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DESTRUCTIVE EXAMINATION OF A SNAP HEAT SOURCE

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ABSTRACT

A detailed examination was made of one of three plutonium-238 fueled SNAP 3 heat sources which was shipped from Mound Laboratory in May 1961 and returned in May 1963. Examination of the source included radiography, calorimetry, dimensional inspection, radiation measurements, internal pressure measurements, metallographic examinations of the containers, and recovery and impurity analysis of the plutonium fuel.

Results showed that the inner tantalum container of the source failed at the weld, allowing the plutonium to escape and to attack the Haynes Alloy 25 outer container. Failure at the weld was attributed to the fact that the plutonium had become molten at some time during its use and to poor welding of the liner during fabrication of the source, and not to corrosion of plutonium on the tantalum. It was concluded that tantalum is a satisfactory container material for plutonium at temperatures below the melting point of plutonium (~ 640°C).

INTRODUCTION

Three SNAP 3 heat sources fueled with plutonium-238 metal were fabricated by Mound Laboratory in 1961. One of these, fabricated and shipped on May 16, 1961, had a weld on the inner tantalum liner which had been difficult to make, and therefore, was suspected of being unsound. However, due to the urgent need for the source at that time, it was shipped to the Martin Company with the other sources. On May 20, 1963, the source was returned for destructive examination and a replacement source was furnished.

The pertinent data on the returned source is presented in Table 1.

Table 1

DATA ON SNAP 3 HEAT SOURCE

Fuel	Pu^{238} Metal
Date Shipped from Mound	May 16, 1961
Amount of Pu^{238} when shipped [a]	52.52 ± 0.26 watts (91.29 ± 0.46 g)
Date Source returned to Mound	May 29, 1963
Amount of Pu^{238} when returned	51.58 ± 0.26 watts (89.66 ± 0.45 g)
Inner Container Material	Tantalum
Outer Container Material	Haynes Alloy 25
Total Internal Volume (Tantalum)	~ 13.0 cc
Volume [b] of Pu^{238} Fuel	~ 7.1 cc
Free Volume	~ 5.9 cc
Pressure at time of loading (5/16/61)	~ 1 atm. (15 psia STP)
Calc. Max. Pressure [c] at time of examination (10/1/63)	~ 28.5 atm. (428 psia STP)
Design operating temp.	950°F (510° C)

[a] Amount of Pu^{238} determined calorimetrically; data based on half-life of 86.4 years.

[b] Based on density of 16 g/cc Pu (80% Pu^{238})

[c] Maximum pressure not expected unless the fuel form was finely divided or molten.

SOURCE EXAMINATION

The returned source was subjected to the following tests and examinations:

External contamination check
Calorimetry and plutonium-238 analysis
Dimensional inspection
Radiography
Dosimetry measurements
Total neutron emission rate
Gamma ray spectrum
Helium leak check
Measurement of internal helium pressure due to alpha decay
Macroscopic examination of Haynes Alloy 25 and tantalum containers
Metallographic examination of Haynes Alloy 25 and tantalum containers

Prior to examination, the source was allowed to attain thermal equilibrium to determine if the external temperature would approach or exceed the melting point of the metal (~640°C). If the melting point had been reached, the data to be collected in the various examinations would not be reliable. However, the temperature of the surface of the source was determined to be between 220° and 250°C when left standing in air in an insulated support.

CHECKS FOR EXTERNAL CONTAMINATION

The SNAP 3 heat source as returned to Mound Laboratory on May 20, 1963, showed no external radioactive contamination. The source and the surfaces of the shipping container were wiped with a soft filter paper and counted. No alpha counts were detected on these wipes.

PHOTOGRAPHS

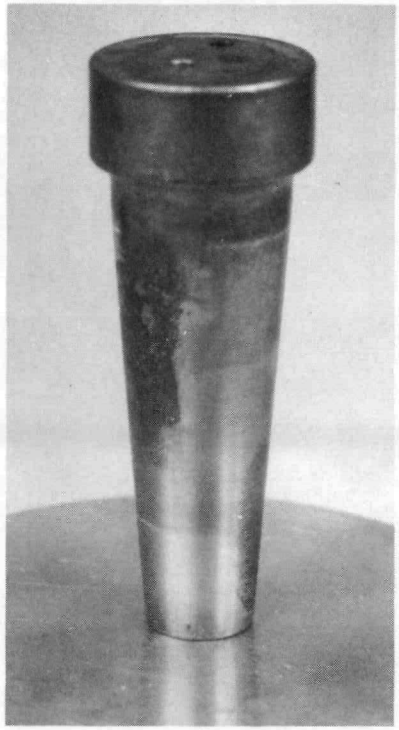
The external appearance of the source indicated that it was not subjected to any physical abuse during use. Slight stains on the tapered surface, shown in Figure 1, were found which were carbonaceous residues left by gloves and other materials used in handling the source during examination. The tool marks on the surface of the specimen are clearly visible and well defined. This indicated that no significant amount of oxidation took place on the outer container during use.

CALORIMETRY

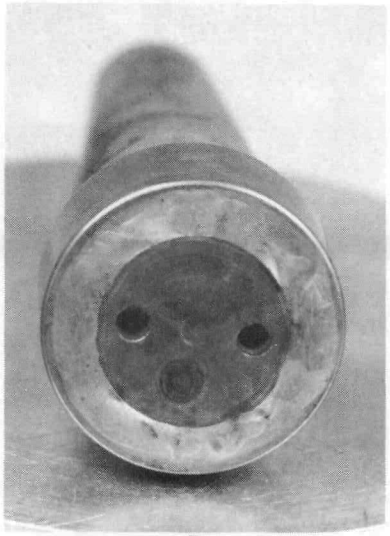
When the source left Mound Laboratory on May 16, 1961, the amount of plutonium-238 was 91.29 ± 0.46 grams as determined calorimetrically (52.52 ± 0.26 watts). On May 29, 1963, the source contained 89.66 ± 0.45 g of plutonium-238 (51.58 ± 0.26 watts) a decrease of 1.79 per cent. The measured decrease is equal to the calculated decrease of 1.62 per cent, within the experimental error of the calorimetric measurements.

DIMENSIONAL INSPECTION

Inspection revealed that the outer Haynes Alloy 25 container met dimensional specifications given in Martin Drawing 356-029 1119. The diameter of the head and the overall length of the specimen were within the specified limits.



A.



B.

C.

Figure 1

SNAP 3 Heat Source as Returned in May, 1963

RADIOGRAPHY

Four radiographs were taken at 45° intervals around the longitudinal axis and are shown in Figure 2. The radiographs show that at some time after fabrication the plutonium, which was originally inserted as four individual precast slugs, had melted. The radiographs suggest that the space between the tantalum inner lid and the outer Haynes Alloy 25 cap became filled with the plutonium metal. The figures also indicate a large void in the top of the source just below the tantalum lid.

The radiographs also suggest that the Haynes Alloy 25 reacted with the plutonium. Portions of the sharp inside circular shoulder in the head of the outer container disappeared into a hazy line in the 0°, 45°, and 135° radiographs.

DOSIMETRY MEASUREMENTS

Dosimetry measurements were taken to determine the neutron flux and the x-ray intensity. The results of these measurements at varying distances from the source are tabulated below:

Distance From Source (cm)	Neutron Flux ($n°/cm^2/sec$)	Gamma Ray Intensity (mr / hr)
0	220	100
10	32	12
50	-	1.5
100	2	0.5

NEUTRON COUNT

The total neutron count of the source was measured as 1.0×10^6 neutrons per second (±5 per cent).

GAMMA SPECTRUM

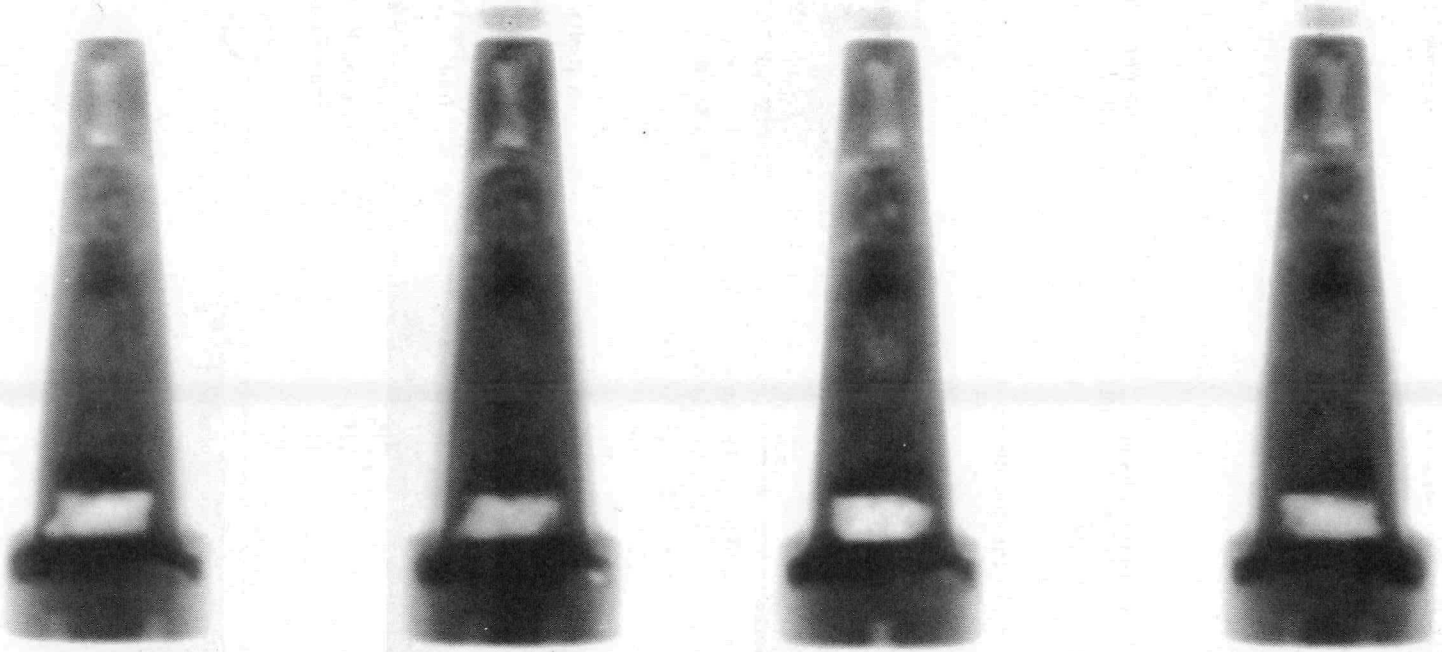
Results of a gamma spectrum of the source are presented in Table 2.

Table 2

GAMMA RAY SPECTRUM: OBSERVED PEAKS AND IDENTIFICATION

Peaks[a] (Mev)	Isotope	Comments
0.026	Th^{234}	
0.063	Th^{234}	
0.101	Pu^{238}	
0.150	Pu^{238}	
0.200	Pu^{238}	
0.512	Tl^{208}	Annihilation Radiation
0.582	Tl^{208}	
0.760	Pu^{238}	
0.870	Pu^{238}	
1.000	- - -	Not Identified
1.09	Tl^{208}	
1.27	Tl^{208}	Back Scatter
1.57	Tl^{208}	Double Escape Peak
2.10	Tl^{208}	Single Escape Peak
2.59	Tl^{208}	

[a] Some Pu^{239} peaks were also detected in the spectrum.



0° 45° 90° 135°

Figure 2

RADIOGRAPHS OF THE SNAP 3 HEAT SOURCE AT VARIOUS RELATIVE POSITIONS AROUND THE LONGITUDINAL AXIS OF THE SOURCE

HELIUM LEAK CHECK

A helium leak check on the source using a Veeco leak detector capable of determining a leak rate of 10^{-10} cubic centimeters (STP) per second indicated that helium was not leaking from the source.

MEASUREMENT OF INTERNAL PRESSURE

The capsule was punctured to measure the increase in internal pressure due to helium from alpha decay. Figure 3 is a schematic of the pressure test system which was used for this purpose. The source was fitted inside a sample block which was connected to the pressure measuring system. A standard volume, a gas collection bottle and a Wallace and Tiernan precision dial manometer were also connected to this system.

The volume of the sealed, drill(sample) block containing the source, and the valving and tubing leading to the pressure side of the manometer was determined. Using a known volume and standard gas expansion techniques this volume was found to be 103.14 cc and was the volume into which the gas from the source expanded when the source was punctured.

The source was placed in the drill block and a hole was drilled into the cylindrical head 1/2 inch from the top. The hole just penetrated the lip of the inner tantalum container. The observed pressure increase was only 10 torr. The source was left in the system for approximately 20 minutes after drilling. No additional increase in pressure was measured. The volume of gas collected from the system was 1.23 cc (STP).

The source was removed from the system and the top end removed to allow macroscopic examination. An abrasive cutoff wheel was used to cut a slot into the head half way between the top and bottom shoulders around the entire circumference just below the weld penetration. However, the cap did not come loose from the body until it was sharply rapped. Upon inspection, it was found that the Haynes Alloy 25 cap had fused to the plutonium which had escaped the inner tantalum liner. Figure 4 shows the plutonium metal in the top of the capsule and the extent of corrosion to the Haynes Alloy 25 cap.

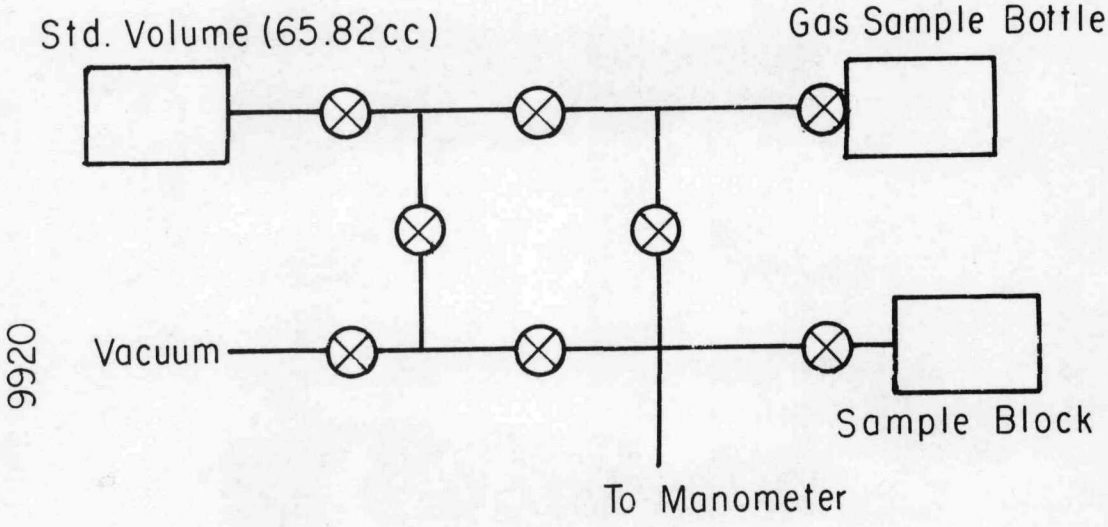
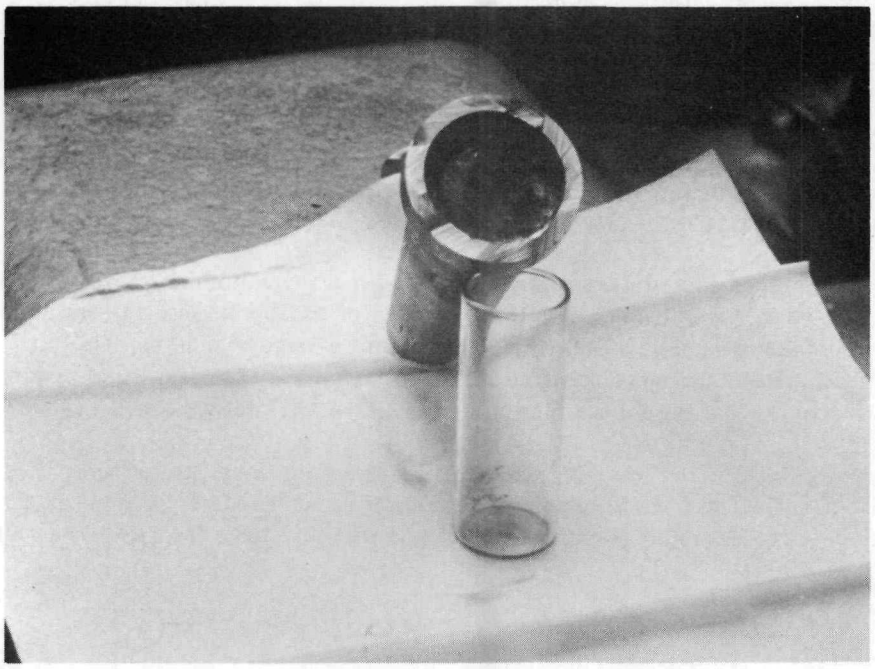
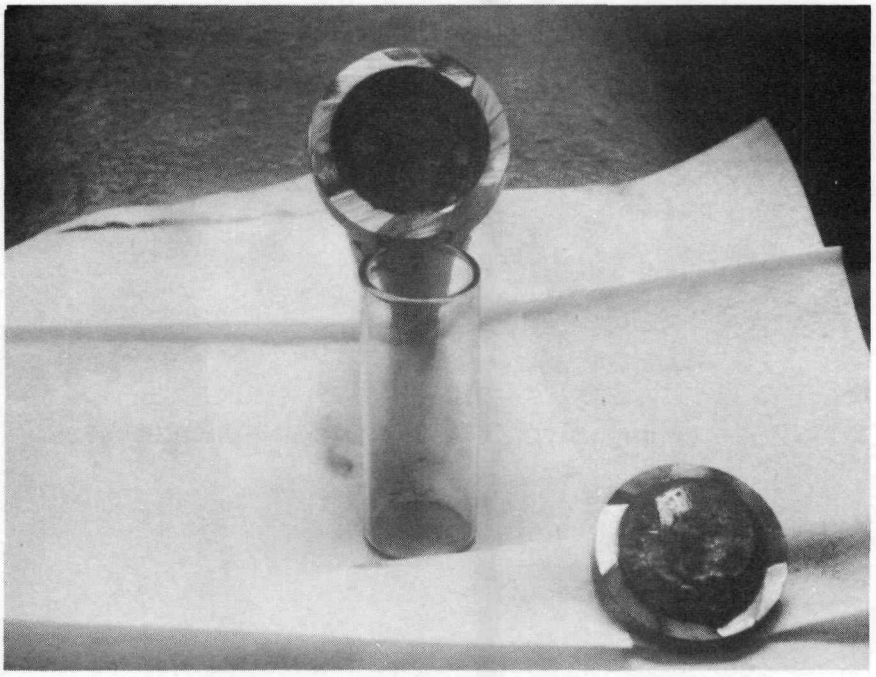


Figure 3

HEAT SOURCE INTERNAL PRESSURE MEASURING SYSTEM



a. Note corrosion inside outer container



b. Note corrosion of outer container cap

Figure 4

VIEWS OF INSIDE OF SOURCE IMMEDIATELY AFTER OPENING

All of the cap was not wetted by the plutonium, and hence the cap was not corroded over the entire surface. The source had to be removed from the pressure measuring device to remove the cap, to examine the cap and contents, and to take the desired photographs. After returning the source to the pressure device, it was noted that gas was slowly leaking out. As will be discussed later, a discrepency exists between the measured and calculated helium accumulation which was caused from this operation. After returning the source to the measuring device, the pressure increased from three to 193 torr in 110.5 cc, equivalent to 25.5 cc (STP) of gas.

The next step consisted of drilling into the large void space in the top of the capsule just under the tantalum cap. This operation yielded 6.23 cc (STP) of gas.

The source was next placed in a stainless steel container and the container with the source sealed inside was inductively heated to approximately 900°C for 20 minutes to liberate any gas entraped in the solid plutonium metal. This operation yielded 72.30 cc (STP) of gas. A large amount of metal was ejected from the source during this operation and was deposited in the bottom of the stainless steel vessel. The container was reheated to above the melting point of the plutonium metal, but no additional gas was evolved.

The total volume of gas collected was 105.3 cc (STP) compared to a calculated amount of 168.0 cc (STP) (162.1 cc (STP) in helium due to delay plus 5.9 cubic centimeters of gas present in the volume during fabrication). The only conclusion to be drawn is that gas was lost when the cap was removed from the source.

DISASSEMBLY

RECOVERY AND ANALYSIS OF FUEL

Six normal hydrochloric acid was used to dissolve the plutonium metal which covered the top of the tantalum liner and sealed it into the outer Haynes Alloy 25 container. This material was easily dissolved and the tantalum liner was removed from the Haynes Alloy 25 outer capsule. The tantalum liner still contained a large amount of plutonium-238 which was removed through the drill hole in the side of the liner and dissolved in six-normal hydrochloric acid. As the fuel was shaken from the liner, it oxidized and glowed a bright red, indicating that this fine powder was not plutonium dioxide but either metal or some lower form of the oxide. It is unlikely that this fine material was plutonium metal since it would probably have agglomerated during the previous melting. After the removal of the fine material, the tantalum liner still contained heat-producing material. Additional plutonium was found in the bottom of the tantalum liner. This metal was dissolved in six-normal hydrochloric acid. When all of the metal had been removed, the bottom of the liner was cut off and the inside of the liner was washed with the six-normal acid. Both the tantalum liner and the Haynes Alloy 25 container were free of all plutonium metal and oxide after these dissolution operations.

Approximately three liters of six-normal hydrochloric acid were used to remove the plutonium and plutonium oxide from the source. The first liter contained the material which had leaked out of the tantalum container and corroded the Haynes Alloy 25. A sample of this solution was analyzed for plutonium and impurities, as were samples of the other two solutions consisting of the remaining material which was removed from the source. Table 3 lists the impurities found in each of the three solutions.

Table 3

ANALYSES OF PLUTONIUM SOLUTIONS

		Impurity (mg/ml)											
Sample Blank	Pu (mg/ml)	B 10	Si 8	Mn -	Mg -	Cr -	Fe <10	Co -	Ni <10	Al <10	Mo -	Cu <10	Ta -
1	5.1×10^3	20	8	18	40	100	120	>100	120	40	10	20	Trace
2	2.1×10^3	20	10	-	-	40	40	<100	14	40	-	14	-
3	8.2×10^3	20	10	-	-	40	40	<100	20	20	-	10	-

As expected, the first solution contained significant amounts of the constituents of Haynes Alloy 25*. The fact that only a trace amount of tantalum was found indicates that the closure weld on the tantalum container was poor, and that corrosion was not the primary cause of failure of the weld.

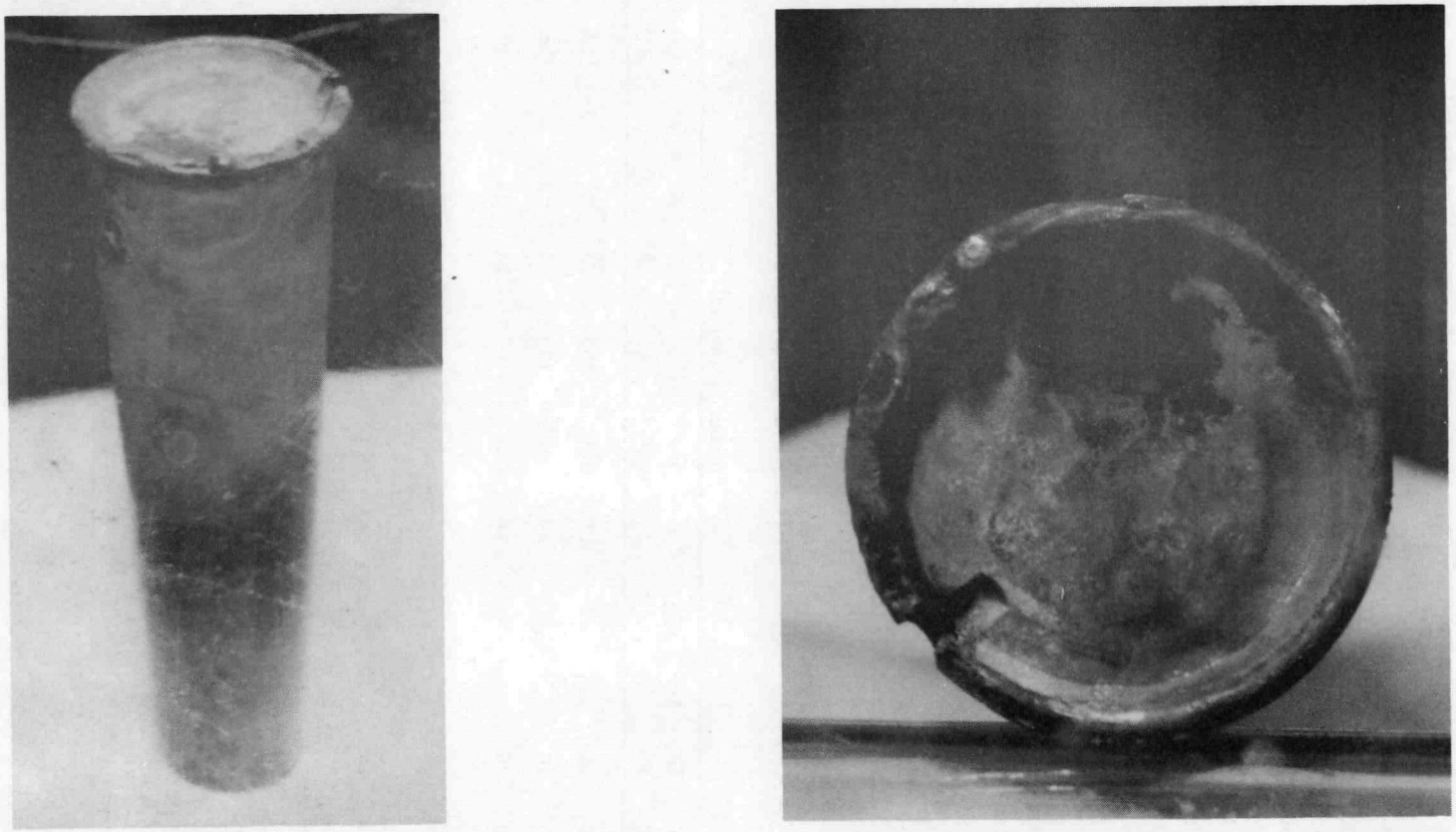
MICROSCOPIC EXAMINATION OF THE HAYNES 25 ALLOY AND TANTALUM CONTAINERS

The failure of the tantalum weld can be seen quite clearly in Figure 5. Half of the weld was in good condition. The plutonium metal penetrated the weld in several places without lifting the cap. Figure 6 shows the extent of corrosion to the Haynes Alloy 25 container.

METALLOGRAPHIC EXAMINATION OF THE HAYNES ALLOY 25 AND TANTALUM

Metallographic sections were taken of various parts of the tantalum and Haynes Alloy 25 containers. These sections were polished, etched, and photographed, as shown in Figures 7 and 8. Inspection of these photographs confirm the poor quality of the weld in the tantalum container. Figure 7A is a photograph of the sound

*Haynes Alloy 25 composition: Nickel 9-10%, Chromium 19-21%, Tungsten 14-16%, Iron 53%, Carbon 0.05-0.15%, Silicon 51%, Magnesium 1-2%, and Cobalt balance.

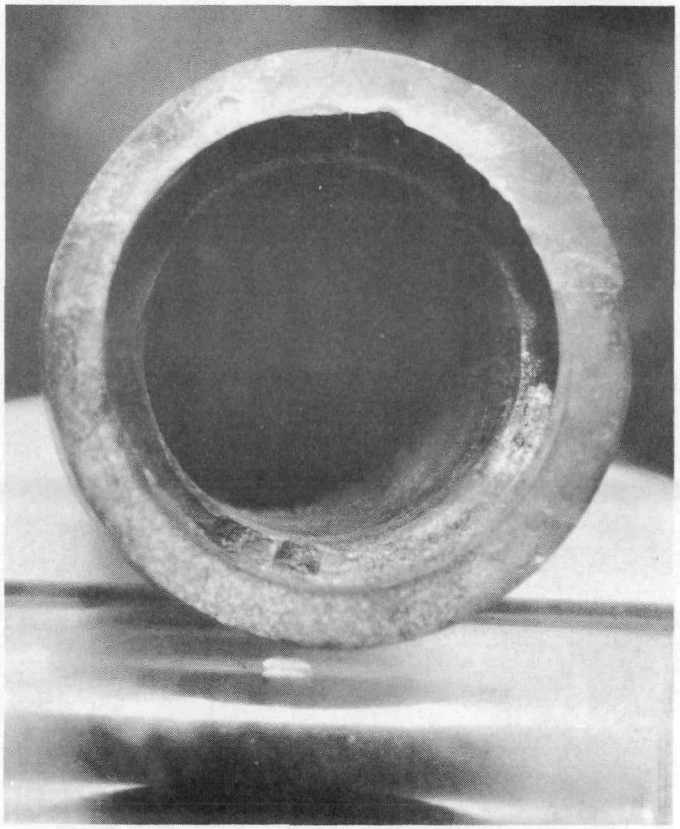


A. B.

Figure 5

TANTALUM LINER OF SNAP 3 HEAT SOURCE
(NOTE BAD WELD ON HALF OF CIRCUMFERENCE OF CAP)

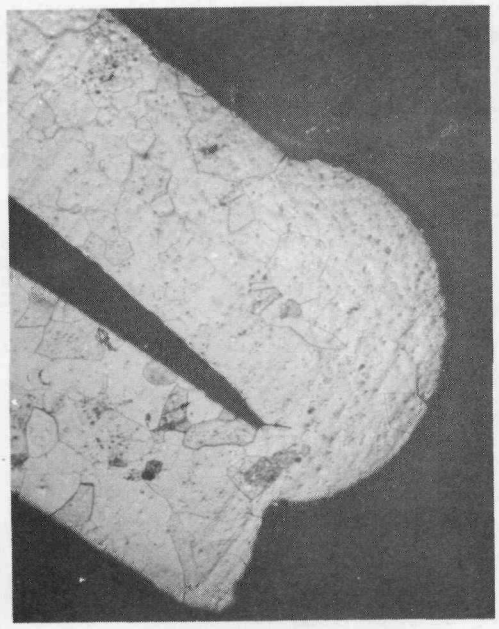
A.



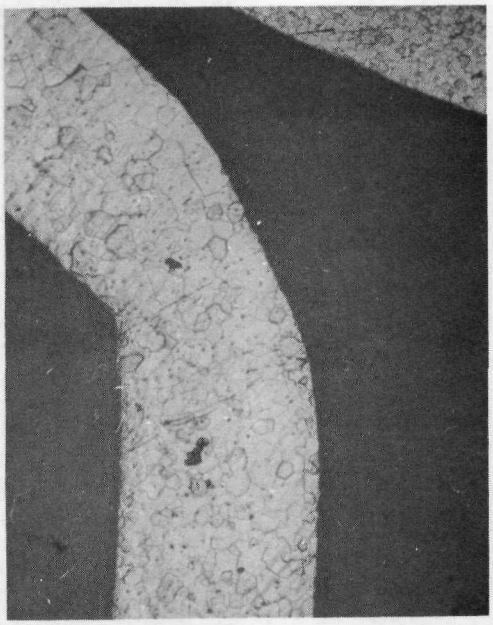
B.

Figure 6

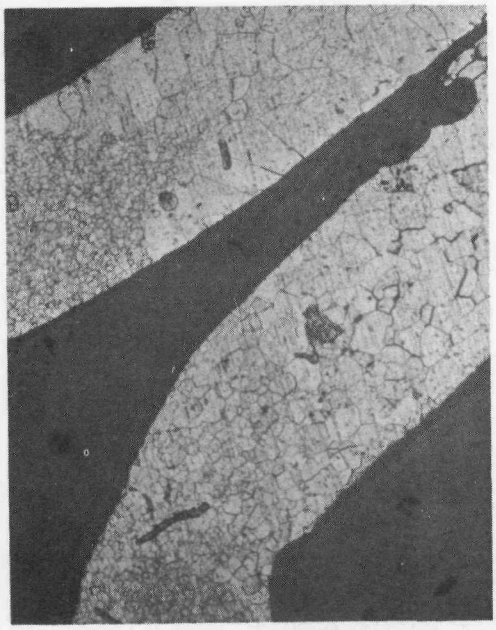
HAYNES ALLOY 25 OUTER CONTAINER OF SNAP 3 SOURCE
(NOTE CORROSION ON INSIDE OF CONTAINER)



A.



B.



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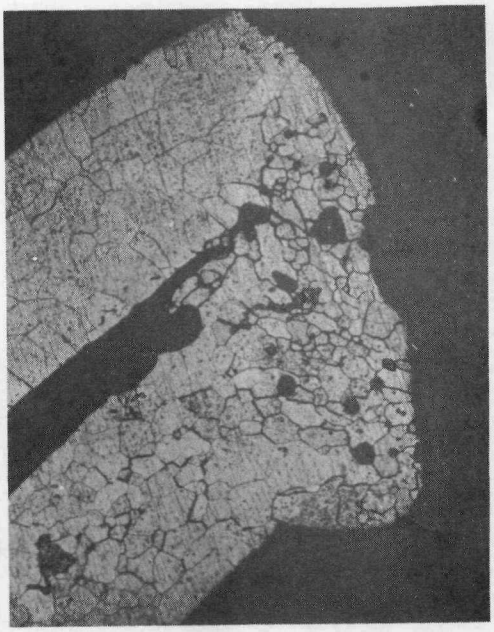
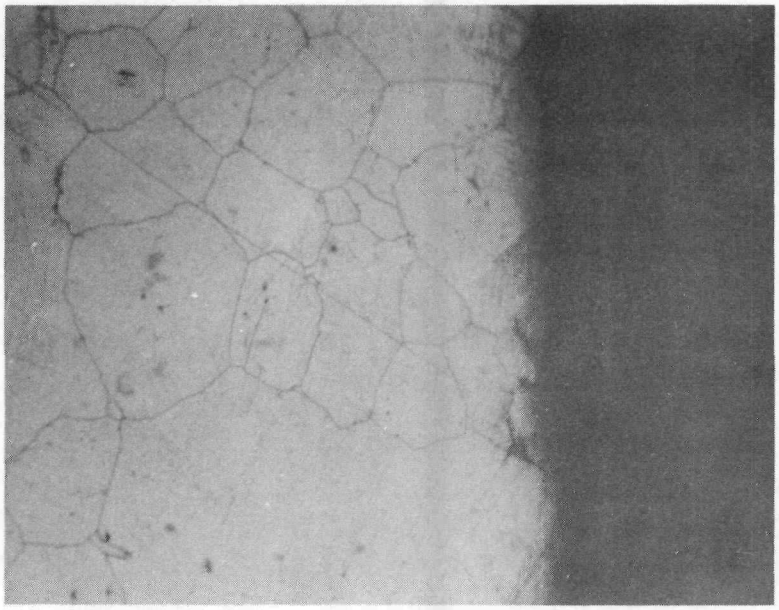


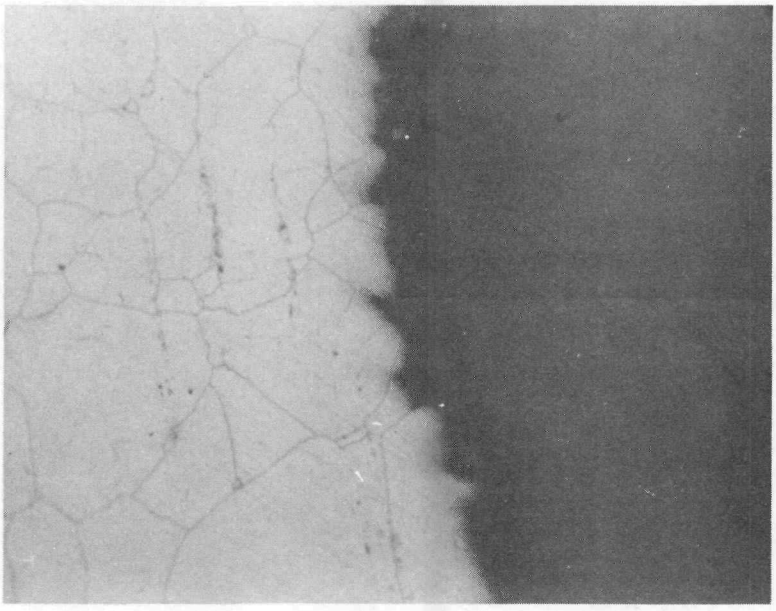
D.

Figure 7

MICROMETALLOGRAPHS OF WELD ON TANTALUM LINER OF THE SNAP 3 SOURCE



A.



B.

Figure 8

MICROMETALLOGRAPHS OF (a) THE SOUND PORTION AND
(b) THE CORRODED PORTION OF THE HAYNES ALLOY 25 OUTER CONTAINER

portion of the weld. Here the weld penetration is only 0.010 inch. Figure 7B shows the tantalum wall and inside cap surfaces where no corrosion is evident. Figures 7C and 7D are two views of the same weld area which show evidence that there was plutonium in the vicinity of the tantalum lip prior to welding. The plutonium occulsions in the weld were responsible for the poor quality of the seal. Figures 8A and 8B are photographs of the sound and corroded portions of the Haynes Alloy 25. As mentioned previously, the section shown in Figure 8B was adjacent to the part of the tantalum container where weld failure occurred.

CONCLUSIONS

These data collected during this examination showed that at some time after fabrication the plutonium metal melted, escaped the tantalum liner through a faulty weld, and attacked the Haynes Alloy 25 outer container. The fuel may have been molten only a short time; once the Haynes Alloy 25 was wet by the fuel, the reaction would continue in the solid state.

The plutonium analysis shows only small amounts of tantalum. Metallographic examination of the tantalum in other than the faulty weld zone showed no attack by the plutonium. It may be concluded that tantalum is a suitable container material for plutonium-238 metal for use at temperatures below the melting point of plutonium.

As expected, not all of the helium due to radioactive decay is released from the solid fuel to the void volume of the container. From the amount of gas collected on melting the fuel, 72.30 cc (STP), compared to the calculated total amount of helium from decay, 162.1 cc (STP), it is suspected that the fuel was melted approximately one year prior to the time the source was opened, or about September or October 1962. Whether the fuel was melted prior to this time would not be revealed by these data.

The data also indicated that for design purposes, unless temperature excursions above the melting point of the plutonium-238 metal can be absolutely avoided, the container must be designed to contain the total pressure expected from radioactive decay.