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MILLIWATT GENERATOR HEAT SOURCE
PROGRESS REPORT

October 16, 1976 - January 15, 1977

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I. Introduction

Activities at MRC associated with the Milliwatt Generator Heat Source efforts over the period October 16, 1976, to January 15, 1977, are presented below.

II. "H" Type Capsule Evaluation (P. E. Teaney and R. L. Wise)

Metallographic evaluation of the "H" type compatibility specimens was completed. The tests consisted of six small (approximately 1 watt) compatibility specimens with T-111 liners and hafnium as a getter for oxygen. Specimens were tested at 600 and 900°C for time periods of 60 days, 240 days, and two years.

The specimens were cross-sectioned and examined on the metallograph in both the as-polished and etched condition. No T-111/fuel compatibility reactions were observed, except for indications of capsule embrittlement in two of the specimens tested at 900°C, and signs of slight intergranular attack at the fuel/T-111 interface of the specimen tested for two years at 900°C. The specimen that was tested for 240 days at 900°C contained an intergranular crack which propagated across the width of the liner side-wall. The specimen that was tested for two years at 900°C contained grain boundary cracks and a void area approximately midway in the cap and toward the inside edge of the liner. The area is shown in the photomicrograph in Figure 1. Electron microprobe analysis at fifteen different sites across the cracks revealed no reaction products or compositional differences between the matrix and crack area. The cracks in both specimens most likely resulted during metallographic sectioning of the very brittle samples.

Microhardness traverses were conducted across the liners of the specimens at various locations and the results are shown in Figures 2 through 7. The data indicated that the liners of the specimens tested at 600°C were ductile and the liners of the specimens tested at 900°C were embrittled. Most of the traverses show a microhardness gradient with highest values toward the inside edge of the specimens indicating diffusion of oxygen into the liners of the specimens.

Anomaly in Liner Cap of "H" Type Specimen
after Two Years at 900°C

FIGURE 1

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**MICROHARDNESS OF "H" TYPE
SPECIMEN AFTER 60 DAYS
AT 600 °C**

FIGURE 2

**MICROHARDNESS OF "H" TYPE
SPECIMEN AFTER 240 DAYS
AT 600 °C**

FIGURE 3

MICROHARDNESS OF "H" TYPE SPECIMEN
AFTER 2 YRS. AT 600°C

FIGURE 4

MICROHARDNESS OF "H" TYPE SPECIMEN
AFTER 60 DAYS AT 900°C

FIGURE 5

**MICROHARDNESS OF "H" TYPE SPECIMEN
AFTER 240 DAYS AT 900°C**

FIGURE 6

MICROHARDNESS OF "H" TYPE SPECIMEN
AFTER 2 YRS. AT 900°C

FIGURE 7

Samples from the liners of the specimens were submitted for oxygen and nitrogen analyses. The results are shown in Table I along with average Vickers hardness values for the specimens. Increasing oxygen content and hardness is indicated with increasing time and temperature. The data also indicate that some of the liner embrittlement can be attributed to nitrogen diffusion through the clad.

To compare the oxygen pick-up in the "H" type capsules with previous data for the "F" type capsules, the controlling activation energy was determined from the slope of $\log(r)$ versus $1/T$, where r is the rate of oxygen absorption. The oxygen values in Table I are mean values of several determinations. The value of r was obtained from the relation

$$r = \frac{C_t - C_o}{t}$$

where C_t is the concentration at time t , and C_o is the initial concentration or blank of 92 ppm oxygen. Assuming oxygen absorption is a linear function of time, the data were fitted to straight lines using least squares. The slope of $\log r$ was plotted versus $1/T$ in Figure 8. The indicated least squares line corresponded to an activation energy of 21.4 kcal/mole. This value is slightly less than the 22.6 kcal/mole obtained for the "F" type capsules previously reported and the 22.9 to 27.3 kcal/mole reported for oxygen diffusion in tantalum.

III. Pressure Burst Test Evaluations

A. Fabrication (C. E. Burgan)

Studies are underway to 1) compare pressure burst test data from Lot 7 T-111 with previous lots of T-111 and 2) to compare data from the conventionally welded (continuous arc) capsules with the pulse arc welded capsules. All the strength member bodies for twenty-five pressure burst capsules (MPT-1F through -25F) of the 25-year single channel design were fabricated from the same sheet of Lot 7 T-111, and were processed through the fabrication and heat treatment operations at the same time. The odd numbered units were welded conventionally while the even numbered units were pulse arc welded.

TABLE I
OXYGEN, NITROGEN AND AVERAGE VICKERS HARDNESS VALUES
FOR "H" TYPE COMPATIBILITY TESTS

<u>Temp (°C)</u>	<u>Specimen</u>	<u>Avg. VHN</u>		<u>Oxygen (ppm)</u>	<u>Nitrogen (ppm)</u>
		<u>Bottom</u>	<u>Sidewall</u>		
600	60 days	220	249	143	9
	240 days	228	225	172	39
	2 years	243	246	351	22
900	60 days	624	515	1071	13
	240 days	982	814	1246	70
	2 years	900	949	6312	75
---	Blank	261	260	92	23

B. Evaluations (D. R. Schaeffer)

Eight of the capsules have been tested to date, with the results limited by the number of units that had stem failures. The results are shown in Table II. The data from the capsules that did not exhibit stem failures are shown in Figure 9, where the log of the internal pressure is plotted as a function of the log of the time the capsule retained its integrity. The results from the previous pressure burst tests of the simulated 25-year, single channel design units are compared to the present data in Figure 10.

Photomicrographs of the failed capsules are shown in Figures 11 through 14. The two 7000 psia capsules (MPT-7F and -8F) failed by developing small leaks in the downslope portion of the weld (Figures 11 and 12), while the two 8000 psia capsules (MPT-3F and -4F) resulted in 30 to 90% separation of the cap from the body (Figures 13 and 14).

The data in Table II show that the pulsed arc welded capsules contained the pressurized helium longer than the continuously welded capsules by factors of 1.23 and 1.83 for the 7000 and 8000 psia tests, respectively. Although this initial data indicates the pulse arc welding is the preferred method, more tests are needed to verify the results.

IV. Examination of Lot 8 T-111 (E. Stacy)

As part of the on-going program to evaluate T-111 sheet material, an attempt is being made to correlate pressure burst capsule failures with sheet imperfections. Presently underway is an examination of Sheet 1 T-111 from Lot 8, which along with Lot 7 T-111 is planned on being used for production units.

Random samples were cropped from each end and middle of the approximately 40 mil thick sheet. A light chemical etch (maximum material removal of 5 mils) was used to remove the surface film. SEM evaluation revealed numerous particles randomly dispersed over the total surface of each sample (Figure 15) and localized concentrations of the same particles (Figures 16 and 17). Surface voids were noted primarily in or adjacent to these areas of concentration, indicating removal of included material during sample preparation (Figure 18). X-ray analysis showed these particles and concentrated masses to be hafnia.

TABLE II

1000°C PRESSURE BURST RESULTS FOR
COMPARISON OF CONTINUOUS AND PULSE ARC WELDED TECHNIQUES

<u>Capsule No.</u>	<u>Internal Pressure (psia)</u>	<u>Time to Failure (Hrs)</u>	<u>Location of Failure</u>
1F	7501	23.34	Stem
2F (P)	7503	<1	Stem
3F	7997	9.74	Cap-Body Weld (Figure 13)
4F (P)	7998	17.82	Cap-Body Weld (Figure 14)
5F	5995	86.17	Stem
6F (P)	5997	98.61	Not Presently Known
7F	7000	52.66	Cap-Body Weld (Figure 11)
8F (P)	6994	64.53	Cap-Body Weld (Figure 12)

(P) = Pulse Arc Welded

PRESSURE BURST DATA SHOWING
CONTINUOUS ARC WELDS VS.
PULSE ARC WELDS

FIGURE 9

PRESSURE BURST RESULTS COMPARING
PREVIOUS DATA WITH PRESENT DATA

FIGURE 10

MPT-7F after 52.66 hours
at 7000 psia, 1000°C

FIGURE 11

MPT-8F(P) after 64.53 hours
at 6994 psia, 1000°C

FIGURE 12

MPT-3F after 9.74 Hours
at 7997 psia, 1000°C

FIGURE 13

MPT-4F(P) after 17.82 Hours
at 7998 psia, 1000°C

FIGURE 14

Typical HfO_2 Dispersion Over
the Total T-111 Surface after Initial Etch

FIGURE 15

Localized Concentration of HfO_2 in T-111

FIGURE 16

HfO₂ Particles Imbedded in T-111

FIGURE 17

Voids Surrounding HfO_2 Concentration,
Indicating Material Removal during Sample Preparation

FIGURE 18

An additional etch was performed (material removal of 2 mils/sample) with SEM/EDXRA evaluation after the etching. Although concentrations and random dispersions of HfO_2 were still present, it was not of the magnitude observed after the initial etch (Figure 19). Areas relatively free of HfO_2 were observed in many instances (Figure 20).

Included material, other than the aforementioned HfO_2 , was not observed after the initial etch. After the second etch, a few inclusions (3 to 5 per square inch) were noted (Figure 21). These were identified by x-ray analysis as primarily silicon, with minor constituents of iron and calcium.

Planned are more etches and examinations of five additional sheets of T-111. From each of these sheets, five strength member bodies will be fabricated. The resulting pressure burst test data can then be correlated to the quality of T-111 sheet.

V. Impact Gun (C. G. Anderson and J. E. Newland)

The impact gun installation in Building 50 has been completed. Figure 22 shows the operating console, Figure 23 the gun breech and part of the barrel, and Figure 24 the vacuum box and fume hood area. Figure 25 shows the muzzle opening of the gun, the catch tube damper (at the top) and one-half the clam shell heater for heating the capsule prior to firing.

To date, eleven shots have been fired, with nine of these for acceptance checking. From these data, it was concluded that there will be no major modification required to the basic gun design. Two shots were made as part of the development efforts to better define the catchtube taper and to allow use of the lowest possible sabot weight. Three more shots in this five-shot series will permit defining final dimensions for fabrication of catchtubes and sabots, which will be followed by impacting simulant and fueled capsules.

HfO₂ Concentration after Second Etch
(showing area surrounding concentration relatively free
of HfO₂ particles)

FIGURE 19

Area Containing Very Few HfO_2 Particles
after Second Etch

FIGURE 20

Inclusions (mostly silicon with iron and calcium)
Imbedded in T-111 Surface

FIGURE 21

Impact Gun Operating Console

FIGURE 22

Impact Gun Breech and Barrel

FIGURE 23

Impact Gun Alpha Box and Fume Hood Area

FIGURE 24

Impact Gun Heater Area

FIGURE 25

VI. Process Engineering

The FY-77 formal process engineering efforts are continuing as follows:

A. Thermodynamic Studies (D. R. Schaeffer)

A program related to $\text{PuO}_2/\text{Y}_2\text{O}_3/\text{T-111}$ equilibria has been designed to provide for better understanding toward capsule compatibility. The effort will concentrate on measuring the oxygen partial pressure in the capsule after fabrication, during pretreatment, and during a subsequent fire test. Accurate measurement of the oxygen partial pressure after the pretreatment will be possible for oxygen pressures of greater than about 10^{-28} atmospheres.

The experimental effort will include the construction of an electrolytic cell consisting of a reference electrode inside a solid electrolyte. This cell will be placed inside a T-111 capsule containing the $\text{PuO}_2/\text{Y}_2\text{O}_3$ mixture. The entire set-up will be heated through a pretreatment thermal cycle while continuously measuring the cell voltage and thus the oxygen partial pressure. A subsequent temperature increase duplicating the fire test would indicate if a rise in the oxygen pressure occurs resulting in further oxidation of the T-111 capsule.

B. Compatibility Studies (P. E. Teaney, R. L. Wise, E. Stacy, and D. L. Roesch)

Metallographic processing and analyses of the samples listed in Table III are continuing. Metallographic examination in the as-polished and etched condition, microhardness and electron microprobe analyses were completed for Groups A through I. The tantalum-10 weight percent tungsten liners of Groups J and K are planned to follow. The data for the tantalum alloys are being correlated and preliminary reports will be initiated during the next month.

Metallographic processing of the second priority samples of molybdenum-50 weight percent rhenium (Groups L, M, and N) will begin during February, with evaluation of the third priority samples (Groups 1 through 10) to follow.

TABLE III
COMPATIBILITY SAMPLE MATRIX

<u>Group</u>	<u>Metal</u>	<u>Fuel</u>	<u>Test Temperature (°C)</u>
A	Ta	SRP	1200
B	T-111	Production ^a	1200
C	Ta-10W/T-111	SRP	1200
D	Ta	PuO _{1.81}	1200
E	Ta-10W	SRP	800
F	Ta-10W	SRP	1200
G	Ta-10W	PuO _{1.81}	1200
H	Ta-10W	SRP + Additive ^b	1200
I	Ta-10W	PuO _{1.81}	1000
J	Ta-10W	Production ^a	1700
K	Ta-10W	Production ^a	1900
L	Mo-50Re	SRP	800
M	Mo-50Re	SRP	1000
N	Mo-50RE	SRP	1200
1	Pt	PuO _{2-x}	1000
2	Pt-20Rh	SRP	1000
3	Pt-40Rh	SRP	800
4	Pt-40Rh	SRP	1000
5	Pt-40Rh	SRP	1200
6	Pt/Pt-40Rh	Production ^a	1200
7	Ti	SRP	800
8	Ti	SRP	1000
9	Hastelloy X	Production ^a	800
10	Haynes 25	Production ^a	800

^aAt Mound

^bYttrium, Tantalum, or Molybdenum

C. Moisture Content of Plutonia (W. A. Zanutelli)

A decision was made with Sandia concurrence that the moisture analysis study would be altered by following the plutonia moisture content through the actual steps of the production operations. This is tentatively planned to involve four to six moisture determinations at seven process points: 1) after calcining, 2) after oxygen-16 exchange and sintering, 3) before calorimetry, 4) after calorimetry, 5) immediately before encapsulation, 6) after encapsulation, and 7) after 1 to 6 months storage. With this approach the humidity and environmental conditions can be followed as the fuel is actually produced for encapsulation.

VII. MF-58C Evaluation (R. L. Wise)

During fabrication of fueled capsule MF-58C, dye penetrant inspection showed a small crack in the liner body sidewall near the dome area, and radiography detected a thin liner weld in one area. For these reasons the unit was considered non-acceptable, and post-mortem evaluations were initiated.

The liner was sectioned and visually examined. Confirmation of the crack in the sidewall could not be obtained, probably because the indication was obscured by handling the unit with tools during the sectioning operation. The thin area of the weld, however, was obvious (Figure 26). Also revealed during visual inspection were four yttrium chips fused to the shim of the capsule on the fuel side (Figure 27). This indicated that the off-gassing of the yttrium in this localized area probably increased the capsule's inner pressure, forcing the fillet out of the overlap area of the weld.

VIII. Tooling (J. E. Newland)

Twenty-five sets of pressure burst hardware are in fabrication. These units will be used to qualify the recently received Lot 8 T-111. Also in fabrication are 10 complete sets of hardware, which are planned on being used for GEND's capability units.

The hardware manufacturing process has been changed for clad bodies, with heat treatment now being performed after the sizing operation rather than before. This will eliminate

MF-58C - Thin Weld Area (Arrow) on Liner

FIGURE 26

MF-58C - Yttrium Chips Fused to Shim

FIGURE 27

the roundness problem of the body before welding. There will be some development required to get the parts back to the drawing dimensions.

Difficulties are being experienced in swaging clad caps because of dimensional variability from part to part. Present production plans call for fabricating clad caps by a machining operation. Swaging development will continue, and if the current problems can be resolved, the swaged caps will be incorporated into production at that time.

Gages and fixtures have been designed and are in fabrication for inspecting the machining operations in the lathe. All in-process gages for the bodies are in fabrication for rework. This was caused by the 0.97 millimeter reduction in length of all bodies.

IX. Inconel 600/T-111 Alloy Compatibility Tests (P. E. Teaney)

Due to the commercial non-availability of Hastelloy C, Inconel 600 is being evaluated as an alternate material for clad bodies and caps. This alloy, if favorable compatibility, machinability, and weldability data are attained, will only be used if the unit requirements exceed the present program projections.

Ten Inconel 600 specimens were fabricated for T-111 compatibility tests according to Table IV.

TABLE IV

INCONEL 600/T-111 COMPATIBILITY MATRIX

<u>Temperature (°C)</u>	<u>Time (Hr)</u>		
1225	1	4	16
1100	10	40	160
1000	100	400	1600

One specimen is to be tested at each time period. In addition, a duplicate specimen with no heat treatment will also be evaluated. Heat treatment of the 1100 and 1225°C specimens was completed and testing of the 1000°C specimens is now underway.