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LOS ALAMOS SCIENTIFIC LABORATORY

OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

QUARTERLY STATUS REPORT OF THE LASL PLASMA THERMOCOUPLE DEVELOPMENT PROGRAM FOR PERIOD ENDING OCTOBER 20, 1962

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PLASMA THERMOCOUPLE DEVELOPMENT

RADIATION SHIELD EXPERIMENT

A thermionic converter has been constructed for the purpose of evaluating the effect of radiation shields in the interelectrode space. If the radiation losses to the anode or collector can be diminished, while electrons and ions are allowed to pass through small holes in the shields, the efficiency of the converter may be increased. The problem is to achieve a balance between the two phenomena. If the electrons undergo many collisions in passing through the shields their energy loss may override the gain achieved by shielding from thermal radiation.

A cell was constructed in which the emitter (or cathode) is the inside surface of a hollow tantalum tube, with an i.d. of 0.5 in. and an effective area of 12.6 cm^2. It is heated by radiation from a coaxial bayonet type tantalum tube heater, instead of by the usual electron bombardment. The nickel anode, 0.25 in. o.d., projects into the cathode and is cooled by circulating liquid metal (NaK). The cell is easily assembled and disassembled, thus permitting ready variation in the type of radiation shield to be investigated.

The first radiation shield studied consisted of eight cylindrical layers of 0.001-in. thick molybdenum which had been tightly wound on a 0.40-in. diameter tantalum mandrel. An outer layer of niobium (0.002-in. thick) surrounded the molybdenum layers and was spot-welded to the outermost one. The molybdenum and niobium layers had roughened surfaces to prevent formation of conduction paths by diffusion welding during high-temperature operation. A series of 1/16-in. diam holes were drilled through the shields to give a total hole area of 1.0 cm^2.

Measurements were made at various emitter temperatures varying from 1000 to 2000°C and at cesium equilibrium temperatures of 300, 250, and 200°C corresponding to pressures of 2.0, 0.5, and 0.1 mm Hg, respectively. Voltage vs current characteristics were taken at each set of conditions. From these curves short-circuit current, open-circuit voltage, total power, and cell efficiency can be determined.

No significant difference was found in the total power and short-circuit currents with and without the radiation shield, although there was evidence that the cell efficiency was higher at intermediate emitter temperatures (1200 to 1800°C). It is felt that, as in past radiation shield experiments, the cell operation was probably limited by the emission area especially at high temperatures. It is planned, therefore, to reduce the hole area in the radiation shields.

IN-PILE TESTING

An in-pile test was made with a cell containing a radiation shield surrounding the emitter and connected electrically to the collector. The shield consisted of ten layers of niobium, each 0.005-in. thick, pierced by 24 holes with a total area of 0.5 cm^2. The highest efficiency obtained with this cell was 5%, representing no improvement over standard emitter cells. This result suggests that the latter are operating at or near the plasma-limited case. The effect of a radiation shield of decreasing, by a factor of 4 to 5, the fission power required to heat the fuel pin to a given temperature is then counteracted by the increase in plasma resistance along the devious path between emitter (fuel pin) and collector which electrons must take in the radiation shielded configurations.

Another in-pile test was made with a cell having an emitter formed by vapor plating a 0.0005-in. layer of tungsten on a $Mo-UO_2$ cermet fuel pin. Inadequacy of the cesium heater in this test assembly prevented the attainment of appreciable cesium pressures and so significant electron emission was not obtained at the highest emitter temperature reached (2300°K). The short-circuit current data obtained indicated that the emitter work function was about 5.0 volts initially, presumably because of oxygen contamination of the tungsten surface. When the fuel pin was heated to a temperature sufficiently high to clean the emitting surface, the short circuit currents of the cell were consistent with Langmuir's data for cesium coated tungsten.

Two attempts to conduct vacuum environment in-pile tests resulted in failure of the cells through the development of a short circuit in times of about 1 h. In one case the fuel pin was cracked and swollen to such an extent that it was touching the collector and hence shorting the cell. Disassembly of the second of these tests revealed cracking and distortion of the fuel pin had again occurred. However, the extent of this type of damage was masked by the presence of severe erosion from the center of the pin. This erosion occurred when the fission power generated in the fuel pin was increased by 20% above the design point for a period of 90 min, although the pin had shorted to the collector prior to the beginning of this over-test.

The failure of the fuel pins is believed to be due to the presence of impurities in the $U^{235}C-ZrC$ powder used in their fabrication. In order to permit the study of the behavior of pins in the reactor environment, simple "drop-in" assemblies have been constructed. The pins are mounted on normal nickel pin bases so that the temperature distribution should be little different from that of a pin in the vacuum environment test configuration. The chamber containing the pins was evacuated so that the cell condition being mocked up was open circuit voltage. No cesium is used in these drop-in tests and so the Peltier cooling of the pin was negligible.

Radiographic inspection of the drop-in assemblies subsequent to irradiation revealed that in several cases the active sections had broken away from the U^{238} sections, the latter apparently being unaffected. Although only a small portion of the active secti ons of the pins was visible in the radiograph it was evident that each section had broken into several pieces.

Metallographic examination of some of the pins made with the same material revealed the presence of unconnected void spaces in the UC-ZrC grain boundaries. In addition, each of the carbide grains (approximately 50μ diam) contained a number of small gas pockets, about 2-3 μ diam. These cavities accounted, however, for only a small fraction of the void space. It is apparent that most of the observed swelling of the pins test was caused by the collection and retention of a major portion of the fission gases in the carbide grain boundaries.

Other pins examined showed inclusions of impurities. However, insufficient data are yet available to determine the nature and amount of impurities which can be tolerated in the UC-ZrC solid solution.

VARIABLE SPACING CELL

The purpose of the experimental program using the variable spacing cell is to investigate the effects of CsF on the operating characteristics of the Cs-W system. The CsF is contained in a glass ampule which is located in a separate arm of the cell system. Data are first taken for the Cs-W system alone, the tungsten being sprayed on the tantalum emitter. The CsF ampule is then crushed and measurements are made on the Cs-CsF-W system. The emitter-collector gap is initially about 1 mm, but as the emitter temperature is raised, the spacing changes. The cell is provided with adjustments that can be made while operating to keep a constant spacing, which is monitored continuously through the sapphire windows provided for this purpose.

Anomalously high electron emission was observed in this experiment. One possible cause might be that the crystal orientation of the tungsten coating influenced the emission. To check this possibility, samples of sprayed tungsten on tantalum polished and unpolished, and of polished tungsten bar stock were heated for 8 h at 2000°C in a vacuum. An x-ray diffractometer scan of the surface of each sample before and after heating was made. Marked changes were observed in the sprayed surfaces while no change occurred in the bar stock; however, there is no evidence to date that this accounts for the anomalous behavior.

SPECTROSCOPIC STUDIES OF CESIUM PLASMAS

The object of this work is to study the emission and absorption properties of a hot quiescent cesium plasma that is known to be in thermal equilibrium without currents or transient effects. The apparatus (Fig. 1) is set up inside a 20-in. vacuum chamber and heat is supplied by a cylindrical resistance furnace. The cell is mounted so that its main body, which can be heated to 2000°C, is inside the furnace. Two sapphire windows protrude from top and bottom, respectively. The main body of the cell (B-C)has thick walls (0.080-in. tantalum) while the transition pieces (A-B and C-D) have thin walls (0.015 in. tantalum). The radiation shield S serves to flatten the temperature distribution over the main body section. Points A and D are cooled to a temperature of approximately 500°C. The cesium vapor pressure in the cell is adjusted by controlling the pod condensation temperature at levels below 500°C.

All components of the cell have been assembled and two runs completed without cesium. These first runs are being used to measure temperature profiles in both the thick-walled and thin-walled sections and to adjust cooling rates for the end pieces.

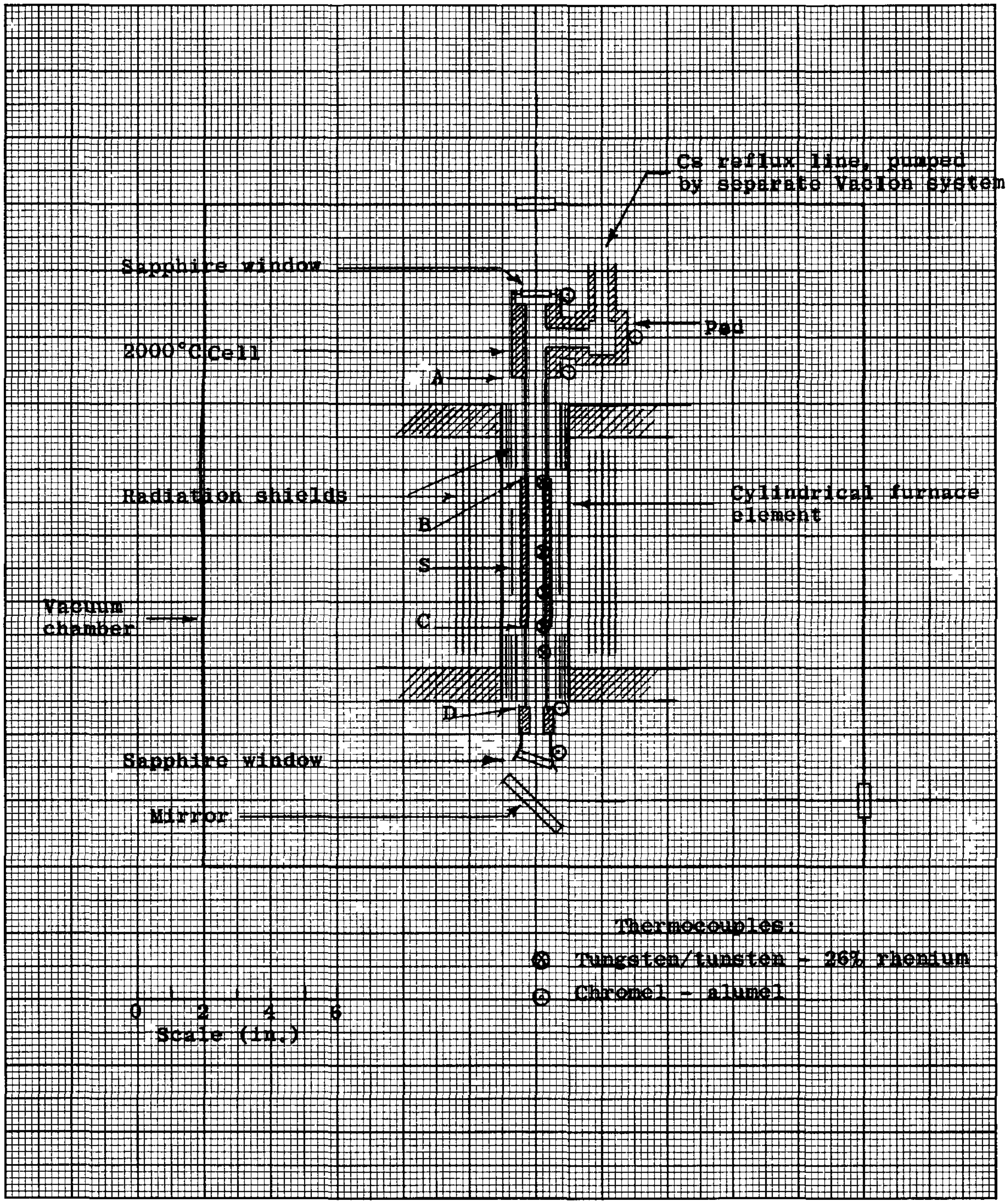

Fig. 1. Cell for Spectroscopic Studies

MATERIALS AND FABRICATION DEVELOPMENT

UC-ZrC Preparation and Pin Fabrication

The preparation of the UC-ZrC powder used in the hot pressing of fuel pins is a batch process, and so it is subject to variations in quality. The basic materials used are high-purity U, crystal-bar Zr, and spectrographic grade graphite. The U and Zr are hydrided with high purity H to produce U and ZrH_2 powders. A mixture of -325 mesh powders of U, ZrH_2 and C is heated to somewhat above $1000^{\circ}C$ in vacuum to form a partially sintered mass of UC-ZrC solid solution. Although other procedures have been explored, the reaction of U, ZrH_2, and C has been found to be best. Because of the considerable handling involved, there is inevitably the possibility of contamination. Since some contaminants in ppm can impair the quality, efforts are being made to improve the purity of the product.

The hot pressing of individual 0.250-in. diameter fuel pins in graphite dies has been superseded by the hot pressing of a relatively large diameter solid cylinder (or slug) from which some 15 pins can be cut by the Elox process. The latter procedure may result in a higher material loss but there is a compensating gain from uniformity in pin quality. The Elox process offers less opportunity for surface contamination of each pin than the separate pressing method. Pins so produced can be ground and micro-finished to secure a smooth surface and accurate dimensions. Further, the length to diameter ratio, ~ 5.5, for individual pins does not favor uniform density over the pin length. Production of a single slug should be beneficial in this respect.

Metallographic examination of hot-pressed pins has shown various phases which should not appear in the UC-ZrC solid solution, e.g., UC_2, U_2C_3, UO_2, graphite, and others. Incomplete reaction of the raw materials, errors in weighing the ingredients, and introduction of contaminants may be responsible.

The objective hitherto has been to produce pins having a density as close to theoretical as possible. Preliminary results of radiation tests suggest that a more porous structure, which could release fission products and reduce internal pressure, would be advantageous. Consequently, an attempt is now being made to produce fuel pins having a uniform density between 80 and 90% of theoretical.

Electrochemical Techniques

An electrochemical technique has been devised for cutting a fuel pin from a solid UC-ZrC cylinder, using a hollow-wall electrode with the electrolyte passage in the wall. A pin, 0.30 in. in diam by 0.50 in. long, was produced in this manner. It is proposed to develop the method to produce pins closer to the desired final dimensions (0.250 in. diam by 1.375 in. long). Since the product possesses an uneven surface, either the process will require improvement or grinding may be necessary.

Holes can be drilled in UC-ZrC by electrochemical milling. By feeding the electrode at a rate of 10 mils/minute, holes of 20 to 50 mils diam have been produced. Such holes are desirable to permit insertion of a thermocouple in the center of the fuel pin for temperature determination during experimental runs.

Bonding of Fuel Pins

Re-examination of the PTC fuel element design has resulted in a change in the collector body. The earlier design is compared with the one now being considered in Fig. 2.

The carbide fuel pin will be brazed to the metal collector (Nb) in the same manner as before with the depleted U-base attached to the collector by a V braze. The diameter of the fuel pin remains the same, with a clearance of 30-40 mils between pin and collector wall. Consideration has been given to reducing the diameter of the depleted U section of the pin by possibly 0.050 in. This would mean a gradual narrowing of the pin

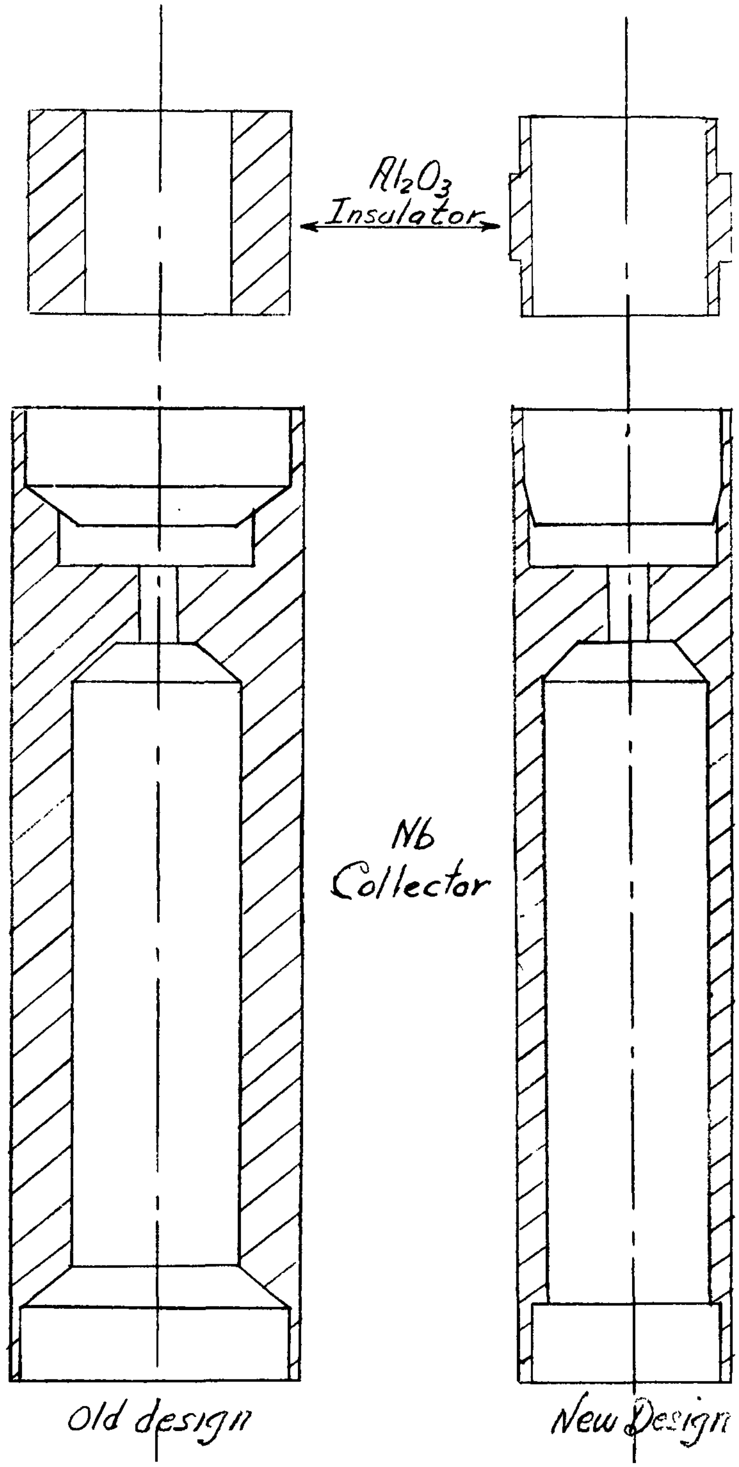

Fig. 2. Plasma Thermocouple Collector and Ceramic Insulator

base from the 0.250 in. diam enriched U section to the reduced depleted U section.

A carbide fuel pin, maintained at 700°C, brazed with V to the old design Nb collector has undergone 400 h test in Cs vapor at 400°C. Brazing tests are underway to join fuel pins to the newly designed Nb collector, using V foil as the braze metal. The brazing technique for this joint seems to be well developed, and no difficulties are expected.

Ceramic-to-Metal Seals:

Two W metallized Al-13 (Al_2O_3) insulators brazed to Nb collectors with Pd foil continue to survive thermal cycling treatments from room temperature to 1450°C in vacuum.

With the Nb collector design change shown in Fig. 2, alteration was required in the insulator shape. The new insulator has a step on the exterior surface that mates with the two collector walls which it separates.

The insulator will be joined to the collectors using the technique in which a WO_3 spray coat is heat treated to form a W metallized surface on the insulator. The metallized insulator is then joined to the Nb-1 w/o Zr collectors with a Pd braze. In the new insulator design only the surfaces of contact with the collectors need be metallized.

Coating Development

To prevent an electrical short between the internal and external components of a fuel rod, the series of joined plasma cells require insulation between the collectors and the enclosing Nb tube sheath. Experimental coatings have been placed on the collector surface by arc spraying Al_2O_3. These coatings have been sufficiently porous to make their electrical insulating capability doubtful. To improve the coating density, powders of different particle size will be tried in the arc-spray with subsequent vacuum sintering.

Magnetic Swaging

It has been difficult to pot electrical coils in plastic materials that will withstand the energy of deformation when the coil is subject to a high energy pulse. The present potting procedure uses a fiberglass and epoxy tube, 12 in. long, 1/2 in. i.d., with a 3/32-in. wall. Over the tube is wound a 6-in. long coil of No. 6 Cu wire, and the assembly is potted in fiberglass and epoxy.

A potted coil of this type has withstood 25 pulses with no visible damage. During one 12-kV pulse, Al will heat to 100°C and Nb to 175°C. Successive pulses increase these temperatures and result in breakdown of the potting materials. Present practice is to pulse the coil once and cool down so that excessive temperatures will not be experienced. New coils are being prepared using fiberglass and silicone rubber tubes to permit higher operating temperature. Experience has shown that a close fit must be maintained between the outer Nb tube and the inner ceramic surface during swaging otherwise the swaged tube possesses wrinkles and an irregular surface.

Cermet Fuel Pins

There has been interest in the use of a cermet fuel pin in place of the carbide pin. The components must have low neutron absorption cross-sections, and because of the high operating temperature (~ 2000°C) the choice is restricted to refractory metals.

A few pins were produced of Mo-40 v/o UO_2 having the same physical dimensions as the carbide pins. Their overall density was rather low and considerable UO_2 loss was experienced when the pins were heated for 10 h at 2050°C in vacuo.

It appears that the 40 v/o UO_2 content was greater than necessary and new fuel pins, containing enriched UO_2, are being fabricated of a Mo-20 v/o UO_2 composition. The final pin will possess a thin W coating on the emitting surface because of work function characteristics. The W coat also will help to contain the UO_2 during high-temperature operation.