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LAMS-2951

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AEC RESEARCH AND DEVELOPMENT REPORT

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LOS ALAMOS SCIENTIFIC LABORATORY
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 JULY 20, 1963
(Title Unclassified)

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LAMS-2951
C-44c, NUCLEAR TECHNOLOGY-
REACTOR ENGINEERING AND
TECHNOLOGY
M-3679 (30th Ed.)

This document consists of 11 pages

No. 132 of 266 copies, Series A

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

REPORT COMPILED: August 1963

REPORT DISTRIBUTED: August 20, 1963

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Exempt from CCRP Re-review Requirements
(per 7/22/82 Duff/Caudle memorandum)
MK 3/17/05

Classification cancelled (or changed to) UNCLASSIFIED
by authority of DIV OF DECLASS. WASH DC
by GG TIC, date 10/26/72

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

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IN-PILE EXPERIMENTS

Low-Density Pin

A plasma thermocouple cell (DIM-13), containing a $(\text{UC})_{0.3}(\text{ZrC})_{0.7}$ pin of 81.5% theoretical density manufactured by General Atomic, was removed from the Omega West Reactor after an exposure of 1005 h because a radiograph indicated that the pin had swollen. This conclusion was confirmed by radiography after removal, which also revealed a longitudinal crack in the interior, although the pin was intact. During disassembly the pin broke into seven pieces but the interface between the oralloys loaded and depleted sections of the pin remained intact despite the fact that there was a marked change of diameter at this point due to the swelling of the oralloys loaded section. This swelling corresponded to a volume increase of 34% for a calculated total atomic burning percentage of 0.16%. The observed swelling is approximately two thirds the projected swelling for similar porous pins manufactured by CMB Division at LASL and is comparable with the swelling rate obtained for dense $(\text{UC})_{0.3}(\text{ZrC})_{0.7}$ pins of local manufacture.

Long-Term Test

An operating cell (HSR-591-1), with a $(\text{UC})_{0.18}(\text{ZrC})_{0.82}$ pin of 85% theoretical density, was removed for examination after 4 weeks in the Omega West Reactor. The initial power output of the cell was 53.5 watts (10.7 watts per cm^2) but it dropped to 27 watts after 2 weeks; during the third week the power decreased sharply to 1 watt, and fell further during the fourth week. Radiography prior to disassembly showed that the fuel pin had broken into four pieces and that a break had also occurred in

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the Ag-Mn brazed joint between the Ni collector body and the Nb bushing to which the insulator was brazed. During disassembly another break occurred in the pin.

Comparison of pin dimensions taken from the post-test radiograph with those obtained by direct measurement during disassembly indicate that an overall volume increase of about 16% occurred but that some of the surface near the center (lengthwise) of the pin spalled off during disassembly.

The most significant observation, however, was that the pin had grown in length to an extent sufficient for it to make contact with the collector bottom when at operating temperature. This suggests the possibility that the Ag-Mn braze joint in the collector may have been broken by tension caused by the pin expansion.

Arc-Melted Powder Pins

Two $(\text{UC})_{0.18}(\text{ZrC})_{0.82}$ pins, made from carefully prepared arc-melted powders, were irradiated for 324 h in test DIM-16. One pin had an oralloy section density of 80% of theoretical with 20% open porosity. The volume increase of 41% in the oralloy loaded section occurred with a total atom burnup of 0.005%. The second pin was of higher density, 87.5% with an open porosity of 8.4%. This pin changed from a cylinder of uniform density to a relatively high-density shell with a low density core. A volume increase of 42% for 0.049% total atom burnup was observed. The swelling in both pins is about three times that expected on the basis of earlier tests. Using the best of arc-melted powders in the preparation of either high-density or porous pins evidently does not solve the swelling problem. The controlled addition of impurities such as tungsten may provide escape paths for the fission products and reduce the swelling.

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Molybdenum-UO₂ Pins

Two 40 v/o UO₂ in Mo cermet pins of 80% theoretical density, which were irradiated for 319 h in the Omega West Reactor, have been examined metallographically. One pin was bare whereas the other had a 0.5-mil W coat. In comparing photomicrographs of sections of both pins with that of an unirradiated pin of the same composition, it is impossible to distinguish between them except near the surface. Depletion of the UO₂ near the surface is evident in the bare pin and there appears to be sintering of the Mo in both pins. This sintering, rather than evaporation, probably accounts for the shrinkage observed previously in similar pins. From the metallographic examination of the two pins it appears that the thin W coat has a beneficial effect in reducing evaporation.

VARIABLE SPACING CELL

Uranium Carbide on Tungsten Emitter

Although UC would be desirable for use as a thermionic emitter, because of its high work function, it has not been considered seriously in the past because of its poor structural properties. Recent fabrication techniques may permit UC to be used on a W supporting surface; with this in mind, an investigation of the UC system has been undertaken. A UC emitter was constructed in the following manner: A W layer, 0.005 in. thick, was sprayed on a Ta button by plasma jet. On top of the W surface a UC powder coating was deposited electrophoretically and then melted in vacuum in an r.f. induction furnace. X-ray scans of the emitter surface, before mounting in the cell, revealed a predominantly UC surface. An extremely small amount of W showed in the pattern but this may be due to the subsurface material. Some UO₂ was present but would disappear on bakeout at 1600°-1700°C as revealed by tests made on other samples with UC surfaces.

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Vacuum electron emission currents taken after bakeout and before the introduction of Cs into the cell indicated an effective work function of 3.3 to 3.4 eV, in agreement with earlier measurements at LASL and elsewhere.

A cell with a UC emitter was run for some time at temperatures in excess of 1500°C. At these temperatures, the evaporation rate of UC is known to be fairly high. During the cell operation, observations of the collector guard-ring surfaces indicated that UC was building up on them. The emitter was consequently removed from the cell, but an x-ray scan of the surface showed no UC on the emitter, but only W. There was no U, C, or WC present, as might be expected if there had been reaction of UC with W, indicating that the evaporated material was UC. When the emitter was replaced in the cell and emission curves rerun, the emission was still that of UC. This result might be explained either by U coverage of the W emitting surface that is too thin to be detected by the x-ray scan or that the last few layers of UC, like O, may be difficult to remove from the W emitter at the temperatures used. When the experiment is completed, the emitter will be sectioned in an effort to resolve this question.

The high current densities from the UC emitter presently in the cell provide an interesting region for spectroscopic measurements of the Cs plasma. Preparations have been made to determine spectroscopically the ion densities, electron temperatures, etc.

CESIUM PLASMA STUDIES

Window Cell

The electrical characteristics of the window cell (LAMS-2852, p. 4) have been examined and some preliminary spectroscopic data have been taken. With the diode spacing set at 0.100 in. and at a pressure of 1 torr, cell currents of several amperes and power outputs of about 1 watt/cm² are obtained at emitter temperatures of 1600°C and below. These values are approximately

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a factor of ten higher than results obtained earlier from the same cell under similar conditions. The emitter material is W sprayed on Ta and the role of impurities and the effect of emitter aging on diode performance is uncertain.

Preliminary spectroscopic data indicate qualitatively a large change in electron temperature between open circuit and closed circuit when operating at the conditions given above. The cell seems to be operating in the so-called "arc mode," but here it appears to be the natural mode of the cell and does not require an applied voltage for initiation.

During operation of the cell several unrelated leaks have developed and it is currently operating with a small leak in the stainless steel bellows; the latter will shortly be replaced by new bellows with welded flanges. When the cell is down for this purpose, the remaining Ag-Mn brazes may also be replaced by electron-beam weld joints, if such welds now being tested on the 2000°C cell prove satisfactory.

CESIUM VAPOR TUBE

A see-through Cs vapor tube has been designed and partially assembled. It will be operated at temperatures up to about 400°C with vapor at a uniform temperature in the entire region between the two sapphire windows. The purpose of the tube is to measure oscillator strengths for the resonance lines (principal series) of Cs, since the data in the literature show great disagreement except for the first few lines. The technique will be based on simple absorption measurement in Cs vapor that is almost optically thin for each line. The lines will be pressure-broadened by the addition of an inert gas so that the widths will be comparable to the resolution of the spectrograph.

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MATERIALS

UC-ZrC Powder Preparation

It has been demonstrated that solid solution UC-ZrC can be made by arc-melting the elements together then thermally treating the as-cast material to eliminate composition variations. Either as-cast buttons or powder ground from buttons may be solution treated. The latter procedure seems to offer the more efficient way to secure homogeneity.

Metallographic examination of vacuum-arc-melted UC buttons has in some instances shown free U in the grain boundaries of the carbide. When striking the arc on the charge, the sudden heating expands residual gas in the crucible which evidently blows out a small amount of C. The net result is to increase the U beyond the stoichiometric UC composition. A small amount of excess C could be added to compensate for the loss, but estimating the proper quantity to add would be uncertain. Consequently, the manner in which the charge is loaded in the crucible is being re-examined.

Careful control of the procedure used to crush and grind UC arc-melted buttons has yielded 64.2% of the initial quantity in the desired -140 +325 mesh particle size range. This size is desired as feed material for coating by plasma-arc spraying. It has been found that if the feed consists of very fine particles (< 325 mesh), material loss tends to increase, whereas larger particles do not feed well and usually form poor surface textures.

While trying to spheroidize UC-ZrC powder by using an induction-stabilized plasma arc in an inert atmosphere dry box, the plasma tended to ionize the He or Ar gas with a resultant breakdown across turns of the induction coil. This effect was not noticed when experimenting in air. To eliminate the arcing, the induction-arc apparatus will be mounted outside the dry box with the discharge into the inert atmosphere.

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Pin Fabrication

Hot-pressed slugs have been made from recently prepared UC-ZrC powder. One pressing was made from powder ground from solution treated UC-ZrC buttons, and two pressings from the same lot of powder which had received further solution treatment in vacuum at 2000°C for 24 h. These slugs will be compared in order to determine the effect of the additional thermal treatment.

Production is being continued of UC-ZrC and Mo-40 v/o UO_2 pins for a variety of tests. The most practical method of securing sound UC-ZrC pins consists in eloxing the pin from a hot-pressed slug. A small hole is drilled through the pin on the axis by the elox method to provide for thermocouple positioning. The hole also accepts a refractory metal tack which is brazed to the pin. The tack protrudes from the pin and this protruding section is used to connect the fuel pin to the collector. The tack method of joining will also be applied to the Mo- UO_2 pins, of which eleven have been made for such tests. These pins were cold pressed and sintered in H_2 at 1700°C for 3 h plus 6 h at 2000°C in vacuum.

Bonding of Fuel Pins to Metal Components

In the present plasma thermocouple cell design, the fuel pin is joined to the collector base with a refractory metal tack. Experiments are planned in which W-2 w/o Th, Mo-50 w/o Re, and Nb will be used as the tack material. Various tack designs are being considered for use with UC-ZrC or Mo- UO_2 pins and Nb collectors. The type of pin material probably will influence the design of the assembly and the tack.

Diffusion bonds between Mo- UO_2 fuel pins and W- ThO_2 tacks using ~ 50 micro in. of Ni plate on the tack were not successful. Further tests will be made using a heavier Ni plate.

An alloy of 77% Nb-23% V has been rolled to 1-mil foil for experimental use as a braze material in the joining of Mo- UO_2 to the various tack materials. The melting point of 2100°C places the alloy in a useful temperature range for plasma thermocouple (PTC) brazing application.

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The use of active carbide formers, such as Nb or V, as braze materials seems to deplete the carbides in joining UC-ZrC to the Nb collector. Since Pd is not a carbide former and no reaction with UC-ZrC would be expected, it is being considered as a braze metal. If it shows a suitable wetting tendency, Pd should yield a satisfactory braze.

Ceramic-to-Metal Seals

Yttria has high-temperature properties which make it attractive for PTC use. One problem has been to fire the oxide to high density. A study of the firing characteristics of Y_2O_3 in wet H_2 , dry H_2 , and air at $1750^\circ C$ for a 15-h soak showed the highest density to be obtained in wet H_2 .

The fabrication of fired ceramic bodies to close tolerances frequently requires considerable diamond grinding. Slip casting and isostatic pressing need careful control of slip and powder mix if parts are to be near final dimensions after firing. Samples of commercial resin-bonded Al_2O_3 bodies are capable of being machined to a close final dimension and then fired to vitrification. These pieces require less final grinding to meet tolerances. The process offers the possibility of producing the PTC collector insulator body to closer accuracy than is obtainable with the present isostatic process. The feasibility will be examined of resin bonding certain compositions of interest that are not available commercially.

Palco braze metal was applied to W metallized ceramics as a foil or a powder and fired. A bright continuous layer of braze metal was formed. Since the powder procedure tends to give a more uniform layer, this method will be examined further. W has a moderate solubility in Palco, indicating a change in composition of the braze layer with time at elevated temperature. Although the effect may be unimportant, further examination seems warranted for the possible changes in the properties of the metal layer joining the collector to the ceramic insulator. It may be necessary to make some type of compensation for the loss of W in the Palco.

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Coating Development

Tungsten cladding of UC-ZrC fuel pins by arc spraying has not been too satisfactory. The coat appears coarse and uneven although it seems adherent to the carbide base. The cladding or "can" should decrease carbide loss at operating temperature and offer structural support to a brittle interior component.

The texture and structure of the W coat can be improved by forming it from WF_6 or $W(CO)_6$ by thermal decomposition. These two methods appear to yield a smoother, more even coat. The coating produced from the fluoride seems to give a finer grain size. Both processes as well as a combination are under examination.

Cladding Development

In an assembly of PTC cells, an outer sheath is placed over the assembly to separate the cells from the liquid coolant. To provide the required electrical insulation between cells and sheath, Al_2O_3 has been flame sprayed on the cells to form a 10-mil thick coat. This coat has been somewhat porous and therefore not as reliable as a solid, high-density layer would be in preventing a short between cell and sheath. To improve the situation, an overspray of a low-melting ceramic eutectic was placed on the Al_2O_3 and fired to flow the eutectic into the pores. This procedure has been reasonably satisfactory except for irregularity of surface and some attack on the underlying Nb collector. Other ceramic compositions will be tried to reduce Nb attack.

The outer sheath must fit tightly over the Al_2O_3 , affording intimate contact so that heat generated by the cell can be conducted outward to the coolant. In general, Al_2O_3 coats formed by flame spraying are uneven and may need grinding to secure the required smoothness. To avoid grinding and improve contact with the sheath (metal-to-metal), a spray coat of Nb or a coat formed by the thermal decomposition of a Nb halide has been placed on the Al_2O_3 .

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Early attempts to obtain good contact between the sheath and Al_2O_3 coat used magnetic swaging as a means to collapse a Nb tube onto the Al_2O_3 . The shock during forming was quite severe and it was feared that fracture of the ceramic insulator-collector joints would occur. Consequently, magnetic swaging was discarded in favor of less drastic forming methods.

Recently stretch forming has been used to shrink the Nb sheath onto the cell. A section of Nb tubing was fitted closely over the cell assembly then stretched and decreased in diameter so that it clamped onto the cell. Forming should be done at a suitably elevated temperature to avoid the elastic springback that would probably occur if cold-forming was used. The stretch forming procedure has been tried, and tests are underway to see if adequate contact was made with the Nb- Al_2O_3 composite coat.

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