MASTER 70-3634-1

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THERMO ELECTRON ENGINEERING CORPORATION

**QUARTERLY PROGRESS REPORT OF RESEARCH AND DEVELOPMENT
OF VACUUM FOIL-TYPE INSULATION FOR RADIOISOTOPE
POWER SYSTEMS**

February - April 1966

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Printed in USA. Price

Available from the Office of
Technical Services, Department of Commerce, Washington 25, D. C.

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OF VACUUM FOIL-TYPE INSULATION FOR RADIOISOTOPE
POWER SYSTEMS

February - April 1966

J. B. Dunlay

Contract No. AT(30-1)-3634

U. S. ATOMIC ENERGY COMMISSION

Issued May 27, 1966

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Progress Report

VACUUM FOIL-TYPE INSULATION FOR RADIOISOTOPE POWER SYSTEMS

by

J. B. Dunlay

ABSTRACT

This report is the first quarterly progress report for the subject insulation contract and covers the work period February - April 1966.

The designs of the material compatibility screening and the planar thermal conductivity test assemblies were completed. Fabrication of the test assemblies was initiated. Preliminary designs of the cylindrical thermal conductivity and the edge heat loss test assemblies were established.

I. INTRODUCTION

Past experience with thermal insulations in the temperature range from 600°C to 1700°C has shown that multi-layered thermal radiation shields provide the most effective insulation materials. The purpose of the present high-temperature thermal insulation development program is to evaluate, and where necessary develop, improved insulation materials and concepts to the point where they can be adapted with confidence for engineering applications in future radioisotope and reactor power systems. In particular, the objectives are to develop concepts capable of providing insulation around high temperature heat sources, insulation adaptable to geometric shaping around energy conversion components, and to improve insulation performance where possible.

The insulation to be investigated under this program consists of thin foils of refractory metals separated by thin layers of high purity refractory oxides. The metal foils serve as thermal radiation shields and the oxide layers provide thermal resistance barriers to prevent conduction between the metal foils.

The program is divided in two phases. The Phase I effort (18 months) will:

- a. Evaluate the chemical compatibility of promising insulation materials.
- b. Evaluate the thermal performance of promising insulation materials in planar geometry and in insulation support combinations.

- c. Evaluate on a preliminary basis the thermal performance of promising insulation materials in cylindrical geometry, the magnitude of edge heat losses, and the insulation outgassing characteristics.
- d. Evaluate the insulation geometric shaping capability to meet system integration requirements.
- e. Obtain extended operation test data to evaluate the insulation lifetime capability.

The Phase II effort (26 months) will:

- a. Complete the evaluation of insulation thermal performance in cylindrical geometry, edge heat losses, and insulation outgassing characteristics.
- b. Evaluate the static and dynamic load capabilities of the insulation.
- c. Test insulation demonstration systems to demonstrate fabrication feasibility and techniques, demonstrate correlation between predicted and actual insulation performance, and demonstrate and evaluate long term insulation performance and materials compatibility with a 10,000 hour test objective.

II. SUMMARY

The design of the material compatibility screening test assembly was completed and fabrication of component parts for two test units was initiated. Approximately 90% of the parts are complete and assembly of both units was started. Compatibility testing at temperatures of 1700°C and 1900°C will begin during the next quarter.

The planar thermal conductivity test unit was designed and fabrication of the component parts is under way. Approximately 10% of the parts were completed for two test units. Assembly, calibration, and initial testing of insulation samples will begin during the next quarter.

Preliminary designs were established for the cylindrical thermal conductivity test assembly and the edge heat loss test assembly. Procurement of long lead time items was initiated for both test assemblies.

III. MATERIAL COMPATIBILITY SCREENING TESTS

The maximum operating temperature of metal foil-oxide layer type insulation is determined by the compatibility limit between the foil and the oxide. To achieve the very low effective thermal conductivities which are possible with this insulation, reactions between the refractory metal and the oxide must be minimized to prevent the metal foils from bonding or sticking together. If bonding occurs between the foils, conduction heat transfer through the insulation increases significantly. The bonding takes place on the high temperature side of the insulation and the foils on the high temperature side become thermally shorted together. The overall effect of bonding is a reduction in the number of effective radiation foils and correspondingly an increase in the heat flow through the insulation.

Insulation material compatibility will be investigated under this program for continuous use of the insulation at temperatures up to 1700°C. Compatibility screening tests are planned at 1700°C and also at 1900°C. Insulation samples will be held at temperature for periods up to 2000 hours or greater. At periodic intervals (100, 500, 1000, and 2000 hours) samples will be withdrawn from the test assemblies and examined to determine the long-term stability of the insulation materials. The materials to be investigated consist of the refractory metals: tantalum, molybdenum, tungsten, and tungsten-rhenium in combination with thorium and yttrium oxides. The test combinations and conditions are summarized in Table 1.

All of the compatibility screening tests will be conducted in vacuum with pressures in the order of 10^{-5} to 10^{-6} mm Hg. Dynamic vacuum pumping will be employed to minimize the possibility of contaminants.

TABLE 1
COMPATIBILITY SCREENING TESTS

Foil Material	Oxide	Test Temperature (°C)	Test Period (hours)
Ta	ThO ₂	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Ta	ThO ₂	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Ta	Y ₂ O ₃	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Ta	Y ₂ O ₃	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Mo	ThO ₂	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Mo	ThO ₂	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Mo	Y ₂ O ₃	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
Mo	Y ₂ O ₃	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W	ThO ₂	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W	ThO ₂	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W	Y ₂ O ₃	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W	Y ₂ O ₃	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W-(25)Re	ThO ₂	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W-(25)Re	ThO ₂	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W-(25)Re	Y ₂ O ₃	1700	100
"	"	"	500
"	"	"	1000
"	"	"	2000
W-(25)Re	Y ₂ O ₃	1900	100
"	"	"	500
"	"	"	1000
"	"	"	2000

3.1 Test Assembly Design

The compatibility screening test assembly design is shown in Figure 1. Two test assemblies are employed (one operating at 1700°C and the second at 1900°C). The insulation test samples are located within a cylindrical black body vessel. This vessel is heated by tungsten heater elements running axially along the exterior of the vessel wall. Six elements are used around the circumference of the wall to obtain uniform heating of the vessel. The black body vessel is closed at each end with conical shaped plugs to obtain heating at the ends of the vessel. The operating temperature of the black body vessel and the insulation samples within the vessel is monitored by pyrometer. A black body hole penetrates the top conical plug to permit direct viewing of the interior of the black body vessel.

The black body vessel and heater assembly is completely surrounded by foil type thermal shields to minimize the heater input power requirements. Low input power is desired to reduce the operating temperature of the tungsten heater wire. The heater elements protrude through ports in the thermal shields with electrical connections completed outside of the shield assembly. Each element is supported individually from the top of the assembly. The elements are free to expand axially through holes in the bottom of the shields. The entire test assembly is designed to be easily disassembled for periodic removal of the compatibility screening samples. A port in the top thermal shields provides access to the black body hole for pyrometer measurements.

3.2 Fabrication Status

Fabrication of the component parts of the compatibility screening test assemblies is nearly complete and the test fixtures are being

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Figure 1. Compatibility Screening Test Assembly Design

assembled. Figure 2 shows the two vac-ion test stations which will be used for the compatibility tests. Each system has a pumping capacity of 75 liters/second. The test assemblies will be housed in six inch bell jars with access to electrical power and thermocouple leads made by ceramic-to-metal leadthroughs located in the base plate configuration shown in Figure 3.

The foil samples are illustrated in Figure 4. Fifteen metal foils of each of the refractory metals (Ta, Mo, W, and W-(25)Re) are sandwiched together as a test sample. Four test samples containing ThO_2 and four test samples containing Y_2O_3 are contained within the black body vessel. One ThO_2 and Y_2O_3 test sample will be removed for inspection at the conclusion of each test interval. A black body vessel is shown partially disassembled in Figure 5 and the operating configuration is illustrated in Figure 6.

High purity commercially available materials will be used for the compatibility screening tests. Table 2 lists the major impurities present in each of the test materials.

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Figure 2. Vac-Ion Test Stations

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Figure 3. Test Assembly Base Plate

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Figure 4. Compatibility Test Samples

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Figure 5. Black Body Vessel Components

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Figure 6. Operating Configuration of Black Body Vessel

TABLE 2
MATERIAL ANALYSIS (ppm)

Element	Ta	Mo	W	W-(25)Re	ThO ₂	Y ₂ O ₃
Aluminum	15	10	< 6	10	4	10.5
Calcium	-	30	< 3	10	-	10
Silicon	10	17	< 7	10	5	44
Molybdenum	10	99.95%	30	50	< 1	-
Iron	10	40	20	20	25	39
Chromium	10	10	4	5	10	-
Nickel	10	10	10	15	15	-
Copper	2	12	5	10	20	-
Tungsten	50	90	99.95%	74.80%	-	-
Sodium	-	8	4	5	-	-
Potassium	-	3	3	-	-	-
Manganese	-	10	< 6	1	2	-
Magnesium	-	< 10	< 3	1	< 1	< 4
Tin	-	10	< 6	-	-	-
Lead	-	< 10	< 6	-	< 1	-
Cobalt	-	< 8	< 3	-	< 5	-
Silver	-	< 1	< 3	-	< 0.1	-
Titanium	10	< 10	< 6	-	3.5	-
Zirconium	20	< 10	< 3	-	5	-
Vanadium	10	-	-	-	< 2	-
Tantalum	99.93% Min	-	-	-	-	-
Columbium	50	-	-	-	-	-
ThO ₂	-	-	-	-	99.99%	-
Y ₂ O ₃	-	-	-	-	-	99.99%
Carbon	30	< 10	< 10	-	-	-
Oxygen	50	25	20	-	-	-
Hydrogen	3	2	1	-	-	-
Nitrogen	20	5	1	-	-	-

IV. PLANAR THERMAL CONDUCTIVITY TESTS

Foil insulation arranged in a planar configuration is required at the ends of cylindrical heat sources. Planar thermal conductivity tests measure directly the heat flow through these planar insulation sections and provide a means of studying the effects of structural loading between the foil layers without introducing thermal expansion uncertainties present in cylindrical geometry.

For minimum heat transfer through multi-layer foil insulations, the dominant heat transfer mechanism must be radiation. Any conduction between foil layers reduces the thermal resistance of the insulation and increases the heat flow through the insulation. The oxide layer located between the metal foils prevents direct metal contact between foils. The thermal conductance path formed by the oxide, however, must be sufficiently low to prevent a significant contribution by conduction to the overall heat transfer. The thermal conductance of the oxide layer itself is generally too large to prevent conduction heat transfer. Since extremely thin oxide layers ($\sim .001$ inch) can be used effectively in foil insulations, it appears that the surface resistance between the metal foil and the oxide layer is the controlling conduction heat transfer mechanism.

The planar thermal conductivity tests will investigate the heat transfer rates through insulation combinations of varying thickness and material composition. The materials' compositions will in general be the same combinations listed in Table 1 if the early compatibility test results indicate that these materials are satisfactory from compatibility considerations. Additional tests will be made with structural loading applied across the insulation to investigate the effect of compressing

the insulation which results in greater surface contact area and contact pressure between the foil and the oxide. Further tests will examine the effects of a helium operating environment which could be present in a thermionic generator fueled with alpha decay isotopes. The helium would increase the heat transfer through the insulation by conduction directly between foils and by conduction between the foils and the oxide layer at the surface contact areas.

4.1 Test Assembly Design

The design of the planar thermal conductivity test assembly is shown in Figure 7. The insulation sample is located between the heater block and the calorimeter collector. A guard collector surrounds the calorimeter collector and is operated at the same temperature as the calorimeter collector to minimize extraneous heat loss from the top of the calorimeter. Heat flows from the heater block through the insulation sample to the calorimeter collector and then along the calorimeter section to the water cooled heat sink. An electrical heater brazed to the calorimeter collector is used to calibrate the calorimeter under actual test operating conditions. The heater block and the calorimeter sections are extensively insulated to limit the amount of heater input power required and to minimize heat losses which bypass the calorimeter.

A gas operated actuator is located at the top of the assembly and provides the force for pressure loading the insulation sample. The force is transmitted by means of a pressure rod which acts on the top of the heater block.

The entire test assembly is operated in vacuum. For the helium environment tests, the circumference of the insulation sample package

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Figure 7. Planar Thermal Conductivity Test Assembly Design

is sealed by welding and a helium inlet tube is attached to the sample package. Helium is introduced only to the insulation samples. The heater block, calorimeter, etc. continue to operate in vacuum and are not disturbed by the helium. When helium is used in the sample package, the pressure actuator provides sufficient restraining force to the sample package to prevent distortion of the walls due to internal pressure and high operating temperatures.

The heater block temperature is monitored by pyrometer. Platinum-platinum rhodium thermocouples are used for the collector and calorimeter temperature measurements.

4.2 Fabrication Status

Fabrication of two planar thermal conductivity test assemblies was initiated. The test assemblies will be operated in 12 inch bell jars pumped by cold trapped diffusion vacuum systems. These vacuum stations are illustrated in Figure 8.

Refractory metal foils were obtained in the sizes listed in Table 3. The two inch width of the tungsten-(25) rhenium foil is the widest available from commercial sources. Figure 9 illustrates samples of these foil materials.

TABLE 3
REFRACTORY METAL FOIL DIMENSIONS

Material	Thickness Inch	Width Inch	Length Feet
Ta	.0005	6	25-100
Mo	.001	6	10
W	.001	6	2-3
W-(25)Re	.001	2	1-1 1/2

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Figure 8. Thermal Conductivity Test Stations

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Figure 9. Refractory Metal Foil Samples

V. CYLINDRICAL THERMAL CONDUCTIVITY TESTS

The curved surface area of the thermal insulation surrounding a cylindrical heat source constitutes a large fraction of the heat source area subject to heat loss. The curved geometry favors a continuous spiral wrapped multi-layer foil configuration.

Heat transfer through the cylindrical insulation is dependent on the compressive loading between foils in a similar manner as in planar type insulation. Radial thermal expansions or applied forces increase the contact area between the foils and the oxide layers with a resultant decrease in the effective thermal resistance of the insulation. The extent of the compressive loading is primarily controlled by the tightness of spiral wrapping or the average spacing allowed between each foil layer.

The cylindrical thermal conductivity tests will investigate the heat transfer rates through insulation test samples of varying number of wraps and spacing between wraps. Test material combinations will be selected from promising combinations evaluated in the compatibility screening tests.

5.1 Test Assembly Preliminary Design

The preliminary design of the cylindrical thermal conductivity test assembly is shown in Figure 10. The cylindrical insulation test sample surrounds a cylindrical heater block. High-temperature electrical heaters consisting of tungsten-rhenium center conductors, beryllium oxide ceramic insulation, molybdenum-rhenium sheaths are brazed to the heater block. The components of the heater are bonded together to provide a good thermal conduction path between the center conductor and

Figure 10. Cylindrical Thermal Conductivity Test Assembly Preliminary Design

the sheath. Surrounding the insulation test sample are the calorimeter and guard collectors. Guard collectors are located at each end of the test assembly to minimize heat losses from the top and bottom of the calorimeter section. The outside of the test assembly is surrounded by thermal shields to reduce radiation losses from the calorimeter collector under high temperature test conditions.

5.2 Design Status

The preliminary design of the cylindrical thermal conductivity test assembly was completed. Electrical heater components and other long lead time materials are being procured.

VI. EDGE HEAT LOSS TESTS

The interface between the planar and cylindrical geometry insulations forms a potential leakage path for heat loss from the insulation assembly. To form a continuous radiation shield at the intersection between the end and sides of a cylinder, the foil layers would have to be fabricated in the form of concentric cups. Fabrication of a large number of close fitting thin wall cups with tight tolerances presents an extremely difficult and exacting task. Fabrication is greatly simplified by providing a joint between the planar and cylindrical sections.

The edge heat loss tests will investigate the magnitude of the heat losses through the insulation planar-to-cylindrical interface joint. Techniques of construction will be evaluated for reducing these heat losses to levels which are small compared to the heat losses directly through the insulation. Tests will be made using promising material combinations selected from the results of the compatibility screening tests.

6.1 Test Assembly Preliminary Design

The preliminary design of the edge heat loss test assembly is shown in Figure 11. The insulation test sample consists of planar and cylindrical sections connected by the test joint configuration. The calorimeter collector surrounds the lower portion of the insulation test sample and collects heat transferred through the planar and lower cylindrical insulation section plus the insulation edge. A guard collector is used to minimize end effects at the top of the test assembly. The edge heat losses are established by comparing the results of these tests with the direct heat losses measured by the planar and cylindrical thermal conductivity tests.

Figure 11. Edge Heat Loss Test Assembly Preliminary Design

6.2 Design Status

The preliminary design of the edge heat loss test assembly was completed. Electrical heater components and other long lead time materials are being procured.