MLM-ML-77-70-0001

**MILLIWATT GENERATOR SOURCE
PROGRESS REPORT**

July 16, 1977 - October 15, 1977

Wayne R. Amos

Wayne R. Amos
Program Manager

RECEIVED

JUL 19 1977

OSTI

Monsanto

MOUND FACILITY

Miamisburg, Ohio

operated by

MONSANTO RESEARCH CORPORATION

a subsidiary of Monsanto Company

for the

U. S. DEPARTMENT OF ENERGY

Contract No. EY-76-G-04-0053

AC04-76DP00053

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MILLIWATT GENERATOR HEAT SOURCE PROGRESS REPORT

I. Introduction

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

II. TMS

Tool Made Sample activities were conducted from July 11, 1977, to September 23, 1977. Design agency personnel were present during a significant portion of this period to review and evaluate the process and associated documentation. Approximately 60% of the operations were observed by the TMS team as they were performed.

Once sufficient fuel and yttrium had been prepared and enough hardware had been manufactured, fabrication was initiated on eleven TMS heat sources (MAD-0001-I77 through -0011-I77). All met specification requirements except MAD-0007-I77, which had a dye penetrant indication on the liner body after fuel encapsulation. Further evaluation showed a small inclusion was evidently removed from the liner body surface during the decontamination operation. This is a relatively uncommon occurrence, it had happened previously on only one of the many development units fabricated to date (Section V, MWG Progress Report, April 16, 1977-July 15, 1977).

Three of the units (MAD-0001-I77, -0003-I77, and -0004-I77) were destructively tested and evaluated, two (-0001 and -0003) after impact at approximately 150 meters/second. The photomicrographs and data on these units are discussed in Section III.

Also fabricated as part of the TMS activities were ten pressure burst capsules (MPT-64F through -73F). Pressure burst capsules are planned on being fabricated periodically during WR production and maintained at RTG operating temperatures. At designated time periods the units will be burst tested as part of the surveillance tests and evaluations. All the TMS pressure burst capsules met the necessary fabrication requirements. Post mortem evaluation was performed on one unit (MPT-64F), which is described in Section IV.

On October 6, 1977, an acceptable TMS rating was released by Sandia Laboratories, Albuquerque, along with the authorization for Mound to produce WR units.

III. Post Mortem Evaluations of MAD-0004-I77, MAD-0001-I77, and MAD-0003-I77 (P. E. Teaney)

Post-mortem evaluations were completed on TMS units MAD-0004-I77 (non-impacted), MAD-0001-I77 (impacted), and MAD-0003-I77 (impacted). Results showed that the heat sources met the design criteria.

A. MAD-0004-I77 (Non-impacted)

Photomicrographs of the cross-sectioned liner sidewall and dome of non-impacted TMS unit MAD-0004-I77 are shown in Figures 1 and 2. Metallographic examination revealed no unusual observations. Yttrium was observed bonded to the liner and shim at a few locations. A site where yttrium was bonded to the shim is shown in Figure 3. This phenomenon was observed in development units and is not detrimental to the integrity of the heat source. Microhardness values averaged 237 DPH across this location of the shim with values near 200 DPH at the yttrium chip.

Microhardness traverses were conducted across the cross-sectioned liner at the cap, sidewall and dome. The traverses were conducted at 2 mil intervals using a 50 gram load. Diamond Pyramid Hardness averaged 275, 278, and 250 across the sidewall, dome and cap, respectively. Values for the raw material range from 237-245 DPH. Oxygen values for the sidewall and dome were 205 ppm and 363 ppm, respectively. The oxygen content of the raw material is approximately 42 ppm. The slight increase in microhardness and oxygen content of the liner is normal during the fabrication process and is primarily due to the localized oxygen enriched zones that form along the fuel/liner interface during the pretreatment operation. Nitrogen values were 20 ppm for the dome and 9 ppm for the sidewall indicating no contamination of the material by air during the fabrication process.

125X (as polished)

MAD-0004-I77 Liner Sidewall

FIGURE 1

125X (as polished)

MAD-0004-177 Liner Dome

FIGURE 2

600X (as polished)

Site where yttrium was bonded
to the shim of MAD-0004-I77

FIGURE 3

All observations and data for the heat source liner were typical when compared to previous development data. Therefore, detailed evaluation of the strength member and clad was not necessary.

B. MAD-0001-I77 (Impacted)

TMS heat source MAD-0001-I77 was impacted at 150.2 meters/second, 450°C, at an impact angle of 90°. No fuel was released. Post mortem evaluation revealed that the clad was breached approximately one half way across the cap and one-third way across the dome at the impact face. The breach is shown in Figure 4 at 0° and 90° angles to the impact face. No breach or cracks were observed in the strength member. Two views of the cross sectioned sample (after removal of the fuel and clad) are shown at 0° and 90° to the impact face in Figure 5.

Metallographic evaluation of the liner revealed an apparent breach of the dome near the bottom edge of the impact face. The area is shown in the photomicrograph in Figure 6. Although the liner appeared to be breached, the strength member remained intact. The only other area of severe distortion is shown in Figure 7. The view is 90° to the impact face at the cap weld. Although extremely distorted, both the liner and strength member remained integral in this area.

Microhardness values of 316, 293, and 278 (DPH) were obtained for the liner sidewall, dome, and cap, respectively. These values show a slight increase in microhardness (approximately 28 DPH) when compared to the non-impacted heat source (MAD-0004-I77). The slight increase is most likely due to the liner material being work hardened during impact.

Due to the severe distortion of the dome during impact there was insufficient material to obtain an individual dome oxygen and nitrogen determination. Therefore, samples were taken from both the dome and sidewall and analyzed to obtain average values for the liner. Liner

0° to Impact Face

90° to Impact Face

MAD-0001-I77 after impact at 150.2 m/sec

FIGURE 4

0° to Impact Face

90° to Impact Face

MAD-0001-I77 strength member after
impact at 150.2 m/sec

FIGURE 5

Strength
Member →

↙ Liner

50X (etched)

Apparent breach at liner dome of
impacted unit MAD-0001-I77

FIGURE 6

25X (etched)

Distortion of MAD-0001-I77 liner and
strength member at welds after impact at 150.2 m/s

FIGURE 7

oxygen values averaged 472 ppm. When compared to the non-impacted heat source the liner showed a slight increase of approximately 188 ppm. The increase appeared to be due to fuel embedded into the liner during impact, as is shown in Figure 8. The nitrogen content of the liner was 25 ppm, indicating no contamination of the material by air.

Observations and data were typical for the liner when compared to previous development data. Therefore, micro-hardness, oxygen, and nitrogen determinations were not performed on the strength member and clad.

C. MAD-0003-I77 (Impacted)

TMS heat source MAD-0003-I77 was impacted at 151.2 meters/second, 450°C, at an impact angle of 90°. No fuel was released. On this unit, disassembly and post mortem evaluations were not required by Sandia if no fuel was released. However, visual and metallographic evaluations were made during disassembly of the heat source to obtain additional data on the production design units.

Visual examination revealed that the clad was breached approximately one-half way around the circumference of the cap weld along the impact face and one-third way across the dome. The condition of the heat source after impact is shown in Figure 9 at 0° and 90° to the impact face. Damage appeared more severe than for MAD-0001-I77 which was impacted at approximately the same velocity. Although Figure 9 (0°) suggests the possibility of a longitudinal crack on the impact face, closer examination revealed that it was not a crack. Examination of the strength member revealed an approximate 1/4 inch long crack along the cap weld midway on the impact face.

The specimen was cross-sectioned and examined metallographically. Photomicrographs of the areas of maximum distortion are shown in Figures 10 and 11. Although extremely distorted, the liner remained integral. The approximately 1/4 inch long breach of the strength member weld at the

62.5X (as polished)

Impacted heat source MAD-0001-I77 showing
fuel embedded in liner after impact

FIGURE 8

0° to impact
face

90° to impact
face

MAD-0003-I77 after impact at 151.2 m/sec

FIGURE 9

13X (etched)

MAD-0003-I77, 90° to side-on impact.
(Liner and shim remained integral,
strength member breached at weld.)

FIGURE 10

13X (etched)

MAD-0003-I77 strength member and liner at dome, 90° to impact face. (Liner and strength member remained integral in this area, although extremely distorted.)

FIGURE 11

impact face is verified in the photomicrograph in Figure 10. The view is 90° to the impact face and the crack apparently occurred along the cap side of the weld. The heat source met the requirement of no fuel release. Therefore, no further evaluations were performed.

IV. MPT-64F (Pressure Burst Capsule) (P. E. Teaney)

Post mortem evaluation of pressure burst capsule MPT-64F was completed. This unit was fabricated, tested, and evaluated as part of the TMS activities. The specimen ruptured after 16.4 hours at 1010°C with 8064 psia pressure. The location of the failure was at the weld ramp down and appeared similar to previous failures on development sources.

The specimen was cross-sectioned at the failure site and examined on the metallograph. A photomicrograph of the area is shown in Figure 12. The crack apparently initiated at the sidewall side of the weld at the inside edge and propagated toward the outside of the weld. The mechanism of failure appeared typical when compared to previous pressure burst specimens. Examination of the weld zone 180° from the failure site revealed no evidence of cracks.

Average diamond pyramid hardness values of 284, 294, and 269 were obtained for the sidewall, dome, and cap of the specimen, respectively. The value for the raw material was approximately 247 DPH.

Oxygen values for the sidewall and dome were 54 ppm and 55 ppm, respectively. Nitrogen content was 9 ppm for both the sidewall and dome of the capsule. These values indicate no significant difference from the raw material values of 40-45 ppm oxygen and 26-30 ppm nitrogen.

In conclusion, the evaluation of MPT-64F indicated a normal mode of failure when compared to previous development data.

Cap

Sidewall

30X (as polished)

Pressure burst capsule MPT-64F
showing location of failure

FIGURE 12

V. Encapsulation (C. E. Burgan)

Difficulties were encountered during welding of the TMS strength members. The problem was traced to a malfunction of the torch drive motor, and was resolved by replacing the motor. The three defective strength members were removed and replaced by new strength members.

A clad welding problem was remedied by changing the clad weld collets. The problem was traced to a recent process change, with the present non-heated treated clad bodies behaving in a slightly different manner than the previously heat-treated bodies.

No welding difficulties have been experienced in either area since the process changes have been incorporated.

VI. Fines Analysis (V. L. George)

Fines analyses were completed on two units during the report period, MF-44F, an impacted reimbursable unit, and MAD-0003, an impacted TMS unit. Results are shown in Table I.

Although the fines contents in the 10-20 micron size range are significant, the values are typical in the respirable or <5 micron size range. Fines samples remaining to be analyzed are the second reimbursable unit (MF-45F), the second TMS unit (MAD-0001) and three fuel aging studies samples (MF-33C, -34C, and 36C).

TABLE I
Pu²³⁸ Fines Data

<u>Sample</u>	<u>Weight % Pu²³⁸</u>		
	<u><5 μ</u>	<u>5-10 μ</u>	<u>10-20 μ</u>
MF-44F	3.6 X 10 ⁻²	0.11	7.11
MAD-0003	1.2 X 10 ⁻²	0.18	1.95

VII. Hardware Fabrication (R. W. Saylor and W. C. Wyder)

To have sufficient hardware for TMS operations and WR heat source production, fabrication of WR hardware was initiated in late July, 1977. Planned is the manufacture of seventy-five complete sets of hardware every four weeks. At a 90% efficiency, this will meet production requirements for FY-78 and FY-79.

To date, two pieces of tooling have had to be reworked. The strength member body blanking die has had the carbide punch replaced with a tool steel punch because of excessive carbide chipping. Also the strength member cap first swage die has had the punch replaced with a larger diameter punch for increased strength. All other tooling appears to be holding up very well.

Because of material variations, it was concluded that the diameter of the strength member body blank would be reduced by 1.5 mm. This is necessary in order to place thicker material at the open end of the finished capsule. The present blanks are larger than need be and the thick edge sections of the formed cups are being cut off leaving material too thin to meet specification requirements at the open end of the capsule. Decreasing the starting blank diameter will alleviate this problem.

VIII. Moisture Content of Plutonia (W. A. Zanolli, Jr.)

Completed was the study related to moisture content of plutonia. The study was undertaken to assure that water was not present or being adsorbed at various steps of the process (or storage) that could lead to compatibility problems during pretreatment or long term storage.

The instrumentation (DuPont Moisture Evolution Analyzer, Model 902H) was modified for glovebox usage as shown in Figure 13. The weighed sample is heated and the vaporized moisture is carried to the phosphorous pentoxide detection cell by nitrogen carrier gas. The current involved in the electrolysis of the collected water is proportional to the moisture content of the sample. The current is integrated and displayed on the instrument as micrograms of water.

SCHEMATIC REPRESENTATION OF MODIFIED MOISTURE ANALYZER

FIGURE 13

Instrument calibration was performed prior to modification and glovebox installation. This was done with sodium tungstate dihydrate ($\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$). On four samples an average value of 10.82% H_2O was obtained, which compared to a theoretical value of 10.91%. Good agreement was obtained with additional calibration data after instrument modification and glovebox installation.

The plutonia samples were selected from five steps of the process as follows:

- A. After self calcining (2.0 g samples)
- B. After oxygen exchange, sintering, washing, and calorimetry (10.0 g samples)
- C. Before encapsulation (10.0 g samples)
- D. After encapsulation (heat sources, 10.0 g)
- E. After one year storage (10.0 g samples)
- F. After calorimetry and exposure to moist atmosphere (10.0 g samples)

With one exception, the sample sizes were chosen to represent actual fuel charges. The smaller size (2.0 g) after self calcining was selected because of the higher suspected moisture content and an attempt to avoid saturating the phosphorous pentoxide detection cell.

The moisture results are shown in Table II, while the relationship between moisture content and process steps is shown in Figure 14.

From the results it can be seen that the only significant moisture is present in the fuel after self-calcination. This was expected, as there is no attempt to remove moisture at this step of the process. The moisture found at the other process points was obviously chemically absorbed moisture, since the counts collected on the instrument were in the 400°C to 725°C temperature range. Very few counts were found in the 0-100°C, indicating low concentrations of adsorbed water or surface moisture. This was confirmed by the samples exposed to moist atmospheres picking up very little water. Evidently there is sufficient heat generated from the fuel, even when spread out on a watch glass, that moisture pick-up is not significant.

TABLE II

MWG Fuel Charge Moisture Results

<u>Process Sample Points or Conditions</u>	<u>% Moisture</u>	<u>Av. % Moisture</u>
A. After Self Calcination	3.37, 3.38, 3.51, 3.44, 3.60, 3.53	3.47
B. After Oxygen Exchange, Sintering, Washing, and Calorimetry	.002, .001, .002	.002
C. Before Encapsulation	.001, .007, .001	.003
D. After Encapsulation	.006, .004	.005
E. After Storage for Approxi- mately One Year	.002, .002, .003	.002
F. Opened Fuel Container in Moist Atmosphere for 2 Days	.001	--
Opened Fuel Container in Moist Atmosphere for 5 Days	.001	--
Opened Fuel Container in Moist Atmosphere for 7 Days	.001	--
Fuel Spread on Watchglass in Moist Atmosphere for 7 Days	.001	--

PuO_2 Process Showing Moisture
Content of MWG Charges

FIGURE 14

In conclusion, it can be stated that the water levels of plutonia charges are low enough that moisture problems will not be encountered with the present process.

IX. Pressure Burst Capsule Evaluations (D. A. Pawlak and D. R. Schaeffer)

Sixteen Lot 7 pulsed arc welded capsules have been pressurized burst tested with the results shown in Table III. Two data points were of no value because of stem failures. Twelve of the fourteen other failures occurred in the cap to body weld area, with two of these being the violent cap-body separations, in which 90-95% of the cap separated from the body. The remaining ten failures were small cap to body ruptures located in the center of the weld fusion zone. One exception was MPT-63F, which had a linear rupture similar to those noted on the non-pulse arc welded units. Two failures were not located, even after pressurizing the tested units at 9500-9600 psia. Of significance was the fact that there was no preferential failure location in the weld zone (such as in the downslope area as was noted previously with the non-pulse arc welded specimens.)

Plotted in Figure 15 is the log of the internal pressure vs the log of rupture time (with the exception of the two stem failures). The lines drawn through the points refer to previous pressure burst test data as reported in Section III, MWG Progress Report, October 16, 1976-January 17, 1977. The prior data and recent data can therefore be compared. As evidenced on the plot, the lower pressure tests exhibit greater uniformity with much less scatter.

Table IV shows the pressure burst test data to date on the more recent Lot 7 capsules that were fabricated as part of TMS requirements. Three of the four units failed in the cap to body weld area (no violent failures), the leaks were small and were located in the center of the weld fusion zone. The other failure location (MPT-67F) was not determined by either pressurization or dye penetrant evaluation. Metallographic examination of the destructively tested TMS unit (MPT-64F) is discussed previously in this report (Section IV). Again, these data are plotted and compared to prior pressure burst test data (Figure 16).

TABLE III

Pressure Burst Test Results of Lot 7 Capsules, 1000°C
(Pulse Arc Welded)

<u>Capsule Number</u>	<u>Internal Pressure (PSIA)</u>	<u>Time to Rupture (Hrs)</u>	<u>Failure Location</u>
MPT-44F	8000.2	4.99	Not determined ¹
-49F	8000.2	17.47	Cap to body weld
-50F	8001.1	8.66	Cap to body weld
-51F	8002.2	4.60	Cap to body weld
-52F	8000.8	33.39	Cap to body weld
-53F	8000.8	7.88	Cap to body weld ²
-54F	6500.5	92.92	Cap to body weld
-55F	6500.2	96.37	Cap to body weld
-56F	6500.7	65.04	Not determined ³
-57F	6501.5	71.09	Cap to body weld
-58F	6501.9	116.13	Cap to body weld
-59F	6501.7	4.64	Stem (lower crimp)
-60F	6500.9	122.97	Cap to body weld
-61F	8000.9	27.03	Cap to body weld ²
-62F	6501.7	1.50	Stem (upper crimp)
-63F	8000.9	12.31	Cap to body weld

¹ Internal pressure of 9600 psia did not reveal leak.

² 90-95% separation of cap from body.

³ Internal pressure of 9500 psia did not reveal leak.

LOG INTERNAL PRESSURE, PSIA

LOG RUPTURE TIME, HR.

CAPSULES
LOT 7
X = 1000 C

FIGURE 15

TABLE IV

Pressure Burst Test Results
of Lot 7 TMS Capsules, 1010°C
(Pulse Arc Welded)

<u>Capsule Number</u>	<u>Internal Pressure (PSIA)</u>	<u>Time to Rupture (Hrs)</u>	<u>Failure Location</u>
MPT-64F	8063.9	16.40	Cap to body weld
67F	8063.1	15.42	Not determined
68F	8064.2	1.90	Cap to body weld
69F	8063.1	12.37	Cap to body weld
71F	6551.3	--	Not yet tested
72F	6553.3	--	Not yet tested
73F	--	--	Not yet pressurized or tested

FIGURE 16

Shown in Figure 17 is the comparison of mean rupture times for the Lot 7 capsules with the lines again representing prior data.

X. WR Production

WR production has now been initiated in all phases of the program. To date seven PuO_2 fuel batches, each weighing approximately 220 grams, have been successfully processed through charge weighing and calorimetry. Approximately 80 grams of yttrium has been prepared as chips of which 72 charges have been weighed. Hardware components fabricated to date total 1020. Table V shows the breakdown of hardware and the process yield (98.8%).

Fabrication of production heat sources was initiated with MAD-0012 through -0034. This series of units has been encapsulated through the clad while the second series (MAD-0035 through -0056) has been encapsulated through the liner.

LOG OF MEAN INTERNAL PRESSURE, PSIA

FIGURE 17

TABLE V
Quality Report on Hardware

<u>Component</u>	<u>Number Fabricated</u>	<u>Number Prime</u>	<u>Number Example Weld</u>	<u>Number Scrap</u>	<u>Process Yield (%)</u>
Liner Shim	165	162	--	3	98
Liner Cap	165	165	0	0	100
Liner Body	165	163	2	0	100
Strength Member Cap	180	167	9	4	98
Strength Member Body	105	90	13	2	98
Clad Cap	120	116	2	2	98
Clad Body	<u>120</u>	<u>117</u>	<u>1</u>	<u>2</u>	<u>98</u>
TOTAL	1020	980	27	13	98.8%