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FABRICATION OF THREE 2500-WATT (THERMAL) STRONTIUM-90 HEAT SOURCES

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FABRICATION OF THREE 2500-WATT (THERMAL) STRONTIUM-90 HEAT SOURCES

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ABSTRACT

Three 2500-watt (thermal) heat sources were fabricated by the Oak Ridge National Laboratory (ORNL) for the purpose of fueling a 500-watt (electric) thermoelectric generator as part of the U.S. Department of Energy's Byproducts Utilization Program (BUP). Each of the sources, which are the largest ever assembled, consist of hot-pressed pellets of ^{90}Sr fluoride, doubly encapsulated in three Haynes-25 inner capsules and in a Hastelloy-S outer capsule. The total ^{90}Sr inventory of all three sources is 1.12 million curies.

The sources were fabricated at the ORNL Fission Product Development Laboratory (FPDL), which is a facility that is capable of processing multi-megacurie quantities of radioactive materials, chiefly ^{137}Cs and ^{90}Sr. The facility has been used for the fabrication of sealed sources of radioactive materials since the 1960's.

Since the campaign to produce the sources involved the application of existing source encapsulation technology which has been developed over the last twenty years, the production campaign was relatively straightforward, with some exceptions. These exceptions were chiefly due to the size of the source and the quantity of radioactive material involved. Source encapsulation hardware was supplied by the thermoelectric generator manufacturer, Teledyne Energy Systems, which also developed, together with ORNL, the required quality assurance criteria. These criteria outlined the use conditions, source geometry and tolerances, type of quality control used in the fabrication, and acceptable source performance.

The source was tested to determine compliance with all of the IAEA Safety Series No. 33 requirements. The source fabrication, assembly, and testing will be described in the presentation.

THREE 2500-WATT (THERMAL) heat sources were fabricated by the Oak Ridge National Laboratory for the purpose of fueling a 500-watt (electric) thermoelectric generator as part of the U.S. Department of Energy's Byproduct Utilization Program. The sources, which are the largest ever assembled, were fabricated at the ORNL Fission Product Development Laboratory, which is a large, multi-megacurie, radioactive materials processing facility. The FPDL, shown in a cutaway view in Figure 1, has been involved in fission-product purification and source-fabrication activities for the past 25 years. The facility produced the original strontium sources as part of the Systems for Auxiliary Nuclear Power (SNAP) program and continues to produce strontium heat sources as part of the Isotope Distribution Program.

SOURCE DESIGN

A cross-sectional view of the heat source is shown in Figure 2. It consists of an outer strength member which encloses three liners, which in turn enclose the strontium fuel. Teledyne Energy Systems, Inc., (TES), in cooperation with ORNL was responsible for this source design concept, which is similar to those which ORNL has produced in the past.

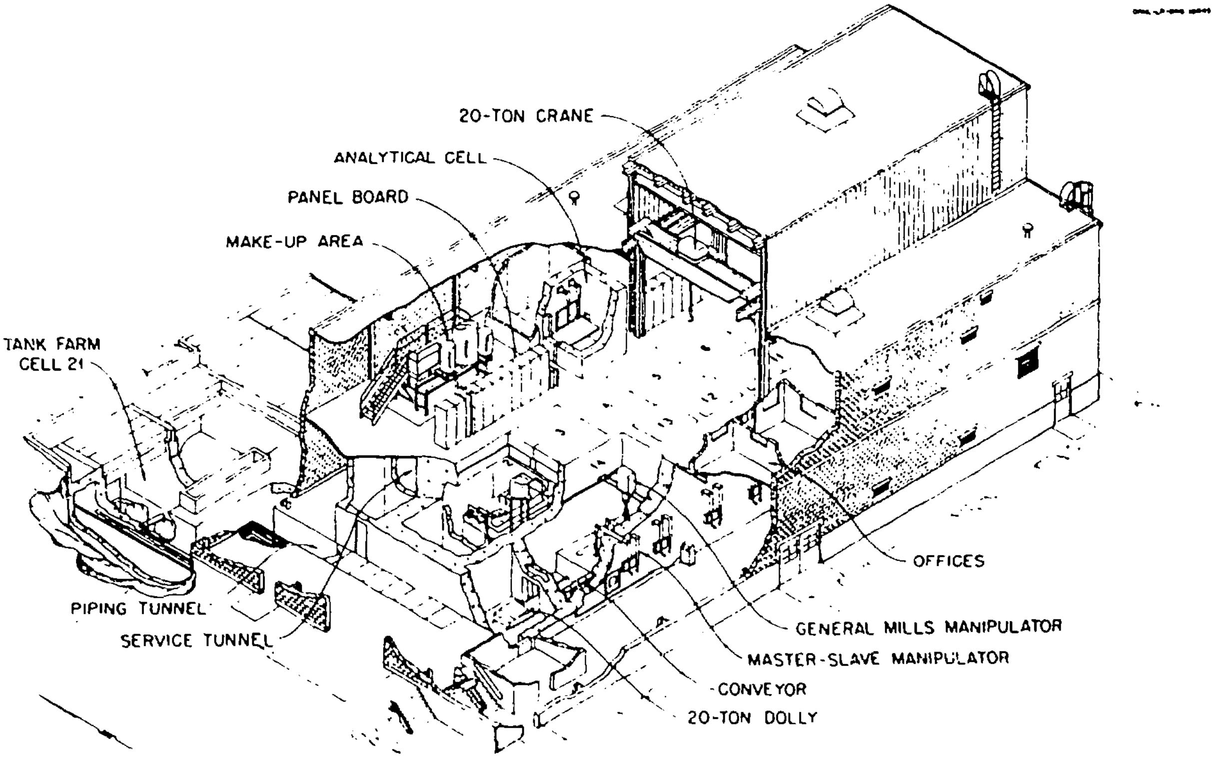

Figure 1. FPDL Facility.

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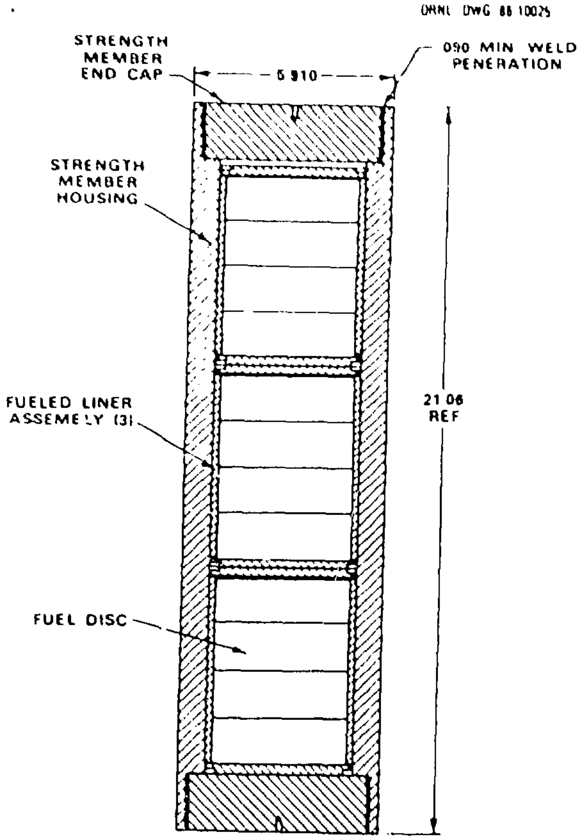

APPROXIMATE FUELED WEIGHT-112 lbs

Figure 2. Cross Section of BUP 2500-Watt Heat Source.

In cooperation with ORNL, TES produced a heat-source specification which covered the design details and quality assurance requirements. A listing of most of these requirements is presented in Table 1.

Table 1 - Source Design Requirements

Fuel	Strontium-90	
Chemical form	$^{90}SrF_2$	
Composition	As synthesized and stored at WESF	
Quantity	Hot-pressed pellets 3.97-in.-O.D. 3.7 to 4.0 g/cm^3 density	
Thermal inventory	Liner	- 833 W_{th} ±3%
	Heat source	- 2500 to 2575 W_{th}
	RTG	- 7500 to 7650 W_{th}
Liner components	AMS 5759 specification Haynes-25 bar stock	
Strength member components	AMS 5711 specification Hastelloy-S bar stock	
Weld test specimen acceptance	Cracks	- no visible cracks
	Leakage	- 1 x 10^{-6} std cm^3 criteria
	Penetration	- liners 0.030 in. str. mbrs. 0.090 in.
	Linear defects total	- <0.015 in.
Production leak testing	1 x 10^{-6} std cm^3 He/s	
Qualification testing	IAEA Safety Series 33	

The chemical form of the fuel is ^{90}Sr fluoride (SrF_2), which is produced at the Waste Encapsulation and Storage Facility (WESF) located at Hanford, Washington. Strontium-90 decays by beta emission to ^{90}Zr and is then contained in the matrix of the fuel. The fuel used in the fabrication of the BUP heat sources contained approximately 19% zirconium fluoride. At FPDL, this material was hot-pressed in a vacuum to produce a fuel pellet that had approximately 90% of theoretical density. The fuel was encapsulated in a liner made of Haynes-25 alloy, which was chosen based on compatibility studies done at Pacific Northwest Laboratories (PNL) - (1).* Haynes-25 was the recommended material at the approximately 900°C use temperature. The three liners were further encapsulated in a Hastelloy-S outer capsule, which was also selected based on PNL recommendations - (2). The liners function as a contamination barrier as well as the inner encapsulation. The strength member serves as a secondary contamination barrier as well as serving as the primary pressure-resisting member.

Source welding methods were at the discretion of ORNL as long as the weld acceptance criteria listed in Table 1 were met. Since nondestructive analysis of the weld joint on each source was not practical, weld test specimens were used to prove weld quality for both the liners and strength members. These specimens duplicated the weld joint on the liners and strength members in every detail.

A weld development program was conducted to define weld parameters, such as starting current, weld current, tail-off current, overlap, and rotational speed for both the liners and strength members. The specification required the welding of four test specimens in a continuous manner with each specimen meeting the weld acceptance criteria before the weld program was certified.

Production welding required a continuous operation using the same welding equipment and parameters established in the weld development program. One test specimen was welded and then destructively analyzed by the methods described below to assess weld quality prior to making any production welds. The weld production run consisted of a batch of five fueled pieces (maximum). Weld test specimens were welded before and after each fueled piece. The last weld test specimen was required to be destructively analyzed per batch for liner welds; however, for strength-member welds, each test specimen was analyzed. If all analyzed test specimens met the weld acceptance criteria, then they were certified. If a test specimen failed to pass analysis, then the fueled piece welded immediately before the failed test specimen was rejected, and the next test piece in sequence was analyzed. If it passed the analysis, then the welds were certified. If not, the process was repeated until a satisfactory test piece was found.

A quality assurance program was conducted to ensure compliance with the specification. This program plan is inherent at ORNL and is the same with all sources that ORNL fabricates. The specification requires that certain information be documented. This information includes fuel chemistry and quantity, liner and strength member weld quality and penetration, surface contamination level on the exterior of the strength member, and the results of the leak tests and metallographic examination. The weld acceptance tests consisted of a dye-penetrant examination of the weld area, a helium leak test with a sensitivity of 10^{-8} std cm^3/s He, and a metallographic examination of each of the four sections cut from the specimen at 90° angles.

Helium leak tests of the fueled sources were done using a procedure involving "bombing" each liner or strength member in an atmosphere of helium for a period of time and determining its leak rate in a vacuum

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*Numbers in parentheses designate References at end of paper.

chamber using a mass spectrometer leak detector having a sensitivity of 10^{-8} std cm³/s.

The heat source capsule design was qualification tested for physical integrity, using the test methods laid out in the publication Safety Series No. 6. (13) A capsule identical in all respects to the actual heat source capsules was fabricated for use in the test series. A dummy heat source was assembled using a simulant for the strontium fuel and the same welding procedures as the actual heat sources. This dummy heat source was subjected to the test series. Helium leak checks were made before and after each test to determine the effect on the source leak rate. A source leak rate in excess of 1×10^{-6} std cm³/s was the criterion for source failure. The test conditions and dummy heat-source leak rates are summarized in Table 2.

Table 2. Qualification Testing Results

Test	Condition	Leak Rate (atm cm³/s) Before	Leak Rate (atm cm³/s) After
Impact	9-m free fall	2.5×10^{-7}	2.4×10^{-8}
Percussion	Impact of a 1.4-kg billet from 5 m	2.4×10^{-8}	1.1×10^{-8}
Thermal	Heat to 800°C and hold for 30 min.	[illegible]	2.5×10^{-7}
Thermal Shock	Heat to maximum operating temperature (800°C) and then quench in 0°C water	4.2×10^{-7}	[illegible]
Pressure	Subject to a pressure of 1000 bars (15 Kpsi)	1.5×10^{-7}	8.0×10^{-8}

During the testing, two situations developed which required adjustment in the testing procedure. An additional thermal shock test was necessary because the first test did not reach the maximum operating temperature. The impact test caused one end of the dummy heat source to become 0.06 in. out of round. It was necessary to remove the out-of-round portion so the dummy heat source could enter the pressure-testing chamber.

As can be seen from the leak test data in Table 2, the highest leak rate measured was 4.2×10^{-7} std cm³/s. Since the result was less than the 1×10^{-6} std cm³/s required by the IAEA test, it was concluded that the dummy heat source met the IAEA criteria.

SOURCE FABRICATION

The exterior of the manipulator cells containing the source fabrication equipment used is illustrated in Figure 3. This equipment consists of a vacuum hot press, powder shipping-can opening and handling equipment, welding and leak-testing equipment, and a calorimeter. Special source-handling equipment was also required as the completed source weight (112 lb) was greater than the weight specifications of the facility manipulators (25 lb). The equipment used to fabricate the 2500-watt heat sources consisted primarily of the same source fabrication equipment which has been developed and used for several years at ORNL with some exceptions. This equipment is described below.

The vacuum hot press, illustrated in Figure 4, was used to fabricate the pellets. It consists of a vacuum-tight enclosure which is water-cooled, is heated with a graphite resistance heater, and has a hydraulic ram attached. The hot-press system has been developed over the years at ORNL and is of a standard design which has been modified for in-cell use. The 3.97-in.-diameter

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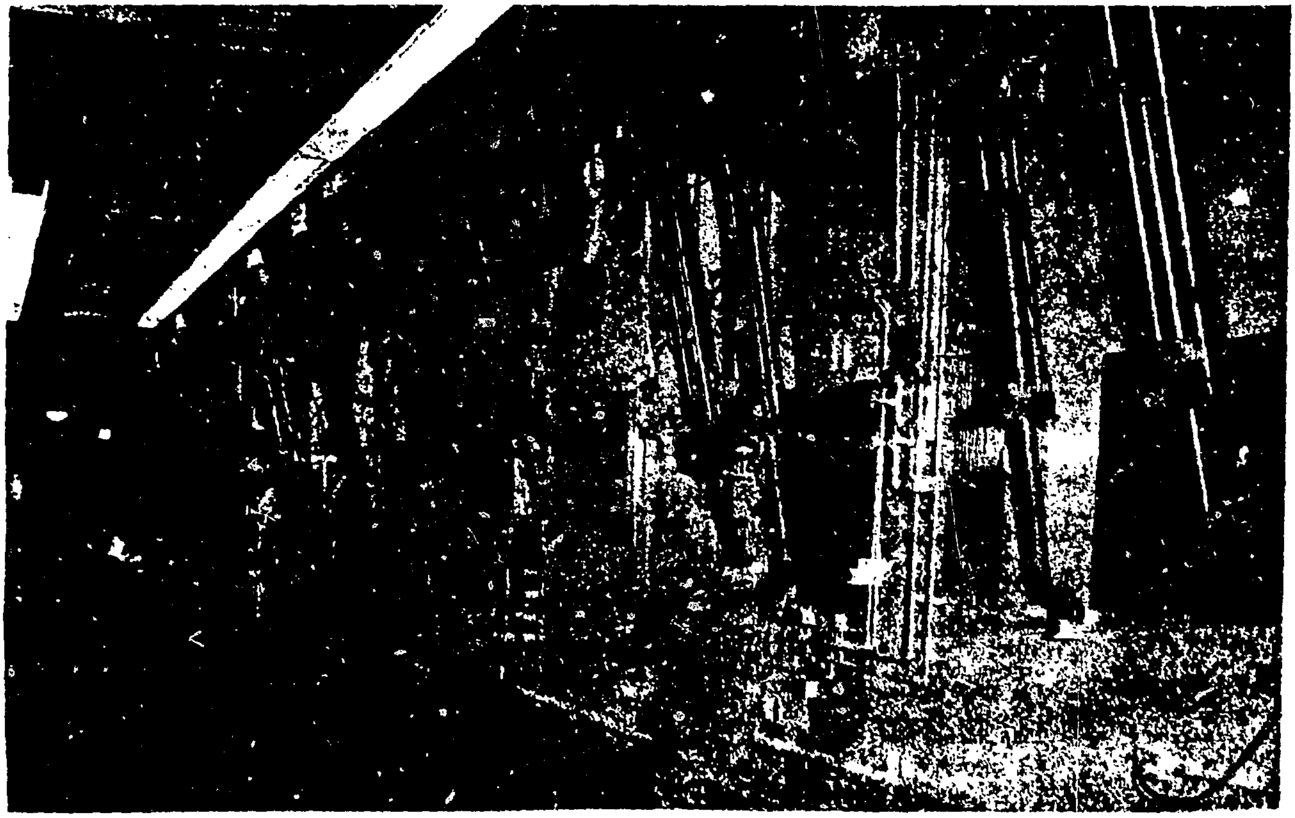

Figure 3. Manipulator Cells 10, 11, and 12 at ORNL.

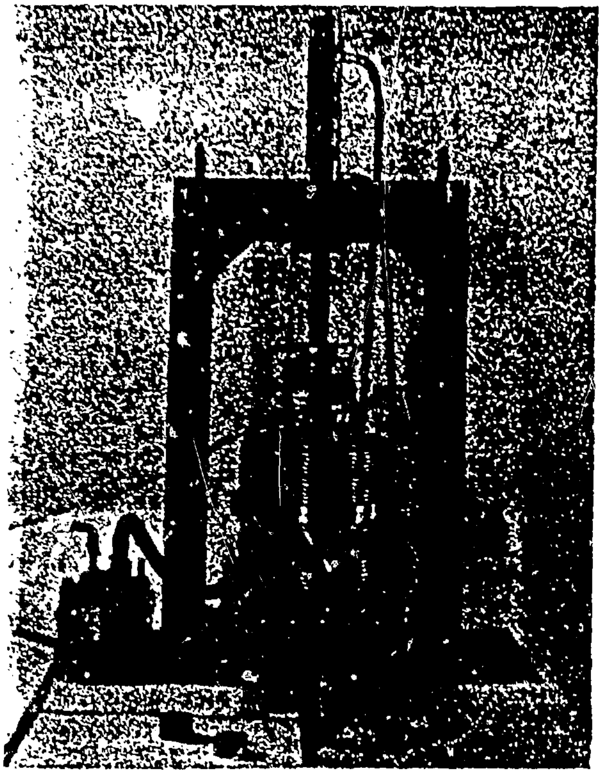

Figure 4. Vacuum Hot Press.

pellet is the largest that can be fabricated in the system. The pellets are cast in a graphite die body which consists of an inner liner made of Type ATJ graphite and an outer casing made of Carbitex™ spun graphite fibers. The operating conditions can be varied from 0 to 10,000 psi in hydraulic ram pressure and ambient to 1600°C temperature. Hot-pressing conditions chosen were 5000 psi and 800°C. The temperature was varied during the campaign to produce specific pellet densities when required.

The calorimeter, which was used to determine the thermal inventory of the sources, consists of an inner chamber surrounded by a water jacket. The inner chamber is thermally insulated from the water jacket except for two stainless steel rings. The rings contain thermocouples, separated by a known distance and are wired together in a thermopile. Flow is controlled at one gallon per minute through the water jacket, and with the heat source in place, it is assumed that the entire heat flow is through the steel rings. The output of the thermopile is measured on a digital voltmeter and recorded. When thermal equilibrium is reached, the millivolt value of the thermopile is read, and then a power output is computed from a predetermined power-vs-millivoltage curve. This curve is derived from a calibration with an electric heater. A diagram of the calorimeter is shown in Figure 5.

The fixtures used for welding both the liners and strength members are identical in form but different in size. They each consist of a turntable which is attached to a torch holder that is movable in both the radial and "Z" (up-dowo) directions. Rotational speed is controlled by the use of a speed controller. The joint centering and the electrode-to-work distance are established visually. A standard tungsten-inert-gas (TIG) weld was done on the strength members because of the penetration requirements. A photograph of the liner welder is shown in Figure 6.

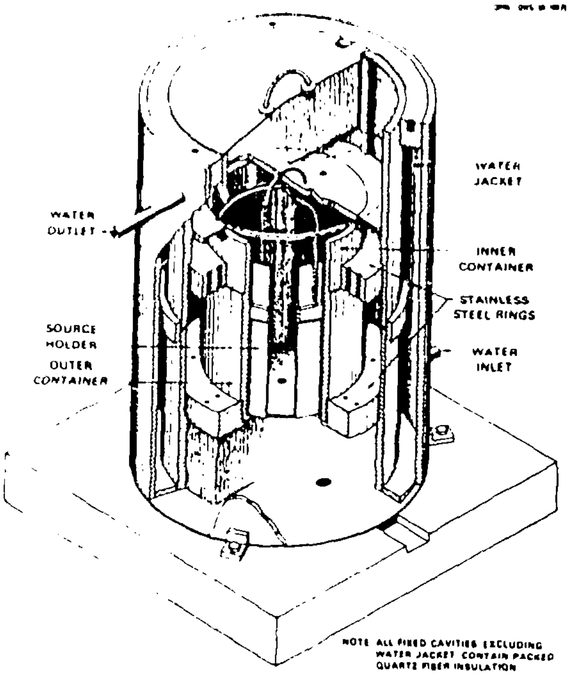

Figure 5. Calorimeter.

Helium leak testing of both the liners and strength members was done. Two sets of pressurization and leak-test vessels were used because of the different sizes. The technique consists of "bombing" the liner or strength member in an atmosphere of helium under positive pressure in one vessel. The tested piece is then taken out of the pressurization vessel and placed into the leak-test chamber where it is tested using a helium leak detector located outside the hot cell.

Although the liners were able to be handled by standard methods, special source-handling equipment was required for handling the strength members since they were too heavy for the manipulators. One piece of this equipment consisted of a dual choker which allowed the handling of an empty strength member in cell prior to loading it. An in-cell hoist was required to be installed in Cell 10E in order to handle the components. Since the transfer port from Cell 10E to 10W is only 12 in. square, while the strength members are 21 in. long, a problem existed in transferring the sources between cells. This was solved by constructing a transfer port insert consisting of an 8-in.-diameter tube, hinged in the middle, and lockable in the horizontal plane. The strength member could be loaded into the tube vertically, the tube turned horizontally, moved through the transfer port, and then uprighted. The strength member could then be removed vertically.

A flow sheet of the movement of materials between cells is shown in Figure 7. This illustrates what operations are required in each cell and the flow of material between hot cells. The vacuum hot press is located in Cell 12; powder handling, weighing, and shipping-can opening equipment are located in Cell 11. Liner welding equipment, a water-cooled storage well, and an in-cell air hoist are located in Cell 10E. All operations were not carried out simultaneously as all

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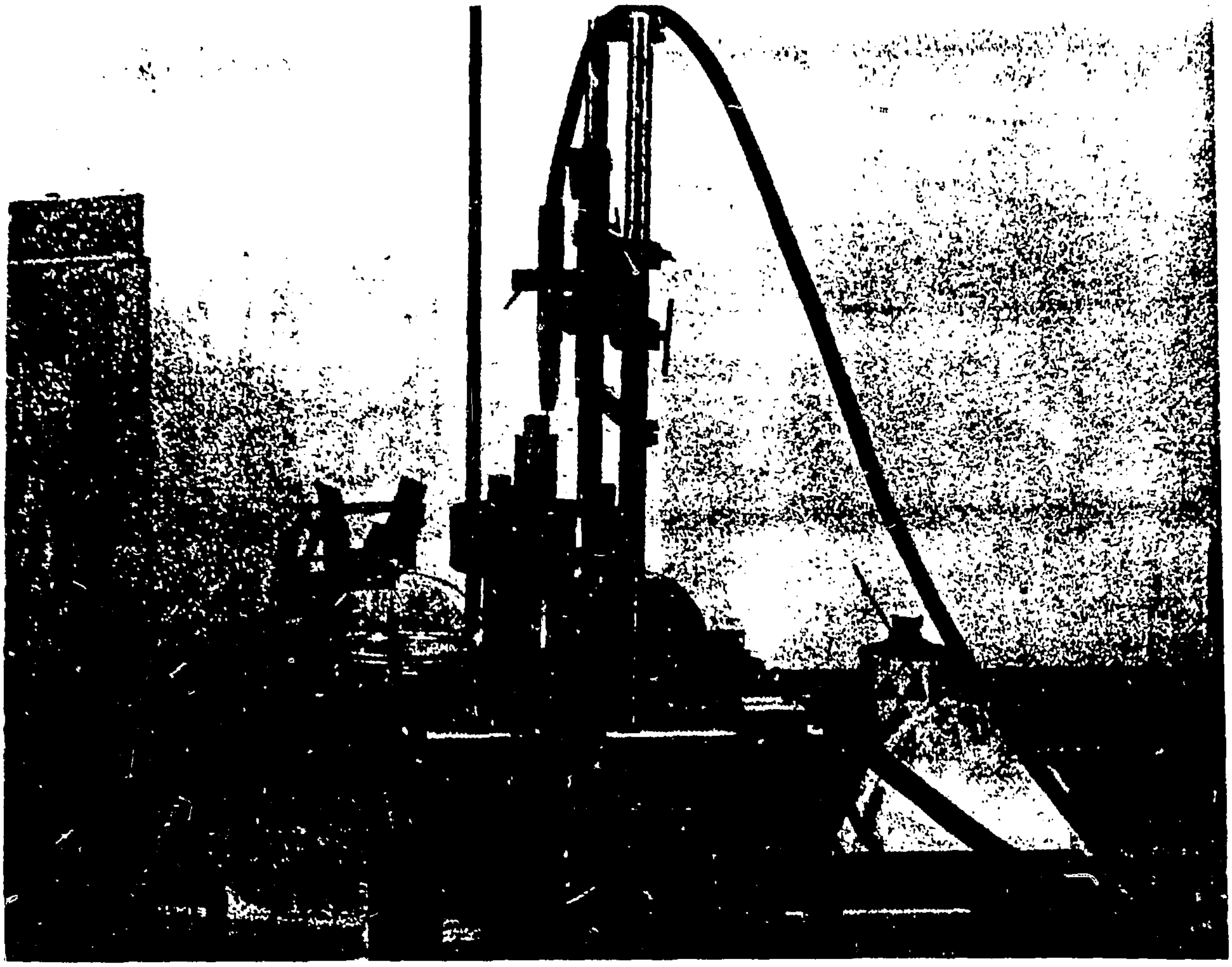

Figure 6. Liner Welder.

equipment could not be contained in the cells at the same time. Generally, the operations were conducted in such a way as to produce the liners in one campaign and the strength members in a separate campaign.

The first operation was to cut open the WESF cans to remove the strontium fluoride. The strontium fluoride was crushed and loaded into the graphite die body which was then loaded into the hot press. After pressing, the resulting pellet was ejected from the die and then loaded into a liner. This process was repeated until the liner was filled with four pellets. This required the WESF can-opening powder-handling equipment to operate along with the vacuum hot press. The end cap was then inserted, and the liner was welded, decontaminated, and assayed in the calorimeter. The liner was then leak tested and stored in the water cooled storage well in Cell 10W. Since there was some variability between the WESF can calorimetry and the actual liner heat output, the calorimetry results from a given liner were used to adjust the fuel loading of the next liner in sequence. Liner data are listed in Table 3.

After all liners were completed, the strength members were assembled. The liners were moved into Cell 10F where they were loaded into the strength member, the end cap was screwed in, and the source was welded. The source was then leak tested and stored. This continued until all three sources were completed.

Table 3. Liner Data

Liner No.	Powder Weight (g)	Calorimetry (W)*	Curies* (000)
85-BUP-1	3726.5	783	115.1
85-BUP-3	4046.5	823	121.0
85-BUP-4	4211.1	898	132.1
85-BUP-5	3863.4	826	121.5
85-BUP-6	3955.3	843	124.0
85-BUP-7	4046.0	855	125.7
85-BUP-8	4004.8	839	123.4
85-BUP-9	3841.3	849	124.9
85-BUP-10	4080.2	905	133.1

*As of September 30, 1985.

Data for the resulting sources are shown in Table 4.

Table 4. Source Data

Source No.	Liner Nos.	Calorimetry (W)
85-BUP-S1	1-7-10	2543
85-BUP-S2	6-8-9	2531
85-BUP-S3	3-4-5	2547

As is commonplace in activities such as this, several problems were encountered. One problem was

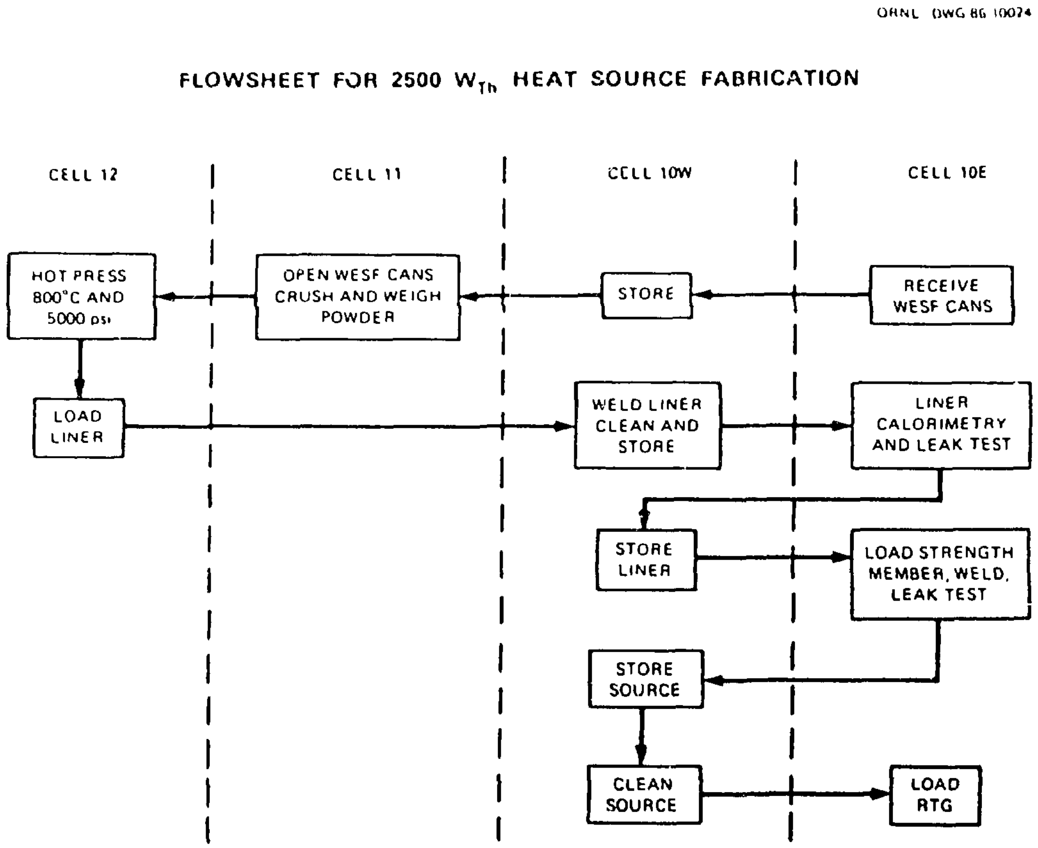

Figure 7. Source Fabrication Flow Sheet.

that of liner welding. Initially, the liners were handled without thermal protection, and the high heat loading produced oxidation in the weld area prior to welding. This was caused by the time (three days) the liner was stored during pellet pressing and insertion. It was thought to be due to the formation of an adherent coating of chromium oxide. This caused the metal not to flow properly during welding. The first two liners fabricated exhibited this problem with the second failing to weld. The problem was solved by fabricating a copper jacket to slip the liner into during the pellet pressing, insertion, and storage to conduct the heat away from the weld lip. Also, an insert was fabricated to fill the inside cap recess at the weld lip to prevent strontium powder from scraping off the pellet during insertion, thus filling the crack between the liner cap and liner. These techniques resulted in the successful welding of subsequent liners.

Another problem encountered was that of variations in calorimetry between the WESF and ORNL. This was significant in that a -5.0 to +7.0% variation was detected and made it difficult to meet the ±3.0% required by the specification. As shown in Table 3, three liners were out of the 808 to 858-W range specified. A deviation was approved by TES since the liners were able to be arranged to meet the total source and RTG thermal inventory specification. Adjustments to strontium specific activity were made during the fabrication of the sources; i.e., the calorimetry results of one liner were used to estimate the correct specific activity of strontium used in subsequent liners in order to encapsulate the proper amount of material. By far, the most serious problem occurred during the welding of the strength members and resulted in the failure of one strength member to weld properly. The problem was caused by stoppage of the welding turntable during the welding of the source. The stoppage was caused by having too long a residence time of the heat source on the welding turntable. This caused binding of the mechanism that drives the turntable due to the heat generated from the source. The source had a burned spot on one part of its circumference and four other blowholes resulting from a subsequent weld pass. Three subsequent weld passes were made to attempt to close the weld, without success.

Because of the size of the source, no equipment existed at ORNL which could have cut open the strength member while retaining the integrity of the liners. Because of this fact, an effort was made to salvage the source by patching the blowholes and proving that the weld met the quality assurance criteria. The blowholes were patched using the following procedure. First, a blowhole was positioned under the plasma-arc welding torch. Then the torch was started, using the parameters shown in Table 5.

Table 5. Patch Welding Parameters

Starting current (amp)	Ramp time (s)	Weld current (amps)	Plasma gas (%)	Shield gas (%)
10	15	70	65	70

Certified Hastelloy-S wire was then added to the work by holding the wire with a manipulator and feeding it in while watching the operation through a telescope. This was continued while the hole was filled. Then the source was rotated to the next hole, and the process repeated. When the last hole was filled, the original weld program (140-amp welding current) was run three times over the patched weld area. When the weld current was insufficient to adequately fuse all of the patched weld metal, one pass was made keeping the same parameters as before but manually raising the weld current to 150 amp in the area of the patch. Two passes were made at this setting. When this current setting proved insufficient, the program was run raising the current to 170 amps in the area of the patch. Two passes were made using this technique. A photograph of the resulting heat source is shown in Figure 8.

Prior to welding the heat source, the technique just described with the exception of raising the weld current at the patch was tried on a weld test specimen which was subsequently examined. Table 6 shows the results of this examination.

Table 6. Results of Test Piece Examination

Helium leak test ($atm-cm^3/s$)	Dye penetrant test	Metallographic section number	Examination penetration (mils)
		A1	100
		A2	110
		B1	105
0.6×10^{-8}	no	B2	120 (patch)
	visual	C1	95
	defects	C2	100
		D1	90 (crack)
		D2	100

The data were reported to the special review board as called for in the heat-source specification, which, after review, determined that the patch was not acceptable for use in the RTG. The reason for the decision was that the weld test specimen analyzed did not adequately represent the weld as it occurred on the source.

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Figure 8. Heat Source 85-BUP-S1 Showing Patched Weld.

Additional work must be done to correct the defective strength member. This consists of cutting open the strength member and its refabrication. RTG loading and testing will be performed by TES personnel on-site at ORNL.

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