELLS

Analysis of the radiative recombination continuum in the spectrum from a PTC cell gives a value of the electron temperature. Substitution of this result into the recombination intensity equation

$$\frac{J(\nu) d\nu}{h\nu} = \nu \sigma_j(\nu) N^2 F(\nu) d\nu,$$

which relates the intensity of light within an increment of the continuous spectrum,  $J(\nu)d\nu$ , to the velocity distribution (or temperature) of the combining electrons,  $F(\nu)d\nu$ , to the ion density,  $N$ , and the cross section for the recombination process  $\sigma_j(\nu)$ , makes possible a determination of the ion density. The cross section for the combination of a cesium ion and an electron to form a cesium atom in its first excited state is given by F. L. Mohler (1937) as

$$\sigma_j(\nu) = \frac{1.5 \times 10^{-56}}{\frac{1}{2} m\nu^2 \left(\frac{1}{2} m\nu^2 + E_j\right)^2} \text{ cm}^2$$

where  $m$  is the electron mass and  $E_j$  is the ionization potential of the first excited state.

Using the absolute values of the ion density obtained in this manner together with the electron temperatures, it has been found that under nearly all operating conditions and positions within the cell, the plasma is always very close to Saha equilibrium. This conclusion must

must be regarded as tentative, but if it is confirmed, analysis of plasma diode behavior will be greatly simplified if thermal equilibrium is known to exist.

#### MASS TRANSFER AND VISCOSITY OF CESIUM VAPOR

Measurements have been made of the transfer rate of cesium vapor and from the results the viscosity was determined. The apparatus consisted principally of a small retort containing liquid cesium which could be vaporized by electrical heating. The vapor pressure of the cesium in the retort was the driving force which moved the vapor through a stainless steel transport tube. The latter served as the resistance element in an electrical circuit and thus could be separately heated. A copper heat sink placed at the receiver end of the transport tube was held at a temperature just above the melting point of cesium. Here the cesium vapor was liquified and the liquid dropped into a graduated cylinder.

The parameters of the experiment were the temperature of the liquid cesium in the retort which determined the driving pressure, and the temperature of the tube which determined the temperature of the transported vapor. The rate of accumulation of the liquid cesium in the graduate determines the mass transfer rate in the experiment. From the data the viscosity of cesium vapor in the tube was calculated, by means of a formula based on the Hagen-Poiseuille equation, and found to be  $1.75 \times 10^{-4}$  poise at 700°K.

## EMISSIVITY OF TUNGSTEN CARBIDE

In order to analyze the results of an experiment on the thermionic emission of a WC-coated wire, the emissivity of WC at 6500 Å was required, but a literature search failed to reveal a value. Measurements were therefore made with a vacuum-pressed cylinder, 1/2 in. diam and 5/8 in. high. The material was heated in an induction furnace and temperatures within holes and at the surface were determined with a micro-optical pyrometer. For a shiny (ground) WC surface, the emissivity was found to be 0.58 in the temperature range from 1300 to 2100°C. A dull surface gave a value of about 0.85.

## EMISSION STUDIES

A redetermination of the electron emission of UC has been completed. The least-square fitted values for the parameters in Richardson's equation were  $\phi = 3.3 \pm 0.1$  volts and  $A = 113 \pm 33$  amp °K<sup>-2</sup>cm<sup>-2</sup>. An x-ray diffraction analysis of the coating on the wire indicated that it was single phase uranium monocarbide. No other phases were detected, in particular no tungsten or uranium metal.

In order to determine whether or not the peculiar results obtained in some previous experiments were due to the presence of tungsten carbide, the electron emission characteristics of WC were studied. The WC was made by admitting about 5 psi of CH<sub>4</sub> into the system and heating a tungsten wire in this atmosphere to 2100°C (observed). The emission data ( $\phi = 3.6$  volts,  $A = 2.7$  amp cm<sup>-2</sup>°K<sup>-2</sup>) indicated that the previous results were probably not due to tungsten carbide.

## PROPERTIES OF URANIUM-ZIRCONIUM CARBIDE

### Vaporization

The principal results of a study of the vaporization of  $(\text{UC})_{0.3}(\text{ZrC})_{0.7}$  are as follows:

1) Evaporating material is appreciably richer in uranium than the residue. X-ray and spectrochemical analysis showed the condensate to be approximately  $(\text{UC})_{0.5}(\text{ZrC})_{0.5}$ .

2) Analyses of the residual material showed significant loss of uranium at the center of the 1 mm radius cylindrical samples. In one case (2525°C for 20 min) some of the surface material was almost pure ZrC and that at the center of the cylinder had been reduced in uranium to  $(\text{UC})_{0.13}(\text{ZrC})_{0.87}$ . Unfortunately it has not been possible to measure the composition as a function of radius with sufficient space resolution to obtain detailed data on the transport of uranium in the sample.

3) Uncertainties in some of the analyses render dubious conclusions about the temperature dependence of the vapor composition and even more uncertain conclusions about the temperature dependence of uranium transport within the solid solution.

Further experiments are under consideration with the object of obtaining additional data from currently available experimental techniques. It is realized, however, that work with larger samples is desirable.

The attempt to measure dissociation pressures of UC-ZrC by means of the mass spectrometer has been discontinued. The difficulty of obtaining constant signals from thin filaments can probably only be circumvented

by changing to a Knudsen cell source, which would necessitate redesign of the source geometry of the instrument. A bakable spectrometer, in which an adequate vacuum can be attained, is necessary for this work.

### Spectral Emissivity

Development of the apparatus for the measurement of spectral emissivity of UC-ZrC was delayed by trouble with the rectifiers in the high-voltage, constant-current power supply used with the electron bombardment apparatus. This has now been overcome and experiments with the equipment are in progress.

A piece of lavite was eliminated from the bombardment apparatus in order to facilitate outgassing and the carbon anode was replaced with one of tantalum of approximately the same type as that which it is proposed to make of UC-ZrC. This anode is a cylinder of about 0.6 cm diameter and about 3.5 cm long with a coaxial cylindrical cavity for about a third of its length near the middle. The interior of the cavity may be viewed through a longitudinal slit about 1.5 mm wide.

## FABRICATION AND DEVELOPMENT

### Ceramic-to-Metal Seals

Firing tests to metallize  $\text{Al}_2\text{O}_3$  with Cu by coating the alumina with  $\text{Cu}_2\text{O}$  and firing in  $\text{H}_2$  have indicated  $1225^\circ\text{C}$  to be better than  $1250^\circ\text{C}$ . At the lower temperature there is less fluidity of the  $\text{Cu}_2\text{O}-\text{Al}_2\text{O}_3$  eutectic; this results in a more even, smoother coating.

Seventeen samples of ceramic-to-metal joints were tensile tested. Three procedures were used to prepare the metallized surface. For one set of samples, one coat of 1 CaO.1 Al<sub>2</sub>O<sub>3</sub>.5 MoO<sub>3</sub> molar composition was fired on a 95% alumina base ceramic. The second set of samples used the same oxide composition placed on the ceramic in two successive coats. The third set of samples was metallized using the standard Mo-Mn procedure. To the first two sets, a Mo-Mn coating was applied. All samples were then brazed to the metal test-piece section.

The average results of tensile tests were as follows:

<u>Sample Set</u>	<u>Tensile Str. (psi)</u>
Single coat	8666
Double coat	9302
Std. Mo-Mn	8293

BeO has been metallized using a mixture of Mo and MnO<sub>2</sub> as the metallizing agent. Firing was done in H<sub>2</sub> at 1550°C for 10 min. The fired coating was plated with Ni then fired at 1000°C. The metallized layer could be brazed satisfactorily but the BeO fractured from thermal stresses during brazing.

A successful high-temperature seal between a ceramic and Nb has been obtained. The test assembly was composed of a Frenchtown alumina insulator, a Cu washer and the Nb test part. A thin coat of titanium hydride was painted on the engaging surfaces. The entire assembly was fired in a vacuum at 1125°C for 5 min. The assembly, which was found to be leak tight, was then baked out at 1000°C in a vacuum for 4 hr, rechecked and found still to be leak tight.

### Cell Container-Tube Protection

In the P.T. cells, an insulating surface is required to separate the Cs vapor from the cell container-tube. The insulator must not be attacked by Cs, it must prevent the Cs vapor from diffusing through it, and must maintain its insulating properties at temperatures in the range 800 to 1000°C. The container-tube material is tentatively stainless steel or Ni. Alumina appeared to meet the requirements of the insulator, so tests were made to deposit an alumina coat on the metal by high-temperature hydrolysis of  $\text{AlCl}_3$ . The base metals were attacked during processing so the method was deemed unsuitable.

Coats of  $\text{ZrO}_2$  on stainless steel and  $\text{TiO}_2$  on Ni were obtained by applying the proper hydride suspended in hexane with a paraffin binder to the metal, then firing in a gas-oxygen flame until red hot. Several successive thin coatings resulted in an adherent, electrically insulating deposit. Heating the  $\text{TiO}_2$ -Ni combination at 1000°C in vacuum caused breakdown of the electrical insulating character of the coat.

A water- $\text{AlCl}_3$  solution was painted on Ni and heated to cause decomposition of the  $\text{AlCl}_3$  and formation of  $\text{Al}_2\text{O}_3$ . An alumina coat, produced by several successive applications of the  $\text{AlCl}_3$  solution and heating, maintained its electrical resistivity after heating at 1000°C for 1 hr in vacuum.

### Anodic Coating

An anodic coating on Be has been produced which is electrically insulating. The Be was treated in a solution containing 3% KOH-1% KBr



at 50°C. The coat, presumed to be oxide, is rather soft and subject to "break-through" if handled roughly. A film thickness of 0.5 mil is readily obtained; thicker films can be made but are quite soft. The coat maintained its structure and electrical resistance after heating in vacuum to 900°C for 1 hr. No test in Cs vapor has been made.

Hard, dense, uniform oxide coats have been produced on Nb by anodic treatment in a 1% H<sub>2</sub>SO<sub>4</sub>-4.5% HF solution at 25 to 40°C. A 1-mil coat can be produced in 15 min. However, these coats do not withstand Cs vapor attack, but they may find application elsewhere.

#### Be-to-BeO Heat Transfer

Although tests to date have indicated that satisfactory heat transfer from Be to a surrounding BeO tube can be achieved by starting with a controlled expansion fit, this technique requires a relatively slow and expensive internal grinding operation to properly size the BeO bore. A possible method of simplifying the procedure is to coat the inside of the BeO tube with a suitable metal lining, just thick enough so that the internal sizing operation can be completed by reaming out metal with no need to grind BeO. Several attempts were made to evaporate metal filaments and deposit the condensed vapor on a BeO tube. Cu, Be, and Ta were all tried but the rates of build-up, under the available operating conditions, were prohibitively slow. However, a promising method of building up a Cu coat was developed; a thin Ag film (for electrical conductivity) was first deposited chemically; Cu was then electroplated on top of the Ag. A dense adherent coat can thus be built up rapidly to any desired thickness.