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SNAP 13
THERMIONIC DEVELOPMENT
PROGRAM
GENERATOR FUELING REPORT

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SNAP 13

THERMIONIC DEVELOPMENT PROGRAM

GENERATOR FUELING REPORT

AUGUST 1967

MND-3060-33

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FOREWORD

This report is a technical summary of the efforts performed to radioisotopically fuel a SNAP 13 thermionic generator system and conduct a post-test examination. The work was performed for the Atomic Energy Commission under Contract AT(30-1)-3060 by the Martin Marietta Corporation. A portion of the effort was subcontracted to Thermo Electron Engineering Corporation under Contract 5E0895. The generator fueling, testing and post-test examination were conducted at the Oak Ridge National Laboratory.

The principal contributors to this report were Eugene Lamb of the Oak Ridge National Laboratory and John Jicha, Winston Seetoo and Walter Haass of the Martin Marietta Corporation.

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SUMMARY

In November 1965, a SNAP 13 thermionic generator was radioisotopically fueled with a curium-242 heat source using a fueling fixture apparatus. The generator fueling was accomplished in a hot cell at the Oak Ridge National Laboratory (ORNL) by remote means and represents the first such operation on a thermionic device. Optimized output obtained was 17 watts (34 amperes at 0.5 volt). Successful, steady operation was realized for a period of approximately 45 hours before generator failure occurred.

Generator failure was characterized by the following events: an electrical arc, a sudden increase in current output, a momentary increase in bell jar pressure, a relatively sudden drop in cesium temperature and a severely reduced generator output voltage. An extensive post-test examination was then conducted by ORNL to permit the cause of failure to be assessed.

Although definitive conclusions could not be reached, failure of the generator most probably occurred as a result of molten fuel leaking from its encapsulation and attacking the thin wall diode spacer causing a rupture of the cesium envelope. Fuel leakage probably took place during the capsule-to-emitter brazing operation which, due to excessive outgassing, required five attempts to accomplish. The characteristics of the fuel form most likely produced fuel melting during the capsule welding and brazing operations since the length of the fuel pellet was approximately 2-1/2 times its design length and the melting point of the fuel composite was estimated to be in the range of 250° to 650° C below its design melting temperature. In addition, leak testing of the capsule following the welding operation indicated doubtful capsule integrity probably due to cracking of the capsule wall. Leakage of fuel during the brazing operation (prior to generator ignition) is further substantiated by the fact that only about one-half the expected generator output was realized from the 290-watt (thermal) fuel loading.

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I. INTRODUCTION

The final portion of the SNAP 13 Thermionic Development program described in this report is concerned with the preparation of the radioisotopic curium fuel form and its encapsulation, the demonstration fueling, assembly and testing of a thermionic generator and, finally, the post-test inspection of the fuel form and generator. These efforts were the responsibility of the Oak Ridge National Laboratory (ORNL). The major objectives of these efforts were to demonstrate the feasibility of the preparation technique for the fuel form and its encapsulation, and the fueling, assembly and 90-day performance testing of the fueled generator, all in hot cell facilities by remote means.

Prior portions of the SNAP 13 Program involved the development of a thermionic generator suitable for use with an isotopic power source, development of a fuel capsule design, development of the apparatus required for remotely fueling and testing a thermionic generator with an isotopic power source and nuclear safety evaluations to assure all phases of the demonstration program could be performed without undue hazard. These efforts have been previously described in the SNAP 13 Final Summary Report (Ref. 1) and were performed by the Martin Marietta Corporation (MMC); Thermo Electron Engineering Corporation (TEECO) was the subcontractor in the area of thermionic generator development.

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II. DESCRIPTION OF GENERATOR, FUEL FORM AND FUELING PROCEDURE

Curium-242 was designated as the isotope to be used in the SNAP 13 thermionic generator because its high power density would provide the operating temperatures required, as well as low total volume and weight of the assembled converter. Oak Ridge National Laboratory (ORNL) was responsible for development of the fuel form based on MMC specifications of maximum internal temperature, thermal conductivity, matrix material, capsule material and capsule configuration. The basic fuel consisted of $(\text{Cm-242})_2\text{O}_3$ and $(\text{Am-241})\text{O}_2$ separated from the fission products formed during reactor irradiation of the Am-241 target. Thorium-oxide was added to the actinide oxide product to form a mixture having a melting point above 2000° C (the melting point of Cm_2O_3 was determined to be 1950° C). Iridium metal was used as a matrix in the volume ratio of 3/1 iridium to actinide-oxide to provide the desired thermal conductivity. The design objectives of the isotope fuel form are listed in Table 1.

TABLE 1

Fuel Form Design Objectives

Heat generation at encapsulation (watts)	315% $\pm$ 10
Geometry	Right cylindrical pellet
Diameter (cm)	1.9
Basic fuel material	Curium-oxide and americium-oxide mixed with metal matrix
Melting point (°C)	2250
Minimum pellet density	70% theoretical
Curium oxide-to-matrix ratio	~1:3 (blend depends on power density and thermal conductivity requirements)
Power density (watts/cm ³)	150 at 100% of theoretical density
Placement of fuel pellet in capsule	Metallurgical bond by brazing with platinum; remelt temperature greater than 2100° C
Chemical compatibility	All compounds, decay products and combinations must be compatible with capsule and braze materials at predicted normal operating temperature

The thermionic generator used in the SNAP 13 program is a cesium vapor, single-diode device housed in a cylindrical stainless steel container. The major features of the final generator design and the anticipated operating performance are presented in Table 2.

The generator fueling and assembly operation is accomplished by utilizing two key brazes--an emitter-to-fuel capsule braze and a casing closure braze--both of which must be made remotely by the fueling fixture. In addition to performing the brazes, the fixture must assemble three basic generator components (the upper and lower casing sections and the fuel capsule as shown in Figs. 1, 2 and 3), into their brazing positions while controlling the temperatures at levels that permit assembly within the design tolerances. These operations are performed in a vacuum due to the high temperature material characteristics of a thermionic generator. The fueling fixture must be located within a hot cell, while the control and monitoring instrumentation is located outside the hot cell during all assembly and fueling operations. Figure 4 shows the fueling fixture undergoing installation within the hot cell at ORNL.

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FIG. 1. THERMIONIC GENERATOR COMPONENTS

FIG. 2. GENERATOR CROSS SECTION

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FIG. 3. SNAP 13 FUEL CAPSULE DESIGN

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FIG. 4. INSTALLATION OF THE FUELING FIXTURE IN A HOT CELL

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TABLE 2

Generator 14 Design Features

Emitter material	Rhenium
Collector material	Molybdenum
Converter area (cm ²)	4.8
Converter geometry	Planar
Emitter-collector spacing (in.)	0.009
Fuel capsule diameter (in.)	1
Fuel capsule length (in.)	1.7
Casing diameter (in.)	2.64
Casing length (not including appendages) (in.)	4.2
Generator weight (lb)	4
End-of-life power level (watts)	12.5
End-of-life emitter temperature (°C)	1400 to 1450
End-of-life efficiency (%)	7
Operational life (days)	130
Environmental load specifications	Surveyor mission

The generator fueling and assembly operation is initiated by manually positioning the upper and lower casing sections in their respective support structures. The encapsulated fuel form contained within a transfer chill block is then placed into a water-cooled chill stand within the fixture by using the hot cell manipulators. The fueling fixture bell jar is remotely positioned and evacuated. The lower casing assembly is preheated to prevent thermal shock of the diode. The isotope fuel capsule is then positioned by the fueling fixture shaft assembly onto the emitter backup plate. An electron bombardment (EB) filament is rotated into position over the fuel capsule and actuated and the heat necessary to braze the capsule to the emitter backup plate is conducted through the length of the capsule to the emitter backup plate. After the braze is completed, the EB filament and brazing thermal shield are moved clear of the generator to permit positioning of the top casing. At this point, the top casing assembly is slowly lowered by the appropriate fueling fixture shaft onto the lower casing assembly and a determination of satisfactory diode operation is made. When satisfactory operation has been demonstrated, a previously positioned EB filament is used to heat the joint between the upper and lower casing and the final casing braze is made. After an extended casing outgassing period using a dynamic vacuum, the outgassing tubulation is remotely pinched off to complete the generator assembly.

Verification of the fueling fixture design was conducted by assembling mockup generators. Mechanical operations were checked by using dummy source capsules and the brazing operations were checked by using simulated capsules that had been modified to permit the insertion of a tungsten wire resistance heater to simulate the radioisotope heat. Qualification of the apparatus was accomplished by the successful assembly of an electrically heated prototype generator (Generator 13) at the MMC Nuclear Division.

Generator 14 and a backup generator (Generator 15) were received by MMC from TEECO and shipped to ORNL in January 1965. The fueling fixture, supporting equipment and instrumentation were shipped from MMC to ORNL in December 1964.

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III. CURIUM-242 FUEL FORM PREPARATION AND ENCAPSULATION

A. PREPARATION OF CURIUM OXIDE

Processing at ORNL of the Cm-242 fuel for the SNAP 13 test started on September 8, 1965, with receipt of the shipment of Cm-242 solution from the Curium Recovery Facility (CRF). Based on analyses of the solution, the Am-241 content of the batch was too high and the level of other contaminants (primarily lithium, calcium and nickel) was high enough to require more processing than the direct oxalate precipitation normally used in preparation of a $(\text{Cm-242})_2\text{O}_3$ source form. Since a different processing scheme was necessary, procedures were modified during the actual fuel processing as information was gained on a day-by-day basis. The originally planned procedure consisted of a series of accumulated oxalate precipitations in which each feed batch would be used to redissolve the actinide oxalate precipitates resulting from precipitation of the previous batch(es). It was then planned to make one final oxalate precipitation and collect the oxalate on a filter for calcination. The procedure was based on the presumption that the feed material would be of sufficient purity as to require no additional chemical separation.

The high level of contaminants in the feed (as shown in Table 3) necessitated considerable chemical processing.

TABLE 3

Feed Composition*

<u>Element</u>	<u>Material Cm-242 (gm)</u>	<u>Total Quantity (gm)</u>
Curium-242	--	3.63
Americium-241	3.98	14.4
Plutonium-238	0.51	1.85
Lithium**	109.0	395.0
Nickel**	2.0	7.26
Calcium**	0.25	0.91

*All quantities of Cm-242 are corrected to the pellet preparation date, 11/26/65, except where noted.

**These contaminants result from processing chemicals, chemical contaminants and corrosion of containment materials.

The final procedure developed is as outlined:

- (1) On each feed batch, a series (usually three) of oxalate precipitations at pH 1.0 to 1.5 was made to remove lithium and other contaminants except for americium, calcium and nickel.
- (2) The separation of curium from americium was done. Calcium was also separated in this operation.

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- (3) Another series of oxalate precipitations was run to remove the potassium introduced during the americium separation. The pH was controlled at 2.5 to 3.0 during these precipitations in order to remove the nickel.
- (4) When the solution analysis showed an acceptable level of purity, a final oxalate precipitation at pH 1.0 to 1.5 was made and the oxalates were collected on filters.

Four batches of feed were transferred to the Source Fabrication Facility (SFF) process equipment by means of a shipping cask; a nitric acid rinse of the cask was made. The first three transfers, a total of 14.6 liters, were routine; however, only 3.7 liters of solution were obtained on the fourth transfer compared to the expected 10.8 liters. The cask was rinsed twice with 2 liters of 4 N HNO_3 and once with 4 liters of 4 N HNO_3 . Then 12 liters of 8 N HNO_3 were added to the cask and allowed to stand for 48 hours. A sample of this solution showed < 0.01 gram Cm-242 remaining in the cask. The total Cm-242 removed from the cask is given in Table 4.

TABLE 4

Batch Quantities of Curium-242

<u>Transfer No.</u>	<u>Quantity of Cm-242 (gm)</u>
1	0.37
2	1.17
3	1.38
4 (plus rinse)	0.71
Total	3.63

The CRF analysis of the solution transferred into the cask was a total of 4.4 grams of Cm-242.

Two of the feed transfers (the second and third) were made without diluting the feed, and samples were taken immediately after transfer. These samples were analyzed radiochemically for Cm-242, Am-241, Pu-238 and Ce-144. Spectrographic and flame photometric analyses were made for the inert constituents. Based on these analyses, the composition of the feed from the CRF is shown in Table 3.

The Ce-144 total content was approximately 9 curies. Other elements present were iron, chromium, molybdenum, manganese, silver and aluminum, all at < 0.05 mg/ml, and copper at ~ 0.1 mg/ml; none of these is a problem in the source due to separation from the curium in the oxalate precipitations. Elements specifically sought and not found were silicon, lead, titanium, vanadium and zirconium.

The separation of curium from americium was accomplished by precipitating $\text{K}_3\text{AmO}_2(\text{CO}_3)_2$ with the use of $\text{K}_2\text{S}_2\text{O}_8$ as the oxidant; separation from other ionic contaminants was accomplished by a series of oxalate precipitations of curium, americium and plutonium. The four batches of product solution were assayed for Cm-242, Am-241 and Pu-238 by radiochemical analysis, and a spectrographic analysis for other components was made. Results of the assay are shown in Table 5.

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TABLE 5

Product Solution Content

Product Solution Content (gm)				
<u>Element</u>	<u>Batch 1</u>	<u>Batch 2</u>	<u>Batch 3</u>	<u>Batch 4</u>
Cm-242	0.60	1.40	0.32	0.31
(Cm-242) ₂ O ₃	0.66	1.53	0.35	0.33
(Am-241)O ₂	0.74	0.73	0.49	1.47
(Pu-238)O ₂	0.08	0.04	0.11	0.03
CaO	0.06	0.03	0.02	0.03
ThO ₂ ***	0.39	0.58	0.24	0.47
Aluminum	*	--	--	--
Boron	--	--	--	--
Chromium	0.04	< 0.02	--	< 0.04
Copper	0.04	< 0.01	0.04	0.16
Iron	0.04	0.16	--	0.14
Potassium	--	0.26	0.14	0.13
Magnesium	< 0.01	< 0.06	< 0.04	< 0.04
Manganese	--	--	--	--
Molybdenum	--	--	--	--
Nickel	0.06	< 0.01	< 0.04	0.35**
Silicon	--	--	--	--
Tin	--	--	--	--
Titanium	--	--	--	--
Zirconium	--	--	--	--
Yttrium	< 0.01	< 0.06	< 0.04	< 0.04

*Very fine trace.

**Two oxalate precipitations followed.

***Added to raise melting point.

These analyses were done just before the final oxalate precipitation in all cases except the last, which was analyzed during the next-to-last precipitation. Based on these results, the amount of ThO₂ needed was calculated and the thorium was added to the acidic Cm-242 solution in the nitrate form. An oxalate precipitation was then made and the precipitate was collected on stainless steel filter thimbles.

The self-heating of the precipitated oxalate was sufficient to effect calcination to the oxide. Temperatures measured at various times by optical pyrometry ranged from 900° to 1200° C; the minimum required calcination temperature was 700° C. Since the measured temperature was probably low due to measurement through the cell windows and was a surface temperature, the inner temperature of the fuel mass was quite likely above 1200° C.

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After all four fractions had been processed to obtain $(\text{Cm-242})_2\text{O}_3$, they were assayed for Cm-242 by calorimetry. The required amount of matrix metal was calculated based on the total oxide content of the four fractions. Since the ratio of Cm-242 to other oxides was known by prior analysis, the Cm-242 content was a measure of the total oxide content. Proportional amounts of iridium matrix metal (as oxide) were added to each of the fractions and each was manually stirred very briefly--a few seconds. Each of the four fractions was blended in the blender four times (two of 15 seconds each; two of 30 seconds each) individually and then all together. Although the theoretical density of IrO_2 is 3.15 gm/cm^3 versus $\sim 11.7 \text{ gm/cm}^3$ for curium, americium and thorium oxides and $\sim 10.0 \text{ gm/cm}^3$ for PuO_2 , blending efficiency had been demonstrated by cold work on simulated material. The blended material was transferred from the blender vessel to platinum-lined cones for calorimetry; then it was mechanically transferred by spatula from the cones to the die bodies. The cones were checked for Cm-242 by calorimetry after the transfers were made to determine if any material was lost. The composition and theoretical volume of the constituents of the Cm-242 product are given in Table 6.

TABLE 6

Composition of SNAP 13 Pellet (290 watts)

Constituent	Quantity (gm)	Theoretical Volume, cm^3
$(\text{Cm-242})_2\text{O}_3$	2.60	0.24
$(\text{Am-241})\text{O}_2$	3.30	0.28
$(\text{Pu-238})\text{O}_2$	0.26	0.02
CaO	0.13	0.04
ThO_2	1.58	0.16
Matrix metal	34.50	1.54
Platinum sheath	<u>13.33</u>	<u>0.62</u>
Total	55.70	2.90

B. PELLET PRESSING

The preparation of the pellets consisted of the following steps:

- (1) Cold pressing of the mixture of actinide and iridium oxides at $\sim 15,000$ psi to form unsintered, partially densified pellets.
- (2) Insertion of the cold-pressed pellets into a cylindrical platinum jacket formed from five-mil foil.
- (3) Heating of the cold-pressed pellets in the loose-fitting platinum jacket contained in the hot-press die body to reduce IrO_2 to iridium metal. The pellets must be heated to a minimum of 1050°C for a minimum of two hours in a helium atmosphere.
- (4) Application of pressure at the required temperature to complete the hot-pressing operation. The hot-pressing conditions are 3500 psi under an argon atmosphere for two hours at a temperature of 1450°C .

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Two cold-pressed pellets were formed in a 0.700-inch diameter die and placed in a 0.718-inch ID platinum cup. However, the total length of the two pellets was ~1-3/8 inches--a height greater by approximately a factor of two than had been calculated, and exceeding the available length in the cup. This excessive length can most probably be attributed to lack of attainment of the hot pressing objectives and the presence of undetected contaminants (see Section VI-A-3). In addition, the pellets were observed to have grown in size within a 30-minute period. Provision for the greater length was made by placing one pellet in a shorter 0.740-inch ID platinum cup which was positioned in the alumina sleeve of the hot-press die set. The original cup with one pellet was then lowered in an inverted orientation into the ceramic sleeve until it nested inside the larger cup and rested on top of the lower pellet. The top ceramic punch was inserted into the ceramic sleeve, and the die set assembly was installed in the hot-press furnace. The use of the 0.740-inch ID platinum cup necessitated the use of a 0.783-inch diameter die rather than the originally intended 0.750-inch diameter die.

After the die set containing the cold-pressed pellets was loaded into the induction furnace, the furnace and press frame were raised to a vertical position, and heating was begun at 7:50 PM, November 26, 1965.

The temperature of the pellet was indicated by a thermocouple of Pt-10%Rh:Pt-30%Rh located between the bottom alumina die punch and the platinum-jacketed pellet. The temperature as indicated by this thermocouple was increased to ~1050° C over a period of 2.5 hours and held at this temperature without application of pressure for 2.2 hours to permit decomposition of the IrO_2 to iridium metal. The calculated maximum internal temperature of the pellet was 1350° to 1500° C during this time (assuming a thermal conductivity value of less than 0.1 watt/cm-°K) well above the 1050° C minimum IrO_2 decomposition temperature in air. In addition, a flow of helium into the die body was maintained during the entire heating cycle to exclude air and to provide dilution and sweeping of the oxygen resulting from the IrO_2 decomposition.

After the IrO_2 decomposition period, a pressure of 3500 psi was applied to the die while the temperature was increased to 1320° C over a one-hour period. At this point the thermocouple in contact with the pellet gave erratic indications of temperature, and further heating over a 20-minute period to raise the temperature an additional 45° C was monitored by a thermocouple in the die body. Although the desired temperature and time of 1450° C for two hours was not attained according to the reading on the erratic internal thermocouple, ORNL personnel estimated on the basis of the indirect temperature measurement that the pellet was at a minimum temperature of approximately 1370° C* for at least 1 hour and 25 minutes. The pressure was released and the pellet was maintained between 1000° and 1050° C as determined by the die body thermocouple for approximately 10 hours until the electron beam welder checkout was completed.

Power to the hot press was shut off at 12:00 noon, November 27, and the die assembly had cooled sufficiently to permit its removal from the hot press at 4:00 PM. Temperature readings at time of removal were 418° C on the internal thermocouple and 370° C on the die body thermocouple.

A photograph of a curium-242 pellet at one stage of its development is shown in Fig. 5. The picture was taken through a two-foot water shielding window and shows the pellet to be at a temperature of about 1000° C even though resting on a chill block.

*Based on a die body temperature of 1225° C and a calculated ΔT to the fuel pellet of 225° C, the pellet temperature was 1450° C.

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FIG. 5. CURIUM-242 PELLETT

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C. TRANSFER OF PELLETT TO CAPSULE

After the die body was removed from the hot press, the ceramic insert (high purity Al_2O_3) was found to be stuck tightly in the graphite outer shell (Type ATJ). The alumina liner assembly was removed from the graphite die body by hammering on a steel rod with a steel cylinder held by a manipulator. The removal operation required approximately 45 minutes in an air environment.

Measurement of the pellet length with the ceramic insert and punch assembly intact showed the length of the pellet to be 0.730 inch. The volumes and calculated densities (based on the weights of material for the pellet shown in Table 6) are listed in Table 7.

TABLE 7

Volumes and Calculated* Densities of Cold and Hot Pressed Pellet

<u>Pellet</u>	<u>Volume (cm³)</u>	<u>Density (gm/cm³)</u>		<u>Calculated Effective Density (%)</u>
		<u>Calculated</u>	<u>Theoretical</u>	
Cold pressed	8.7	6.4	--	--
Hot pressed with platinum	5.7	9.8	19.2	51.0
Hot pressed without platinum	5.1	8.3	18.6	44.6

*All calculated densities assume total weight of added material only.

The ceramic liner assembly was then placed in a machinist's vice, contained in a platinum tray, and broken by tapping with a hammer. Both punches broke loose smoothly from the ends of the pellet but a section of the alumina liner adhered to the side of the pellet. Most of the ceramic material was removed by tapping with the hammer, and several small pieces were removed with a stainless steel screw driver used as a chisel. This operation required approximately 40 minutes. The general appearance of the pellet was good. The ends were flat and the side area was smooth except for two or three very small pieces of ceramic which were difficult to remove. A Dewar flask containing liquid argon and equipped with a platinum-lined nickel crucible suspended near the top of the flask was used to transfer the pellet from the hot-press cell to the adjacent transfer cell.

A cylindrical section of a tungsten source capsule was used as a go-no go gage. Initial attempts to pass the pellet through the gage were not successful due to the small pieces of ceramic material adhering to the pellet, but pressure on the pellet caused the ceramic pieces to flake off and the pellet passed freely through the gage. The outside diameter of the pellet was approximately 0.783 inch. The pellet was placed in a chill block with a cover, the pellet cavity was purged with argon, and the line admitting argon was closed with a valve to maintain an argon atmosphere around the pellet during its transfer to the welder. Ascertaining pellet dimensions consumed approximately 15 minutes in an air environment. For a discussion of the possible reasons for the low effective pellet density attained, see Section VI-A-3.

The chill block was removed through the rear of the transfer cell and placed in a stainless steel vessel with a friction fit cover contained within a lead-shielded cask.

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Liquid argon in the stainless steel vessel served as a coolant and provided an inert atmosphere around the chill block. The assembly was transported by lift truck from the SFF to the cell block in the adjacent Radioisotope Development Laboratory in Building 3047.

D. ENCAPSULATION

Conditions were first developed for EB welding of the SNAP 13 capsule on a bench setup. The welding conditions were as follows:

Welding speed (rpm)	10
EB spot location	Bottom edge of spot at joint
EB angle of incidence	15° (fixed by welder design)
Low beam current (ma)	~ 100 (fixed by high beam setting)
High beam current (ma)	240
Initial capsule temperature (°C)	600 (based on observing first capsule in similar chill block)

The procedure required one revolution at low beam current and an immediate increase to welding current (high beam) for one revolution. The beam current was then gradually decreased over about one revolution to a sufficiently low value to prevent crater formation upon shutting off the beam. These conditions and procedures resulted in welds which met the conditions of 0.010-inch maximum radial weld overhang, full penetration, helium leaktight and no indications of cracking.

After completion of the welding development, the EB welder was transferred to Cell D in the Radioisotope Development Laboratory. The short 10-foot cables were replaced with 65-foot cables for operation in the cell. The additional length of cable caused an unacceptable 10% loss of beam current. Modification of the originally developed procedure resulted in regaining the desired beam current but with a loss in sensitivity. A change in the gun was also made to achieve a smaller spot size with the objective of reducing the weld overhang.

A practice capsule was welded using these revised conditions. This capsule had a somewhat narrower weld, which decreased the general weld overhang to approximately 0.003 inch on the diameter. However, a 0.020-inch protrusion of weld metal occurred in one spot which was thought to be due to a small instability in the beam during welding and, therefore, was not expected to be typical. The narrower beam also penetrated better. It was observed, however, that it made a narrow weld bead trace on the opposite inside diameter wall of the capsule during welding.

On November 26, at approximately 1:00 PM, a test weld was made under these conditions using set No. 4 of the wrought tungsten-molybdenum capsules. The result was almost identical to the previous one, including the 0.020-inch protrusion and the EB trace on the capsule inside diameter. This capsule was fluorescent-penetrant inspected and found to have four cracks in the longitudinal direction, each of which had one end at the weld bead but did not extend into the weld. The appearance of these cracks after welding was surprising in view of the fact that many simulated weld joints had been made on the wrought W-2%Mo material with no evidence of cracks of any kind. The cause of the cracks could not be established but was suspected to be related to the potential thermal stress caused by the narrower electron beam striking the opposite wall and/or the possibility that cracks existed in the capsule although it had been previously inspected twice by fluorescent-penetrant inspection. Two actions were immediately taken. One was to return to the wider beam and the other was to reinspect the remaining capsules using the fluorescent-penetration technique.

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The remaining capsules included four of wrought material and one of cast material. Two of the wrought capsules exhibited minute surface cracks on the inside surface. The cast capsule exhibited gross pitting on the inside surface and also had a prominent 1/4-inch long crack at the lip of the capsule.

On the morning of November 27, a test weld was attempted with the cast capsule. Upon reaching welding current, the welder arced and shut off with a resulting over-weld in one spot. This same cast capsule was used again in order to re-establish the stability of the welding conditions.

At approximately 9:30 AM on November 27, a successful test weld was made with one of the wrought capsules (one which exhibited surface cracks on the interior surface). The weld metal overhang was 0.015 inch on the diameter, and sectioning revealed full penetration. On the basis of this inspection, the technique for welding of the fuel capsule was approved.

At approximately 5:10 PM, the pellet was moved to the welder, but immediate insertion into the capsule was prevented by some protrusions (apparently alumina pieces) on the pellet. During the next 40 minutes, attempts were made to gently force the pellet into the capsule by alternately lifting, dropping and rotating the capsule in the chill block. At approximately 5:50 PM, the pellet was inserted by pressing it toward the bottom of the capsule with a stainless steel rod. It was not known whether the alumina pieces fell inside or outside the capsule and to what extent the pellet contacted the capsule bases. A flow of argon gas into the welder was maintained during the time required for pellet insertion.

The welder was evacuated to 2×10^{-5} torr and maintained at this vacuum for one hour without a cap on the capsule to reduce oxides on the pellet and capsule which may have formed during the transfer and loading operations. Previous experimental data obtained on a hot-pressed platinum-wrapped iridium pellet in air indicate that very little oxidation occurred during the period required for removal from the die body and transfer to the capsule. A weight gain of 0.4 mg/hr was observed on a full-size dummy SNAP 13 pellet at 1000° C in air, the worst oxidation condition.

At approximately 7:00 PM, the weld chamber was backfilled once again with argon and the cap on the capsule opened to position. The welder was evacuated; however, the cap did not appear to be positioned properly and the welder was again backfilled with argon and opened to reposition the cap. The welder was evacuated to 2×10^{-5} torr prior to performing the weld. Adjustment of the capsule cover took approximately 20 minutes.

Optical pyrometer measurements through the cell and welder windows indicated that the capsule temperature was 840° to 900° C in the evacuated welder. The capsule temperature was estimated by applying a correction factor of $+ 190^\circ \text{C} \pm 20$ for the shielding window. No correction factor was available for the window on the welder. Based on previous work, welding conditions developed on the basis of a 600° C starting capsule temperature may not result in a desirable weldment if used on a capsule originally operating at 900° C. Therefore, a first pass at 100-milliamperes current was made to establish base conditions. Visual inspection indicated that the joint was sealed but that the penetration was insufficient. A second pass was made at 175 milliamperes and significant penetration was indicated by weld bead width, weld contour and vapor plating of tungsten on the weld chamber window--conditions associated with acceptable welds made previously. A third pass of approximately 200 milliamperes was not attempted because it was judged that the resultant weld overhang could exceed specifications.

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The helium leak detector test of the welded capsule indicated a release rate of 10^{-7} cm³/sec. A previous leak detector determination of helium release from the unsealed capsule had also shown a rate of 10^{-7} cm³/sec. The helium background in the welder was determined to be negligible before the pellet was inserted and after the welded capsule was removed from the welder (as indicated by a leak rate measurement of 3×10^{-8} cm³/sec at both times). These measurements indicate that the leak tightness of the weld was questionable. A portion of this apparent leak rate could be attributed to a contaminated capsule resulting from the difficulties encountered during the placement of the pellet into the fuel capsule.

The welding chamber was then brought up to atmospheric pressure with argon, and the capsule was removed from the welder and transferred to a tungsten chill block on the cell tray. A go-no go gage was passed over the weld area, which indicated that total overhang in the weld area was approximately 20 mils. Visual inspection indicated this overhang to be present on one side only. A platinum foil smear was taken of the weld area, and a bell jar with an argon purge was placed over the chill block and capsule. The platinum smear was removed from the cell and determined to contain 250 counts/min of alpha activity.

The capsule was taken out of the chill block and examined in front of the cell window in an air environment for 5 to 10 minutes. No cracks could be discerned. Doubt was expressed by ORNL personnel that a tighter capsule could be obtained in a subsequent encapsulation attempt. Therefore, a decision was made to continue with the subsequent generator fueling operations rather than remove the pellet and encapsulate it in the spare capsule. This decision was made with the concurrence of New York Operations Office, the Martin Marietta Corporation and ORNL personnel.

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IV. GENERATOR FUELING AND OPERATION

Generator 14 which was used in the fueled test and a backup generator (Generator 15) were received by MMC from TEECO and shipped to ORNL in January 1965. The fueling fixture, supporting equipment and instrumentation were shipped from MMC in December 1964 and were set up outside Cell C in Building 3047 at ORNL.

A. PREFUELING CHECKOUT

In order to familiarize ORNL personnel with the operation of the fueling fixture, a series of prefueling checkout runs was performed using thermal mockup generators. This effort took place during March 1965 and involved the capsule-to-emitter and casing brazes for two thermal mockups. For the initial mockup, difficulties such as high bell jar pressure and arcing of the high voltage filaments were encountered during the capsule-to-emitter braze; these difficulties are attributable to the expected outgassing of the thermal mockup parts. A successful capsule-to-emitter braze was performed on March 9. After similar difficulties, a successful casing braze was performed on the initial mockup on March 15. A remelt of this braze was also performed.

During brazing of the second thermal mockup, high voltage arcing occurred. Following appropriate fueling fixture repairs, both the capsule-to-emitter and casing brazes were successfully accomplished on March 22 and 23, respectively.

Upon completion of the preliminary checkout operation in April 1965, the fixture was installed in Cell C. The thermocouple and heater leads for Generator 14 were rewired to multicontact plugs in May 1965 to facilitate connecting and disconnecting the generator with the hot cell manipulators.

The generator was installed in the fixture and checked out by the Martin Marietta-ORNL test team in July 1965. During this test the generator was powered with the capsule-to-emitter EB filament. Review of the data realized from this test indicated normal generator operation but inconsistent thermocouple readings. This necessitated a retesting effort which was conducted in October. Instrumentation difficulties persisted and a retest with a radiation-type heater (Generator 11 heater) was scheduled for November. At this time, it was discovered that several thermocouples were incorrectly wired into the plugs. The leads were resoldered, hooked up correctly and checked out.

A final prebraze checkout of the fixture was performed in accordance with the Technical Manual (Ref. 2) on November 22, 1965. During the period of November 27 to 28, Generator 14 was fueled.

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B. CAPSULE TRANSFER TO FUELING FIXTURE

The transfer of the welded capsule containing Cm-242 from the welder in Cell D to Cell C where the fueling fixture was located was done in an argon atmosphere and took approximately three minutes. The transfer of the fuel capsule from the conveyor to the chill stand in the fixture was accomplished in air in about three minutes.

Once the capsule was in the chill stand, the bell jar was lowered and a vacuum established. Approximately two hours were required for the vacuum to reach 2×10^{-5} mm Hg. While the system was pumping down, the capsule heated up to approximately 1000° C (estimated) in the chill stand. The generator base was preheated to prevent thermal shock to the diode during the capsule transfer.

When the capsule was transferred, the T_1 (cesium tip temperature) was 330° F, T_2 (cesium reservoir temperature) was 400° F and T_3 (generator casing) was 490° F. During the transfer, the capsule touched the temporary braze shield as it was lowered and resulted in a slight shifting of the shield. When the capsule was on the emitter, the cesium and casing heaters were shut off and the temperatures were allowed to float. The open circuit voltage began to climb immediately and stabilized at approximately 0.2 volt. About two hours of additional outgassing were required to regain a pressure of 2×10^{-5} mm Hg. Just prior to the capsule braze, T_1 was 335° F, T_2 was 410° F and T_3 was 500° F.

C. CAPSULE-TO-EMITTER BRAZE

When the capsule-to-emitter braze filament was rotated over the capsule, the top of the capsule was parallel to the filament, indicating that the capsule was fairly well seated and not cocked. Because of outgassing (efforts were made not to exceed 8×10^{-5} torr inside the bell jar), the specified braze schedule could not be followed. Five attempts over a 2-hour and 20-minute period were required to achieve the braze input level. In two of these attempts, a braze power greater than that required to braze mockup generators was attained. A positive indication* of a braze was not realized. Repeated additional attempts at this braze temperature were deemed inadvisable since severe thermal cycles to the diode would result.

During the braze attempts, some arcing occurred between the filament leads along the ceramic leadthrough and along the ceramic heat shield. The maximum temperatures recorded during the braze attempts are given in Table 8.

*During some of the mockup and electrically heated generator assembly operations, instability in brazing power supply amperage occurred or a sharp rise in bell jar pressure was observed. The excessive outgassing could have either masked the usual pressure rise or resulted in an earlier outgassing of the braze material.

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TABLE 8

Maximum Temperatures During
Capsule-to-Emitter Braze Attempts

Braze Attempt	Temperature (° F)			Heating Time (min)
	T ₁	T ₂	T ₃	
1	430	565	690	50
2	405	455	612	20
3	405	460	638	25
4	388	425	565	15
5	400	472	675	10

D. HEAT SHIELD REMOVAL

When the heat shield pickup arms were lowered into the generator to remove the heat shield, they did not extend fully into the heat shield. (Actually, the shield had sagged into the generator either as a result of the prolonged heating necessary to effect the capsule braze or because the shield was shifted during the capsule transfer or both.) As a result, the bottom edge of the shield would not clear the top of the fuel capsule by approximately 1/32 inch even with the pickup switch in the manual mode of operation.

The shield was slowly rotated until the edge touched the top of the capsule. Further rotation caused the shield to cock and clear the capsule. The pressure to the pickup arms was increased to prevent the cocked shield from slipping and falling and possibly break the casing braze filament. The heat shield removal operation took approximately 15 minutes.

Since heat losses would increase when the braze shield was removed, the cesium heaters were turned on prior to heat shield removal. A record of the various temperatures is given in Table 9.

TABLE 9

Temperature Changes During Heat Shield Removal
and Cover Lowering Operations With Cesium Heaters On

Time (min)	Temperature (° F)			Remarks
	T ₁	T ₂	T ₃	
0	425	425	500	Initiate shield removal
30	530	480	590	Cover lowering
65	550	550	660	Cover lowering

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E. CASING COVER LOWERING

As the cover was lowered, it became apparent that the shield assembly, contained within the cover, would not clear the capsule. This was noted when the shield touched the capsule. The apparent factors which caused the interference were capsule weld overhang (approximately 20-mils occurred all on one side) and misalignment of the cover. (The holding screws probably loosened during the capsule-to-emitter braze cycles.) Cover alignment therefore became extremely critical since a 20-mil clearance exists between the capsule and thermal shield. Alignment of the casing cover was achieved by using the capsule pickup arms in the manual mode to provide a slight lateral movement allowing the shield to clear the capsule. The cover was then lowered in 1/4-inch increments and the open circuit voltage observed in each step. As the cover was lowered, there was difficulty in maintaining bell jar pressure. When the upper casing reached a level approximately one inch from its fully assembled position, the diode was ignited when the open circuit voltage pegged the voltmeter. During this time T_1 crossed T_2 (see Table 9). At the final 1/8-inch position, the cover hung up and would not close. The cover was raised, then lowered again, and it settled into place on the generator base. Prior to ignition, the open circuit voltage was increasing as the cover was lowered. The generator was ignited at approximately 3:30 AM, November 28. The entire cover lowering operation took about 80 minutes.

F. GENERATOR IGNITION

The ignition power supply was used to ignite the generator at $T_1 = 550^\circ \text{ F}$, $T_2 = 550^\circ \text{ F}$ and $T_3 = 655^\circ \text{ F}$; the output was 0.500 volt and 30 amperes. The expected optimum output with a 290-watt loading based on previous generator operational experience is 0.5 volt, 60 amperes. During the subsequent optimization procedure, a maximum of 17 watts electrical output was achieved (0.500 volt, 34 amperes). A tabulation of temperature versus output is given in Table 10 while a plot of temperature versus time is given in Fig. 6.

TABLE 10

Time of Day, AM	Diode Optimization Data			Output		Remarks
	Temperature ($^\circ \text{ F}$)			(volt)	(amp)	
	T_1	T_2	T_3			
5:15	550	550	655	0.5	30	Ignition
6:10	475	585	700			Temperatures varied to obtain optimum output
6:35	570	615	742			
7:05	509	628	751			
8:25	565	725	860	0.5	34	

G. CASING BRAZE

The water to the generator support stand was turned on to prevent excessive heat from the EB filament from adversely affecting the ceramic-to-metal seals in the diode. The minimum temperatures were recorded 15 minutes after the water was turned on and were $T_1 = 500^\circ \text{ F}$, $T_2 = 470^\circ \text{ F}$ and $T_3 = 582^\circ \text{ F}$.

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FIG. 6. SNAP 13 ISOTOPIC GENERATOR OPERATION

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When the casing filament was energized, severe outgassing occurred. Three attempts to reach braze temperature were unsuccessful with less than half the braze schedule being attained. The final braze attempt is given in Table 11 with pertinent comments.

TABLE 11

Generator Temperatures During Final Casing Braze Attempt

Time of Day	Temperature (° F)			Remarks
	T ₁	T ₂	T ₃	
9:10 AM	530	550	738	
9:25 AM	498	555	745	
9:55 AM	510	580	775	
10:24 AM	520	600	792	
10:25 AM	1030	605	800	
10:26 AM	1020	605	795	Arc occurred; high voltage tripped
10:35 AM	1050	612	812	
10:37 AM	1015	585	725	Filament off
11:00 AM	880	490	632	Filament on; steady state
11:30 AM	1045	632	838	Steady state
12:00 Noon	1062	638	835	Filament off
1:40 PM	900	525	705	T ₁ dropped suddenly, then recovered
1:45 PM	725	535	720	
1:50 PM	970	545	740	
2:40 PM	972	575	760	Stabilized
3:30 PM	955	555	725	Stabilized
3:31 PM	508	572	705	Helium leak check
3:40 PM	688	542	710	
4:30 PM	718	510	665	Stabilized

During the braze run, an arc occurred from the braze filament to the generator case. This caused the braze power supply (high voltage) to automatically shutdown. However, damage to the load resulted, which changed the operating characteristics of the load transistors (collector-to-emitter voltage of the transistor was exceeded). It should be noted that complete failure of the load would result in short circuit operation of the generator, i.e., the load fails safe. At this point, the cesium tip temperature showed a step increase of 510° F (520° to 1030° F, see Fig. 7) indicating that the thermocouple had either suffered damage by the arc or was reflecting a stray voltage pickup. However, after five hours of operation, the T₁ reading was reduced to 740° F with no effective change in thermal input to the cesium reservoir. The generator output remained at 15 watts electrical; this was achieved at 37 amperes which indicated that the emitter temperature was reduced by additional electron

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FIG. 7. SNAP 13 ISOTOPIC GENERATOR OPERATION

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cooling. Further attempts to complete the casing braze at this time were discontinued to allow the generator to outgas further during the next few days, at which time another braze attempt could be made.

H. HELIUM LEAK CHECK

A helium leak check was performed to determine if the casing braze was leak-tight. The generator had stabilized at $T_1 = 955^\circ \text{ F}$, $T_2 = 555^\circ \text{ F}$ and $T_3 = 725^\circ \text{ F}$ and at an output of 0.246 volt and 37 amperes with an external vacuum of 5×10^{-6} torr and an internal vacuum of 2×10^{-7} torr.

The diffusion pumping system was isolated from the Vac-Ion pump-generator system, and a short burst (valve open approximately 0.3 second) of helium was admitted to the bell jar. The Vac-Ion pump tripped immediately, indicating a helium leak. The system was immediately pumped out and in 50 minutes had stabilized as shown in Table 12 and in Fig. 8.

TABLE 12

Generator Parameters Before,
During and After Helium Leak Check

Time of Day, PM	Temperature ($^\circ \text{ F}$)			Output	
	T_1	T_2	T_3	(volt)	(amp)
3:29	955	555	725	0.246	37
3:30	Helium admitted, leak indicated				
3:31	508	572	705		
3:40	635	542	710		
4:30	718	510	665	0.250	43

I. GENERATOR OPERATION

Normal operation of the generator was realized from 4:00 PM, November 28, 1965, to 1:05 AM, November 30, 1965, at which time a glow-type flash was observed. Concurrently, the fueling fixture pressure increased rapidly from 10^{-6} to 10^{-4} torr and then recovered immediately. The cesium tip temperature (T_1) decreased over a 15-minute period from a steady-state temperature of 770° to 580° F as shown in Fig. 9.

The generator output current indicated full scale on the 50-ampere range for a short period, but generator output voltage was not observed. The cesium heater power input was unchanged at the next data taking point, while the cesium temperature leveled off at 565° F . Subsequent normal cesium heater operation was observed which indicated no difficulties with this heater. The casing heater and casing thermocouple indicated normal operation before, during and after the "flash".

Typical operating data are given in Table 13 and in Fig. 9, both before and after the "flash" occurred.

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FIG. 8. SNAP 13 ISOTOPIC GENERATOR OPERATION

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FIG. 9. SNAP 13 ISOTOPIC GENERATOR OPERATION

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TABLE 13

Generator Parameters Prior to
and Following the Failure Incident

Time of Day		Temperature (° F)			Output	
		T ₁	T ₂	T ₃	(volt)	(amp)
4:00 PM	11/28	740	520	680	0.250	36.8
	to	to	to	to	to	to
1:03 AM	11/30	780	530	690	0.343	43.0
1:04 AM		770	530	684	0.343	36.7 ^a
1:05 AM ^b		832	532	690		
1:06 AM		738	534	700		
1:07 AM		678	538	700		
1:08 AM		640	539	700		
1:09 AM		612	540	700		
1:10 AM		598	540	698		
1:11 AM		585	540	694		
1:12 AM		580	539	690		
1:15 AM		565	528	688		
1:30 AM					0.07	9.4

^aAt 12:30 AM

^bFlash occurred

Remedial action recommended by MMC to increase the cesium temperature was initiated by increasing the heat input to the casing heater, cutting off the generator support stand cooling water and heating the casing with the EB filament. At 8:00 AM, a cesium temperature of T₁ = 670° F was achieved and an output of 16.5 amperes and 0.105 volt was realized. It was feared that the capsule would overheat at this output with subsequent damage to the capsule and diode and the generator was flooded with helium at 8:10 AM, November 30. An attempt to operate the generator at 1:35 PM on November 30 resulted in a maximum output of 0.095 volt and 15.3 amperes with T₁ = 660° F, T₂ = 700° F and T₃ = 845° F. Figure 10 presents a temperature history during this operation. The generator de-ignited at 3:00 PM and was flooded with helium from November 30, 1965 at 4:00 PM to 7:05 AM, December 1, 1965. The system was evacuated and heated for a final attempt at re-ignition on December 1, 1965. No open circuit voltage was obtained with maximum temperatures of T₁ = 650° F, T₂ = 772° F and T₃ = 892° F at 4:45 PM. The generator was flooded with helium at 4:50 PM on December 1 and purged overnight. The temperature history for this final attempt to operate Generator 14 is shown in Fig. 11.

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FIG. 10. SNAP 13 ISOTOPIC GENERATOR OPERATION

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FIG. 11. SNAP 13 ISOTOPIC GENERATOR OPERATION

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V. POST-OPERATIONAL INSPECTION

A. GENERATOR SYSTEM DISASSEMBLY

At 9:15 AM on December 2, 1965, the bell jar was evacuated and the casing heater was turned on to permit removal of the generator cover. Prior to cover removal, a visual inspection from inside the cell was conducted to determine if a short was external to the generator. Contamination surveys revealed no surface contamination on the outside surfaces of the bell jar or on horizontal surfaces in the cell. Radiation readings of 700 mr/hr beta-gamma and 300 mr/hr neutrons at one-foot from the bell jar were obtained. The inspection was conducted by ORNL personnel and consisted of looking inside the bell jar for evidence of shorting at the output leads by loose wires or by absence of the shorting bar. No evidence of shorting was observed. Next, the leads external to the bell jar were inspected; again no evidence of shorting was observed. Finally, the base plate of the fixture was inspected for evidence of cesium. Some irregular white beads and specks were scattered at random on the fixture base and could not be positively identified from the visual inspection. It was speculated that the material might be cesium oxide, Sauereisen or chips of ceramic insulation.

The emitter lead was disconnected from the load and a reading taken directly across the generator output leads. No voltage was recorded, indicating a direct short. The generator was then checked for continuity and read 0.75 ohm with the leads in one position and 1.1 ohms when reversed.

The emitter lead was reconnected and the visual inspection of the fueling fixture and generator was complete. At 9:55 AM, the cover was removed from the generator base while the casing braze filament heated the casing braze. The temperatures recorded at this point were $T_1 = 585^\circ \text{ F}$, $T_2 = 795^\circ \text{ F}$ and $T_3 = 955^\circ \text{ F}$.

Since the cover was maintained under tension during heatup, at the moment the braze melted, the weight of the cover caused it to fall onto the base plate.

When the shield in the cover cleared the base, it was noted that the fuel capsule was not on the base but lodged inside the thermal shield. Inspection of the base showed that the emitter plate was still brazed to the capsule and that jagged edges of the emitter spacer remained (i.e., failure of the emitter spacer had occurred).

At 11:00 AM, the bell jar was backfilled with helium and allowed to purge until 12:50 PM at which time the bell jar was removed. An air sample from the cell containing the generator was taken from 10:35 to 10:45 AM on December 2, 1965. These read 15 dis/min alpha and 50 dis/min beta-gamma. Another sample taken from 11:25 AM until 12:20 PM, read 66 dis/min alpha and 260 dis/min beta-gamma. The air sampler was started again at 12:20 PM. At 1:20 PM, the air sampler filter was checked with an alpha counter and was found to be highly alpha-contaminated.

The thumb screw holding the cover was tightened with the manipulator to keep the cover in position while the capsule recovery attempt was made. A tray was placed over the generator base to keep the capsule from falling more than 1/4 inch in the event it was dislodged. Attempts to dislodge the capsule by tapping on the cover and by pushing on the capsule base were unsuccessful. When the capsule base was pushed, it moved relative to the shield. When the bell jar was raised and the system exposed to air, black smoke was observed rising from the generator base. It was believed that this was cesium vapor.

During the attempts to remove the capsule, material fell from the space between the capsule and the thermal shield. This material appeared to be flaky to some ob-

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servers and fluid droplets to others. When the material struck the tray, it looked like a grey-white ash.

The attempts to remove the capsule involved a six-minute exposure to air. Since the efforts were unsuccessful, the bell jar was lowered and the system was evacuated. An extremely high (off scale) alpha contamination level was recorded on the cell floor after these efforts.

The high alpha contamination of the air inside the cell during the attempts to remove the capsule from the generator cover indicated leakage of Cm-242 from the capsule; therefore, it was decided to transfer the capsule to the Source Fabrication Facility (SFF) for further operations.

The generator cover and capsule were removed from the generator cell in a specially made chill block and stainless steel container and transferred to the SFF. The bell jar was replaced on the fueling fixture and the system was evacuated.

The generator top was held in a machinist's vise and several attempts were made to insert a knife between the tantalum heat shields and the fuel capsule. The capsule could not be pried free, and several taps on the generator top with a hammer failed to dislodge the capsule. Finally, the screws holding the heat shield assembly to the generator top were removed, and the capsule was pushed out of the heat shields at 6:00 PM, December 3, 1965. The capsule was visually inspected for leaks and cracks in the wall or weld, but none was apparent. The outside edge of the braze appeared sound with a good fillet and there was no evidence of capsule-to-emitter plate misalignment. Figures 12 and 13 are photographs of the capsule taken through the cell window. The capsule was placed in a tungsten chill block under a bell jar and purged continuously with argon.

The tantalum heat shield assembly by this time was a mass of tantalum oxide powder and small pieces of metal. A calorimetric check of this material revealed the presence of 52 watts of Cm-242. A calorimetric measurement of the fuel capsule indicated 83 watts were present inside the capsule.

An inspection by means of a mirror of the generator base remaining in the fueling fixture under vacuum revealed a glowing red ring around the collector plate. Because of the evident accumulation of a substantial quantity of Cm-242 in the generator base, it was decided to also transfer the base to SFF for further operations.

Temperature readings of the generator under vacuum were taken with the cooling water shut off and were $T_1 = 300^\circ \text{ F}$, $T_2 = 330^\circ \text{ F}$ and $T_3 = 380^\circ \text{ F}$. At 9:00 AM, December 8, 1965, the vacuum system was shut down, and the bell jar pressure was raised to atmospheric pressure using helium. The bell jar was raised and a temporary cap was placed over the generator base. The casing braze element was removed, the strap was taken off the cesium reservoir, and all leads were disconnected. A lifting ring was attached to the generator base, and the base was removed from the fueling fixture and sealed within two concentric stainless steel containers for transfer to the SFF. This operation took 3 hours and 40 minutes to complete.

Close inspection of the diode showed the following:

- (1) The casing braze had melted ~25% around the periphery
- (2) Approximately 50% of the emitter support plate was covered with a dark material to a thickness of ~1/16 inch. This was removed on December 17, 1965, and found to contain 45 watts of Cm-242 (see Fig. 14)

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FIG. 12. CAPSULE IN CHILL BLOCK AFTER REMOVAL FROM GENERATOR TOP SHOWING BRAZED-ON EMITTER PLATE

FIG. 13. CAPSULE IN CHILL BLOCK AFTER REMOVAL FROM GENERATOR TOP

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FIG. 14. LOWER PART OF GENERATOR SHOWING HALF-TORUS OF LEAKED OUT FUEL AROUND DIODE

FIG. 15. CAPSULE BASE AFTER REMOVAL OF EMITTER PLATE

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- (3) The ridge of the emitter support base was clean and shiny.
- (4) A light grey deposit was evident on parts of the interior wall of the generator.

The temporary thermal shield used during the capsule brazing operation was inspected for evidence of fuel, etc. There was no evidence of fuel or oxidation. However, a uniform light grey-white deposit was present on the interior walls which were adjacent to the capsule.

B. COMPONENT DISASSEMBLY AND INSPECTION

Following the system disassembly, additional inspection of the components was performed. These are detailed in the following.

1. Fueling Fixture

- (1) The transistorized load was removed from the fueling fixture and tested for electrical continuity. The test, performed by Martin Marietta personnel, revealed that the load was still in operating condition, but that the V_{ce} of one of the four transistors was variable from 0 to <0.5 volt dc rather than 0 to 3.5 volts. This indicated that the collector-to-emitter junction of the transistor was damaged. This was probably the result of the high voltage arc which occurred during the generator cover brazing attempt.
- (2) A sample scraped from deposits on the inner surface of the temporary thermal shield component used during the capsule-to-emitter braze was spectrographically analyzed and only tantalum was found to be present.
- (3) The white powdery particles which fell from the interior of the upper portion of the generator during its removal from the fueling fixture were spectrographically analyzed. The analytical results are tabulated. This material contained approximately 40 milligrams of Cm-242 (5 watts).

<u>Element</u>	<u>Occurrence (gm/gm Cm-242)</u>
Aluminum	0.056
Calcium	0.070
Chromium	0.021
Cesium	0.131
Copper	0.021
Iron	0.021
Potassium	0.035
Lithium	0.003
Magnesium	0.007
Sodium	0.139
Nickel	0.007
Platinum	0.348
Silicon	0.056

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- (4) Material deposits located on the base of the fueling fixture were collected by scraping and a sample was spectrographically analyzed. The results are tabulated. This material contained approximately 40 milligrams of Cm-242 (5 watts).

<u>Element</u>	<u>Occurrence (gm/gm Cm-242)</u>
Aluminum	0.755
Calcium	0.121
Chromium	0.151
Cesium	<0.050
Copper	0.352
Iron	0.040
Potassium	0.302
Lithium	<0.005
Magnesium	0.003
Sodium	1.007
Nickel	0.071
Platinum	0.202
Silicon	0.081

- (5) The interior of the Vac-Ion pump could not be checked for curium. However, gamma radiation readings taken at various points in the cell were consistently higher in the vicinity of the pump. These readings indicate that some curium with the residual gamma emitting fission products was in the pump but no quantitative determination was possible.
- (6) The multipoint temperature recorder was recalibrated using a millivolt input to the recorder corresponding to the expected thermocouple output at even hundred degree readings from 100° to 1900° F. All points on the recorder were found to indicate correctly within 5° F.
- (7) The cesium heater control system was checked and found to be operable.

2. Thermionic Generator (No. 14)

- (1) The continuity of the electrical circuit associated with the thermistor located on the cesium reservoir was established by determining its resistance to be 5000 ohms.
- (2) Upon inspection of the internal surface of the lower generator casing, an apparent material deposit was observed. On closer inspection, this material deposit was found to be an apparent discoloration of the surface. Additional analysis was not performed.
- (3) A leak check of the cesium reservoir and lower casing assembly by the helium leak detector technique revealed no detectable leaks.
- (4) A leak check of the diode assembly after removal of the cesium reservoir tubulation revealed no leaks.

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3. Fuel Capsule

- (1) The capsule was removed from the emitter plate relatively easily by prying it apart (see Figs. 15 and 16). The capsule tip did not break off.
- (2) The capsule was cut across the diameter in three places to permit removal of the fuel. The pellet was easily removed from the bottom of the capsule, indicating that no bond was present.
- (3) Visual examination of the capsule under low power magnification (5X) did not reveal any cracks in the capsule walls or in the weld region. A representative view of the capsule is shown in Fig. 12. There was no evidence of material which may have seeped through a crack and solidified on the walls of the capsule. As illustrated in Fig. 17, foreign material was found adhering to one region of the capsule wall about one inch below the weld. A cross section through the material is shown in Fig. 18; X-ray diffraction analysis indicates the presence of tantalum in this material.
- (4) Measurements of the weld overhang revealed that it was in one region and 0.001 inch beyond the allowable 0.020 inch. No evidence was found to indicate that the weld overhang remained in contact with the heat shield although it probably contributed to the interference problem encountered during the cover lowering operation.
- (5) A bubble leak test prior to sectioning the capsule indicated a possible defect in the wall 1-1/2 inches from the bottom. The section cut from this region cracked while removing the fuel material, and further metallographic examination of this particular section could not be made.
- (6) The typical appearance of the weld bead at the joint between the cap and body of the capsule is shown in Fig. 19. A Zygo inspection of the weld revealed no cracks and microscopic examination revealed full penetration of the weldments. As shown in Fig. 20, an intergranular foreign phase was found in the weldment. The dark chunk of material embedded in the crevice of the joint was identified as alpha-emitting material by an autoradiograph.
- (7) The metallographic examination revealed gross attack on the interior of the W-2%Mo capsule. Figure 21 illustrates the intergranular nature of the attack with penetration to a depth equivalent to 50% of the wall thickness. A macrophotograph (Fig. 22) shows the thinning of the capsule walls in a localized region. A view of two sections of the fuel mixture provided by a transverse cut through the pellet is shown in Fig. 23. The cavity visible is typical of shrinkage cavities frequently associated with solidifying masses. There was no evidence of platinum in the capsule samples examined metallographically.
- (8) The platinum braze between the capsule and emitter plate achieved ~60 to 70% coverage based on an examination of available photographs and experience with previous braze samples. The remaining area was void space evenly distributed at the interface. The outline of the disc was quite evident and did not cover the full diameter of the capsule. A shadow (probably material plating out during the braze cycle) formed almost out to the edge of the emitter plate. There was no evidence of braze material running over the edge onto the emitter spacer. Figure 24 is a macrophotograph of the surface of the emitter which had been attached to the capsule. The appearance is typical of brazes realized during the braze development program and ascertained on generators which performed properly.

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FIG. 16. EMITTER PLATE

[0.8 inch]

FIG. 17. FOREIGN DEPOSIT ON SIDE OF SNAP 13 CAPSULE

FIG. 18. TRANSVERSE SECTION THROUGH CAPSULE SHOWING PORTION OF TANTALUM HEAT SHIELD ADHERING TO CAPSULE WALL

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b. Section of termination portion of weldment (6X)

FIG. 19. VIEWS OF WELDED JOINT AFTER CLEANING OPERATIONS

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FIG. 20. PHOTOMICROGRAPH OF WELDED JOINT SHOWING INTERGRANULAR ENTRAINMENT OF FOREIGN PHASE AND REDISTRIBUTED OXIDE FUEL

FIG. 21. MICROSTRUCTURAL DAMAGE TO THE INSIDE WALL OF THE SNAP 13 CONTAINER

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[——— 0.7 inch ———]

FIG. 22. LONGITUDINAL VIEW OF TOP SECTION OF SNAP 13 CAPSULE SHOWING ATTACK ON INTERIOR WALLS

FIG. 23. SURFACE APPEARANCE OF FUEL MIXTURE IN SECTIONS Cm-2 AND Cm-3 CUT FROM SNAP 13 CAPSULE

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FIG. 24. SURFACE OF EMITTER SHOWING PLATINUM BRAZEMENT

[0.05 inch]

FIG. 25. INTERGRANULAR FISSURE IN WALL OF SNAP 13 CAPSULE

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- (9) An intergranular fissure penetrating 80% of the capsule wall was found ~0.1 inch above the top of the pellet as shown in Fig. 25.

4. Curium Fuel

- (1) The melting point of the fuel mixture remaining in the capsule was determined to be essentially the same as that of iridium. Attempts to analyze samples for the chemical constituents of the fuel mixture remaining in the capsule proved unsuccessful due to the difficulties encountered in dissolution and handling of the material.
- (2) The fuel material which leaked from the capsule was analyzed spectrographically and the results are tabulated here. This material contained about 0.38 gram of Cm-242 (45 watts). The melting point of this material was determined to be 1220° C.

<u>Element</u>	<u>Occurrence (gm/gm Cm-242)</u>
Aluminum	0.128
Calcium	0.026
Chromium	0.128
Cesium	0.110
Copper	0.029
Iron	0.292
Potassium	0.018
Lithium	0.002
Magnesium	0.029
Sodium	0.066
Nickel	0.029
Platinum	--
Silicon	0.011

- (3) The generator thermal shield remnants were spectrographically analyzed with the results tabulated. This material contained approximately 0.45 gram of Cm-242 (55 watts).

<u>Element</u>	<u>Occurrence (gm/gm Cm-242)</u>
Aluminum	17.67
Calcium	41.20
Chromium	17.67
Cesium	29.45
Copper	11.78
Iron	--
Potassium	29.45

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<u>Element</u>	<u>Occurrence (gm/gm Cm-242)</u>
Lithium	2.945
Magnesium	17.67
Sodium	117.80
Nickel	17.67
Platinum	--
Silicon	--

- (4) When the capsule was cut open, the fuel pellet was found to be about 3/8 inch longer than its original length and was not brazed to the capsule base. A meniscus-shaped deposit of material adhering to the interior capsule wall immediately above the top of the pellet was observed (Fig. 26).
- (5) The top of the fuel pellet remaining in the capsule presented an appearance similar to that of solidified lava (Fig. 26).
- (6) No evidence of the platinum pellet cover was observed.
- (7) The distribution of the remaining Cm-242 fuel was as follows:

	<u>(watts)</u>
Generator base	45
Remaining in capsule	83
Thermal shields	50 to 55
Thermal shield drippings	5
Base of bell jar	1 to 5
Total	~190

Approximately one-third of the original fuel loading is unaccounted for and is probably associated with the Vac-Ion pump and fueling fixture equipment. These wattage values were measured in December 1965.

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FIG. 26. CAPSULE WITH TOP SAWED OFF AND PELLET EXPOSED

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VI. DISCUSSION

A. FUEL FORM

1. Encapsulated Curium-242 Content

The quantity of Cm-242 in the pellet was determined by calorimetric measurement of the $(\text{Cm-242})_2\text{O}_3$ fuel prior to its placement in the pellet press die. No calorimetric measurement of the pressed pellet was made; however, the vessels, tools and die parts were checked for residual fuel after the pressing operation and were found to contain no significant amounts. Therefore, the value of 290 watts should be considered the encapsulated fuel content.

2. Ionic Contaminants

The analysis of the Cm-242 product for ionic contaminants was performed on samples of the product solution immediately prior to precipitation of curium as the oxalate and calcination to the oxide (analysis of the oxide has proved unreliable due to difficulties in handling the intensely alpha-active material and obtaining a quantitative dissolution of the oxide without introducing additional contamination from the containment vessel). Based on this analysis (see Table 5), all ionic contaminants were within the limitations imposed by the volumetric specifications. The possibility of subsequent contamination of the curium oxide by mechanical or chemical degradation of the stainless steel filter cannot be completely discounted. Even though contamination due to filter material was not observed in test runs, the combined effects of the intense radiation and heat may have caused accelerated attack of the filter.

3. Pellet Volume

The observed pellet volume was approximately twice the expected volume (see Tables 6 and 7). The discussion of possible causes of this additional volume is presented below.

a. Possible incomplete reduction of IrO_2

Prior to the hot pressing operation, the iridium was present in the fuel mixture as IrO_2 . After the fuel mixture was loaded into the die in a platinum envelope, the temperature was raised to 1050°C which was maintained for 2.2 hours without application of die pressure. Heating of IrO_2 at 1050°C for 2 hours in air has been demonstrated to provide complete decomposition to the metal. In addition, the interior of the pellet was well above the temperature indicated by the thermocouple and a flow of helium was maintained through the die body; therefore, the iridium probably existed as the metal in the hot-pressed pellet. The iridium metal was protected from oxidation after the hot pressing operation by the platinum jacket.

b. Possible presence of undetected contaminants

As discussed in 2. "Ionic Contaminants," the possibility of contamination of the curium oxide fuel mixture by the stainless steel filter cannot be completely discounted. However, the assumption of a volume of contaminant from this source equal to the volume of the fuel mixture is probably incorrect.

c. Possible low pellet density

If it is assumed that no contaminants were present in the fuel mixture, the observed volume of the pellet would indicate that its effective density was 44.6% of theoretical.

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Possible contributing conditions to attainment of low pellet density could have been:

- (1) Holding the pellet at 3500 psi and 1450° C for insufficient time
- (2) Erroneous indication of temperature and/or pressure
- (3) Sticking of the die plunger so that full pressure was not transmitted to the pellet.

Based on similar hot pressing operations, holding the required pressure for 1 hour and 25 minutes instead of the 2-hour period called for in the procedure would have allowed attainment of much greater than the observed density. Sticking of the die plunger due to low-melting material melting and running from the pellet has been observed in other hot pressing operations.

Of the three possible contributors to the high pellet volume discussed above, only the incomplete reduction of IrO_2 to iridium metal can be eliminated. The attainment of low density during hot pressing is the most likely cause, but the possibility of some volume taken up by undetected contaminants (including excessive iridium) must also be considered.

4. Melting Point

The melting point of a mixture of actinide oxides [$(\text{Cm-244})_2\text{O}_3$, $(\text{Am-241})\text{O}_2$ and $(\text{Pu-239})\text{O}_2$] and 20% ThO_2 simulating the SNAP 13 concentrations was determined to be greater than 2250° C prior to the SNAP 13 fueling. Experiments conducted following SNAP 13 fueling with a mixture of $(\text{Cm-244})_2\text{O}_3$, $(\text{Am-241})\text{O}_2$ and $(\text{Pu-239})\text{O}_2$ contaminated with 7.5 mol % CaO (the concentration shown in the analysis of SNAP 13 product solutions) showed that the melting point would be lowered by 100° to 200° C. Even the presence of 50% CaO (the eutectic mixture) would have lowered the melting point a maximum of 300° C. Based on these observations, the melting point of the SNAP 13 fuel mixture (without contaminants) should have been ~ 2000° C. Subsequent to the SNAP 13 experiment, the melting point of the SNAP 11 $(\text{Cm-242})_2\text{O}_3$ fuel (with no ThO_2 added) was determined to be ~ 1600° C. The addition of ThO_2 to this fuel mixture would probably raise the melting point, but the amount is unpredictable due to the unknown effect of contaminants. Based on the above observations, it is estimated that the melting point of the SNAP 13 fuel mixture was in the range of 1600° to 2000° C, 650° to 250° C below the melting point given in the fuel specification.

Following the termination of the SNAP 13 experiment, the melting point of the fuel which had leaked into the generator base was determined to be 1220° C. This fuel cannot be considered representative of the mixture in the capsule because of known contamination with aluminum, cesium and iron which probably occurred after the fuel was released from the capsule. The melting point of the pellet remaining in the capsule was determined to be essentially that of iridium (2450° C).

5. Fuel Temperature

The greater-than-anticipated fuel pellet length results in a maximum fuel temperature of 2060° C (see Fig. 27) during the capsule-to-emitter brazing operation based on a fuel form thermal conductivity of 0.35 watt/cm-°K (i.e., if the highest anticipated fuel form thermal conductivity were achieved). If the increased pellet length was due only to the achievement of a very low theoretical density, a severe reduction in the thermal conductivity may be expected, which could result in a maximum conductivity value of 0.15 watt/cm-°K and a peak fuel temperature of 2380° C. Both of these temperature levels exceed the estimated melting point of the fuel.

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$\Delta T_{\max}$ assumes 30% contact after brazing

Fuel inventory: 290 watts (t)

T_s = temperature of emitter backup plate $\geq 1550^\circ \text{C}$ during normal operation
= 1800°C during brazing

FIG. 27. FUEL PELLETT TEMPERATURE

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Even if the maximum thermal conductivity were achieved, the fact that the pellet length was approximately twice the expected length would alone account for an increase in fuel temperature, since the heat would have to travel through a longer path to heat the diode (or to be dissipated). It appears reasonable to conclude, therefore, that melting of the fuel form did occur during the capsule-to-emitter brazing operations (brazing temperature of 1800° C).

6. Compatibility

A compatibility test, using Cm-244, was run at 1650° C for 21 days; the description of the material used in this test is tabulated below.

<u>Sample</u>	<u>Fuel Composition</u>	<u>Source Form</u>	<u>Braze</u>	<u>Capsule Material</u>
1	80 wt % Cm_2O_3 - 20 wt % ThO_2	Oxide only	None	W-2% Mo
2	Same	30 vol % fuel - 70 vol % Ir	10-mil Pt	W-2% Mo

The test was terminated after 21 days because of the failure of the brazed closure. The results of the test are not conclusive, due to the possible effects of the braze material, but some pertinent observations were made. In Sample 1 a uniform penetration zone to a depth of a few mils was seen, but no catastrophic attack on the capsule wall occurred. In Sample 2, which contained a cermet pellet brazed to the capsule with platinum, the platinum had completely diffused into the capsule wall, and preferential local attack of the capsule wall immediately above the liquid surface had occurred.

In other tests, W-2% Mo was exposed to molten platinum at 2200° C for periods of 1, 4 and 16 hours. The molten platinum was found to dissolve the W-2% Mo with a preferential attack to a depth of 35 to 45 mils just above the liquid surface in all cases.

The above studies indicate that a pure Cm_2O_3 - ThO_2 fuel form would be compatible with the W-2% Mo and that molten platinum would attack the capsule wall within the duration of the SNAP 13 test. The metallographic examination of the SNAP 13 capsule following the test indicates attack by a liquid rather than a solid-state diffusion. The extent of the removal of the capsule wall also indicates a lengthy exposure to molten material, possibly for the entire duration of the test.

B. CAPSULE

1. Material

The W-2% Mo alloy was chosen for the SNAP 13 capsule material because of its refractory properties and its high-temperature strength. Possible disadvantages of this alloy are its large grain structure, particularly in the cast form, and its susceptibility to propagation of surface cracks under stress. First generation SNAP 13 capsules, made of cast alloy, exhibited such large grains that only a few crystals spanned the wall thickness. The capsules used in the SNAP 13 test were fabricated from wrought alloy to reduce crystal sizes and were polished by honing to reduce the possibility of stress-propagated cracks. However, surface cracks were found to develop while the capsules were stored for approximately one month after fabrication.

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2. Possible Fuel Leakage Routes

The helium leak detector test of the welded capsule indicated a leak rate of 10^{-7} cm³/sec. Experimental determinations of leak rates from calibrated orifices show that a fissure with a diameter of 0.02 mm would release helium at the observed rate. The same helium release rate was observed when a test of the unsealed capsule prior to welding was made; therefore, there is a possibility that a fissure larger than 0.02 mm in diameter existed after welding and that the helium release rate observed was that characteristic of the pellet at that time.

The capsule weld was leak tested during the post-test inspection and found to be leak free. Metallographic examination of the weldment indicated minute amounts of curium oxide contamination but no leakage paths. A single leak point through the capsule wall was detected by a bubble test during the post-test examination, but that area of the capsule was cracked during removal of the fuel pellet and could not be examined further. Metallographic examination of the sectioned capsule revealed an intergranular fissure extending through most of the wall. This observation presents the possibility that one or more fissures may have developed and extended through the capsule wall during the thermal cycles imposed during welding and brazing.

3. Capsule Corrosion

During the loading of the fuel pellet and placement of the capsule cover, there was a possibility that both the fuel and capsule suffered some degree of oxidation. Oxidation also occurred after welding during the smear and go-no go tests and the visual inspection.

Another factor of importance is the effect of alumina on the capsule. If alumina were present on the pellet surface, there is the possibility of its melting during the weld (or subsequent braze) cycle. If so, the alumina would react with the W-2 Mo capsule. (This occurred with several of the MMC pressure-tested capsules and weakened the wall by attack at the grain boundary, see Ref. 1.)

C. CAPSULE-TO-EMITTER BRAZE OPERATION

1. Possible Cause of High Bell Jar Pressures

During the capsule-to-emitter braze attempts, gassing within the bell jar prevented attainment of the desired vacuum level. Five attempts over a period of 2 hours and 20 minutes were required to accomplish the braze. The possible causes of the observed gassing are discussed below.

a. Oxide coating on the capsule

A thin oxide coating was formed on the capsule while exposed to the air for a few minutes during inspection and transfer to the bell jar. It is doubtful that this layer of oxide contributed to the gassing, because tungsten oxide is stable up to 1750° to 1900° C. The tungsten oxide would vaporize from the heated surfaces of the capsule at ~1300° C without decomposition and deposit on cooler adjacent surfaces.

b. Loss of fuel from the capsule

The possibility of loss of fuel from the capsule during various phases of the test is discussed below in Sections E and F of this chapter. If the possibility of fuel release during the capsule brazing operation is assumed, the release of entrapped gases from the fuel would contribute markedly to the observed high bell jar pressures.

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c. Outgassing of capsule material

During the welding operation, the bottom part of the capsule was kept considerably cooler than the temperature achieved during brazing. The brazing temperature would cause additional outgassing and high bell jar pressures.

Based on this discussion, a definite conclusion regarding the sources of gases causing high bell jar pressures cannot be made; however, both outgassing of the capsule material and leakage of fuel from the capsule were possible contributors.

2. Possible Damage to Diode Spacer

Three possible causes of damage to the diode spacer during the brazing operations are as follows:

a. Oxidation during outgassing of the capsule

Such attack is possible; however, outgassing during test runs on prototype thermionic generators has not necessarily resulted in catastrophic attack on the generator spacer.

b. Thermal shock to the diode spacer during the five high-heat braze cycles

Weakening of diode structural brazes could possibly have occurred.

c. Attack by fuel which leaked from capsule

It cannot be definitely established that fuel which may have leaked from the capsule during the brazing operation remained in contact with the diode spacer during the operation of the generator. The observed deposit of fuel, other than in the thermal shield, was in the ring below the diode spacer and was not in contact with the spacer. It should also be noted that embrittlement of the diode spacer by contaminants in the atmosphere due to the presence of fuel could have occurred.

3. Possible Damage to Capsule

The five braze attempts could have propagated intergranular fissures in the capsule wall due to the repeated heating and cooling. Peak braze power levels on two cycles were 3.4 and 3.57 kw, compared to the specified input of 3.34 kw. The peak temperatures were not known during the heating cycles, but with the added heating from the concentrated Cm-242 source, the temperature of the inside capsule wall could have greatly exceeded the melting point of platinum, 1770° C, and was possibly > 2000° C. Evidence found during the post-test examination shows that the platinum covering of the pellet was completely melted away. Corrosive attack of the capsule wall by the molten platinum (and possibly molten fuel) could have occurred during the extremely high temperatures experienced over the 2 hour and 20 minute brazing period.

4. Capsule Braze Efficiency

The platinum braze between the emitter plate and the capsule achieved ~ 60 to 70% coverage. The braze was brittle, as evidenced by the relative ease of prying the emitter plate from the capsule during the post-test examination. How much force was required is not pertinent to the degree or type of bond achieved, since the bond itself is quite brittle. Bonds achieved with thermal mockup diodes were usually brittle after the braze was made. No bonds were strength tested after aging for 45 hours (the time the generator operated) at ~ 1500° C, but it is believed that the bond

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could have become further embrittled. Hansen (Ref. 3) indicated that an intermetallic ordered (brittle) phase occurs between platinum and tungsten. This is where the separation occurred, as would be expected, and not between the platinum and the tantalum.

D. CASING COVER LOWERING AND BRAZING

1. Interference of Thermal Shield with Capsule

Trouble was experienced during the lowering of the thermal shield over the capsule, so that completion of this operation was achieved only after joggling the shield up and down and lowering in steps. The weld bead passed the go-no go gage test, but the post-test examination revealed the overhang to be 0.021 inch (versus 0.020 inch allowable) and to be mainly on one side of the capsule. Also, a small vertical band of foreign material on the side of the capsule following disassembly of the generator gave an X-ray diffraction analysis indicating tantalum, which was the thermal shield material. Thus, the thermal shield was probably pulled to one side by the weld overhang and in conjunction with fuel leakage resulted in bonding of the thermal shield material to the capsule.

2. Outgassing During Brazing of the Cover

The possible causes of outgassing during the cover brazing operation would be similar to those discussed in Section C of this chapter. Added sources of gas would be the generator casing sections, surrounding parts of the fueling fixture and the shaft seals in the bottom of the fueling fixture.

3. Possible Damage to System During Cover Lowering and Brazing

During the cover lowering and brazing operation, damage to the generator, controls or capsule could have occurred by:

- (1) Oxidation of the diode spacer during the outgassing periods
- (2) Possible shorting of the cesium tip thermocouple, T_1 , when the high-voltage arc from the filament to the generator case occurred
- (3) Damage to the load transistors when the high-voltage arc occurred.

E. GENERATOR OPERATION

Prior to the casing braze attempts, the ignition power supply was used to ignite the generator, and an output of 0.500 volt and 30 amperes was obtained. During subsequent optimization procedures, a maximum of 17 watts electrical output was achieved (0.500 volt, 34 amperes) compared to an expected 30 watts (0.500 volt, 60 amperes) with a 290-watt thermal loading in the capsule.

The generator operated at ~ 50% of anticipated output for ~ 45 hours, until a flash was observed in the cell and the output of the generator changed drastically. After ~ 7 hours of nonstandard operation, the generator was flooded with helium, because of the possibility of overheating the capsule and diode due to the low power output.

The possible causes of low power output during the 45 hours of normal operation are: loss of part of the fuel from the capsule during the brazing operation; loss of

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part of the thermal output through the thermal shield due to high temperature capsule operation and/or partial loss of insulating capability of the thermal shield; and inefficient operation of the generator-transistorized load system due to damage sustained during brazing operations. These possible causes are discussed further below.

1. Loss of Fuel During Brazing

As discussed in Section B of this chapter, a fissure with a diameter of ~ 0.02 mm was indicated by the helium leak detector test of the welded capsule. However, a single fissure of this size would have allowed only microgram quantities of the actinide oxide to escape by vapor transfer during the brazing period. In fact, as shown below, calculations indicate that a fissure with a diameter > 5.0 mm would have to be available for vapor transfer of $\sim 40\%$ of the actinide oxides during the time required for the capsule and casing brazes.

Recent work at Savannah River Laboratory (January 1967) shows that the vapor pressure of Cm_2O_3 lies between 5×10^{-6} and 1.5×10^{-5} atmospheres at 1800°C . The higher vapor pressure can be used to calculate the open surface evaporation rate for Cm_2O_3 into vacuum at 1800°C to be $1.1 \text{ gm/cm}^2 \text{ hr}$. (It should be noted that the presence of an inert gas would greatly decrease this rate.) The calculated rates of diffusion of Cm_2O_3 through small circular holes at 1800°C into a vacuum are tabulated below. Certain simplifying assumptions were made which could cause these calculated rates to be somewhat higher than actual values.

<u>Hole Diameter (mm)</u>	<u>Rate of Diffusion (gm/hr)</u>
0.01	8.9×10^{-7}
0.05	2.2×10^{-5}
0.1	8.9×10^{-5}
0.5	2.2×10^{-3}
1.0	8.9×10^{-3}
2.0	3.5×10^{-2}
5.0	2.2×10^{-1}

If transfer of the Cm_2O_3 from the capsule as a vapor is assumed, the 0.02-mm diameter fissure indicated by the $10^{-7} \text{ cm}^3/\text{sec}$ helium leak rate would pass Cm_2O_3 at a rate of only $3.5 \times 10^{-6} \text{ gm/hr}$. Since this rate is far too small to account for the transfer of $\sim 3.0 \text{ gm}$ of actinide oxide within the brazing period, a much larger fissure, probably greater than 5.0 mm diameter, would have to exist. An opening of this size would have been readily visible; however, the post-test examination of the capsule revealed no visible opening. Multiple fissures of smaller diameter could also be postulated; however, only one leak was detected during post-test examination.

Another possibility would be the ejection of molten fuel through the small fissure detected by the leak test. One can postulate the existence of a fluid fuel phase during the brazing operations with some certainty. If heat conduction only through the pellet to the emitter is assumed, a maximum fuel temperature of 2060°C would result during the capsule-to-emitter braze (Fig. 27) based on the observed pellet length

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and on a fuel form thermal conductivity of 0.35 watt/cm-°K (i.e., if the highest anticipated fuel form thermal conductivity was achieved). This calculation is based on an emitter temperature of 1800° C during brazing. Since higher power was supplied to the electron beam heater during two brazing attempts than was called for in the procedure, the emitter temperature probably exceeded 1800° C. In addition, the thermal conductivity of the pellet could be lower than 0.35 watt/cm-°K due to the low density of the pellet or the presence of undetected contaminants.

For 40% of the Cm-242 to be transferred from the capsule during the brazing operations, ~ 1 gm/hr of molten actinide oxide would have to flow through a small fissure under a pressure differential of ~ 15 psi. Although the viscosity of molten Cm₂O₃ has not been measured, the required viscosity for the transfer of 1 gm/hr through various size holes can be calculated and, by comparison, the credibility of leakage of molten fuel can be judged. The results of this calculation are given below.

<u>Fissure Diameter (mm)</u>	<u>Viscosity of Fluid if 1 gm/hr is Transferred Through 0.1-in. Long Fissure (centipoise)</u>
0.01	0.35
0.05	2.2
0.1	3.5 x 10 ³
0.5	2.2 x 10 ⁶
2.0	565.0 x 10 ⁶

By comparison, the viscosity of water at 20° C is one centipoise and that of a cold, heavy-grade oil would be in the 10⁵ to 10⁶ centipoise range. Since molten salts do not usually exhibit high viscosity, it is reasonable to assume that a small fissure, perhaps < 0.1 mm in diameter, existed and that a portion of the fuel was transferred through this fissure as a fluid during the period of high temperatures which existed during the capsule braze attempts.

2. Loss of Heat Through the Thermal Shield

The low power output of Generator 14 can be partially explained by the loss of heat through the thermal shield. The additional heat loss would be 68 watts(t), with 222 watts(t) available to produce an output of 16.9 watts(e) as determined in Appendix A. This appendix defines the capsule temperature required for dissipation of the 68 watts(t) additional through the thermal shield to be ~ 2000° C (350° to 400° C higher than the emitter temperature of 1600° C).

A calculation to determine the temperature gradient as a function of the percent of complete surface contact achieved by the braze indicates that, with only 10% contact, the temperature drop across the braze required to transfer all the isotope heat (290 watts) is less than 5° C. This, however, is only part of the increased temperature gradient resulting from increased surface contact resistance. There exists an increased temperature gradient in the base of the W-2% Mo fuel capsule as a result of distortion of the heat flow lines and the isotherms perpendicular thereto. This effect increases the thermal gradient across the fuel capsule base by 40° C. A similar, but slightly higher, increase in the emitter backup structure temperature would also be encountered, i.e., an increase of 53° C. Thus, the total effect is less than 100° C for an extremely poor braze providing only 10% contact. The selection of a 10% braze is conservative in view of the observed 60 to 70% effective brazed area observed in the post-test examination.

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It can, therefore, be seen that a fuel capsule temperature of $\sim 2000^{\circ}\text{C}$ during generator operation could not be achieved. In fact, based on the effective brazed area, the fuel capsule temperature was close to expected and of the order of 1700°C . It is concluded that very little of the unaccounted for 68 watts(t) was lost through the thermal shield. This analysis further substantiates the loss of fuel during the capsule-to-emitter brazing and cover lowering operations.

The calculations in Appendix A do not take into account the effects of contact of the thermal shield with the capsule and the possible distortion of the multilayered tantalum heat shield to provide high thermal conductivity paths. These effects were probably present, but their contribution to the heat leak cannot be calculated.

3. Inefficient Operation of Generator-Transistorized Load System

As noted in Section D of this chapter, damage to the transistorized load and cesium tip thermocouple occurred during the brazing operation. In addition, the diode system may have suffered damage due to the repeated thermal cycling and high temperatures. Inefficient operation of the generator-transistorized load system due to this damage may have contributed to the low electrical output.

4. Conclusions

In view of the above discussion, the cause of low electrical power output cannot be assigned to any one circumstance. Rather, it was probably a combination of the following:

- (1) Loss of fuel during the brazing cycles and cover lowering operation (most probably the primary cause)
- (2) Higher-than-normal temperature operation of the capsule due to the increased temperature of the fuel form (due to the greater length)
- (3) Increased heat conduction through the thermal shield due to contact with the capsule and distortion of the shield
- (4) Inefficient operation of the generator-transistorized load system.

It can be concluded that $\sim 70\%$ (210 watts, based on an estimated 8% efficiency) of the original fuel was present in the capsule during the 45 hours of operation, because 17 watts(e) output was obtained, compared to an expected 30 watts(e). No fuel was lost from the capsule during the operational period, as evidenced by the steady electrical power output.

F. ANALYSIS OF GENERATOR ABORT

The generator abort was characterized by the following events:

- (1) A flash of light from the generator system
- (2) A momentary increase in bell jar pressure
- (3) Increase to full-scale (50 amperes) generator output current reading from the steady-state value of 36.7 amperes, followed by a decline to 10 amperes over several minutes

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- (4) Reduction of generator output voltage to 0.07 volt from the steady-state value of 0.343 volt over several minutes
- (5) Active control of cesium heater input power
- (6) Reduction in load resistance
- (7) Reduction in output of cesium pinch-off thermocouple.

A post-test examination of system components was conducted to provide clues to the cause of the observed phenomena during and after the abort. It was generally found that the available evidence would give an indication of causes but would not permit definitive conclusions. A discussion of the pertinent events and probable causes is given below.

1. Light Flash

The technician on duty did not observe the exact origin of the flash within the manipulator cell because his attention was focused on the board at the time. An electrical arc as a result of a short in one of the electrical leads could have caused the flash; however, post-test examination revealed no evidence of an electrical short (see Section V-A). It is more likely that release of the cesium produced a momentary ionized atmosphere, resulting in a low-energy discharge across the cesium and/or casing heater leads. A similar failure mode was experienced with Generator 12A.

2. Loss of Fuel from Capsule

Immediately prior to the abort, more than 70% of the fuel loading was in the capsule, as evidenced by the electrical output. The post-test examination revealed that 29% (83 watts) of the original loading remained in the capsule. Therefore, at some time subsequent to the abort, at least 41% of the fuel loading was expelled from the capsule. The precise timing of this release cannot be established; however, the temperatures induced in the capsule by loss of electron cooling in the thermionic generator subsequent to the abort would have been quite high, probably ~ 1700° C. Melting and vaporization of the fuel would result, as during the brazing operations, and loss of molten fuel could have occurred in the six hours before the bell jar was flooded with helium.

The analyses of the released fuel are listed in Section B of Chapter V. The materials found in the released fuel are not indicative of their presence in the encapsulated fuel, due to contamination after release. For example, the cesium contamination undoubtedly came from the breached cesium reservoir.

3. Breaching of Cesium Reservoir

The post-test examination revealed the emitter sleeve to be badly oxidized. The fact that the fuel was molten and a leakage path existed for the release of the fuel to the base of the capsule would readily account for contact between the fuel and the tantalum alloy (T-111) diode spacer. Chemical compatibility has not been demonstrated between the molten fuel complex (Cm-242, Am-241 and Pu-238) and T-111 at 1500° C. At best, embrittlement of this 5-mil spacer should be expected, which would result in diode failure as a result of the high thermal stresses associated with this heat choke. Thus, the combination of embrittlement and chemical attack by the released fuel over a period of approximately 50 hours and the effects of the copious outgassing observed during the generator assembly operations could have resulted in the violation of the diode spacer and loss of the cesium. This, in turn, would account for the momentary increase in bell jar pressure. The same pressure rise

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was experienced with Generator 12A upon its failure after approximately 45 hours of operation. It was pointed out, in the case of Generator 12A, that the lowest vapor pressure that cesium would have would be that resulting from its operation at the effective fueling fixture temperature ($\sim 50^\circ \text{C}$). The vapor pressure at this temperature is only 1×10^{-5} mm Hg and is not in itself high enough to account for the sudden rise in bell jar pressure. However, it should be noted that the bell jar pressure was measured with a Bayard-Alpert ionization gage. The fact that the sensitivity of an ionization gage varies with the gas species present is well established (Refs. 4 and 5). It has been recently determined by Summers (Ref. 6) that the sensitivity of such a gage with cesium is 13.7 times the gage sensitivity for air. Thus, even the minimum vapor pressure of cesium, 1×10^{-5} mm Hg, would appear like a pressure of 13.7×10^{-5} mm Hg and result in the large pressure observed at the time of the diode failure.

4. Cesium Tip Thermocouple (T_1) Behavior

The generator abort was also characterized by an apparent rapid decrease in cesium temperature. This occurrence must be evaluated with respect to the data realized during the optimization of the generator (see Fig. 6) and that ascertained before, during and after the casing braze arc (Fig. 7 and Table 14).

TABLE 14
Cesium Tip, Cesium Reservoir and Casing Temperatures
at Various Times During Operation of Generator 14

<u>Operating Condition</u>	<u>Temperature ($^\circ\text{F}$)</u>			<u>Remarks</u>
	<u>T_1</u>	<u>T_2</u>	<u>T_3</u>	
Running normal (optimum)	545	590	805	(from TEECO data sheet)
Prior to casing braze	520	600	792	
After arc occurred	1020	605	795	
Before helium leak check	955	555	725	
Immediately after leak check	508	572	705	
Stabilized after leak check	718	510	665	
Just prior to failure incident	770	530	684	
At failure	832	532	690	
10 minutes after failure	565	532	680	
1 hour after failure	565	528	688	
7 hours after failure	670	705	845	
Startup attempt (11-30-65)	670	705	845	
Startup attempt (12-1-65)	575	710	840	

The optimum cesium temperature (as determined by T_1) was 550°F which was anticipated as a result of the prefueling performance operation of Generator 14. The input power to the cesium heater was also at the level specified by the prefueling tests. From Fig. 7 it is evident that Thermocouple T_1 was drastically influenced by the arc

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which occurred between the EB filament used for the casing braze and the generator casing (ground potential). Three alternatives exist to account for the step increase in the T_1 thermocouple output, i.e., apparent temperature reading. The first is damage or other chemical reaction to the thermocouple bead (e.g., by oxidation). Damage of this type, however, could not account for the extremely large change in output that was experienced and, in almost every case, a chemical change would result in a decrease in Seebeck voltage and not the large increase realized. The second alternative, that of the thermocouple picking up a spurious voltage, appears more probable, since an increase of only 11.7 millivolts is required to raise the temperature reading from 550° to 1050° F. This thermocouple was attached directly to a ring encircling the cesium pinch-off tubulation ~1/4 inch from the crimp. The thermocouple bead was peened into a hole drilled in the ring. Thermocouples T_2 and T_3 were sheathed thermocouples and apparently were not affected by the arc. The spurious voltage might have been produced by the generator itself, since the casing and cesium pinch-off were part of the collector circuit (collector is grounded).

The third alternative is that T_1 was reading correctly and that the generator itself acted as a resistor during the arc and heated up during the time it took for the short to open. If this occurred, then the temperature would come down very slowly, due to the mass of the system. The fact that the cesium control system did not shut off the heaters at this point indicated that the thermistor did not sense a real change in temperature at that point, as T_1 indicated. Thus, the second alternative appears the most probable.

Upon completion of the helium leak check, during which period thermocouple T_1 reached a low point reading of 505° F, the temperature leveled off at ~710° F. Over the next 40 hours, the cesium temperature increased by 60° F to ~770° F. During this same time period, both the casing temperature (T_3) and the cesium reservoir temperature (T_2) also increased, by ~20° F. At or during the "failure incident," a relatively sharp increase in cesium temperature to 830° F was experienced.

The gradual increase in system temperature (T_1 , T_2 and T_3) could just be a reflection of the redistribution of the fuel, i.e., as fuel accumulated on the diode support ring, an increase in the collector, generator casing and cesium reservoir temperature should be expected. The sharp increase in T_1 "at or during the incident" could be explained by the fact that, when all the cesium is distilled from a reservoir, the large heat input to the reservoir can only be dissipated by an increase in reservoir temperature. An alternative explanation of the sharp rise in T_1 is associated with the fact that this thermocouple was reflecting a voltage pickup. If the thermocouple were influenced by generator output, the increase in the apparent T_1 reading could be directly related to the increase in the generator output, i.e., the greater than 50 amperes observed at failure. A third possibility for accounting for the recorded rise in T_1 would be a malfunction of either the cesium heater or cesium heater control system. This possibility appears remote, since a post-test check on the heater and control system indicated that both components were operable. Their operability was also demonstrated during the attempts to ignite the generator after the "failure incident."

Since T_1 decreased from 770° to 565° F in a relatively short period of 15 minutes at the same time that the generator output was rapidly degrading, it may indicate

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that cesium was distilling from the diode. The temperature would drop from evaporative processes while generator output would degrade because of loss of cesium.

5. Generator Output Behavior

The increase in generator output at the time of failure remains one of the most difficult observations to reconcile. If the generator were operating at a cesium pressure which was above optimum, it would be possible during the transient operation associated with a cesium leak to pass through this optimum, whereby an increase in generator output would be observed. Another possible explanation might be that the cesium escaped suddenly from the diode near the diode spacing, allowing the space to "see" many cesium ions at the instant of rupture. This "cloud" would give a momentary increase in output, since it would be almost equivalent to a direct short across the space. It should also be noted that a change in the load characteristic could also account for the increased output current. The load resistance did change from 9.25×10^{-3} to 7.44×10^{-3} ohm from the time that the generator output current changed from 36.8 to 7.4 amperes. However, reduction in load resistance of this magnitude should be expected due to the decrease in operating temperature of the transistors employed in the load circuit.

Immediately after the "failure incident," when the generator output was less than 1.5 watts, the cesium temperature T_1 assumed a level consistent with the specified initial cesium temperature. In addition, the cesium heater was operating at the specified initial input power. Thus, it would appear that conditions were approximately as required for normal operation of the diode. However, normal operation of the diode could not be achieved, even though all remedial action resulted in the establishment of conditions which would have ensured diode operation. It is thus concluded that the cesium reservoir had been breached at this point, obviating further normal operation.

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VII. CONCLUSIONS

Failure of the SNAP 13 generator most probably occurred as a result of molten fuel attacking the diode spacer, resulting in violation of the cesium envelope.

Major events in the development of the above conclusion are given below. Note that, in most cases, only the most probable or most reasonable conclusion can be given, since absolutely definite information was not available.

1. Fuel Form

- (1) The length of the hot-pressed pellet was 2.5 times that expected, due to inadequate hot pressing to achieve the desired density and possible inclusion of additional contaminants.
- (2) The presence of ionic contaminants in the fuel composite resulted in a melting point in the range of 1600° to 2000° C, considerably below the desired melting point of 2250° C.

2. Fuel Capsule

- (1) Due to poor thermal conductivity and excessive pellet length, the fuel could have melted during the electron beam welding operation and initiated reaction with the capsule material.
- (2) If alumina were present on the fuel form, it would react with the tungsten -2% molybdenum (W-2 Mo) capsule at 1550° C (Martin experience) and result in attack at the grain boundaries.
- (3) The leak tightness of the capsule was doubtful, as determined by a helium leak test following the welding operation. The weld was found to be leak free, but cracks may have been present in the capsule wall since the W-2 Mo is susceptible to surface cracking.
- (4) Fuel was found in the generator outside the capsule. A single leak point was found in the capsule wall from which the fuel probably was released as a liquid.
- (5) The possibility exists that several fissures developed and extended through the capsule wall as a result of the thermal cycles imposed during the welding and braze attempts.

3. Capsule-to-Emitter Braze Operation

- (1) The five braze attempts probably accelerated any fuel-capsule reaction with possible loss of fuel.
- (2) Outgassing and the loss of molten fuel from the capsule would account for the high bell jar pressures experienced during the braze attempts.
- (3) The possible presence of fuel in contact with the 5-mil-thick spacer and the severe outgassing encountered could have resulted in a deleterious reaction to the spacer.
- (4) An estimated braze contact area of 60 to 70% was achieved.

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4. Casing Cover Lowering and Brazing

Severe outgassing difficulties were encountered during the casing brazing operations to the extent that a satisfactory braze could not be obtained. A leak check of the casing braze revealed that it was not leaktight.

5. Generator Operation

- (1) Generator operated properly for approximately 45 hours, but at half the expected output, most probably indicating loss of fuel. The fuel was probably lost as a liquid (hole size < 0.1 mm dia) during the capsule brazing operation.
- (2) An arc occurred, which resulted in damage to the load. However, the generator output amperage increased, thereby providing greater electron cooling for the fuel capsule.

6. Generator Abort

Generator failure was characterized by an electrical arc, a sudden momentary increase in current output, a sudden momentary increase in bell jar pressure, a relatively sudden drop in cesium temperature and a severely reduced generator output voltage.

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APPENDIX A

FUEL CAPSULE HEAT TRANSFER ANALYSIS

This appendix presents an analysis to determine the fuel capsule temperature which must exist in order to account for the lower-than-expected diode output based on the 290-watt initial fuel loading.

The analysis is based on the performance data realized for Generator 13, which demonstrated a generator efficiency of 7.6% at 17.5 watts output instead of the higher performing system, Generator 11A, which demonstrated an efficiency of 8.7% at 20 watts output. By using Generator 13 data, more of the total thermal input is accounted for by normal diode operation, and less of a burden is placed on the ability of the fuel capsule system to dissipate the additional heat.

$$\text{Required heat input to the generator} = \frac{16.9 \text{ watts}}{0.076} = 222 \text{ watts.}$$

$$\begin{aligned} \text{Additional heat which must be} \\ \text{dissipated by the fuel capsule} &= 290 - 222 = 68 \text{ watts.*} \end{aligned}$$

The corresponding diode emitter temperature based on TEECO research data (Ref. 7) is 1870° K (1597° C). **

Determination of the fuel capsule temperature which must exist to dissipate the additional 68 watts of heat is based on the thermal shield experimental data presented in Fig. II-7 of Ref. 1 and shown in Table A-1. The heat loss gradients with respect to temperature, based on the 1800° to 1900° K temperature range, are 0.0040 watt/cm²-°K (linear extrapolation) and 0.0051 watt/cm²-°K (experimental extrapolation).

TABLE A-1

Thermal Shield Data

Capsule Temperature (°K)	Thermal Shield Heat Loss (watts/cm ²)
1800	1.29
1840	1.48
1900	1.69*
1900	1.80**

*Linear extrapolation.

**Experimental extrapolation.

*Based on Generator 11A data (~ 8.5% efficiency at ~17 watts(e) output), the additional heat which must be dissipated would amount to 91 watts.

**It should be noted that recent undocumented reports have indicated that this temperature is too high, due to experimental error. However, it has been used in this analysis to provide further conservatism.

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Utilizing a thermal shield heat transfer area of 40 cm^2 , the final fuel capsule temperature required to dissipate the 68 watts of additional heat is calculated as shown in Table A-2. The required final fuel capsule temperature lies in the range of 2203° to 2295° K .

TABLE A-2

Fuel Capsule Temperature Required
to Dissipate 68 Watts

	Type of Data Extrapolation	
	<u>Linear</u>	<u>Experimental</u>
Additional heat loss (watt/ $^\circ\text{K}$)	0.16	0.204
Required capsule temperature rise ($^\circ\text{K}$)	425	333
Required capsule temperature ($^\circ\text{K}$)	2295	2203

Considering the $\pm 5\%$ possible variation in the fuel inventory, the required fuel capsule temperature lies in the range of 2132° to 2385° K as shown in Table A-3.

TABLE A-3

Required Capsule Temperature Considering
 $\pm 5\%$ Fuel Inventory

	<u>-5%</u>	<u>+5%</u>
Required capsule temperature rise		
Linear extrapolation ($^\circ\text{K}$)	334	515
Experimental extrapolation ($^\circ\text{K}$)	262	404
Required capsule temperature		
Linear extrapolation ($^\circ\text{K}$)	2204	2385
Experimental extrapolation ($^\circ\text{K}$)	2132	2274

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