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FABRICATION AND QUALITY CONTROL TESTING OF SNAP-15C CAPSULES

D. L. Prosser
D. P. Kelly
J. A. Powers

AEC Research and Development REPORT

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MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

U.S.GOVERNMENT CONTRACT NO.AT-33-1-GEN-53

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SNAP Program

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Date: July 1, 1965

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SUMMARY

In cooperation with the 3M Company, Mound Laboratory fabricated and evaluated heat source capsules for the SNAP-15C program. These capsules were fueled with sufficient plutonium-238 metal to provide approximately 7.3 thermal watts of power each. Fabrication procedures for these sources, fuel properties, and fabricated capsule parameters are presented.

INTRODUCTION

In 1963 Mound Laboratory entered into a program with the Minnesota Mining and Manufacturing Company (3M Company) to provide fueled heat source capsules for a large milliwatt generator. After prototype units were successfully tested under prescribed environmental conditions, Mound Laboratory fabricated six fuel-units to be used in demonstration models of the generator.

The generator capsule was originally designed to contain 6 watts (thermal) of plutonium-238 metal (~80% Pu^{238}) and to operate for two years with an electrical output of 125 milliwatts. During the course of the program the fuel requirements were increased by the 3M Company to 7.3 thermal watts without any increase in capsule dimensions. The change reduced the free volume in the capsule, which required a refinement of fuel loading and welding techniques to provide a sealed unit with good integrity.

The fuel capsule consisted of an inner and an outer housing as shown in Figure 1. The inner housing and cap were machined from tantalum-10% tungsten bar stock and the outer housing and cap from Hastelloy C.

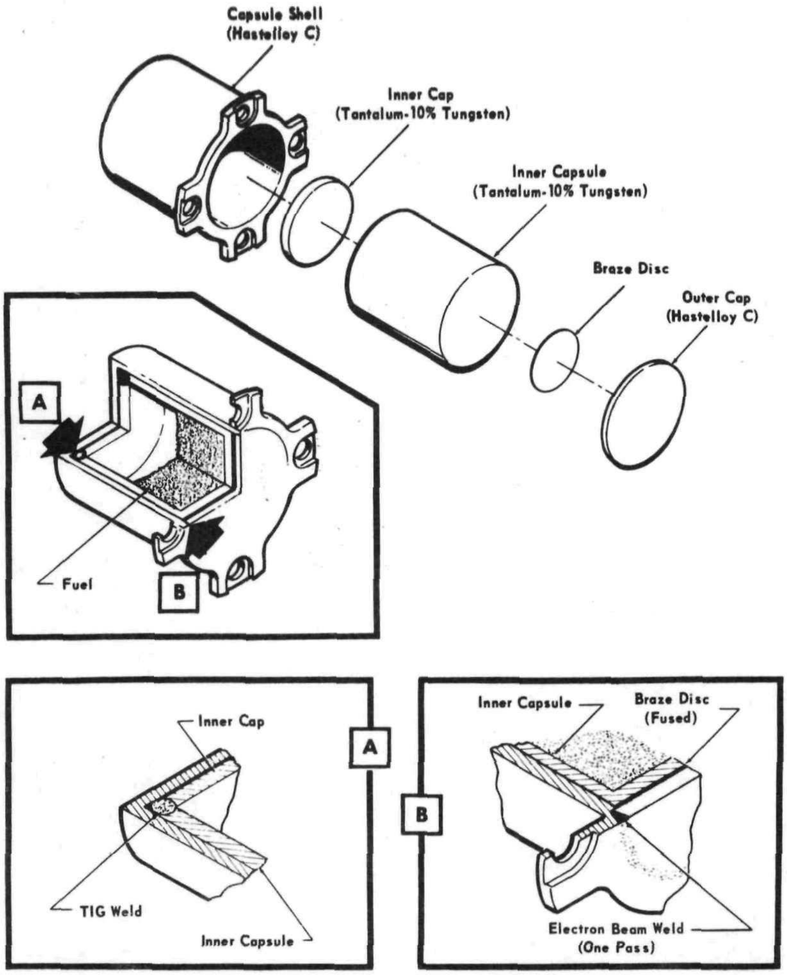


Figure 1. *SNAP-15C Fuel Capsule*

A large number of capsules were assembled for the environmental tests specified by the program prior to fabrication of the fueled units. At Mound Laboratory the work on the test capsules included effecting the welds on the inner capsules and brazing the inner and outer capsules together. For the fueled units the fuel was sized to fit into the capsule and then was melted into position after the capsule was sealed.

FABRICATION PROCEDURE

At each stage of fabrication both the test capsules and the fueled capsules were subjected to a thorough inspection program developed by Mound Laboratory. For the fabrication of the test capsules the following operations were performed:

1. Dimensional check of capsule components.
2. Dye penetrant test of capsules.
3. Visual inspection of inner and outer welds.
4. Sectioning and metallographic examination of inner and outer welds.
5. Radiographic examination of inner and outer welds.
6. Mass spectrometer helium leak test of inner and outer welds.
7. Dye penetrant test of welds.

For the fueled units the following additional operations were completed:

1. Chemical analysis of capsule materials.
2. Chemical analysis of fuel.
3. Differential thermal analysis of fuel.
4. Calorimetric determination of fuel.
5. Total neutron flux measurement.
6. Decontamination check.

All capsules, upon receipt, were gauged by conventional methods to check their dimensions against the specified values.

The dye penetrant used to examine the outer surfaces of the capsules for defects and cracks was a Zyglo compound supplied by the Magnaflux Corporation. The surface to be inspected was cleaned by applying the Zyglo cleaner ZC-7 from a spray can. The cleaner was removed with a cloth and the penetrant ZL-22 was sprayed on the part. The cleaner was applied again and wiped off to remove the excess penetrant. The developer ZP-9 was applied in a thin film. The part was placed under ultraviolet light, and cracks or defects were indicated by fluorescent lines or spots.

After the proper welding parameters had been established, the inner and outer capsules were welded using the following procedure:

> The inner capsule was sealed using a Hobart 300-ampere AC-DC Tungsten Inert Gas Welder, Model ADI-364. The housing was held in a steel collet which was mounted in an aluminum cooling block 4 in. in diameter and 5.5 in. long. The block was positioned in the chuck of a turntable. The housing extended out of the collet by 3/16 in. A 2% thoria-tungsten welding electrode, 1/16 in. in diameter, with the tip ground to a 60° included angle, was used. The electrode was held in an Airco water-cooled torch, Model H-35-B, which was mounted in a fixture to keep the electrode normal to the top of

the capsule. The electrode extended 0.25 in. beyond the gas cup; the tip was positioned 0.040 in. above the capsule and directly over the interface between the cap and the housing. The capsule was rotated at a rate of 13.5 sec/rev while helium gas was passed over the work-piece at a rate of 15 cfh. During the rotation the arc was struck by starting the current and bringing it immediately to 85 amp with the capsule at positive d.c. polarity. The welding was continued while the capsule was rotated for 16 sec. The current was then turned down quickly with the fine control and then shut off with the foot switch. The part was allowed to cool and was removed from the collet. These parameters were found to give the required penetration of 80% of cap thickness.

The outer capsule was sealed using a Hamilton Standard high-voltage, 3-kw welder equipped with a Zeiss gun. The cap was placed on the capsule and six of these units were clamped down in a holding fixture. The fixture was placed in the welder, which was then evacuated. After the beam was focused, the cap was spot welded at six different points around the circumference. An electron beam at 90 kv and 1 milliamp was used for 1 to 2 sec to make each tack. The tacking procedure was necessary to prevent the cap from warping during the closure operation. The welder was brought up to atmospheric pressure and the fixture was removed.

Each part was placed in a chuck in the welder, which was again evacuated. The electron beam was adjusted to focus on the capsule and was aligned to fall directly on the interface between the cap and the housing. The position was checked for concentricity with the interface. Welding was done with an electron beam of 95 kv and 1.5 milliamp and was continued for 5 sec with the capsule rotating at 3-3/4 sec/rev. The beam was brought up to full power immediately to start the welding, then was reduced to zero over a period of 2 to 3 sec at the end of the welding cycle. The welder was brought up to atmospheric pressure and the part was removed. These parameters met the requirements for obtaining a penetration equal to 95% of the cap thickness.

The inner and outer welds were visually examined under 30-power magnification. Control of the depth and quality of the welds was maintained by welding two inner capsules before and after each set of inner test capsules was sealed. One each of the pre- and post-weld inner capsules was sectioned and the others were placed in outer capsules and used as weld controls for welding the set of inner test capsules into the outer capsules. These outer capsule weld controls were also sectioned, mounted, polished and photographed to facilitate the examination of the grain growth in the weld area and the depth of the weld.

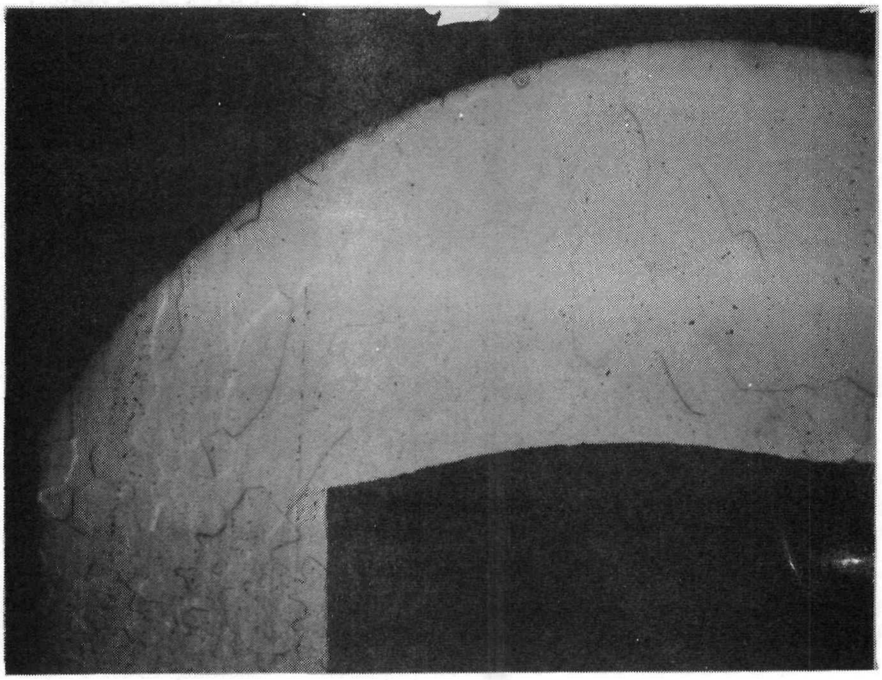
The preparation of both the inner and outer metallographic samples was accomplished by cutting the capsule in half along the diameter with a Buehler cut-off wheel. The sample was mounted in Selectron 5026 mounting resin which was allowed to harden at room temperature. The sample was ground using first a 50-grit silicon carbide paper grinding disc and water as a coolant and lubricant; for finer grinding a 180-grit silicon carbide paper grinding disc was used, and finally 600-grit silicon carbide. For the rough polishing 1-micron alumina on silk was used and, for the final polish, 5-micron alumina on microcloth. The inner tantalum-10% tungsten capsule was etched with a solution consisting of three parts lactic acid, one part nitric acid and one part hydrofluoric acid. The outer Hastelloy C capsule was electroetched at 4 to 6 v for about 20 sec in a 10% chromic acid solution. The weld area of the etched samples was photographed at 30-power magnification and the braze interface at 500 power. Typical photographs of the braze interface and of the inner and outer welds, before and after welding of the samples, are shown in Figures 2, 3 and 4.

Figure 2. *Cross-Sectional View of Braze Bond Between Inner and Outer Capsules (500X)*

The welds were also examined by radiographic techniques to detect flaws. After the radiography parameters had been established the following radiography procedures were employed to examine the inner and outer welds properly.

For the inner weld an iridium-192 gamma source, mounted in a Budd Multitron unit, was used to provide the required penetration of the dense metal in the inner container. The distance from the center of the source to the film is 36 in. An exposure of about 45 curie-hours was used. Type M radiographic x-ray film was sandwiched between sheets of lead to give improved definition. The lead and film were arranged in the sequence 5-M-1½-1½-M-5, where the numbers represent the thickness of the lead in millimeters and M represents the film. The parts were held in intimate contact with each other either under vacuum or in a hard-back cassette while the film was being exposed. Both methods have been used for the inner capsules with equal results. The developed images were examined with a 7-power eye comparator. Four views of each capsule were taken - at 0°, 45°, 90°, and 135°.

The outer capsule weld was examined using a 250-kv Picker X-ray Machine. A lead filter was used in the head. A hard-back cassette holding type M film



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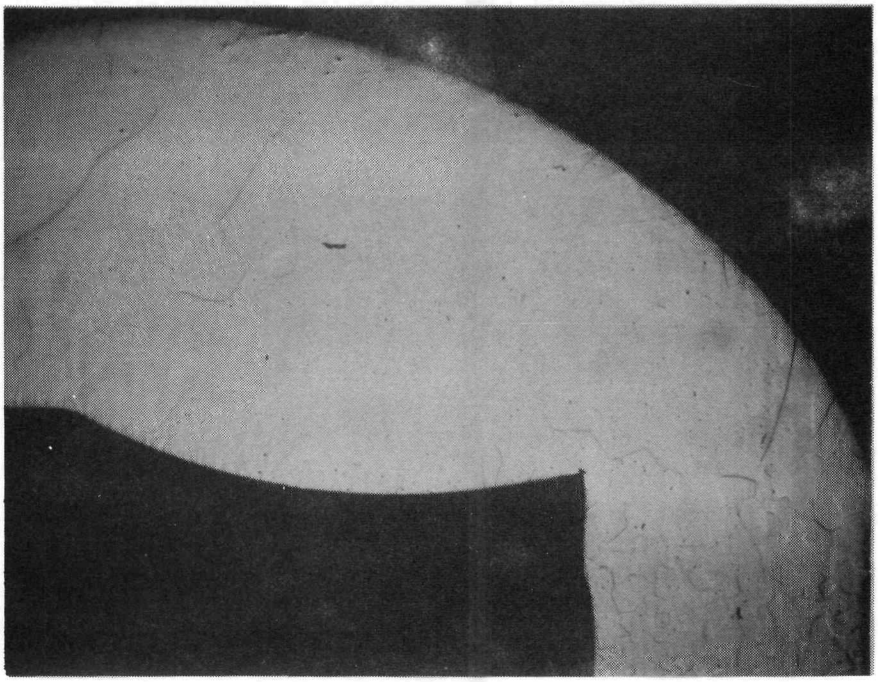
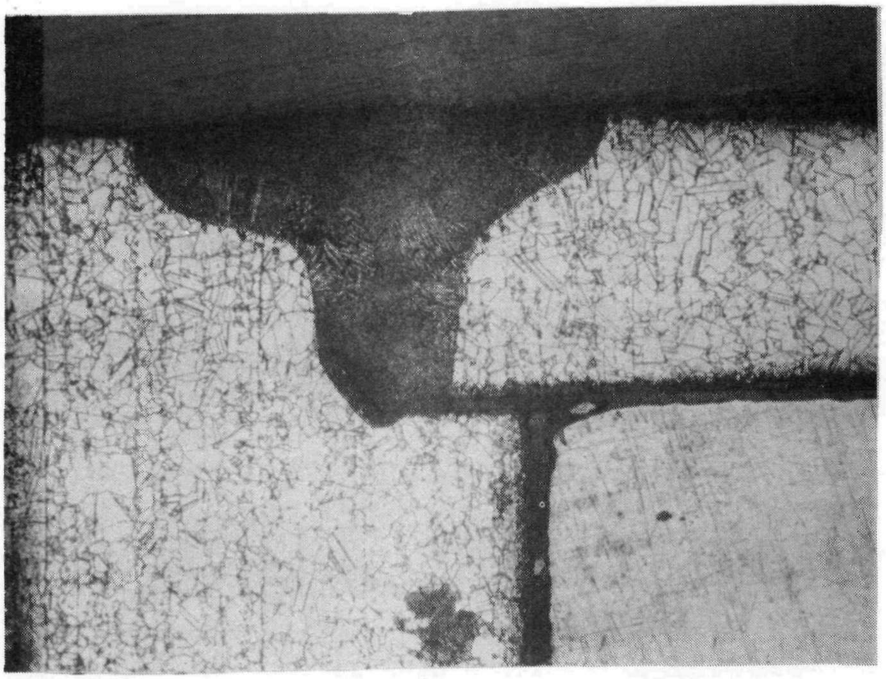


Right Half

Figure 3. *Cross-Sectional Views of Cap Welded into Inner Capsule (30X)*



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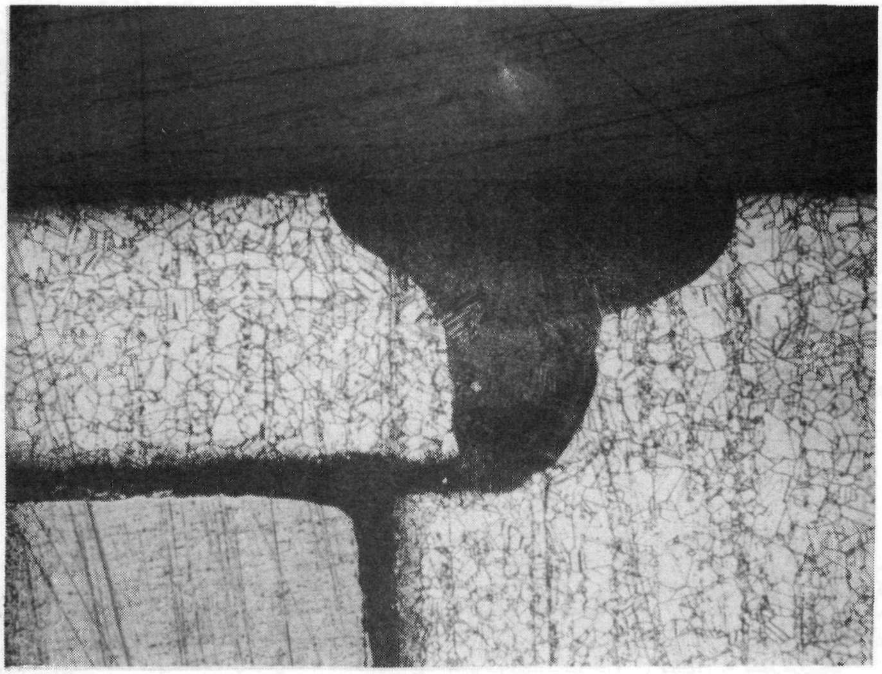


Right Half

Figure 4. *Cross-Sectional Views of Cap Welded into Outer Capsule (76X)*

interspersed with lead sheet as described previously was used. The exposure time was 113 sec at a 15-microamp current. The same developing procedure as for the iridium source exposures was used. The film was examined under a 10-power binocular microscope. The exposure had to be made between the lugs on the capsule to obtain clear radiographs of the weld. For this reason only two views were made for each capsule. Representative radiographs for the inner and outer capsules are shown in Figures 5 and 6.

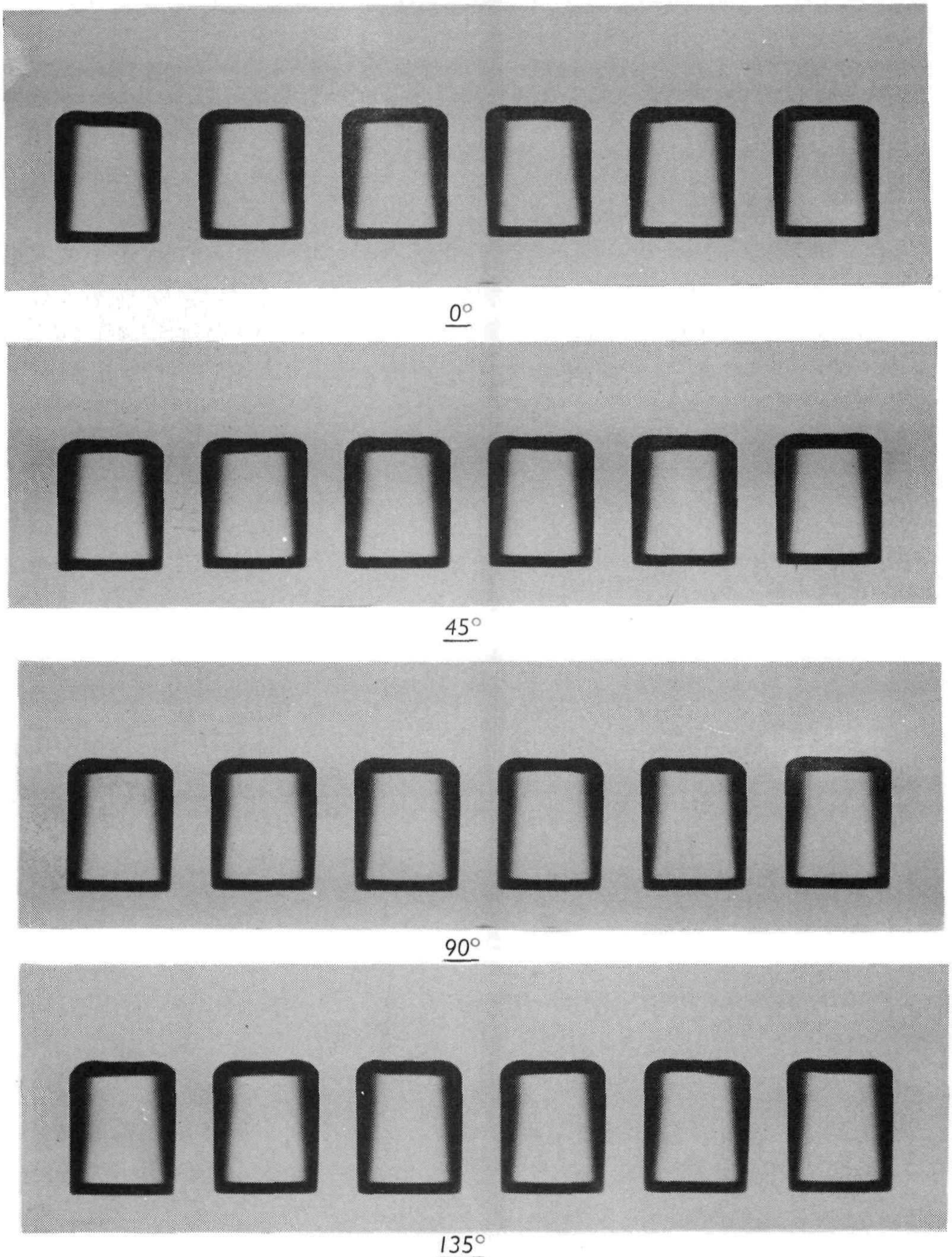
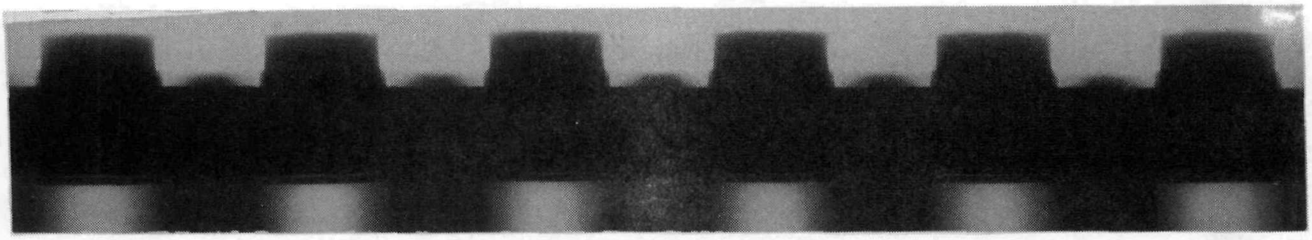
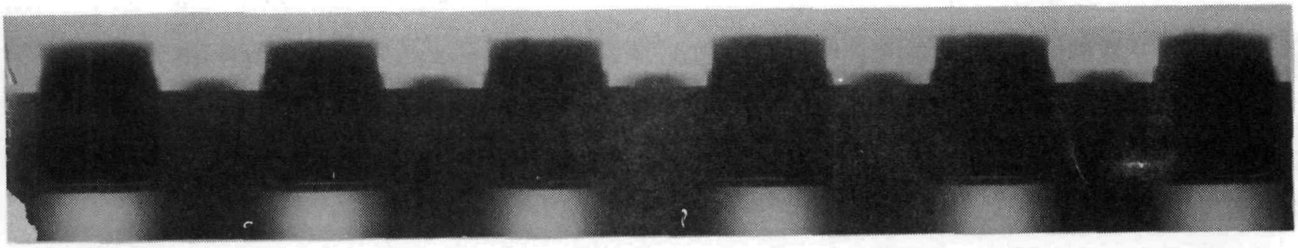


Figure 5. *Radiographs of Inner Welds*



0°



90°

Figure 6. *Radiographs of Outer Welds*

The mass spectrometer helium leak test was performed on both the inner and outer sealed capsules by placing them under helium at a pressure of 600 psig for 30 min. The pressure was reduced and the capsules were placed under vacuum and connected to a leak detector to search for helium escaping from the capsules. The detector was calibrated with a standard leak and the leak rate of each capsule was calculated. The maximum leak rate specified for the capsules was 1×10^{-7} cc/sec, STP.

The inner and outer welds were inspected for surface defects using the dye penetrant method described earlier.

CAPSULE FUELING PROCEDURE

When the test capsules had passed the environmental requirements successfully, the loading of capsules with plutonium fuel was undertaken. A quartz tube (Figure 7) was made with a precise inside diameter and a flat bottom. The diameter was held at a dimension of 0.010 to 0.015 in. less than the inside diameter of the inner capsule. The tube was placed in a high-purity argon controlled-atmosphere glove box and loaded with pieces of plutonium. The tube was inserted in a quartz thimble (Figure 8) which was placed vertically in a furnace, then evacuated and heated. When the temperature reached 800°C the metal was stirred by a tungsten rod extending through a vacuum seal in the top of the thimble. The metal was converted to a compact mass, free of voids, in the narrow part of the tube. The tube was removed from the thimble and, after cooling, was broken away from the metal. The metal was sawed to the approximate lengths required and weighed. The excess weight of the metal slug was further reduced until the weight calculated for the capsule was obtained.

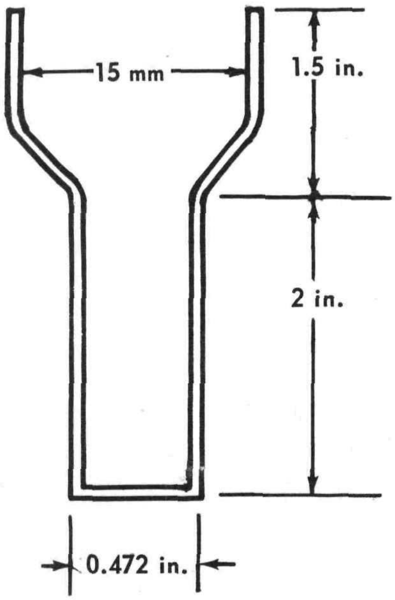


Figure 7. *Quartz Casting Tube*

The slug was placed in the inner capsule and the cap was welded into place by heliarc welding in a controlled atmosphere. The capsule was decontaminated, using wet abrasive powder, to 500 cpm of alpha activity and was placed weld-end first into the outer capsule. A disc of brazing foil 0.375 in. in diameter and 0.003 in. thick was placed on the inner capsule, and the outer cap was inserted.

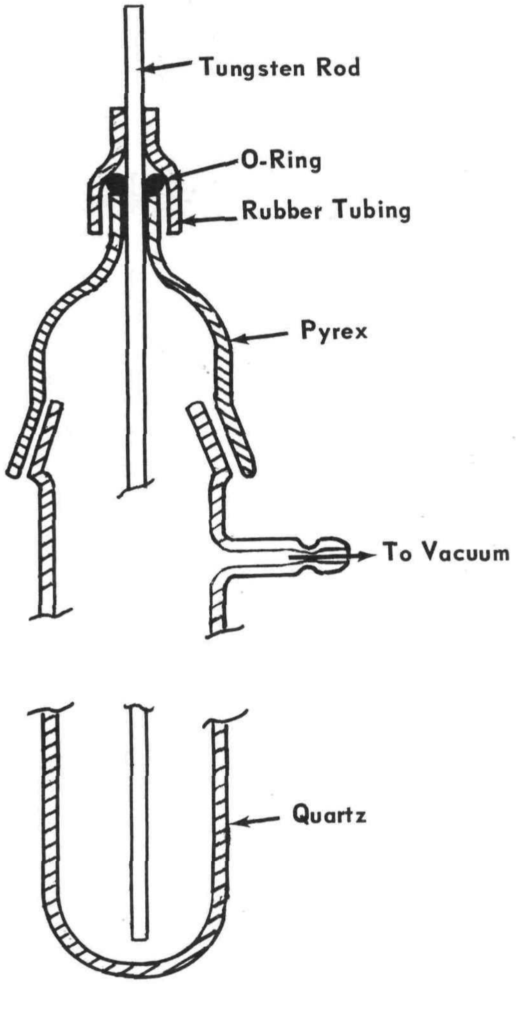


Figure 8. *Quartz Vacuum Thimble*

Both the bottom of the inner capsule and the underside of the outer lid had been plated prior to assembly. The plating consisted of flash-coating each part first with copper and then with silver. The flash coating on the mating surfaces allowed the braze to wet each surface adequately in the absence of a flux. The braze was expected to improve the transfer of heat from the fuel through the top of the capsule to the thermoelectric elements. The braze material was Lithobraze B.T. consisting of 71.8% silver, 28% copper and 0.2% lithium, with a melting point of 1435°F.

The outer cap was sealed into place by electron beam welding. The capsule then was placed flange down in a muffle furnace at 1450°F for 20 min to bond the inner and outer containers together with the braze and to melt and position the fuel into the bottom of the inner capsule. Location of the fuel in the capsule is illustrated by the radiograph in Figure 9. Finally the capsules were decontaminated, if necessary, to less than 10 cpm.

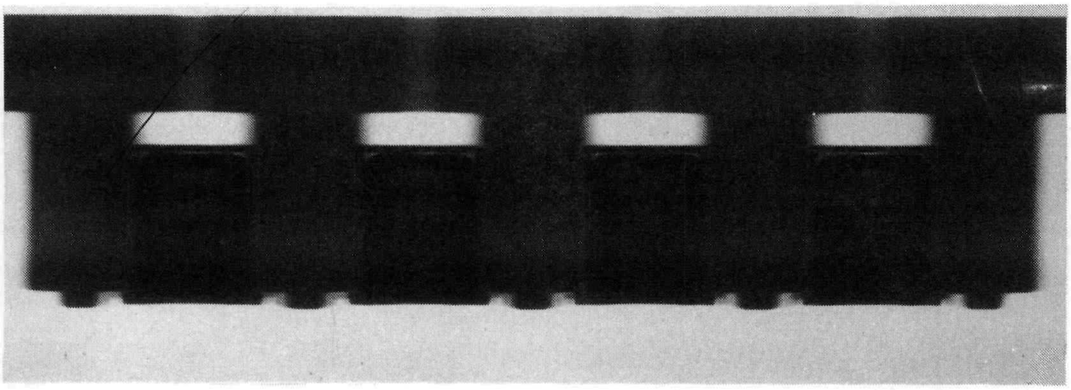


Figure 9. *Radiograph Showing Location of Fuel in Capsule*

The braze was used in the test capsules as well as in the fueled units. The inner and outer fueled capsules were also leak tested with the helium leak detector.

FUEL AND CAPSULE TESTING

Following the fueling of the capsules the testing of the fuel and capsules was started. First the capsule materials were analyzed using conventional analytical techniques. The composition of the tantalum-10% tungsten is given in Table 1. The Hastelloy C analysis is given in Table 2. The analyses for both metals were within the manufacturer's specifications.

Table 1

COMPOSITION OF TANTALUM-10% TUNGSTEN

Element	Content (%)	Element	Content (%)
W	10.5	H	0.0004
Mo	0.05	N	<0.0001
Al	0.008	O	0.0013
C	0.003	Cr	Trace
Mg	<0.0005	Ta	~89.4

Table 2

COMPOSITION OF HASTELLOY C

Element	Content (%)	Element	Content (%)
Ni	55.8	Si	1.0
Mo	17.7	Mn	0.5
Cr	15.6	Sn	0.3
W	4.83	Al	0.1
Fe	4.8	Cu	0.06
Co	1.06	Mg	<0.002
C	1.09		

The metallic plutonium fuel was also analyzed by spectrographic and chemical methods and by differential thermal analysis. A typical chemical analysis for elements in the fuel is given in Table 3.

The differential thermal analysis was obtained from both the heating and cooling curves for the sample. The curves were examined for peaks indicative of unwanted impurities. Typical curves for the samples are shown in Figures 10 and 11.

The heat content of the capsules was determined using a Mound conduction calorimeter. The heat outputs, in watts, for the six capsules fabricated for the program were 6.99, 7.03, 7.06, 7.28, 7.40, and 7.37. Accuracy of the measurements was ±0.2%.

Table 3

COMPOSITION OF METALLIC PLUTONIUM FUEL

Element	Content (%)	Element	Content (%)
Ag	Trace	Mg	0.093
Al	0.036	Na	0.063
B	Trace	Ni	0.005
Cr	Trace	Si	Trace
Cu	Trace	Ti	Trace
Fe	0.094	Pu	~99.7

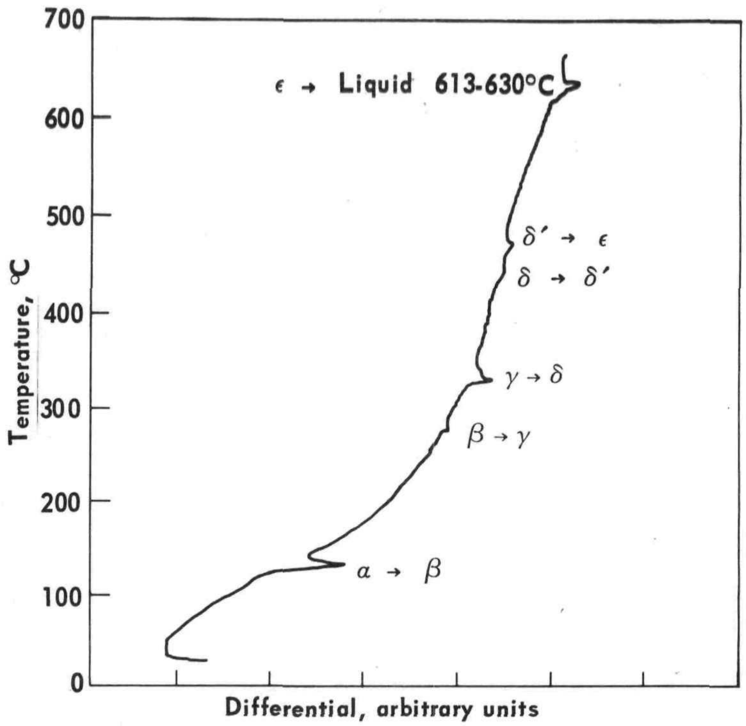


Figure 10. *Differential Thermal Analysis Curve (Heating) for Plutonium Metal*

The total neutron flux was measured against an NBS calibrated standard using a DePangher precision long counter. The flux values, in neutrons per second, for the six sources were: 8.7×10^4, 6.7×10^4, 4.8×10^4, 6.3×10^4, 4.6×10^4, and 4.5×10^4.

In the course of the program, 92 complete sets of capsules consisting of inner and outer units with caps and 36 inner capsules were used. Of these, 57 sets were used for the various environmental tests, and 6 were used for the fueled units. The remaining 29 sets plus 36 inner units were consumed in making welding studies, in preparing welding control samples and in evaluating the compatibility of the various materials

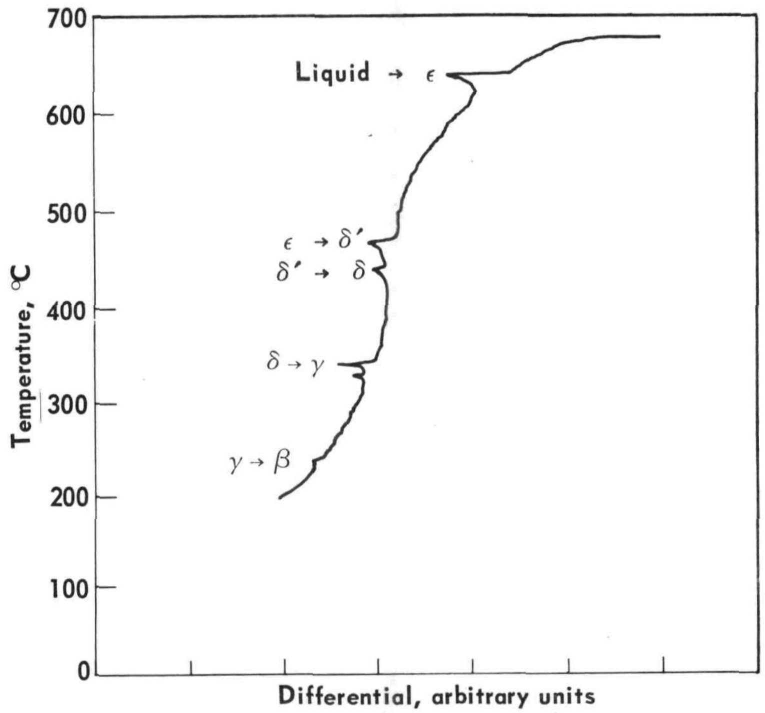


Figure 11. *Differential Thermal Analysis Curve (Cooling) for Plutonium Metal*

in the assembly. The fueled capsules were shipped in a container which was specially designed at Mound Laboratory to dissipate the heat of the capsules; it is shown in Figure 12.

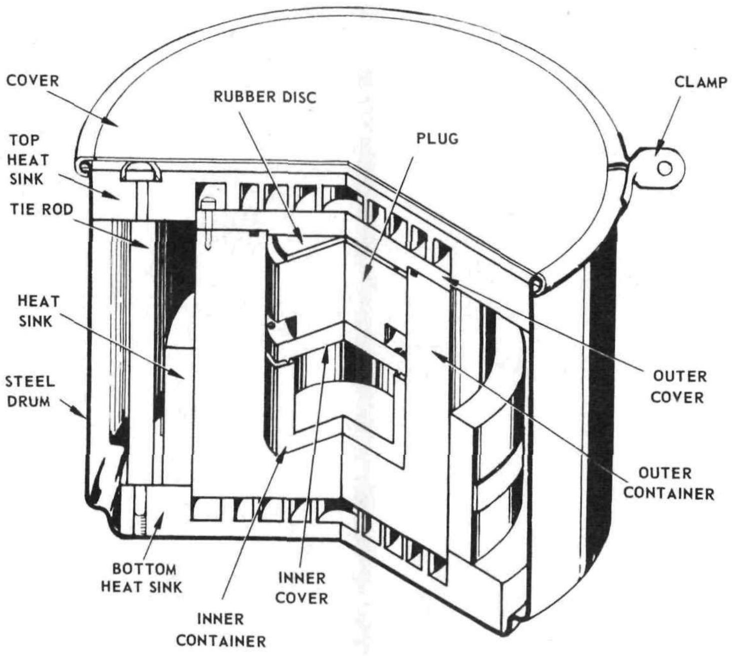


Figure 12. *Shipping Container for Fueled SNAP-15C Capsules*

In addition to the fabrication and testing work described above, a group of capsules to be used for crush tests was sealed with a small radioactive krypton-85 gas source in each one of them. The most minute fracture in the capsule wall or weld would be indicated by the release of gas during the test. The small gas sources were prepared by introducing krypton-85 into small copper tubes, then crimping and soft-soldering the ends of each tube. A heat shield was devised to protect the soft solder from the intense heat present during the welding of the cap and to prevent the premature release of the gas. The gas source was wrapped in quartz wool and placed in a small quartz tube. The tube was then wrapped with six layers of quartz wool and aluminum foil and placed in this heat shield in the bottom of the capsule. When the capsule was sealed it was heated to melt the soft solder and to release the krypton-85 into the interior of the capsule.

In addition to the safety tests performed on the empty capsules, a doubly encapsulated unit containing 16 g (7.3 watts) of plutonium metal was subjected to simulated fire test conditions. The unit was held at 1800°F for 2 hr in a furnace to determine the compatibility of the capsule materials with each other and the effect of heat on them. The cooled capsule was cut in half along its diameter under argon in a glove box. The fuel was removed to expose the interface of the fuel and the inner capsule material more clearly. The interface was polished and etched to determine if the molten plutonium had attacked the tantalum-10% tungsten metal. Figure 13 illustrates the good condition of the inside surface of the tantalum-10% tungsten capsule after it had been polished.

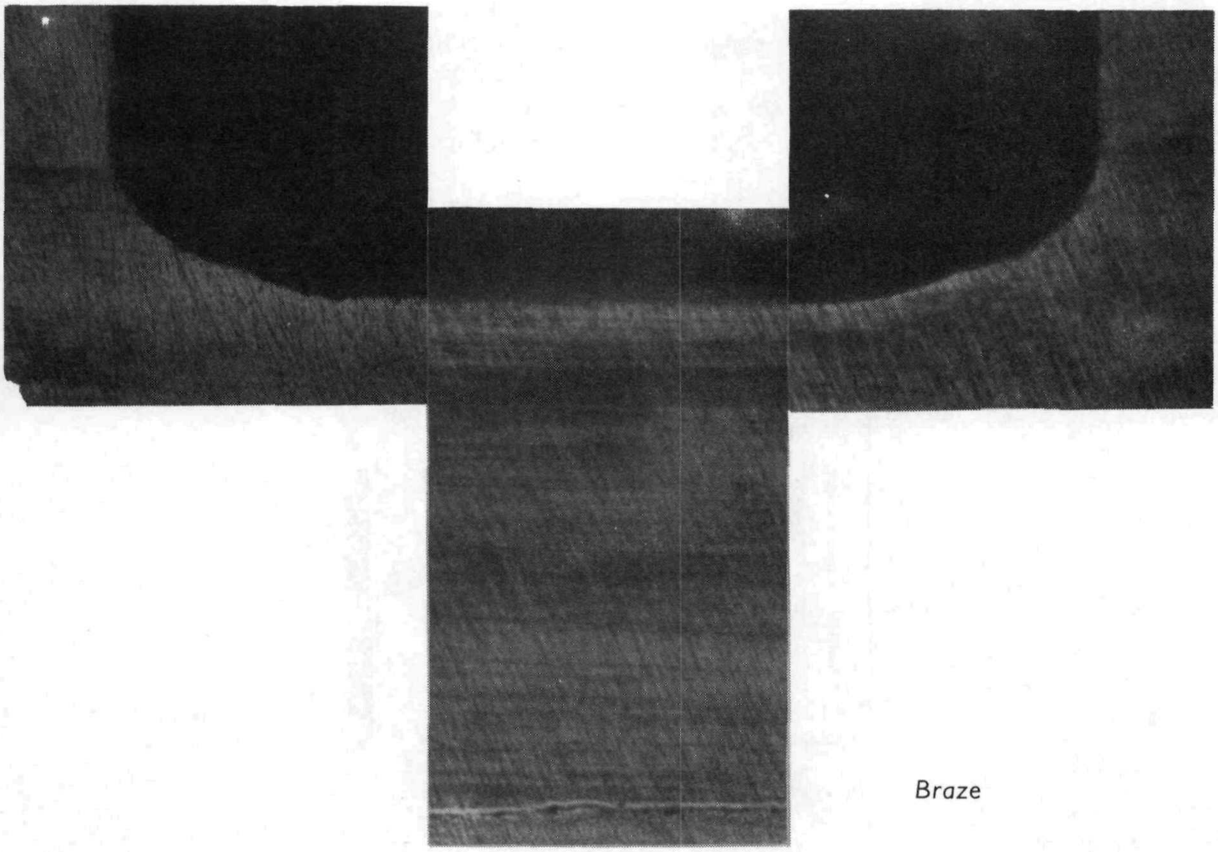
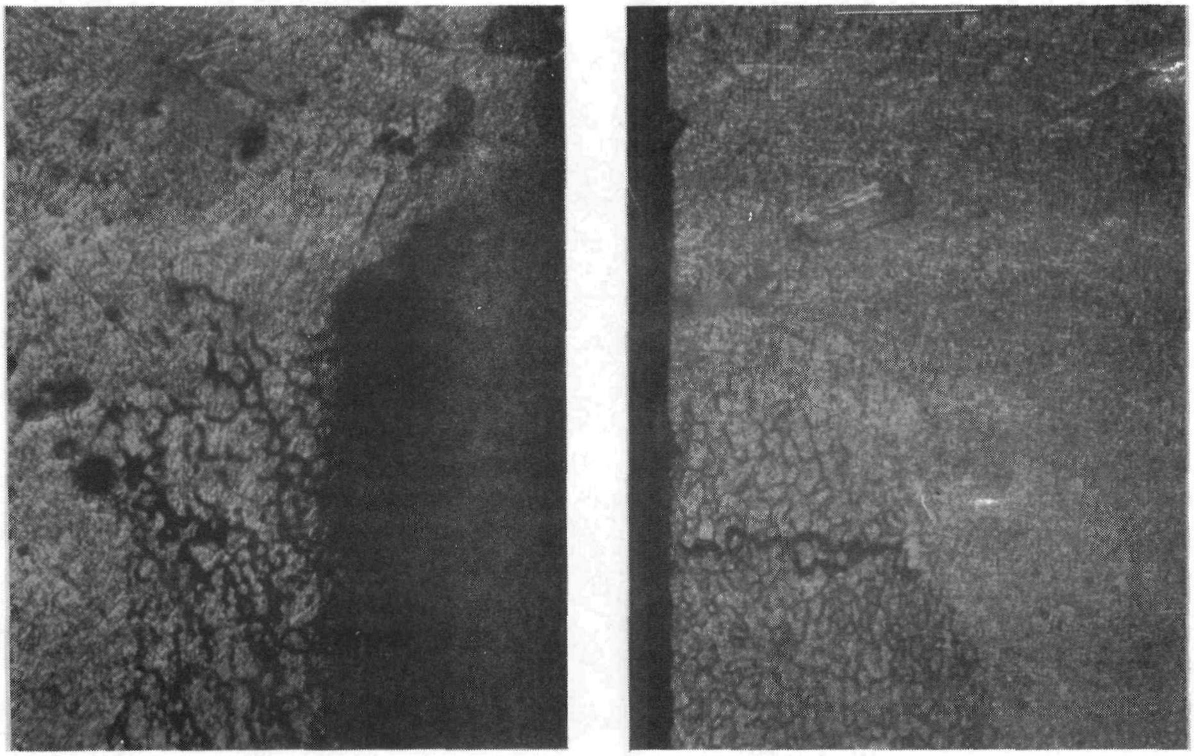


Figure 13. *Inner Bottom of Tantalum-10% Tungsten Capsule after Contact with Molten Plutonium at 1800°F for 2 Hours (Original Magnification 50X, Reduced 25% for Reproduction Purposes)*

The right and left inner side walls of the capsule were also examined to determine if corrosion had occurred. These areas are shown in Figure 14. The right-hand view is for the wall about half-way up the side



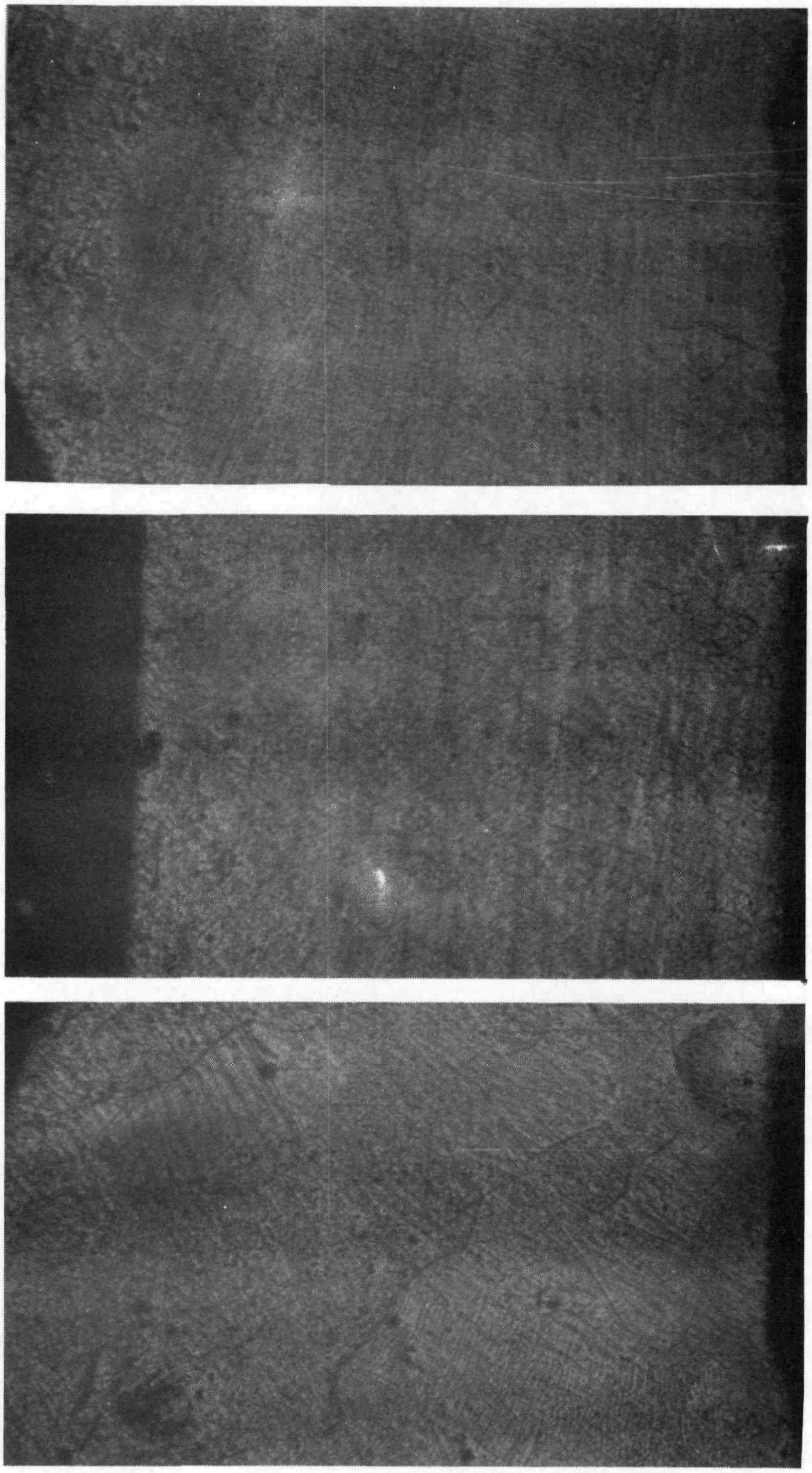
Left Side (50X) — Right Side (80X)

Figure 14. *Inner Side Walls of Tantalum-10% Tungsten Capsule after Exposure to Molten Plutonium at 1800°F for 2 Hours*

of the capsule, whereas the left view is for the wall very close to the TIG-welded top. The plutonium, after being remelted in the capsule, occupies about one-half of the capsule volume. The surface of the plutonium would then be in the vicinity of the apparent corrosion indicated in the right-hand view. The area shown in the left-hand view is above the plutonium surface. The change in granular structure may be the result of plutonium which had been deposited in this area during the fabrication, or which had crept up the wall during the fire test. Figure 15 shows the condition of the underside of the cap and weld at the top of the capsule. This section was not in contact with the molten plutonium. It was exposed only to plutonium vapor at a very low pressure. No attack of the metal is evident.

The outside weld on the Hastelloy C capsule (Figure 16) was inspected for defects after the heating. Although the weld was not exposed to plutonium during the test it was exposed to air. The weld does not appear to have been adversely affected by this exposure.

Additional work was also involved in the preparation and testing of the pressure burst capsules. To permit pressurization of the capsule a small-diameter tantalum-10% tungsten tube was sealed into the cap of the inner housing by electron beam welding. The cap was then sealed into the inner housing by heliarc welding in the same manner as the inner welds for the other capsules were made. The tube was inserted through a hole in the bottom of the outer capsule and the outer cap was welded into place. This assembly

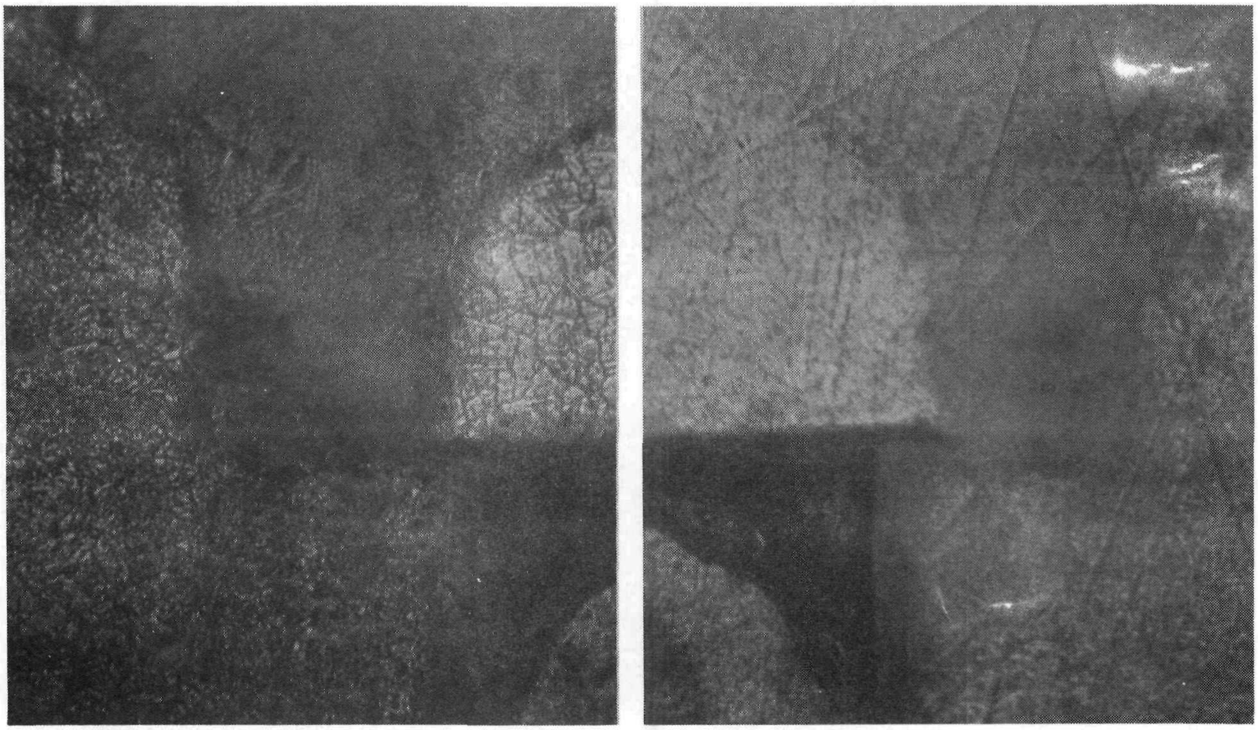


Right

Center

Left

Figure 15. *Underside of Cap and Weld of Tantalum-10% Tungsten Capsule after Exposure to Plutonium Vapor at 1800°F for 2 Hours (50X)*



Left *Right*

Figure 16. *Outer Weld on Hastelloy C Capsule after Being Heated for 2 Hours at 1800°F (80X)*

was suitable for room-temperature pressure testing. However, at elevated temperatures the tube had to be protected from oxidation to prevent it from breaking away from the capsule before the test was completed. To provide this protection a Hastelloy C tube was electron-beam welded to the bottom of the outer capsule. The inner tube was slid inside the Hastelloy C tube when the inner capsule was placed in the outer capsule. Figure 17 presents three views of the capsule. The right view is of the tube sealed to the inner

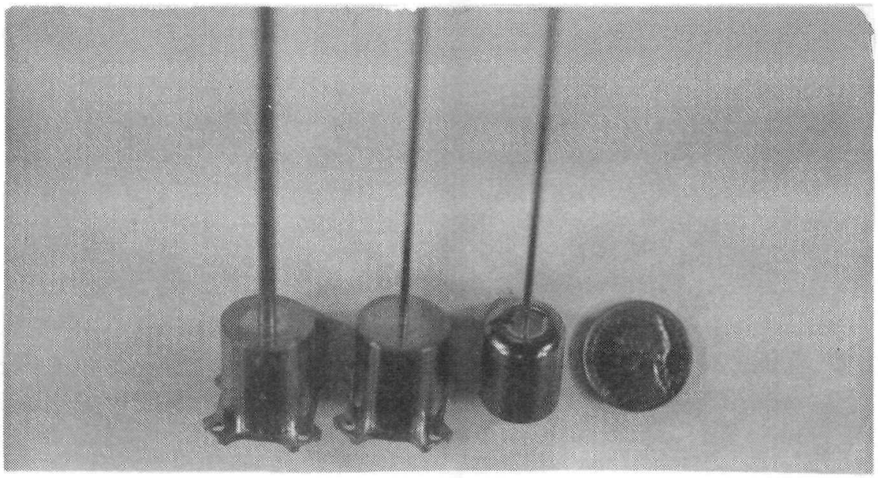
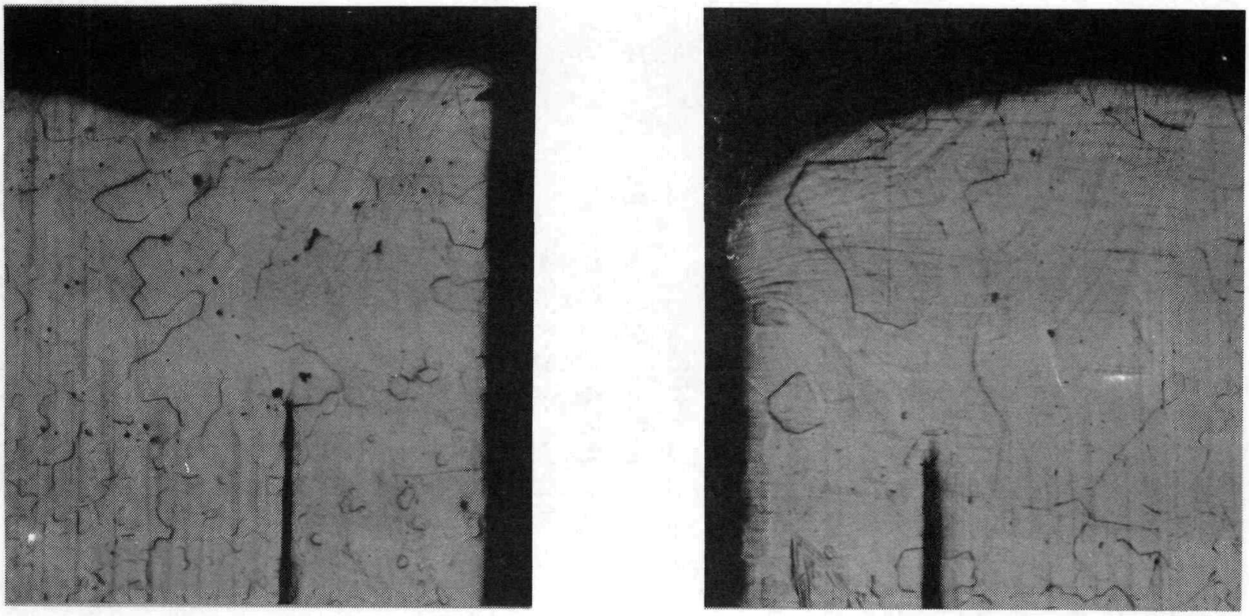


Figure 17. *Inner and Outer Capsules with Inner and Outer Tubes Attached*

capsule, the middle view is of the inner capsule after sealing into the outer capsule, and the left view is of the outer capsule with the protective Hastelloy C tube.

Figure 18 shows a metallographic cross-section of the tantalum-10% tungsten tube welded into the inner cap. The depth of weld penetration is equal to the wall thickness of the tube, which is satisfactory.



Left Right

Figure 18. *Metallographic Views of Tantalum-10% Tungsten Tube after Welding into Cap (76X)*

The capsules could not be tested for leaks using the test method employed for the other capsules. They were tested by introducing helium through the inner tube to a pressure of 300 psi while the capsule was immersed in acetone. Bubbles escaping into the acetone were searched for while the capsule was viewed horizontally during illumination with light at 90° to the line of vision. No bubbles were detected. Independent investigations at Mound Laboratory established that the escape of the finest bubbles was equivalent to a helium leak rate of 10^{-8} cc/sec, STP. This test could not be performed on the outer capsules because of the hole in the bottom through which the inner tube projected. However, all inner capsules for the pressure burst test were examined.

CONCLUSIONS

The preparation of the heat sources for the SNAP-15C program has demonstrated that integral welds can be obtained and reproducibility of fuel content can be achieved. The inspection procedures have maintained good control over the quality of the operations performed on the capsules. It is suggested that this program be considered in the preparation of sources for future SNAP programs.