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**MILLIWATT GENERATOR HEAT SOURCE
PROGRESS REPORT**

January 16, 1977 - April 15, 1977

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MILLIWATT GENERATOR HEAT SOURCE PROGRESS REPORT

I. Introduction

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

II: Pressure Burst Capsule Evaluations (D. R. Schaeffer)

All twenty-five capsules of the test series comparing pulsed arc welding vs. steady arc welding have been pressure tested, with the results shown in Table I. Due to stem failures and one equipment malfunction, six of the twenty-five capsules did not provide any significant information. The results of the remaining nineteen (including a stem failure which does provide useful information) are plotted in Figure I.

The results of the capsules which ruptured in the cap-to-body weld and the one capsule (MPT-24F[P]) which failed in the stem are plotted in Figure 2, where the log of the internal pressure is plotted as a function of the log of the time to rupture. The lines drawn through the points were found by using a least-squares fit.

The coefficients for these lines give the following equations.

$$t_r = \left(\frac{10,700}{P} \right)^{8.90} \quad \text{for the continuous welded capsules}$$

$$\text{and } t_r = \left(\frac{11,300}{P} \right)^{8.58} \quad \text{for the pulsed arc welded capsules,}$$

where t_r = time to rupture (hours)

and P = internal pressure (psia)

The lines indicate an increase in rupture life with the pulsed arc welded units of about 48% at 8000 psia and 37% at 6500 psia.

TABLE I

RESULTS OF PULSED VERSUS STEADY ARC WELDED
PRESSURE BURST CAPSULES

<u>Capsule Number (1)</u>	<u>Internal Pressure (PSIA)</u>	<u>Log Internal Pressure (Log PSIA)</u>	<u>Time to Rupture (Hours)</u>	<u>Log Time to Rupture (Log. Hrs.)</u>	<u>Failure Location</u>
MPT-1F	7501	3.8751	28.34	1.4524	Stem
2F(P)	7503	3.8752	0.		Stem
3F	7997	3.9029	9.74	0.9886	Cap-to-Body Weld, 30% Sep'n
4F(P)	7998	3.9030	17.82	1.2509	Cap-to-Body Weld, 90% Sep'n
5F	5995	3.7778	86.17	1.9354	Stem
6F(P)	5997	3.7779	98.61	1.9939	Stem
7F	7000	3.8451	52.66	1.7215	Cap-to-Body Weld
8F(P)	6994	3.8447	64.53	1.8098	Cap-to-Body Weld
9F	6500	3.8130	72.05	1.8576	Dome
10F(P)	6499	3.8129	0.15		Stem
11F	7496	3.8749	13.97	1.1450	Dome
12F(P)	7500	3.8751	22.31	1.3485	Dome
13F	6502	3.8131	15.63	1.1940	(2)
14F(P)	6497	3.8127	95.10	1.9782	Cap-to-Body Weld, 70% Sep'n
15F	8002	3.9032	19.00	1.2787	Cap-to-Body Weld, 40% Sep'n
16F(P)	8000	3.9031	23.94	1.3792	Cap-to-Body Weld, 40% Sep'n
17F	7999	3.9030	15.18	1.1814	Cap-to-Body Weld, 80% Sep'n
18F(P)	7999	3.9031	18.03	1.2560	Cap-to-Body Weld, 40% Sep'n
19F	6497	3.8127	78.18	1.8931	Cap-to-Body Weld, 40% Sep'n
20F(P)	6498	3.8128	107.35	2.0308	Cap-to-Body Weld, 40% Sep'n
21F	6500	3.8129	63.60	1.8035	Cap-to-Body Weld, 40% Sep'n
22F(P)	6502	3.8130	126.72	2.1028	Cap-to-Body Weld, 40% Sep'n
23F	6500	3.8129	92.34	1.9654	Cap-to-Body Weld, 40% Sep'n
24F(P)	6504	3.8132	124.13	2.0939	Stem
25F	6496	3.8126	16.17	1.2088	Dome

(1) Capsules with a "(P)" following their number are pulse arc welded

(2) Test terminated because of equipment malfunction

FIGURE 1

FIGURE 2

Fourteen of the eighteen capsule ruptures occurred in the cap-to-body weld. Six of these fourteen ruptures were violent, separating from 30% to 90% of the cap from the body as seen in Figure 3. The remaining eight cap-to-body weld ruptures in both the pulsed and steady arc welded capsules were small leaks and were all located in the downslope portion of the cap-to-body weld as shown in Figure 4.

One difference was observed between the leaks in the downslope region of the pulsed and steady arc welds. A linear "stretch" mark was observed at the failure location of the steady arc welded capsules (Figure 4B). However, little or no disturbance of the surface was seen at the point of failure in the pulsed arc welded capsules (Figure 4A).

The four capsule ruptures which did not occur in the cap-to-body weld occurred in the radius portion of the dome as seen in Figure 5. The location of the dome leaks was always indicated by a "stretch" mark.

Hardware is presently being fabricated from Lot 7 T-111 for approximately 20 pressure burst capsules. These units, all to be pulsed arc welded, will be used to qualify the T-111 lot for production.

III. Tantalum/SRP $^{238}\text{PuO}_2$ Compatibility (P. E. Teaney, R. L. Wise, E. Stacy, and D. L. Roesch)

As part of the FY-77 formal process engineering efforts, work was completed on the evaluation of the tantalum-SRP $^{238}\text{PuO}_2$ compatibility specimens. The design of the test capsule is shown in Figure 6, while the material and fuel analysis are given in Table II. The specimens were tested at 1200°C for 30, 60, 90 and 180 days. Blanks were tested for 90 and 180 day intervals. The specimens were tested in duplicate except for the 30-day time period.

Metallographic evaluation of the specimens revealed the formation of a gray phase reaction product located throughout the matrix and along each side of the cylinders which had been embedded in the fuel. The reaction product is shown in Figure 7. Except for the reaction product concentrations along the fuel/metal interfaces of the cylinders, the reaction product was fairly evenly distributed throughout the matrix of the cylinders with preferential formation along grain

3A

MPT-3F after 9.74 hrs.,
7997 psia, 1000°C

3B

MPT-4F after 17.8 hrs.,
7998 psia, 1000°C

Typical Cap-Body Separation Failures

FIGURE 3

4A

MPT-22F after 126.7 hrs.,
6502 psia, 1000°C

4B

MPT-23F after 92.34 hrs.,
6500 psia, 1000°C
(Note "stretch" mark)

Typical Failure in Weld Downslope Areas

FIGURE 4

MPT-25F after 16.17 hrs.,
6496 psia, 1000°C

Typical Dome Area Failure

FIGURE 5

Configuration of Tantalum/SRP²³⁸PuO₂
Test Specimen

FIGURE 6

TABLE II

SRP Fuel Impurity Analysis Report
(%)

Ni - <0.002
Fe - 0.06
Al - 0.07
Ca - 0.03
B - <0.01
Si - Trace
Cr - <0.03
Cu - Trace
Mg - 0.01
Zn - <0.03
Na - Trace

Total - <0.26% (as Metal)

Tantalum Analysis
(ppm)

Fe - 10	Co - Not Detected ²
Si - <5	Cr - 10
Mg - <5	Mn - Not Detected ³
Al - <5	Ni - Not Detected ²
Cu - 10	C - 60
Nb - Not Detected ¹	O - 50
Sn - 100	N - 130
Pb - 10	H - 20
	Ta - Balance

¹ If present, less than 250 ppm

² If present, less than 50 ppm

³ If present, less than 10 ppm

250X as polished

Tantalum after 90 days at 1200°C
(showing gray phase reaction product)

FIGURE 7

boundaries. The reaction product was identified on the electron microprobe as tantalum oxide and most likely is Ta_2O_5 . From the photomicrographs, measurements were made of the reaction product along the outer edges of the cylinders of each specimen. Also, measurements of the percentage of the phase in the matrix was made by quantitative image analysis. These values were then combined to yield estimates of the total percentages of the cylinders which contained the reaction product. The results are given in Table III. Increasing formation of the Ta_2O_5 reaction product with increasing time at $1200^\circ C$ is indicated by the data. (The data suggests that the identification of the 30-day specimen may have been switched with one of the 60-day specimens, but this could not be verified.)

Sequential examination of the specimens revealed a fairly even saturation of the cylinder with oxygen after 60 days at $1200^\circ C$. Small white phase flecks throughout the matrix and white phase zones along the PuO_2 /tantalum interfaces of one of the 60-day specimens suggests the initial formation of substoichiometric tantalum oxide.

The phase turns from white to gray as it forms a stable oxide of tantalum (presumably Ta_2O_5). This apparently occurs first along the fuel-metal interface and is followed by formation within the matrix during 30-60 days at $1200^\circ C$. The reaction product then forms progressively with increasing time at temperature. Photomicrographs of the specimens are shown in Figures 8 through 14.

The reaction product that occurred in the cylinders of the specimens was not observed in the liners. There are apparently two reasons for the lack of tantalum oxide formation in the liners. The first and most obvious reason was because oxygen diffusion was occurring from only one side of the material, thus lessening the potential for saturation of the material by oxygen. The less obvious reason was the considerable nitrogen diffusion into the material from the outside which apparently inhibited oxidation.

Oxygen, nitrogen and Vickers hardness data for the specimens are given in Table IV. The high microhardness values for the liners of the specimens are due predominately to nitrogen diffusion through the 0.250" thick Hastelloy-X outer container into the liner. Although oxygen from the fuel also diffused into the liner, the effects were masked by

TABLE III
PERCENTAGE OF TEST SPECIMEN CONTAINING REACTION PRODUCT

<u>Sample Test Conditions</u>	<u>Volume % of Phase in Matrix¹</u>	<u>Volume % of Phase Along Fuel/Metal Interface²</u>	<u>Total % Ta₂O₅</u>
30 Days at 1200°C	0.0	3.7	3.7
60 Days at 1200°C	0.0	0.0	0.0
	1.9	4.0	5.9
90 Days at 1200°C	2.0	6.3	8.3
	1.9	7.0	8.9
	0.0	0.0	0.0
180 Days at 1200°C	2.4	8.8	11.2
	7.1	5.9	13.0
	0.0	0.0	0.0

¹ Measured by quantitative image analysis

² Measured from photomicrographs

125X as polished

Tantalum Specimen after 30 Days at 1200°C

FIGURE 8

125X as polished.

125X as polished

Duplicate Tantalum Specimens after 60 Days at 1200°C

FIGURE 9

125X as polished

125X as polished

Duplicate Tantalum Specimens after 90 Days at 1200°C

FIGURE 10

125X as polished

Unfueled (Blank) Tantalum Specimen
after 90 Days at 1200°C

FIGURE 11

125X as polished

125X as polished

Duplicate Tantalum Specimens after 180 Days at 1200°C

FIGURE 12

125X as polished

Unfueled (Blank) Tantalum Specimen
after 180 Days at 1200°C

FIGURE 13

250X etched

Light Phase Reaction Layer
(approximately 15 μm thick, presumably a Ta-N Compound)
on Outside Edge of Ta Liner after 30 Days at 1200°C

FIGURE 14

TABLE IV

OXYGEN, NITROGEN AND AVERAGE VICKERS HARDNESS VALUES
FOR TANTALUM/SRP $^{238}\text{PuO}_2$ TESTS

<u>Sample Test Conditions</u>	<u>Oxygen (ppm)</u>		<u>Nitrogen (ppm)</u>		<u>Average VHN</u>	
	<u>Cap</u>	<u>Side</u>	<u>Cap</u>	<u>Side</u>	<u>Liner</u>	<u>Cylinder</u>
30 Days at 1200°C	1602	1997	1953	1703	561	378
60 Days at 1200°C	1618	2210	4658	1848	561	374
	2053	1963	1389	1267	502	478
90 Days at 1200°C	1059	1935	2019	2378	554	419
	2151	2249	2407	3930	827	479
	(Blank) 253	252	3418	4700	844	163
180 Days at 1200°C	1551	2222	3544	2297	623	304
	1746	1690	3669	2426	728	399
	(Blank) 188	335	4105	5104	795	206

the high quantity of nitrogen. In addition to general nitrogen diffusion into the liner, a white phase reaction layer was present along the outside edge which could only be attributed to the formation of a tantalum-nitrogen compound. The reaction layer is shown along the outside edge of the liner in Figure 14. Electron microprobe analysis of the phase indicated the presence of only tantalum. This is not surprising due to the poor sensitivity of the instrument for nitrogen. Microhardness values for the cylinders indicate an increase from base hardness (163-206 VHN for the blanks) to 300-500 VHN as oxygen uptake occurred. Microhardness of the matrix apparently stabilizes as formation of the reaction product progresses.

Date from these tests suggests that tantalum would be unsuitable as a $^{238}\text{PuO}_2$ heat source liner material at 1200°C due to oxygen embrittlement and the formation of tantalum oxide. Compatibility could undoubtedly be improved with the addition of a getter for oxygen in the fuel, such as in the milliwatt generator heat source. However, tantalum would still lack the "pretreatment" advantage of T-111 alloy.

Results from these tests also point out a weakness of previous compatibility tests which was not realized at the time the interim refractory metals report was written and these tests were designed. The excessive nitrogen diffusion through the Hastelloy-X outer container makes PuO_2 /liner reaction determinations impossible. All compatibility determinations from these tests must be made from discs or cylinders within the liner which are shown by the blanks to be relatively free of reactions occurring from outside influence. Similar problems were encountered in initial milliwatt compatibility tests, but the reactions were distinguishable due to the presence of the strength member and to somewhat lower test temperatures.

IV. Moisture Content of Plutonia (W. A. Zanolli)

Additional work in the formal process engineering efforts for FY-77 includes the study of moisture content of plutonia. The du Pont 902H Moisture Analyzer has been received and has been set up for operation. The pyrometer has been calibrated at 150°C and 700°C. With a sodium tartrate sample containing a theoretical moisture content of 15.66%, six samples were heated at 160°C for three hours. Weight loss indicated a moisture content of 15.70%. When eleven of these samples

were analyzed on the moisture analyzer, results ranged from 15.15 to 15.58% moisture. Although an attempt to increase the accuracy and precision by adjusting the carrier gas flow rate was unsuccessful, discussions with du Pont service representatives indicated that this was the problem. Once the problem is pinpointed and corrected, the instrument will be calibrated and then exposed to an alpha box atmosphere where the plutonia studies will be initiated.

V. Capsule Impact and Evaluations (C. G. Anderson and W. B. Cartmill)

Impact of 25-year design simulant capsules (containing a tungsten-molybdenum powder mixture) has been initiated. To date seven capsules have been impacted at the most critical impact angle (side-on, or 90°), with the data shown in Table V. As can be seen from the results, liner rupture only occurred at 178.1 meters/second at 1000°C , well above the safety requirements of 150 meters/second at 450°C .

Typical photomicrographs of an impacted capsule (MT-16F, 450°C , 169.7 m/s) are shown in Figures 15 and 16. As shown in Figure 15A, the capsule does not appear to be impacted at 90° . This is due to the thick strength member cap (0.100"), which causes the impact energy to be transformed toward the dome end of the capsule, which causes some distortion at the dome end. These phenomena (apparent impact at a less than 90° angle and dome end distortion) were not as evident with the previous design which had a 0.060" thick strength member cap.

There will be a slight delay (early May) before impacting the other three simulant units (MF-12F, -13F, and -17F) because of a problem in the catch tube damper. Fueled units will be impacted after completion of the simulant units.

VI. Pre-TMS Activities

In a meeting on March 8, 1977, with Design Agency personnel, the production design was finalized. The heat source will contain 4.0 thermal watts and will have a design life of 25 years. Because of the more favorable pressure burst data from the pulsed arc welded units (Section II), it was concluded that the liner, strength member, and clad would be pulsed arc welded.

TABLE V

90° IMPACT OF 25-YEAR DESIGN SIMULANT CAPSULES (W-Mo)

Capsule No.	Impact Temperature (°C)	Impact Velocity (meters/sec)	Post-Impact Conditions		
			<u>Clad</u>	<u>Strength Member</u>	<u>Liner</u>
MT-3F	1000	178.1	Rupture at weld, complete cap separation, crack from weld to dome area	Ruptured at dome, end, with 0.55" crack	Ruptured with small crack on dome end
MT-4F	1000	98.8	Rupture at weld with 0.30" crack, two 0.10" cracks at dome end	No rupture	No rupture
MT-16F	450	169.7	Rupture at weld end and dome end	No rupture	No rupture
MT-19F	450	146.1	Ruptured at weld end	No rupture	No rupture
MT-20F	450	160.8	Ruptured at weld and dome end	Rupture crack at weld and 0.45" around circumference	No rupture
MT-5F	450	128.5	Rupture at weld end, 0.4" crack	Rupture at weld end, 0.25" crack ¹	No rupture ¹
MT-11F	450	144.9	Rupture at weld end, two small cracks	No rupture	No rupture

¹Metallographic evaluations showed the strength member and liner components to be "different", further evaluations by SEM are planned

15A

15B

Impacted Capsule MT-16F (169.7 meters/second)

FIGURE 15

Strength Member and Liner at Weld End of
Impacted Capsule MT-16F (169.7 meters/second)

FIGURE 16

Operation sheets and manuals are being written, with each document to be reviewed by Sandia prior to TMS. The first operation sheets are presently in the Sandia review stage.

Also being finalized are the flow-sheet/travelers that will provide input to the GEISHA system for transmitting production data to Sandia. The form design is essentially complete and computer debugging has been initiated with forms containing "dummy data". The forms will be implemented during fabrication of GEND's capability units in May to provide "dry run" data and allow for further refinement prior to TMS.

VII. Fuel Aging Studies (P. E. Teaney)

The status of the fuel aging studies is summarized in Table IV. Testing of the final six specimens (originally scheduled for 15 months) has been extended to 27 months. The specimens are now due off test on May 7, 1977. Two specimens are to be impacted before analysis and two are to be thermal ramped before analysis. The remaining two specimens will be analyzed as aged for comparison.

VIII. Inconel 600/T-111 Compatibility (P. E. Teaney)

Testing of the Inconel 600/T-111 alloy compatibility specimens was completed. The specimens were cross-sectioned, metallographically processed, and are undergoing metallographic examination. When metallurgy is completed, the diffusion depths of individual components will be measured by electron microprobe analysis. Although evaluation of the specimens is incomplete, some preliminary observations were made.

Metallographic examination of the specimens in the as polished condition revealed a reaction which progressed with increased time and temperature. The width of the reaction zones were measured for the specimens tested at 1100°C. Reaction width measurements of 26.0, 38.7, and 70.0 microns were obtained for the 10, 40, and 160-hour specimens, respectively. Although diffusion occurred in both directions, diffusion of the Inconel 600 alloy components into the T-111 alloy predominated. As a result, a line of Kirkendall-type voids were present along the Inconel 600 side of the reaction zone.

A thorough evaluation of the specimens, including data for the 1000 and 1225°C specimens, will be given in the next progress report. Reaction rate and activation energy determinations will be made based on these data.

TABLE VI
FUEL AGING STUDIES

Sample Description	Type of Sample	No. Samples	Age	Analyses to be Performed	Status of Metallography and Fines Analysis
Baseline Fuel	A	2	-	<5 μ fines, fuel microstructure	Completed
Pretreatment Effects	B	2	-	<5 μ fines, fuel microstructure	Completed
Impact Effects on Non-Aged Fuel	C	2*	-	<5 μ fines, >5 μ fines distribution	Completed
Aging Effects	C	2	6 mo at 450°C	<5 μ fines, fuel microstructure	Completed
	C	2	$\geq$ 9 mo at 450°C	<5 μ fines, fuel microstructure	Completed
	C	2	$\geq$ 12 mo at 450°C**	<5 μ fines, fuel microstructure	Completed
	C	2	$\geq$ 15 mo at 450°C***	<5 μ fines, fuel microstructure	Off Test on 05/07/77
Impact Effects on Aged Fuel	C	2	$\geq$ 15 mo at 450°C***	<5 μ fines, >5 μ fines distribution	Off Test on 05/07/77
Thermal Ramp (to 1000°C) Effects on Aged Fuel	C	2	$\geq$ 15 mo at 450°C***	<5 μ fines, fuel microstructure	Off Test on 05/07/77

A - PuO₂ shards, no Y

B - PuO₂ shards with Y, encapsulated in liner and strength member, pretreated

C - PuO₂ shards with Y, encapsulated through clad, pretreated in strength member

*Impact data agreed with previous impact data, therefore, second capsule will be reserved for other tests

**Testing extended to 15 months

***Testing extended to 27 months

IX. Tooling-Hardware Fabrication (W. C. Wyder and R. W. Saylor)

Enough additional strength member bodies and strength member caps are being fabricated from Lot 7 T-111 to yield 20 prime sets of pressure burst hardware. These will be used to qualify this lot of material. Twenty-five complete sets of hardware are being fabricated using Lot 7 T-111 material as a pre-TMS exercise. The reworked gages will be used to test the reliability of the system.

The tooling changes for the clad bodies have been completed. They were necessitated by a revision of the heat treatment in the fabrication process. Hardware is presently being fabricated to dimensionally demonstrate this tooling. A carbide insert has been fabricated for the clad cap swaging, and development will continue as time permits.