MLM-MU-76-67-0002

MILLIWATT GENERATOR HEAT SOURCE

PROGRESS REPORT

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April 16, 1976 - July 15, 1976

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Milliwatt Generator Program

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Miamisburg, Ohio.

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MONSANTO RESEARCH CORPORATION

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UNITED STATES ENERGY RESEARCH
AND DEVELOPMENT ADMINISTRATION

U. S. Government Contract No E-33-1-GEN-53

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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 April 16, 1976, to July 15, 1976, are presented below.

II. Yttrium Analysis (W. D. Pardieck and H. S. Carden)

The production yttrium as received from Ames Laboratory was in three lots of platelets, labeled by Mound as Lot 7A, 7B, and 7C. Recertification of Lot 7A has been completed, with a comparison of analytical data between Mound and the vendor shown in Table I.

When nitrogen was initially determined by Mound by vacuum fusion, the values were significantly higher than those reported by the vendor. An investigation led to the conclusion that the conversion of hydrogen to water (passage over copper oxide at 325°C) was incomplete because of the large amount of gas evolved when the sample was alloyed in the platinum bath. Fourteen mole percent hydrogen was found (by mass spectroscopy) in the carbon dioxide/nitrogen mixture after circulation of the gases through the copper oxide for fifteen minutes. This hydrogen content biased the nitrogen results on the high side. Increasing the circulation time reduced the error, but did not eliminate it. However, the hydrogen was completely converted to water by increasing the copper oxide temperature to 360°C. On re-analysis, the nitrogen results by vacuum fusion then compared favorably to Ames vacuum fusion values and Mound values when nitrogen was determined by the independent Pregl-Parnas-Wagner micro-Kjeldahl method.

The distillation unit currently used to separate fluoride from yttrium metal is being modified in efforts to reduce the amount of distilled perchloric acid which interferes

TABLE I
LOT 7A YTTRIUM^a

<u>Element</u>	<u>Platelets</u>		<u>Chips (Mound)</u>
	<u>Ames</u>	<u>Mound</u>	
Y	>99.1 % ^b	99.1 %	99.0 %
Ta	0.81%	0.73%	0.82%
O	340	390	390
C	115	136	121
N	103	124	123
H	14	10	6
F	4	13	16
Fe	32	13	13
Zr	1	<1	<1
Mg	-	<1	<1
Al	-	<1	<1
Si	-	<1	<1
Ca	-	<1	<1
Ti	-	1	1
Co	-	<1	<1
Cd	-	<1	<1

^aResults in ppm unless noted otherwise

^bCalculated by difference

with the colorimetric analysis. If this proves successful, analysis time will be reduced substantially on future samples.

Also evaluated was the possibility that the yttrium would be contaminated during the conversion of platelets to 1/16" square chips. A comparison of analytical data between the chips and platelets is also shown in Table I and, as can be seen, contamination did not occur.

Similar recertifications and platelet-chip comparisons are planned for Lots 7B and 7C.

III. Encapsulation (C. E. Burgan)

Fabricated during the report period were 27 fueled capsules of the 16-year, single channel design. Initial encapsulation of the first series of these units (MF-68C through -78C) was performed on May 12, 1976. All the units met the required acceptance criteria and were shipped to General Electric, Pinellas, on June 24, 1976, as partial fulfillment of Sandia reimbursable requirements. Encapsulation of the second series (MF-79C through -94C) was initiated on June 17, 1976. These units, also planned for shipment to General Electric, are now undergoing post-encapsulation evaluations.

IV. Pressure Burst Testing (D. R. Schaeffer and R. L. Wise)

In a joint Sandia-Mound effort to correlate dye penetrant indications with pressure burst behavior of simulated 25-year design capsules, Mound's portion of the study has been completed. Eleven capsules [MPT-94C(E) through -104C(E)] were fabricated from Lot 6 T-111 (Sheet 11), with dye penetrant evaluations performed after each of the fabrication steps. Ten of these capsules (of which four had dye penetrant indications after at least one of the fabrication steps) were then tested at pressures varying from 6000 to 8000 psia at 1000°C.

Results are shown in Table II. Failure of eight of the ten capsules occurred in the cap-to-body weld, with seven of

TABLE II
PRESSURE BURST TEST RESULTS (AT 1000°C)

<u>Capsule No.</u>	<u>Dye Penetrant Indications</u>	<u>Internal Pressure (psia)</u>	<u>Failure Time (Hours)</u>	<u>Failure Location</u>
MPT-94C(E)	No	7006	69.59	Cap-Body Weld
-95C(E)	Yes	6500	80.20	Cap-Body Weld
-96C(E)	Yes	8006	16.54	Cap-Body Weld
-97C(E)	No	7502	34.57	Cap-Body Weld
-98C(E)	No	6000	54.63	Cap-Body Weld
-99C(E)	Yes	*	-	- - -
-100C(E)	Yes	5995	162.86	Cap-Body Weld
-101C(E)	No	7994	3.12	Stem
-102C(E)	Yes	7003	55.28	Cap-Body Weld
-103C(E)	No	6504	101.09	Cap-Body Weld
-104C(E)	No	7997	20.41	Stem

* Not tested; reserved for inclusion evaluations

these developing small leaks in the downslope region of the weld (Figure 1). The other weld failure [MPT-96C(E)] was a separation of approximately 70% of the cap and body (Figure 2). Two of the ten capsules developed leaks in the tantalum stem, causing premature termination of the test. There was no relationship between the point of failure and the location of the dye indications.

Figure 3 is a log-log plot of internal pressure versus time-to-rupture. The data falls within a reasonable error band, and can be approximated by the equation

$$t_r = \left(\frac{A}{P} \right)^n$$

where t_r = time to rupture (hours)
 P = internal pressure (psia)
 n = constant of 7.5
 A = constant of 11,950

The data are also presented in a Larson-Miller plot in Figure 4 with stress substituted for by internal pressure.

One capsule [MPT-98C(E)] failed prematurely and does not compare favorably with the other results. Scanning electron microscopic analysis of this capsule attributed the early failure to inclusions in the T-111 at the failure site.

The equipment used in the pressure burst tests was calibrated before and after this test series. All the equipment retained its accuracy except for a thermocouple which had degraded, giving a 15°C error. Therefore, over the total testing period of 600 hours the test temperature increased from 1000°C to 1015°C. Because of the thermocouple degradation, another thermocouple has been added which duplicates the one used to determine the capsule temperature. The new thermocouple will be replaced with other calibrated thermocouples at frequent time intervals to check the accuracy of the main thermocouple.

MPT-100C(E) after 162.86 hours at 5995 psia, 1000°C

FIGURE 1

MPT-96C(E) after 16.54 hours at 8006 psia, 1000°C

FIGURE 2

LOG INTERNAL PRESSURE VS LOG RUPTURE TIME

LOG TIME

FIGURE 3

$P = T (15+ \text{LOG } t)$

FIGURE 4

V. Pressure Burst Test Failure Evaluations (R. E. Zielinski, E. Stacy, and C. E. Burgan)

Although most of the pressure burst test specimens fail above what would be considered design minimum, the failure zone is rather peculiar. Rather than fail by a random creep failure in the weld/body-heat affected zone (HAZ) interface, which should be typical for this design, most capsules failed by means of a "blow hole" occurring in the ramp-down area of the weld bead. Because of this unusual failure, a study was initiated to try to pinpoint the cause of the "blow hole" failure.

The first step of the post-pressure burst test examination was location of the failure zone. In all strength members which did not fail in the fill tube region or by cap/body separation, the failure was primarily a "blow hole" which occurred in the weld ramp-down area. Once the failure zone was identified, the strength member cap was sectioned so that the weld and body HAZ were preserved. Visual examination of the interior surface of the failed specimens indicated that a circumferential crack was present at the weld/HAZ interface on the body side of the strength member. The crack was present in all specimens that had been heated to test temperature and it has been demonstrated that this crack occurs during the heat-up to test temperature.

Once the strength members were sectioned and visually examined, they were mounted for metallographic examination. Some of the samples were mounted perpendicular to the weld bead and others parallel. The mounted specimens were ground toward the point of failure. At various times during this procedure, the samples were examined in both the as-polished and etched condition. While this study showed the extent of the circumferential crack (Figure 5) and crack paths through the weld (Figure 6), it did not provide any data which would explain why the failure occurred in approximately the same zone in each tested specimen.

As the next step in the evaluation process, the Scanning Electron Microscope (SEM) was employed. Using this instrument, it was possible to confirm the "blow hole" fracture

Circumferential
Crack in Heat-
Affected Zone

FIGURE 5

Radial Crack which
Propogated after
Initial Circumfer-
ential Crack

FIGURE 6

zone (Figures 7A and 7B), as well as identify the cause of the peculiar type of failure.

Examination of the as-tested specimens using the SEM was helpful in pinpointing the exact point of failure. Using the Energy Dispersive X-Ray (EDX) System which was ancillary to the SEM, high concentrations of iron were detected in the "blow hole" regions. In an attempt to identify the source of the iron signal, it was decided to chemically etch the as-tested specimens and examine them after various times in the etchant bath to see if the source of iron could be identified. This technique had previously been successfully applied to identify inclusions present in the "as-received" T-111 sheet material.

The etchant consisted of 70% HNO₃ and 30% HF by volume. Using this solution, a material removal rate of 0.15 to 0.20 mm per minute was possible. The first specimen which was placed in the solution underwent a violent localized etching in the failure zone and at several isolated areas in the body HAZ. SEM examination of this specimen after the initial etch showed that a foreign material was concentrated in the ramp-down zone of the weld (Figures 8, 9, and 10). Isolated locations of this material were also in the body HAZ (Figure 11). In fact, in every area that had etched violently, a concentration of foreign material was found beneath the surface. Since it was spherical in shape, this material had evidently been molten, presumably during the weld operation. The only location of the impurity in the weld was the ramp-down zone, where it extended through the cross-section as is shown in Figure 12. EDX analysis indicated that the material consisted of iron, silicon, tantalum, and some minor constituents.

Examination of other strength members that failed in the same manner indicated similar results. An impurity concentration was observed in the ramp-down zone of each strength member examined. The shape of the inclusions varied somewhat from capsule to capsule (Figures 13, 14, 15, and 16), depending upon the concentrations of the elements which were present. Iron and silicon were the major constituents in all samples, while tantalum, molybdenum, copper, and zinc were also detected. The elemental concentrations varied among samples.

← 7A

7B →

"Blow-Hole" Fracture
Zones in Center of
Weld Ramp-Down Zone

FIGURE 7

Foreign Material Concentrated in Ramp-Down Zone of Weld

FIGURE 8

Foreign Material Concentrated in Ramp-Down Zone of Weld

FIGURE 9

Foreign Material Concentrated in Ramp-Down Zone of Weld (Primarily Iron and Silicon)

FIGURE 10

Foreign Material in Body Heat-Affected Zone

FIGURE 11

Included Material Extending through Cross Section of Weld Ramp-Down Zone

FIGURE 12

Included Material Slightly Different in Shape than that in Figure 8

FIGURE 13

Included Material with
Shape Different than
that in Figures 8 and
13

FIGURE 14

Included Material
Similar to that in
Figure 13

FIGURE 15

Inclusion with Different
Appearance

FIGURE 16

Included Material
(Primarily Iron and
Silicon) in Starting
Sheet Material

FIGURE 17

By chemically etching random samples of the T-111 sheet material and examining these samples at various steps during the etching process, it was possible to prove the existence of localized concentrations of impurities as inclusions. This type of analysis indicated that there were three basic types of inclusions present in the sheet material (Figures 17, 18, and 19). The inclusions of the type observed in Figure 17 were primarily iron and silicon; while those observed in Figures 18 and 19 consisted of tantalum, silicon, iron, molybdenum, copper, zinc, and potassium. The difference between the inclusions observed in Figures 18 and 19 was their orientation with respect to the rolling direction. The inclusion in Figure 18 was parallel to the rolling direction, while the one in Figure 19 was perpendicular. The inclusion observed in Figure 19 extended approximately 0.25 mm into the sheet material. The distribution of these inclusions was quite random with only the iron-silicon type (shown in Figure 17) found in clusters. The effect of their presence on the hardware forming operation and the mechanical integrity of the strength member bodies has been well documented. Figures 20 and 21 show areas of strength member bodies that have cracked during forming and in each crack the presence of an inclusion just below the surface was noted.

In strength member bodies that have failed in the body sidewall during testing (Figure 22), the presence of included material was also observed (Figure 23).

Knowing that the inclusions were randomly dispersed throughout the starting sheet material and knowing the composition of the inclusions found in the welded capsule, it was possible to postulate the mechanism by which included material became concentrated in a ramp-down area of the strength member assemblies.

A continuous radial gas-tungsten arc (GTA) weld was used to attach the end cap to the strength member body. During the weld cycle, the molten "weld puddle" picked up all the inclusions in its path. These impurities were transported radially around the weld bead. In the ramp-down zone, the weld area where penetration decreased as power was reduced, the impurities precipitated out as a concentrated mass. From capsule

Inclusions in Starting
Sheet Material - Parallel
to Rolling Direction
(Fe, Si, Ta, Mo, Cu, Zn,
and K)

FIGURE 18

Inclusions in Starting
Sheet Material -
Perpendicular to
Rolling Direction
(Fe, Si, Ta, Mo, Cu,
Zn, and K)

FIGURE 19

FIGURE 20
Cracks in Bodies
During Hardware
Forming

20B →

→ 20A

← 21A

21B →

Cracks in Bodies
During Hardware
Forming

FIGURE 21

Capsule that Failed in
Sidewall (See Figure 23)

FIGURE 22

Included Material in
Sidewall of Failed
Capsule

FIGURE 23

to capsule, this mass varied in size, physical appearance, and composition. These variables depended upon the number and type of inclusions which were originally present in the weld zone of each capsule. However, the effect on the mechanical integrity of the strength member was the same. After the initial circumferential crack was formed at the weld/body HAZ at the outset of the test, several crack paths were initiated and as soon as one reached the included zone, it progressed rapidly through the zone and resulted in a linear crack path through the weld. This path terminated in "blow hole" failure on the outside of the weld bead.

Since a similar type of "blow hole" failure had been observed earlier in 16-year design capsules, several of these were examined in the same manner. The 16-year capsule design had a thinner strength member cap, and quite possibly a different type of failure mechanism. The examination of these capsules, however, showed that included material was also concentrated in the ramp-down zone (Figure 24). Thus, these capsules failed by the same mechanism.

To prevent "blow hole" failures from occurring in the future and possibly increase the life of the strength member, a study has been initiated involving a pulse arc welding technique to effect the closure weld. The technique pulses the weld arc through a programmed current range at the rate of two pulses every second. The pulsing should prevent the accumulation of included material in the weld ramp-down area. Another benefit of this type of weld is that the heat-affected zone is minimized.

While no strength member specimens have, as yet, been tested, several example welds have been made and examined. Some of the immediate benefits derived from this new technique were a more uniform weld bead on the interior of the capsule and a smaller, less pronounced heat-affected zone.

SEM examination of the weld zones also showed that the included material was randomly dispersed throughout the weld bead and not concentrated in the ramp-down area. The interior surface of a typical pulse arc weld bead can be seen in Figure 25. Included material was frozen to the bead. Approximately 0.18

Included Material in
Weld Ramp-Down Zone of
16-year Capsules

FIGURE 24

Interior Surface of
Pulse Arc Welded
Capsule

FIGURE 25

Random Disbursement
of Included Material
0.18 mm into the
Pulse Arc Bead

FIGURE 26

mm into the weld bead, inclusions were randomly dispersed (Figure 26) as they were in the starting material. In fact, several of the inclusions seemed to have suffered very little effect from the welding process (Figure 27).

From the standpoint of preventing a build-up of impurity material in the weld ramp-down zone, the pulse arc weld seems to be successful. Also, it provides a more uniform weld bead and a smaller heat-affected zone. This should result in a strength member that is structurally stronger than those previously tested.

Along with the good features, however, some possibly detrimental effects have been observed. In the pulse arc welded capsules, the initiation of the weld ramp-down zone is physically more apparent (Figure 28). Another adverse consequence might be porosity. The pulse arc welded material appears to have more isolated gas bubbles present in the weld zone. The third effect, which is rather isolated, having been observed only twice in the same weld, is a dendritic structure (Figure 29) found at the center of the weld structure. This zone is exposed to the highest temperature during welding. It is quite possible that some unusual inclusions were present in the two areas and the resulting heat input effected the formation of the dendritic structure. This effect was quite isolated in that it was found in only one sample and in only two widely separated areas of that sample.

It is impossible to assess the effects of these unusual features at the present time. However, the first series of pulse arc welded strength member samples will be thoroughly evaluated after test to assess whether these features, or any other peculiar to the welding process, adversely affect the mechanical integrity of the strength member bodies.

VI. Helium Leak Testing (J. D. Hastings, O. W. Dodds, and J. Stockton)

The investigation is continuing to develop a method for detecting helium leaks less than 1×10^{-11} cc/sec in the MWG strength member and clad components. A static accumulation system has

← 27A

27B →

Inclusions in Pulse
Arc Welded Capsules

FIGURE 27

FIGURE 28
Weld Ramp-Down Zone
on Pulse Arc Welded
Capsules

28B →

→ 28A

← 29A

29B →

Dendritic Structures
at Center of Pulse
Arc Weld

FIGURE 29

been developed, and is shown in Figures 30 and 31. The system has dual accumulation chambers, one which collects the helium from the heat sources (A) and the other (B) which collects the helium from the calibrated helium leak standard (C). (The internal volumes of the chambers agree within $\pm 5\%$, since the accumulation measurement is volume dependent.) A commercial Veeco MS-18 (presently MS-12) helium leak detector (D) is utilized for the vacuum system and analyzer. The external vacuum system consists of nominal $1\frac{1}{2}$ -inch high vacuum hardware with pneumatically controlled metal sealed valves (E) operated by the control unit (F, Figure 31) and precision timers (G). At the end of the accumulation cycle (presently two minutes), the pneumatic valves open and helium is "dumped" into the helium leak detector. A transient helium peak is recorded on the chart recorder (H). Typical helium saw-toothed pulses for eight heat sources (in the clad) are shown in Figure 32, with the calibrated helium standard analyzed before and after the heat sources.

The eight sources were measured simultaneously, with a value of approximately 2×10^{-12} atm cc/sec obtained. The value of the calibrated helium leak standard is 1.5×10^{-10} atm cc/sec. Longer accumulation times than two minutes may be used to improve the sensitivity. Reproducibility of the leak rate measurements is $\pm 10\%$, well within the accuracy of $\pm 20\%$ for the calibrated leak rate standards.

One of the reasons that helium leak detectors cannot measure leaks below approximately 10^{-10} cc/sec is that this is the background level from natural occurring helium diffusing through glass and elastomeric O-rings. For this system, therefore, metal seals and metal sealed valves were utilized. To further reduce the helium background, the accumulation chamber was outgassed at 800°C for 16 hours. A 16-hour background count showed helium levels of approximately 10^{-14} atm cc/sec.

Off-gassing the surface of the heat sources assures that small leaks (10^{-8} atm cc/sec or less) are not clogged with liquids. One hour outgassing at approximately 400°C reduces the dynamic helium leak rate to the background levels. The units are then cooled in situ to near room temperature so that the sensitivity is not altered by the increased temperature.

F = valve control unit
 G = precision timers
 H = chart recorder
 I = helium leak rate amplifier

FIGURE 31

FIGURE 32 — TYPICAL HELIUM LEAK RATE DATA FROM EIGHT HEAT SOURCES (2.00 minute accumulations)

VII. T-111 Evaluations (R. E. Zielinski, W. C. Wyder, D. A. Sohn)

During Mound's recertification of recently received Lot 7 T-111, two defects were ultrasonically noted in one piece of the 0.100" thickness. One of these defects was approximately 4" long with 1/4" width, while the second defect was 2" long with a 1/4" width. These portions of the material were cropped from the original sheet and a sample of the defect area was cross sectioned. Optical photomicrographs are shown in Figure 33, while SEM photomicrographs are shown in Figure 34. It was concluded that the defect was a stringer and the ancillary EDXRA of the SEM identified it as tungsten. This type of defect has never been observed in any of the samples previously examined.

Because this was the first Lot 7 T-111 available for SEM examination, the sample was chemically etched and reexamined. Approximately 1.5 mils of material was removed from the surface during etching. Examination of the same cross-section showed that the stringer was extended throughout the 0.38" length of the sample (Figure 35). Also obvious in Figure 35A are the inclusions typically observed in T-111 sheet material. The inclusions represented by the dark spots are well distributed throughout the cross section (Figure 36) and are of the same shape and composition as those observed in previous lots of T-111 (Figure 17).

VIII. Documentation

On June 24, 1976, DTER (Drawing Transfer Engineering Release) was officially received by Mound from Sandia. This permits Mound to change hardware drawings outside the formal ECN system during development phases. Once AER (Advanced Engineering Release) occurs, all changes will be transmitted to Sandia by the TIE System (Technical Information Exchange).

Plans are now being formulated for the development-production transition of other documentation such as operating procedures and specifications. The GEISHA system design is essentially finalized and Sandia concurrence has been obtained concerning format and the type of production information that will be transmitted.

← 33A
(17X)

33B →
(300X)

Lot 7 T-111 (0.100")

FIGURE 33

Lot 7 T-111 Tungsten
Stringer
FIGURE 34

34B →

→ 34A

← 35A

35B →

Lot 7 T-111 Tungsten
Stringer After Etching

FIGURE 35

FIGURE 36
T-111
Inclusions in Lot 7

36B →

→ 36A

IX. Tooling (J. E. Newland)

All the "development dies" (16 year tooling modified to produce 25 year hardware) have been demonstrated successfully and are being used to fabricate reimbursable and development hardware. Dimensions for the 25 year hardware have been established and design has been completed on all production tooling. Fabrication of the dies is 85% complete and the gage fabrication is approximately 75% complete.

Hardware bodies have been fabricated from the blank, draw, and sizing dies (production tooling), and dimensional data are now being obtained to determine whether further refinement is necessary.

In evaluation of the liner cap swage die, there was a problem in reducing the flange to the proper thickness. Grinding the stop blocks to increase the punch travel only caused a bowing of the cap. Shimming of the lower punch (ejector) decreased the nest depth with caps now appearing to be satisfactory (although dimensional data are not yet available).

Difficulties are also being experienced with the liner cap trim die. After five strikes in the die, the part would not eject. Increasing the spring pressure under the ejector did not solve the problem. The die is now being resharpenered and alignment will be checked before additional parts are tried.

Results look favorable with the strength member cap swage die. Some development will be necessary in adjusting stop blocks and the lower punch height, but no problems are expected. Parts are presently being evaluated dimensionally.

The clad cap boss swage die has been completed, but the swaging operation has not yet been demonstrated satisfactorily. Once development has been completed on this die, development work will be initiated on the tenon swage die.