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## SNAP PROGRAMS QUARTERLY PROGRESS REPORT NO. 1

Task 8—Strontium-90 Fueled  
Thermoelectric Generator Development,  
November 1, 1960 Through January 31, 1961

June 1961  
[OTI Issuance Date]

Nuclear Division  
Martin Company  
Baltimore, Maryland

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SNAP Programs  
Quarterly Progress Report No. 1  
Task 8--Strontium-90 Fueled  
Thermoelectric Generator Development  
November 1, 1960, through January 31, 1961

This report is prepared  
under Contract AT(30-3)-217  
with the U.S. Atomic Energy  
Commission

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## FOREWORD

This quarterly report covers the period from October 18, 1960, through January 31, 1961, and has been prepared under the requirements of Contract AT(30-3)-217 with the Atomic Energy Commission, as set forth in Statement of Work MN-SW-1013, Revision A, November 30, 1960, Paragraph 1.6.2 of Part I, and Paragraph 1.5 of Parts II, III and IV.

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## SUMMARY

The SNAP 7 program is being conducted for the purpose of developing four radioisotope-fueled thermoelectric power generation systems. An important phase of this program is the processing of Strontium-90 into heat sources for these systems.

The current effort involves the design and engineering analysis of two of the four thermoelectric generators. Fuel process flow and associated equipment requirements for remote conversion of Strontium-90 feed material to strontium titanate pellets are covered. Further, technical standards have been evolved concerning raw fuel material specification.





## I. INTRODUCTION

The SNAP 7 Program covers:

- (1) Design, fabrication, test and delivery of four radioisotope-fueled thermoelectric generator systems to meet the rigorous environmental requirements of field use by the United States Coast Guard and United States Navy.
- (2) Fabrication of the Strontium-90 fuel for two of the aforementioned generators.

The four deliverable generator systems under the contract are as follows:

- (1) SNAP 7A: 5-watt electric generation system for U. S. Coast Guard light buoy, Subtask 8.1.
- (2) SNAP 7B: 30-watt electric generation system for U. S. Coast Guard fixed light station, Subtask 8.2.
- (3) SNAP 7C: 5-watt electric generation system for U. S. Navy weather station, Subtask 8.3.
- (4) SNAP 7D: 30-watt electric generation system for U. S. Navy boat-type weather station, Subtask 8.4.

Fuel processing was isolated as a separate subtask, Subtask 8.5, after initiation of the program, to permit a more detailed surveillance of this aspect of the program. This report has been divided into three major sections: one covering Subtask 8.1 and 8.3, one for Subtask 8.2 and 8.4 and one for Subtask 8.5; however, it should not be overlooked that this is a highly interrelated program where variations in any subtask may produce significant effects in one or more of the others.



## II. SNAP 7A AND 7C FIVE-WATT ELECTRIC GENERATION SYSTEMS--SUBTASKS 8.1 and 8.3

### A. INTRODUCTION AND SUMMARY OF SIGNIFICANT TECHNICAL ACHIEVEMENTS

The SNAP 7A and 7C generators were engineered during this period. This, though simply stated, is perhaps the most significant single technical achievement covered by this report. Technical milestones leading to this achievement were the successful completion of the following:

- (1) Formulation and selection of a conceptual design.
- (2) Preparation and release of engineering drawings to Manufacturing and Procurement.
- (3) Coordination with using agencies to ensure satisfaction of their detailed requirements.

The generators, as designed, featured many interesting aspects, such as:

- (1) Employment of integral thermoelectric modules, each of which consists of several prewired and pretested thermoelectric couples.
- (2) Use of a four-capsule fuel block.
- (3) Use of depleted uranium as shielding material in the combination fuel shipping cask and generator shield.
- (4) Control of thermal-to-electrical conversion efficiency by adjustment of the thermal conductivity of insulation.

### B. ENGINEERING--EQUIPMENT DESCRIPTION, DESIGN TECHNIQUES AND PROCEDURES, AND TEST FOR SNAP 7A AND 7C\*

The Task 8 program is directed toward the design, development and test of auxiliary power systems for a variety of applications. The objectives of Subtasks 8.1 and 8.3 are to analyze, design and fabricate SNAP 7A and 7C thermoelectric systems.

---

\*J. Keenan

The SNAP 7A system consists of a 10-watt thermoelectric generator, a dc-to-dc converter and rechargeable nickel-cadmium batteries capable of delivering five watts of direct current at a nominal output of twelve volts. It is scheduled to be installed in a Coast Guard buoy to operate a flashing light for a maintenance-free period of two years. The buoy is shown in Fig. 1. During its first year of operation the buoy will be located in Arundel Cove at the U. S. Coast Guard Station, Curtis Bay, Maryland (Fig. 2).

The SNAP 7C system will be used with a U. S. Navy automatic weather station designed for use in Antarctica. Figure 3 shows the weather station. The system will consist of a 10-watt thermoelectric generator, dc-to-dc converter and nickel-cadmium batteries. Normally rejected waste heat from the thermoelectric generator will be used to maintain reasonable battery temperatures during subzero operation. The design life of the power supply system is also two years without maintenance.

Both generators have potential lives of 10 years at rated power, this consideration dictating the amount of radioisotope fuel required and, hence, the overall design characteristics.

Fuel for these generators, in the form of encapsulated Strontium-90, will be provided by Oak Ridge National Laboratory.

The major effort during this report period was the analysis and design of the 10-watt thermoelectric generator which is a basic component of both the SNAP 7A and 7C power systems. The succeeding pages of this section will present pertinent details of the 10-watt thermoelectric generator under the following headings:

- (1) Generator description.
- (2) Isotope fuel.
- (3) Safety and shielding considerations.
- (4) Heat transfer analysis.
- (5) Thermoelectric elements.
- (6) Manufacturing drawings and weights.

#### 1. Thermoelectric Generator\*

The SNAP 7A and 7C generators are of identical design. The differences in the resulting systems lie in the voltage conversion, energy storage

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\*H. Morton

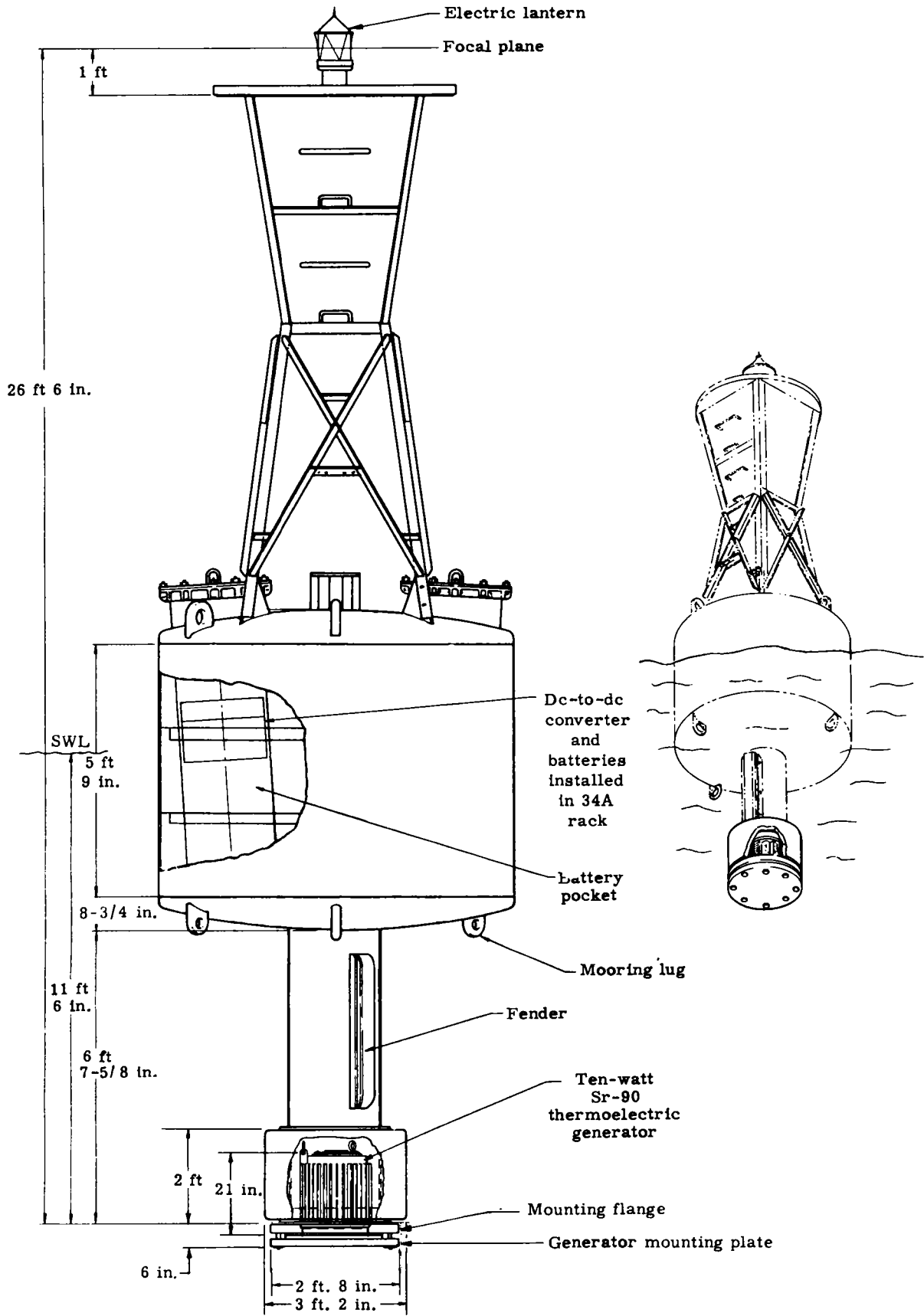


Fig. 1. U. S. Coast Guard Buoy

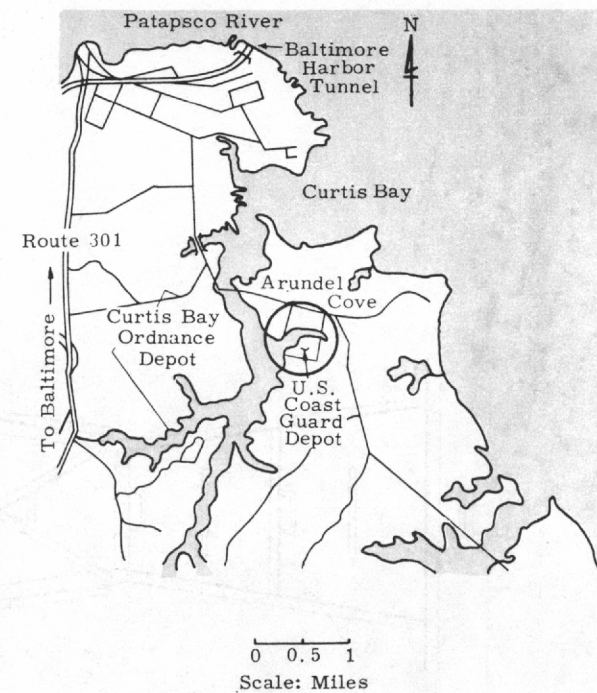
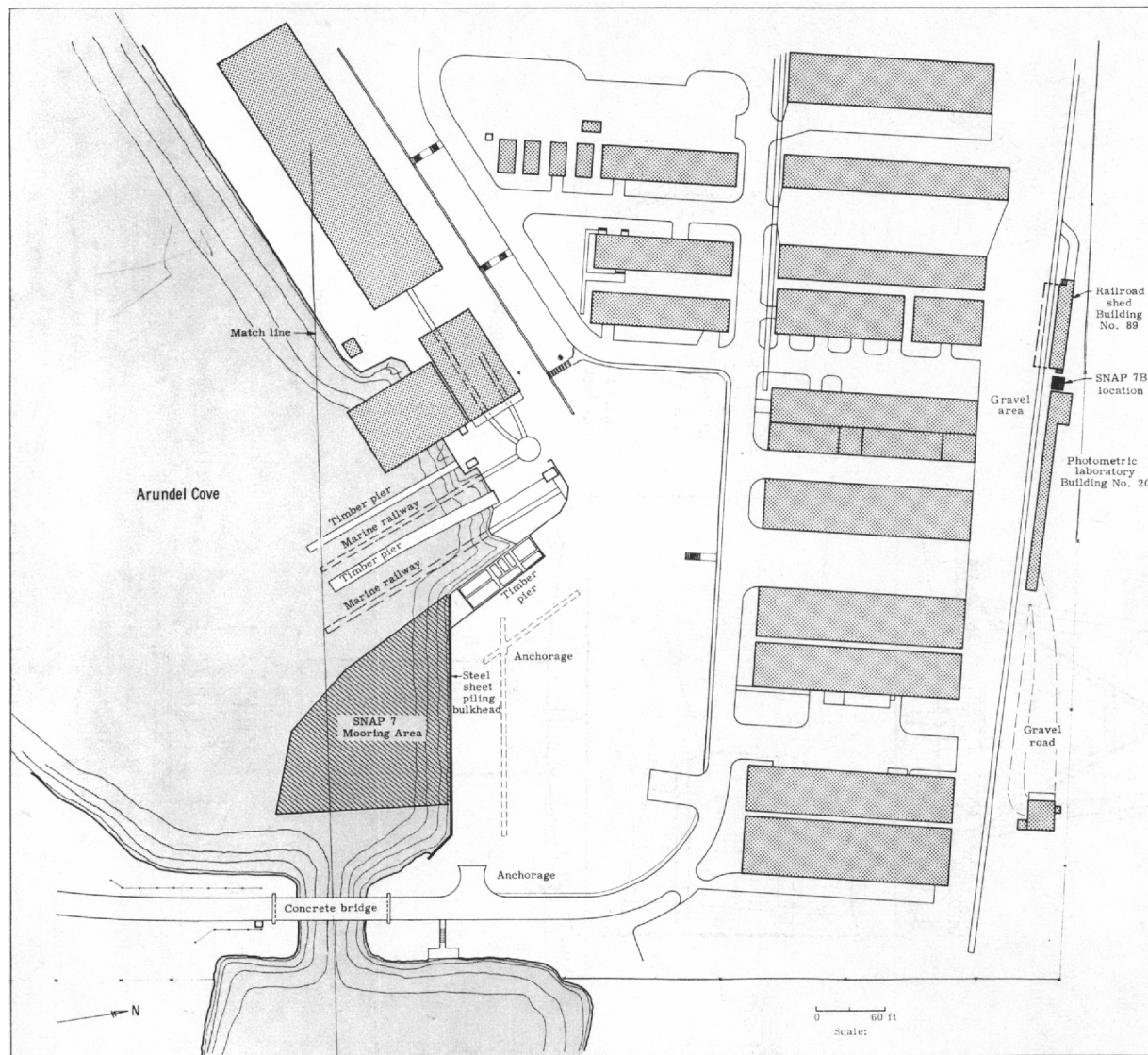


Fig. 2. Locations of SNAP 7A and 7B Within Coast Guard Yard and Location of Yard in Curtis Bay Area

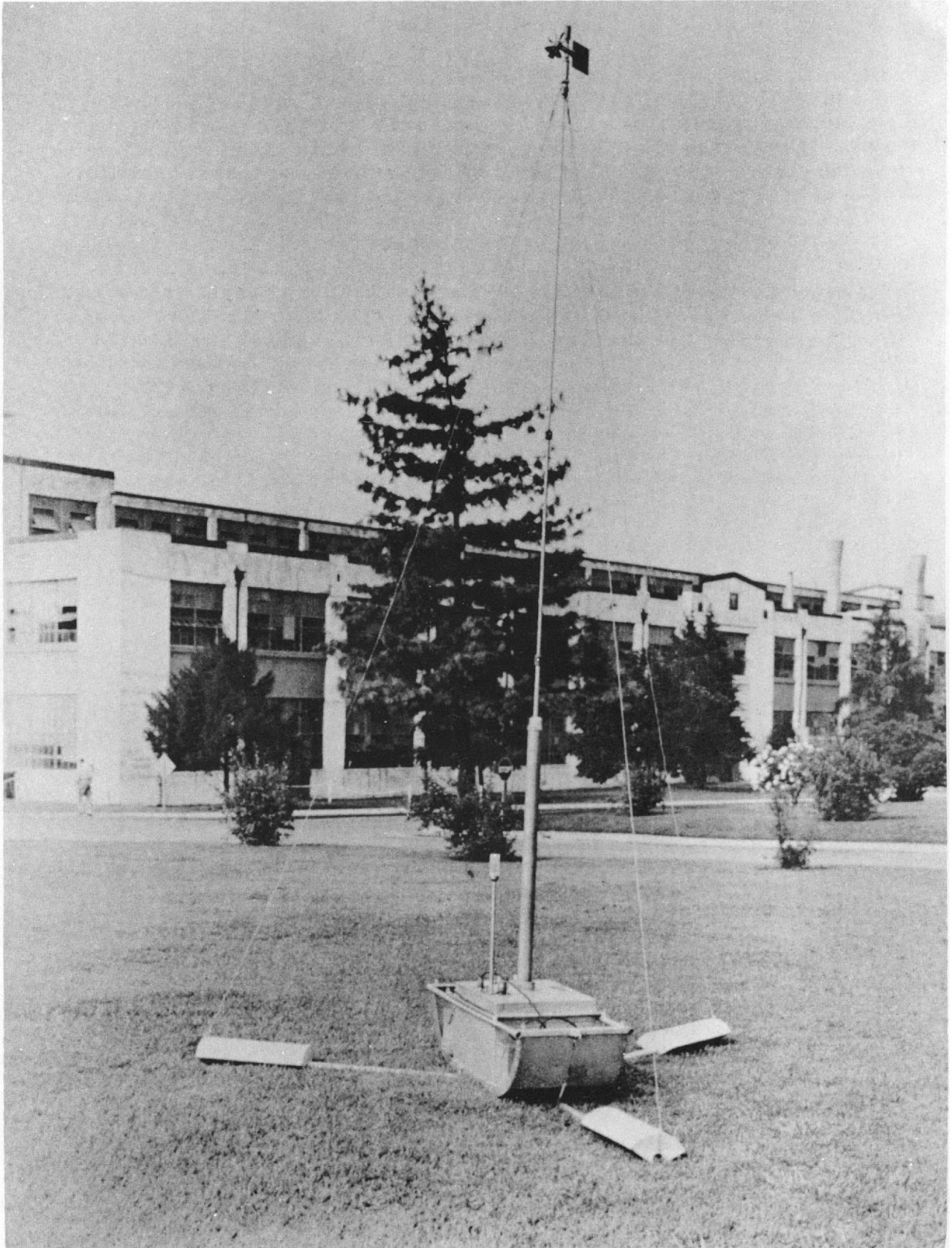


Fig. 3. Automatic Weather Station

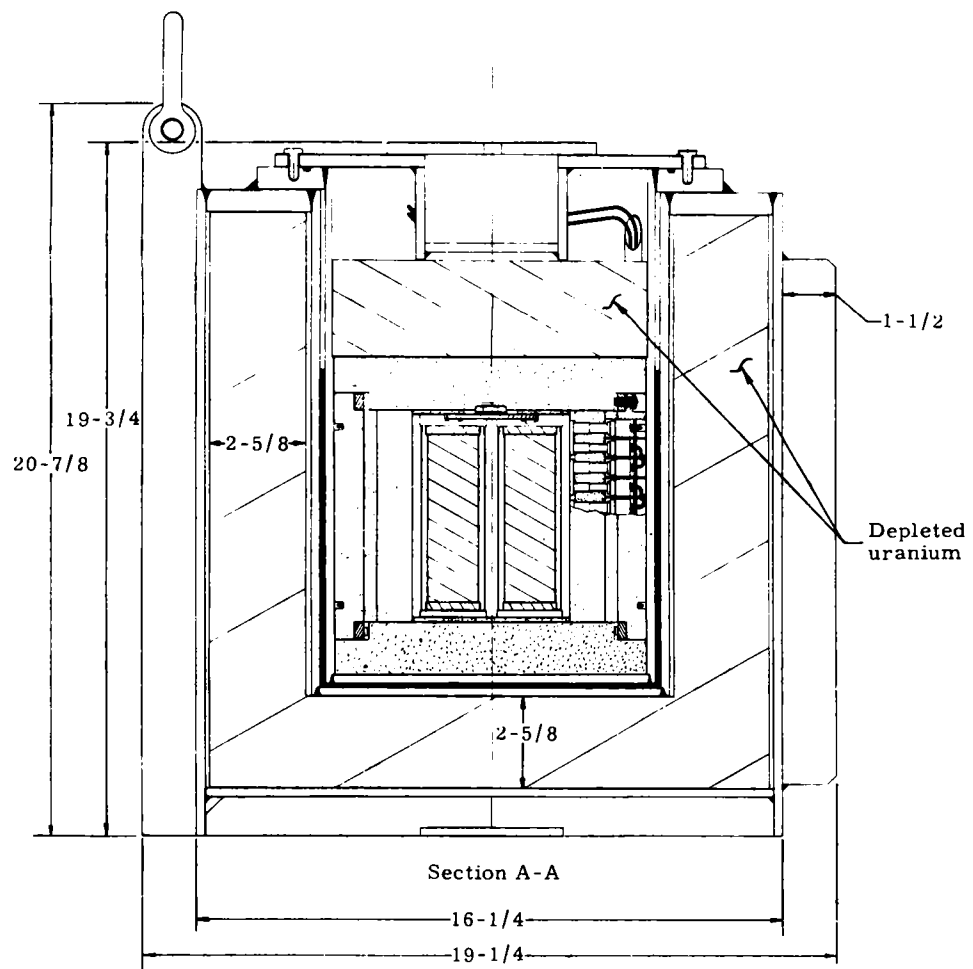
and installation details. Figure 4 shows the design of the 10-watt generator and identifies the major components in final assembly. Basically, the generator consists of an isotopic heat source, a thermoelectric conversion circuit, a heat rejection system and a biological shield. Each component serves a necessary function in the operation and safety of the thermoelectric generator.

Four radioactive fuel capsules containing strontium titanate provide a total of 256.5 watts of thermal output, initially, to power the generator over its 10-year potential life. Next to the Hastelloy C fuel block (see Fig. 5) are 60 pairs of lead telluride thermoelectric elements which produce a minimum of 10 watts of electrical power. Heat which is not converted to electricity is released to the surroundings through the 36 fins welded to the outer shell. Between the inner and outer shells of Hastelloy C are 2.6 inches of depleted uranium. This high density metal reduces the radiation from the isotope fuel capsules to tolerable levels. The maximum dose rates permitted in the shipment of radioactive materials are 200 milliroentgens equivalent man per hour (mrem/hr) at the container surface and 10 mrem/hr at one meter from the source. Adequate shielding is provided within the 10-watt generator to safely meet the requirements of the Interstate Commerce Commission.

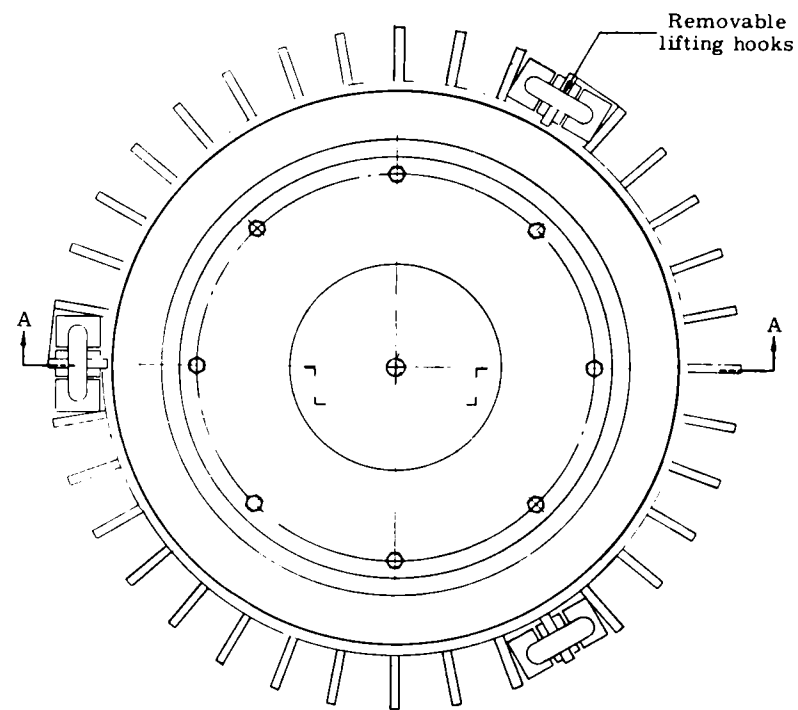
The Hastelloy C fuel block has four bored holes closed at the bottom with circular plates welded in place. The four fuel capsules are maintained in place by a square plate of Hastelloy C bolted to the top of the fuel block. The corners of the fuel block are cut at a 45-degree angle so that adequate thermal insulation may be positioned to support and insulate the high temperature block. The four flat surfaces will be pre-coated with aluminum oxide and then used as the hot surface for mounting the thermoelectric modules.

Lead telluride thermoelectric couples are installed in separate subassemblies called modules. Five pairs of elements are electrically interconnected to form each module, and 12 modules are used in the complete 10-watt generator. Three modules are installed opposite each of the four flat surfaces of the fuel block. Except for the thermoelectric area, the complete fuel block is insulated with 1.4 inches of Johns-Manville Min-K insulation. When a temperature difference of 700° F is maintained across the 60 pairs of thermoelectric elements, the generator will produce 10 watts of electric power. Hot junction temperatures in the range of 900° to 1000° F will produce a larger electrical output. Figure 5 shows the complete thermoelectric assembly within the 10-watt generator.





All dimensions in inches



Total weight of generator and shield = 1840 lb

Fig. 4. Ten-Watt Generator-Biological Shield Assembly

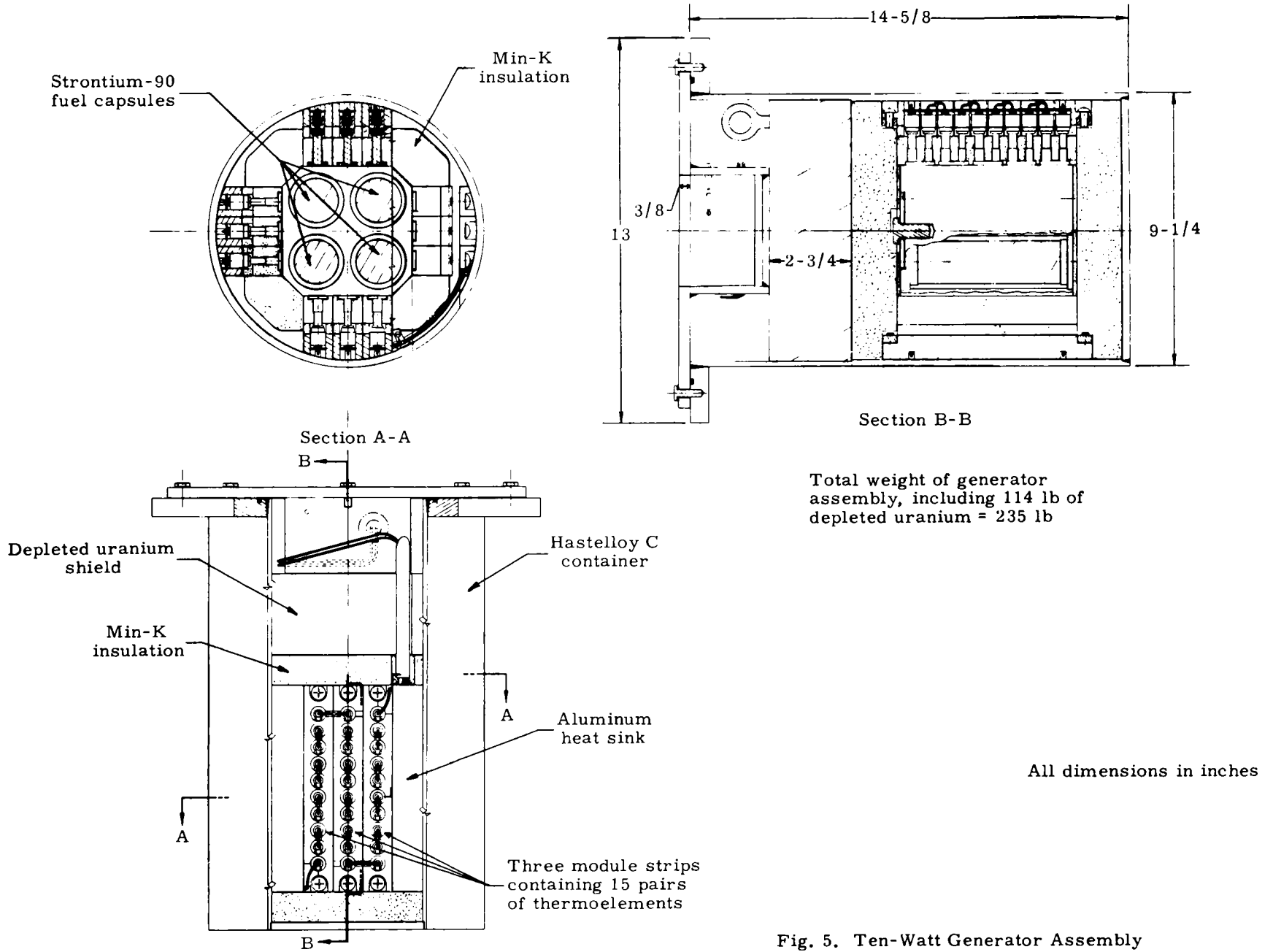


Fig. 5. Ten-Watt Generator Assembly

Thermal power that is not converted to electric power is conducted to the exterior of the generator and released to the surroundings. In the SNAP 7A system the heat will be conducted to the buoy and, eventually, to the water. In the SNAP 7C system the waste heat is used to maintain an internal storage temperature of 40° F or higher, even under subzero ground temperature conditions. The heat, however, is eventually dissipated to the surrounding terrain.

Copper caps are soldered to each of the 120 thermoelements. Waste heat is conducted through the copper caps and receivers to the aluminum pistons and module heat sinks. Each aluminum component will be hard-coated to allow thermal conduction to take place without electrical conduction. Excess heat will then be conducted to the Hastelloy C generator shell and eventually through the uranium shield to the exterior fins. Electric power is obtained through the series circuit connecting the 60 pairs of elements with the output terminals of the generator.

## 2. Isotope Fuel\*

Strontium-90, with its relatively long half life (27.7 years), was selected as the isotope fuel for the SNAP 7A and 7C generators. The chemical form selected was strontium titanate,  $\text{SrTiO}_3$ , a stable ceramic compound that has extremely low solubility in sea water. Specific thermal power from the prepared fuel form was estimated at 0.5 watt per cubic centimeter (watt/cc).

Because of the long lead time required for chemical processing, it was necessary to fix the volume of the fuel prior to detailed design of the generator. This was accomplished by assuming a 5% overall generator efficiency at the end of 10 years. The thermal input required for a 10-watt output is then 200 watts. From the known isotope-decay rate, the input requirement at the beginning of life was calculated to be 256.5 thermal watts. Since the thermal power density of  $\text{SrTiO}_3$  is 0.5 watt/cc, the volume of fuel should be 513 cubic centimeters (31.3 cubic inches).

In order to load 31.3 cubic inches of fuel into the 10-watt generator with a minimum loss of heat, it became necessary to distribute the fuel in four separate fuel capsules containing a total of 47,000 curies of Strontium-90, based on a conservative specific power of 0.0055 watt per curie. Other considerations were that the ceramic pellet diameter be limited to 2 inches or less and that an existing welder at Oak Ridge could be used to seal the capsule if the height did not exceed 6 inches. Based on this reasoning, the four capsules for the 10-watt generator were sized as shown in Fig. 6. Fuel capsule drawings have been submitted to ORNL for concurrence and comment relative to fuel volume.

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\*A. Streb

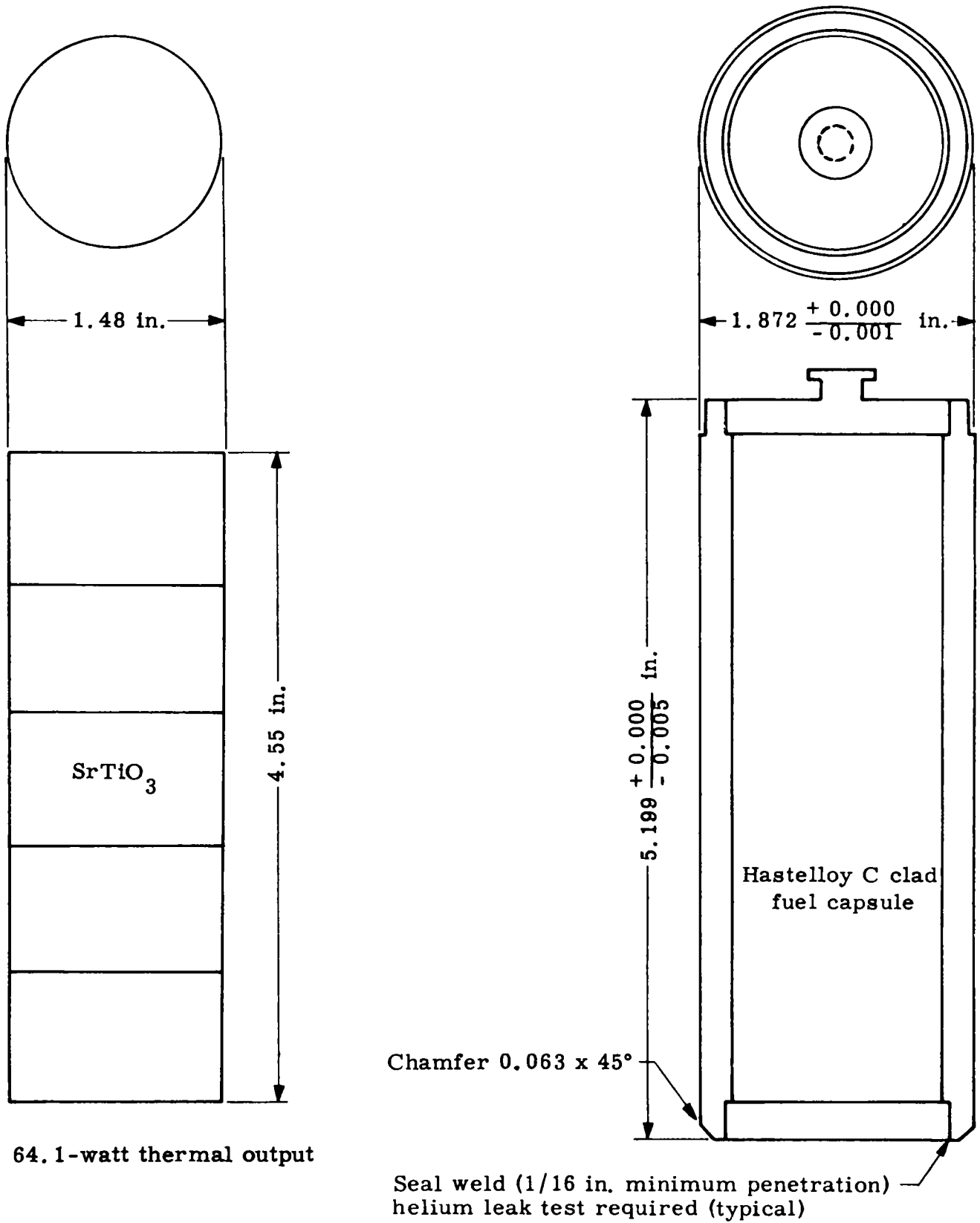


Fig. 6. Fuel Capsule Assembly of Strontium-90 Generator

Each fuel capsule has a button on the top to facilitate the installation or removal of fuel by means of remote handling equipment in a hot cell. All fuel cladding is made of Hastelloy C. This material was selected because of its excellent structural properties and resistance to corrosion. Results from a 10-year corrosion test in sea water at Wrightsville Beach, North Carolina, show that Hastelloy C has an average corrosion rate of 0.1 mil per year without any tendency toward pitting. This ensures safe cladding for over 1000 years, even if the corrosion rate proves to be twice that experienced in the test.

### 3. Safety and Shielding Considerations\*

Radiation dose rates from the unshielded capsules of the 10-watt thermoelectric generator were computed by means of an IBM 709 computer. Each capsule was assumed to contain one fourth of the total 47,000 curies of Strontium-90. On this basis the dose from one capsule (containing 11,750 curies of Sr-90) was as follows:

	<u>Dose at Side (r/hr)</u>	<u>Dose at Top and Bottom (r/hr)</u>
At surface	89,000	62,000
At one meter from surface	136	59

From the four capsules loaded into the fuel block without shielding, the total dose rate is as follows:

At one meter from surface	500	235
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Shielding values were computed for depleted uranium to reduce the dose rate from 47,000 curies of Strontium-90 to 100 mr/hr at 4 feet from the centerline of the 10-watt generator, in accordance with the specifications established at the inception of the program. Uranium was proposed originally as the shield material because its use resulted in generators of lighter weight and smaller dimensions. The minimum thicknesses (inches) of uranium required are as follows:

---

\*A. Spamer and T. Dobry

<u>Shield</u>	<u>Thickness (in.)</u>
Top	1.87
Side	1.54
Bottom	1.67

However, the biological shield for the 10-watt generator was designed with a thickness greater than the minimum to permit the use of the shield as a shipping container for transportation by common carriers in interstate commerce. The designed thicknesses and the resulting dose rates are as follows:

<u>Shield</u>	<u>Thickness (in.)</u>	<u>Dose Rate at 4 Feet (mr/hr)</u>
Top	2.75	5.1
Side	2.625	9.6
Bottom	2.625	7.7

These dose rates would meet ICC shipping regulations for radioactive materials.

More generalized IBM 709 calculations were performed to determine average tenth- and half-value thicknesses of cylindrical shields for various materials between one and five inches thick. The results are as follows:

<u>Material</u>	<u>Tenth-Value Thickness (in.)</u>	<u>Half-Value Thickness (in.)</u>
Uranium	0.88	0.26
Lead	1.46	0.44
Steel	2.40	0.72

Experimental measurements were obtained by Oak Ridge personnel concerning shielding values for a strontium titanate fuel pellet containing about 1000 curies of Strontium-90. A small amount of Cerium-144

(0.3 curie) was later detected as an impurity in the pellet tested. However, the tenth- and half-value thicknesses of flat plate shields were determined for the pellet as follows:

<u>Shield Material</u>	<u>Tenth-Value Thickness of Pb (in.)</u>	<u>Half-Value Thickness of Pb (in.)</u>
1/8-inch Hastelloy C and lead	1.63	0.49
1/2-inch Hastelloy C and lead	1.56	0.47

The experimentally determined values appear to be slightly higher than those resulting from IBM 709 calculations. Possibly, this could be attributed to the cerium content. However, on large thicknesses of shielding, the total actual shielding as measured by ORNL was less than that calculated by IBM 709. In general, it is felt that the shielding for the 10-watt generator is conservative and should adequately serve the purposes of both the U. S. Coast Guard and the U. S. Navy.

Complete safety analysis reports concerning the use of strontium-fueled thermoelectric generators were prepared in greater detail (Refs. 1 and 2).

#### 4. Heat Transfer Analyses\*

Extensive heat transfer analyses were carried out for the 10-watt thermoelectric generator to ensure proper temperature distribution during the total 10-year life of the generator. Considerations were made for the continuous release of unconverted thermal energy to the surroundings to prevent internal temperature excursions. Each component in the generator was reviewed completely for its ability to transfer heat and maintain reasonable operating temperatures. Some of the considerations are reviewed as follows.

##### a. Hot junction temperature

Maximum overall efficiency may be attained for the 10-watt thermoelectric generator at a hot junction temperature of approximately 900° F. Due to the input-power decay, it is impossible to maintain a constant temperature over the 10-year life interval. It would be most desirable to operate the hot junction at 900° F at the end of life in order to achieve

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\*A. Streb

the greatest efficiency possible when the power input is the smallest. However, the hot junction temperature at the beginning of life would be greatly in excess of the maximum temperature tolerated by the thermoelectric materials. Experience with available materials has shown that the hot junction temperature should not be permitted to exceed 980° F. The thermoelectric elements have been sized to produce various hot junction temperatures at the end of life; the resulting beginning-of-life temperature was computed with this geometry until a temperature of 980° F was found. The end-of-life temperature which produces this condition is approximately 800° F.

#### b. Cold junction temperature

The cold junction temperature is established by the environmental temperature, the temperature rise between the environment and the generator shell, and the temperature rise within the generator shell and cold cap assemblies, shown in Fig. 5. The maximum operational environmental temperature occurs in the buoy application, where the surface is in direct contact with sea water ranging in temperature up to 80° F. The temperature rise in the film has been calculated to be approximately 4.5° F. The total temperature rise between the surface and the cold end of the elements has been estimated at 41.5° F. The cold junction should operate at approximately 126° F at the beginning of life. At the end of life, the heat output has decreased to 78% of the original value and the cold junction temperature, correspondingly, has been reduced to 115° F.

#### c. Heat rejection system

Biological shielding must be provided to attenuate the radiation to a tolerable level. The shielding could be located entirely around the heat source or at the outer surface of the generator. A study was made to determine the best location for the shielding, and it was concluded that the shielding should be located entirely on the outside for the 10-watt generator. This was due primarily to the significant performance penalty suffered when the shielding is part of the fuel block.

The outer shell, therefore, must be designed to contain the shield material (depleted uranium) and must also be capable of dissipating the total heat produced by the isotope while maintaining reasonably low temperatures at the generator cold junction. The generator must exist in two distinct environments:

- (1) During shipping, when the air temperature may go as high as 125° F (however, the generator terminals may be short-circuited during shipment, resulting in a significant reduction in temperature drop across the elements).



- (2) During operation, either submerged in sea water not exceeding 80° F or emplaced in the Antarctic.

Condition (2) poses no problem relative to cooling the surface, and the generator will be designed for operation in temperatures resulting in these conditions. Condition (1), however, could result in elevated generator temperatures. The surface must be designed to operate at temperatures no higher than 175° F in order to maintain internal temperatures at a reasonably low level during shipment. The maximum design heat input is 256.5 watts. The heat lost from the surface is given by

$$q_{\text{total}} = q_{\text{conv}} + q_{\text{rad}} = 256.5 \text{ watts}$$

where

$$q_{\text{conv}} = h_c A_c \Delta T, \text{ convection heat loss}$$

$$q_{\text{rad}} = h_r A_r \Delta T, \text{ radiant heat loss}$$

$$h_c = \text{convection film coefficient}$$

$$h_r = \text{radiation heat transfer coefficient}$$

$$T_s = \text{surface temperature of container}$$

$$T_r = \text{environmental temperature}$$

$$A_c = \text{convective heat transfer area}$$

$$A_r = \text{radiant heat transfer area}$$

$$\Delta T = \text{temperature difference between generator shell surface and environment} = T_s - T_r.$$

The generator surface is considered to consist of a cylinder 16-1/4 inches in diameter and 17 inches in height with a number of 1-1/2 inch fins running longitudinally on the cylindrical surface of the shell. It is desired to determine the number of fins required to assure a maximum surface temperature not in excess of 175° F.

The radiant heat transfer area is approximately the area of the cylinder taken over the edge of the fins, including the top of the cylinder but excluding the bottom.

$$A_r = 8.59 \text{ square feet}$$

The convective heat transfer area is approximately the area of the cylinder, excluding the bottom, plus the area of the fins, multiplied by the fin efficiency. The fin efficiency is given by the following expression (Ref. 3):

$$e_s = \frac{1}{\sqrt{2\epsilon}} \tanh \sqrt{2\epsilon}$$

where

$$e_s = \text{fin efficiency}$$

$$\epsilon = W_c^{3/2} \sqrt{h_c/kA}$$

$$A = \text{cross-sectional area of fin} = 2\delta W_c$$

$$W_c = W + \delta$$

$$\delta = \text{one-half fin thickness} = 0.125 \text{ inch}$$

$$W = \text{length of fin} = 1.5 \text{ inches}$$

$$k = \text{thermal conductivity of fin material} = 6.5 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

$$h_c = \text{convective heat transfer coefficient}$$

$$\approx 0.27 (\Delta T)^{0.25} \quad (\text{Ref. 4, p 240})$$

$$= 0.27 (50)^{0.25} = 0.718 \text{ Btu/hr-ft}^2\text{-}^\circ\text{F}$$

Then

$$\epsilon = \left( \frac{1.5 + 0.125}{12} \right)^{3/2} \sqrt{\frac{0.718}{(6.5)(0.00302)}} = 0.331$$

and

$$e_s = \frac{1}{\sqrt{0.662}} \tanh \sqrt{0.662} = 1.228 (0.6725) = 0.826 = 82.6\%$$

therefore,

$$A_c = \text{total convective heat transfer area} = 7.46 + 0.291N \text{ square feet}$$

where N is the number of fins.

The radiant heat transfer coefficient may be determined from Eq 13.29, Ref. 5:

$$h_r = \frac{0.1714\epsilon_s \left[ \left( \frac{T_s}{100} \right)^4 - \left( \frac{T_r}{100} \right)^4 \right]}{(T_s - T_r)}$$

$$= 0.692$$

where

$$\epsilon_s = \text{surface emissivity} = 0.50.$$

The total heat transfer is then given by

$$q_{\text{total}} = (0.718)(7.46 + 0.291N)(50) + (0.692)(8.59)(50)$$

which must equal the total design input of 877 Btu/hr:

$$q_{\text{total}} = 268 + 10.43N + 296.5 = 877$$

gives

$$N = 29.9 \text{ fins.}$$

So a minimum of 30 fins will be required on the shell surface.

In order to provide some degree of conservatism, the number of fins actually incorporated in the 10-watt generator design has been set at 36.

d. Determination of insulation thermal conductivity

A limited program was conducted to measure the effective thermal conductivity of Min-K insulation with various gas fills. This information was not available from the vendor nor from any other known source. Determination of the thermal conductivity was necessary to estimate the composition of fill gas required to maintain design temperatures in the SNAP 7 generators.

The test was performed in a gastight aluminum container with 1-1/2 inches of Min-K insulation covering an electric heater. The hot skin and cold skin temperatures were determined for fixed values of input power. Results indicate that the effective thermal conductivity of Min-K in the temperature range 150° to 900° F is approximately 25% higher than reported by the vendor. The values for various gas fills at a pressure of one atmosphere are as follows:

<u>Gas Fill</u>	<u>Thermal Conductivity (Btu/in.-hr-ft<sup>2</sup>-°F)</u>
Air	0.2865
Argon	0.2285
Hydrogen	0.597
Vacuum	0.1188

The test data indicate that the thermal conductivity of Min-K 1301 can be varied over a wide range to control the temperature of the SNAP 7 generators. The exact type of gas fill to be used in the generators will be determined when the generator heat losses have been measured.

e. Excessive thermal power in Strontium-90 fuel

In the Hanford fission-product waste, the isotope Strontium-89 produces as much as 20 to 80 times the heat of Strontium-90 at reactor discharge, depending on the atomic ratio of the two isotopes. Although Strontium-89 has a half life of only 51 days, current production schedules indicate that sufficient quantities may be present at the time of generator loading to cause temporary overheating and consequent damage to the thermoelectric elements. However, because of its transient nature, the Strontium-89 thermal power cannot be considered part of the total thermal power requirement.

Figure 7 illustrates the combined thermal output of Strontium-89 and -90 for 47,000 curies of the heavier isotope as a function of months after discharge from the reactor. Atomic ratios of Strontium-89 to -90 were arbitrarily chosen as 0.20 and 0.64. Depending on the age of the reactor waste and the initial atomic ratio of Strontium-89 and -90, the fuel for the 10-watt generators may have some thermal contribution from Strontium-89 which would require rejection to the surroundings. This condition has been analyzed; with proper use of conducting gas in the insulation, the excess heat can be released without affecting generator performance. This conforms to the SNAP 7A and 7C fuel requirement which permits an overload of 40 watts due to Strontium-89 content.

#### f. Insulation heat losses

The fuel block is separated from the cold sink by a layer of Min-K 1301 insulation. Min-K was chosen over other high-temperature insulations because of its superior insulating characteristics, good structural qualities and its ability to be accurately fabricated into complex shapes. The insulation fits snugly between the fuel block and the cold sink, providing support for the fuel block. It has a mean depth of 1.38 inches (space between the block and the sink). The insulation depth was selected as a compromise between thermal and fabrication considerations.

An insulation thermal conductivity corresponding to an approximate mixture of 90% argon and 10% hydrogen was selected (thermal conductivity  $k = 0.0175$  Btu/ft-hr-°F) as a nominal value for the generator design.

Heat losses through the insulation were calculated with the relation

$$q = \frac{6 \pi r_1 r_2 k \Delta T}{r_2 - r_1}$$

where

$k$  = thermal conductivity of insulation

$\Delta T_i$  = temperature difference between  $r_1$  and  $r_2$

$r_1$  = radius of fuel block surface

$r_2$  = radius of insulation.

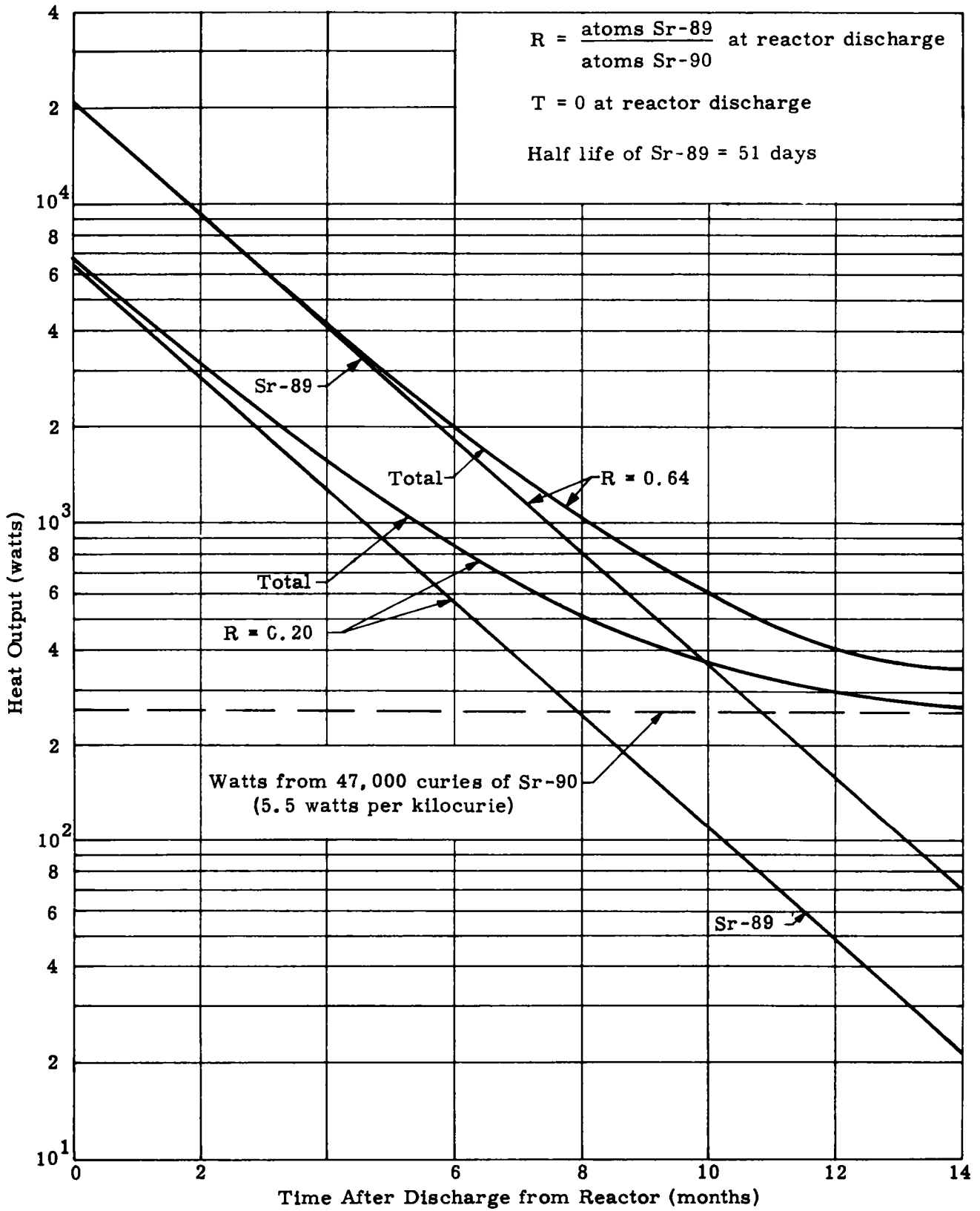


Fig. 7. Effect of Strontium-89 on Heat Output of SNAP 7A 10-watt Generator

This relation holds for a cylinder with a length-to-diameter ratio of 1, surrounded by a constant-thickness layer of insulation. The calculated total heat loss at the end of life is 50.75 watts, including an estimated 2.75 watts lost through the mica insulators surrounding the thermoelements. The mica insulator loss was estimated by treating it and the heat flow through the thermoelement as analogous to current flow through parallel resistors.

### 5. Thermoelectric Elements\*

The relative merit of a thermoelectric material may be measured by the "figure-of-merit" parameter. This may be expressed as

$$FM = \frac{s^2}{k\rho}$$

where

- s = Seebeck coefficient
- k = thermal conductivity
- $\rho$  = resistivity.

The figure of merit for various available materials was compared on the basis of published information. From this comparison, it was apparent that the Martin N-type thermoelectric material and Transitron P-type material would make the best possible combination for an element pair although the Martin P material had almost the same figure of merit as the Transitron P material. However, the Transitron P material was originally selected because of its superior strength. Samples of each of the materials considered were procured, bonded to suitably simulate the hot and cold shoe installation in the generator (Figs. 8 and 9) and operated at temperatures between the limits expected in the generator (115° and 800° F). Measurements made during these tests were used to calculate Seebeck voltage, resistivity and contact resistivity. The properties of the test samples have been averaged and are shown in Table 1 along with thermal conductivity values which were measured independently. Based on the figure of merit determined from the measured properties (also shown in the tabulation), the best combination of thermoelectric materials is Martin N-type with Martin P-type. Long-range testing has shown these two materials to be very durable, displaying very little degradation in performance with time.

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\*A. Streb and H. Miller

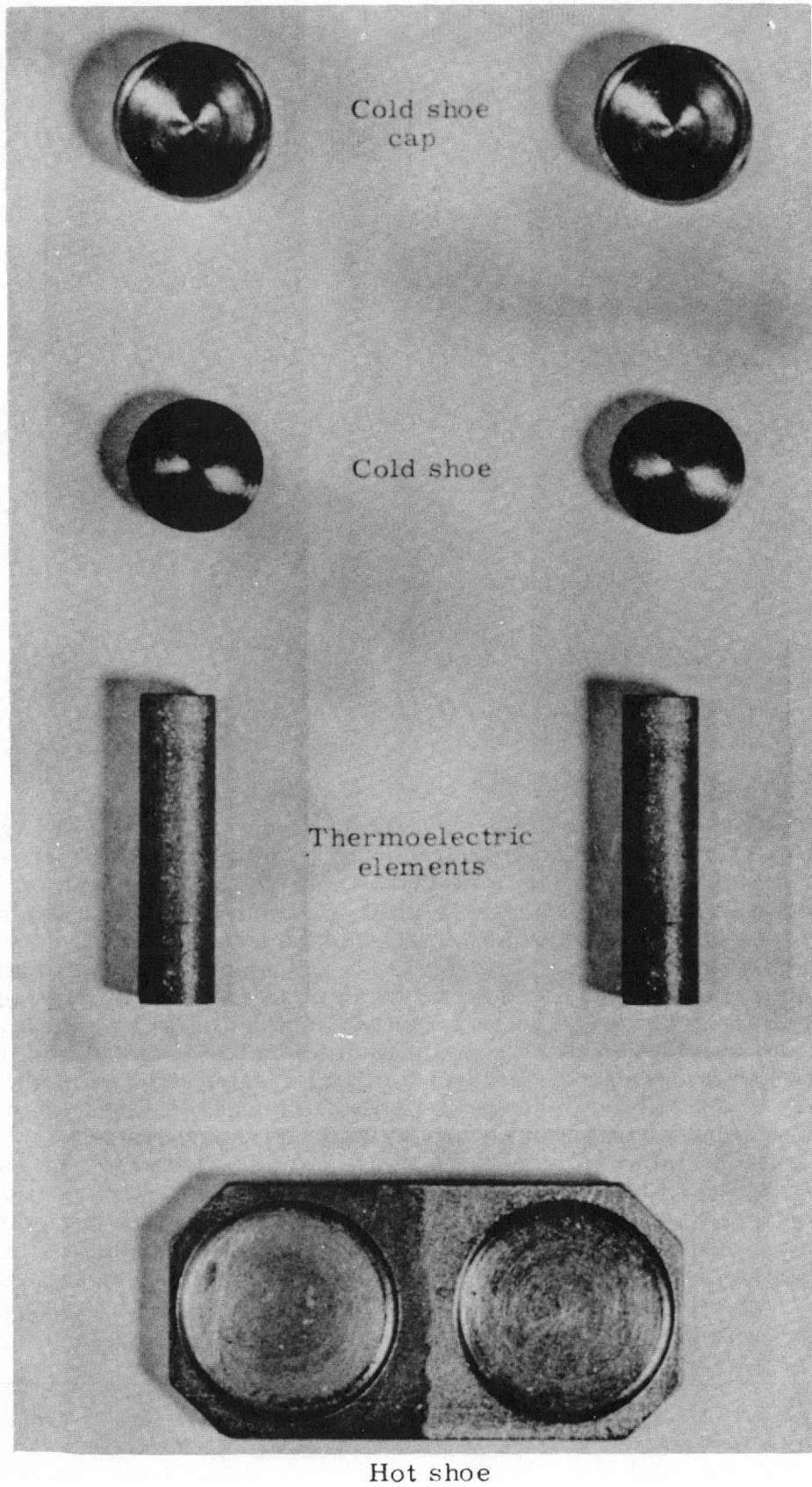


Fig. 8. Components of Thermoelectric Couple



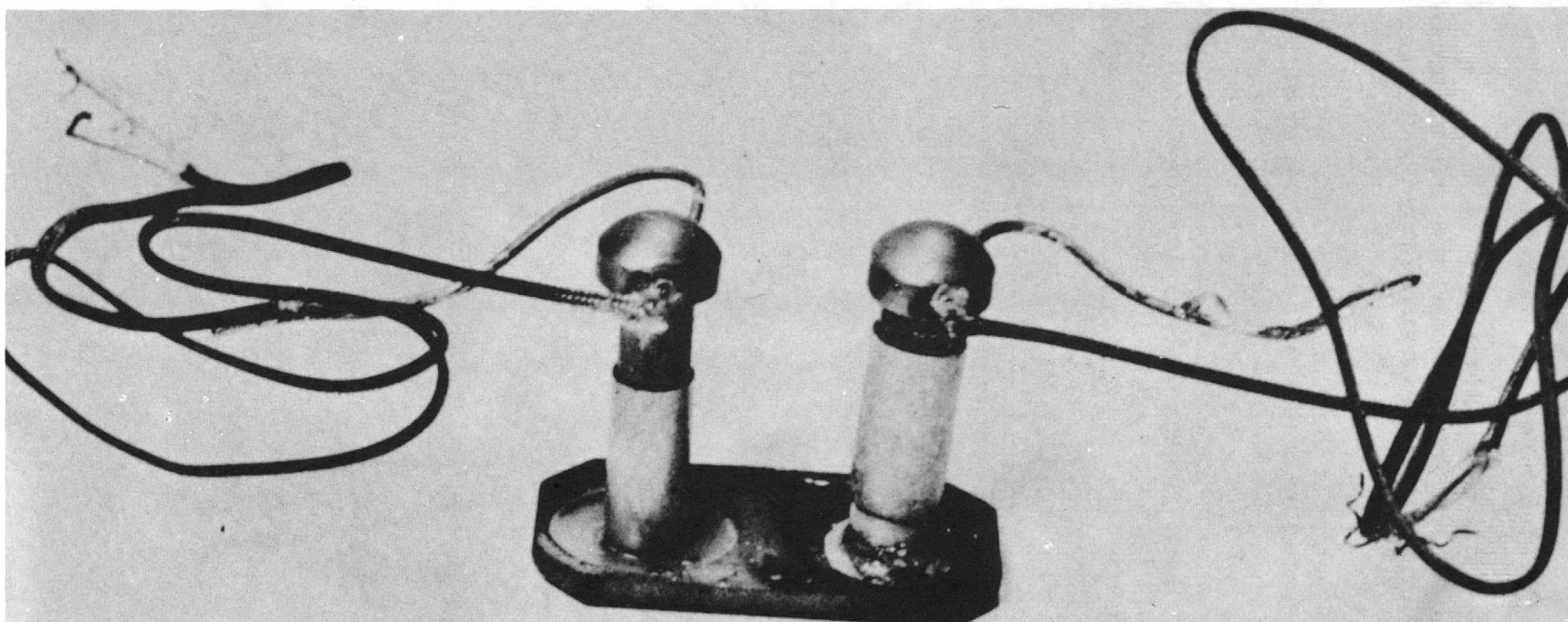


Fig. 9. Bonded Thermoelectric Pair After Testing at 980° F

Calculations were performed to fix the element sizes and generator performance. For an element length of one inch, the equations used below determine optimum element diameters and corresponding generator performance.

The gross thermal output at the end of life is 200 watts; the net available heat input to the thermoelements is 149.25 watts (50.75 watts, insulation losses).

As was previously stated, the elements were sized to operate at temperatures between a hot junction temperature of 800° F and a cold junction temperature of 115° F. Figure 10 shows the test fixture for thermoelectric couple tests.

TABLE 1

Summary of Module Test Thermoelectric Properties

<u>Average Properties</u>	<u>Martin N</u>	<u>Transitron N</u>	<u>Martin P</u>	<u>Transitron P</u>
Seebeck voltage ( $\mu\text{v}/^{\circ}\text{C}$ )	204	257	177	213
Resistivity ( $\mu\text{ohm-cm}$ )	1310	2970	2675	4300
Contact resistivity ( $\mu\text{ohm-cm}^2$ )	386	575	830	760
Thermal conductivity (watts/cm- $^{\circ}\text{C}$ )	0.0199	0.0180	0.0208	0.0196
Figure of merit $\times 10^{-6}$ ( $\text{s}^2/\text{k}\rho$ )	1595	1235	564	538
Advertised $\alpha$ ( $\mu\text{v}/^{\circ}\text{C}$ )(approx)	210	215	188	190
Advertised $\rho$ ( $\mu\text{v}/^{\circ}\text{C}$ )(approx)	1305	2000	1750	2350

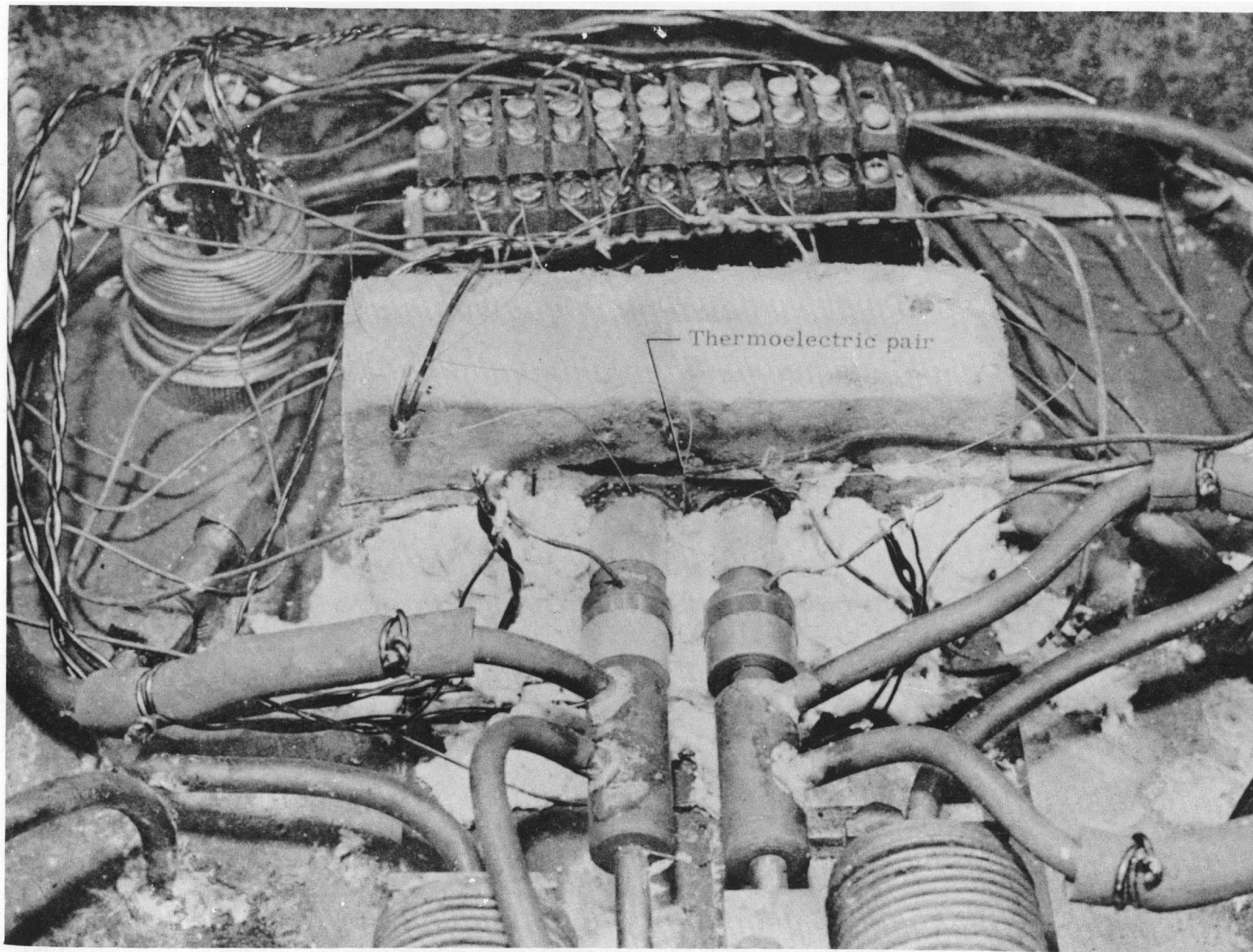


Fig. 10. Test Fixture for Individual Couple Tests

The average element properties used in the calculations are based on the experimental data shown in Table 1 and are listed as follows.

	<u>N-type</u>	<u>P-type</u>
Seebeck coefficient ( $\mu\text{v}/^\circ\text{C}$ )	$s_N = 204$	$s_P = 177$
Resistivity ( $\mu\text{ohm-cm}$ )	$\rho_N = 1345$	$\rho_P = 2735$
Thermal conductivity ( $\text{watts/cm-}^\circ\text{C}$ )	$k_N = 0.0199$	$k_P = 0.0208$
Contact resistivity ( $\mu\text{ohm-cm}^2$ )	$c_N = 386$	$c_P = 830$

The following system parameters used in the calculation are defined:

$l$  = element length = 2.54 cm

$T_I$  = hot junction temperature = 700° K

$\Delta T_t$  = thermoelement temperature difference = 380° K

$R_i$  = thermoelement internal resistance

$R_c$  = thermoelement hot junction contact resistance

$R_l$  = load resistance

$A_P$  = cross-sectional area of P element

$A_N$  = cross-sectional area of N element.

$\alpha$  = total Seebeck coefficient for element pair

$$= s_N + s_P = 381 \times 10^{-6} \text{ v}/^\circ\text{C}$$

$$B = \frac{s_N}{s_P} = \left[ \frac{k_P (\rho_N + c_N/l)}{k_N (\rho_P + c_P/l)} \right]^{1/2} = 0.715$$

where  $S_N$  and  $S_P$  are shape factors (ratio of area to length) for the N and P elements, respectively.

$$K = (k_P + k_N B) S_P = 0.0350 S_P \text{ watts/}^\circ\text{C}$$

$$R_i = \left( \rho_P + \frac{\rho_N}{B} \right) \frac{1}{S_P} = \frac{0.004615}{S_P} \text{ ohm}$$

$$R_c = \left( \frac{c_P}{l} + \frac{c_N}{lB} \right) \frac{1}{S_P} = \frac{0.00054}{S_P} \text{ ohm}$$

$$x = \frac{R_i}{R_i + R_c} = \left[ 1 + \frac{\alpha^2 T_I}{K (R_c + R_i)} - \frac{\alpha^2 \Delta T (R_i/2 + R_c)}{K (R_c + R_i)^2} \right]^{1/2}$$

$$= 1.178$$

The thermoelectric efficiency is given by

$$\eta = \frac{\Delta T}{T_I} \frac{x}{(x+1) + \frac{K (R_c + R_i) (x+1)^2}{\alpha^2 T_I} - \frac{\Delta T (R_i/2 + R_c)}{T_I (R_i + R_c)}} = 0.0623.$$

$$\text{Net power output} = P = (P_{in}) (\eta) = 9.30 \text{ watts.}$$

$$\text{Open circuit voltage} = e = N\alpha\Delta T = 8.70 \text{ volts.}$$

$$\text{Load voltage} = v = \frac{x}{x+1} (e) = 4.71 \text{ volts.}$$

The resistances may be determined as follows:

$$P = \frac{v^2}{R_1} = 9.30 = \frac{22.15}{R_1}$$

$$R_1 = 2.38 \text{ ohms}$$

$$\frac{R_1}{R_i + R_c} = x = 1.178$$

$$R_i + R_c = 2.02 \text{ ohms}$$

$$(R_i + R_c)/\text{pair} = \frac{2.02}{60} = 0.0337 = \frac{0.005155}{S_P} .$$

The shape factor for the P element is then

$$S_P = \frac{A_P}{l_P} = 0.153$$

and the cross-sectional area is

$$A_P = 0.389 \text{ cm}^2 .$$

The P- and N-element diameters are then

$$D_P = 0.705 \text{ cm} = 0.277 \text{ in.}$$

$$D_N = 0.595 \text{ cm} = 0.235 \text{ in.}$$

These diameters have been incorporated already in the SNAP 7A and 7C generator design.

The design power output of 10 watts at the end of life may be achieved by altering the gas fill in the insulation. A change from an argon atmosphere to a krypton atmosphere reduced the apparent thermal conductivity of the insulation, resulting in a slight elevation in hot junction temperature and an increase in the estimated power output from 9.3 watts to 9.85 watts. A further increase in power output to 10.10 watts is possible with a gas fill of xenon. It is believed that the estimate of power output stated above is very conservative and represents minimum values.

A thermoelectric reliability program was established to demonstrate the feasibility of module assembly techniques, to determine thermal and electrical characteristics of the module designs, and to obtain a reliability estimate by a long-term operational test.

A test program outlining a 15-couple reliability test for the 10-watt generator configuration was prepared. In addition, a test container was designed to simulate a segment of the generator. The test container has been fabricated as shown in Fig. 11. A simulated cold sink structure is now being fabricated for a thermal calibration test on the container. The thermal calibration test will be performed to provide a basis for heat loss determinations for the simulated generator segment. The module installation procedure will be monitored to determine possible generator assembly techniques and to locate potential problems. The reliability test will then be performed to provide a preliminary estimate of generator performance and element capabilities.

#### 6. Manufacturing Drawings and Weights\*

The majority of the manufacturing drawings for the 10-watt generator were completed during this period. The complete list of detail and assembly drawings is tabulated in Appendix C.

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\*H. Morton

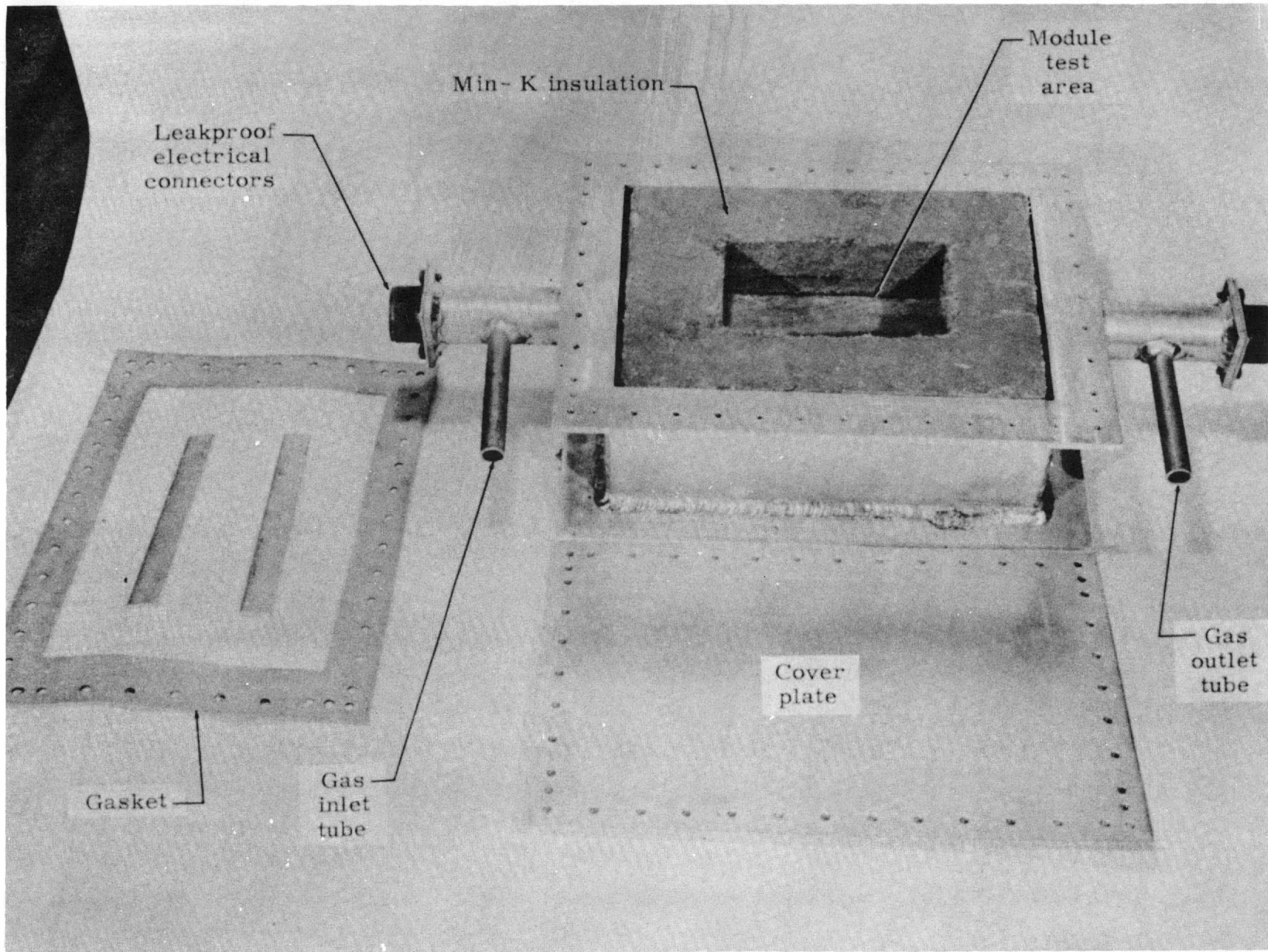


Fig. 11. Test Container



### III. SNAP 7B AND 7D 30-WATT ELECTRIC GENERATION SYSTEMS--SUBTASKS 8.2 AND 8.4

#### A. INTRODUCTION AND SUMMARY OF SIGNIFICANT TECHNICAL ACHIEVEMENTS

The SNAP 7B and 7D generator preliminary design was initiated and the fuel loading requirements were established.

#### B. ENGINEERING--EQUIPMENT DESCRIPTION, DESIGN TECHNIQUES AND PROCEDURES, AND TEST FOR SNAP 7B AND 7D \*

The objectives of these subtasks are to analyze, design and fabricate SNAP 7B and 7D thermoelectric generators for two different 30-watt electrical systems.

SNAP 7B consists of a 60-watt thermoelectric generator, a dc-to-dc converter, and rechargeable nickel-cadmium batteries which are capable of delivering 30 watts of direct current at 32 volts. The complete 7B system is scheduled to be installed in the ground at the U. S. Coast Guard Test Station at Curtis Bay, Maryland, for preoperational evaluation (see Figs. 2 and 12).

The SNAP 7D system will be used with a U. S. Navy boat-type floating weather station. The system will consist of a similar 60-watt thermoelectric generator, a dc-to-dc converter and nickel-cadmium batteries to produce 30 watts of direct current at 24 volts. The complete system must be sized to pass through a 24-inch manhole in the floating weather station. The boat-type weather station is shown in Fig. 13.

Most of the effort during this period has been devoted to the 10-watt generator with only a limited effort concerning the design of the 60-watt generator; however, the thermal power requirement for the 60-watt generator was established. By assuming an overall efficiency of 5% and a 10-year life for the generator, the Strontium-90 fuel requirement was fixed at 1546 thermal watts. The total isotope fuel will amount to 281,000 curies and will be processed at the Martin Nuclear Laboratory at Quehanna, Pennsylvania. Details of this effort are covered in Chapter IV of this report.

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\* J. Keenan

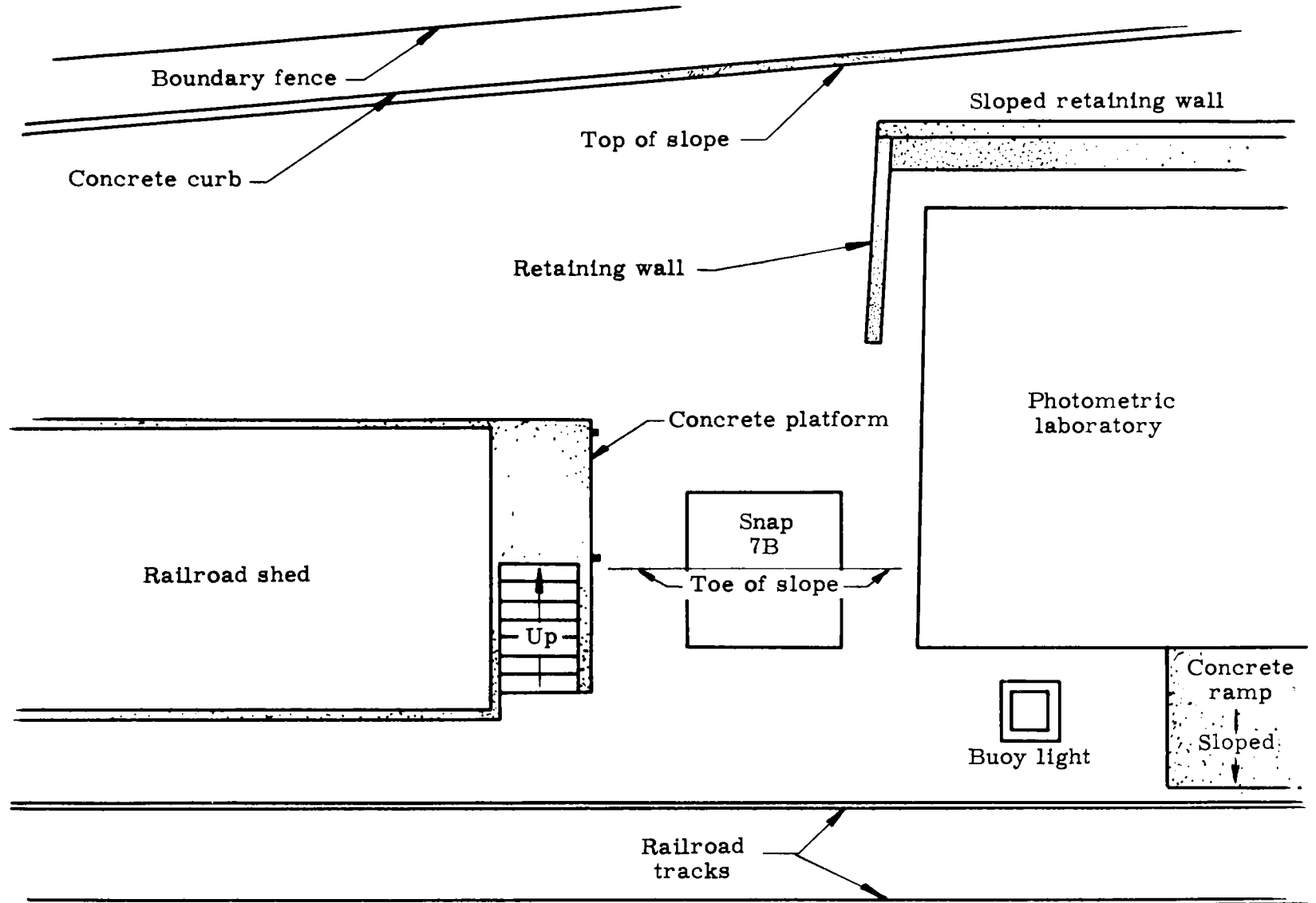


Fig. 12. Site of Snap 7B Installation

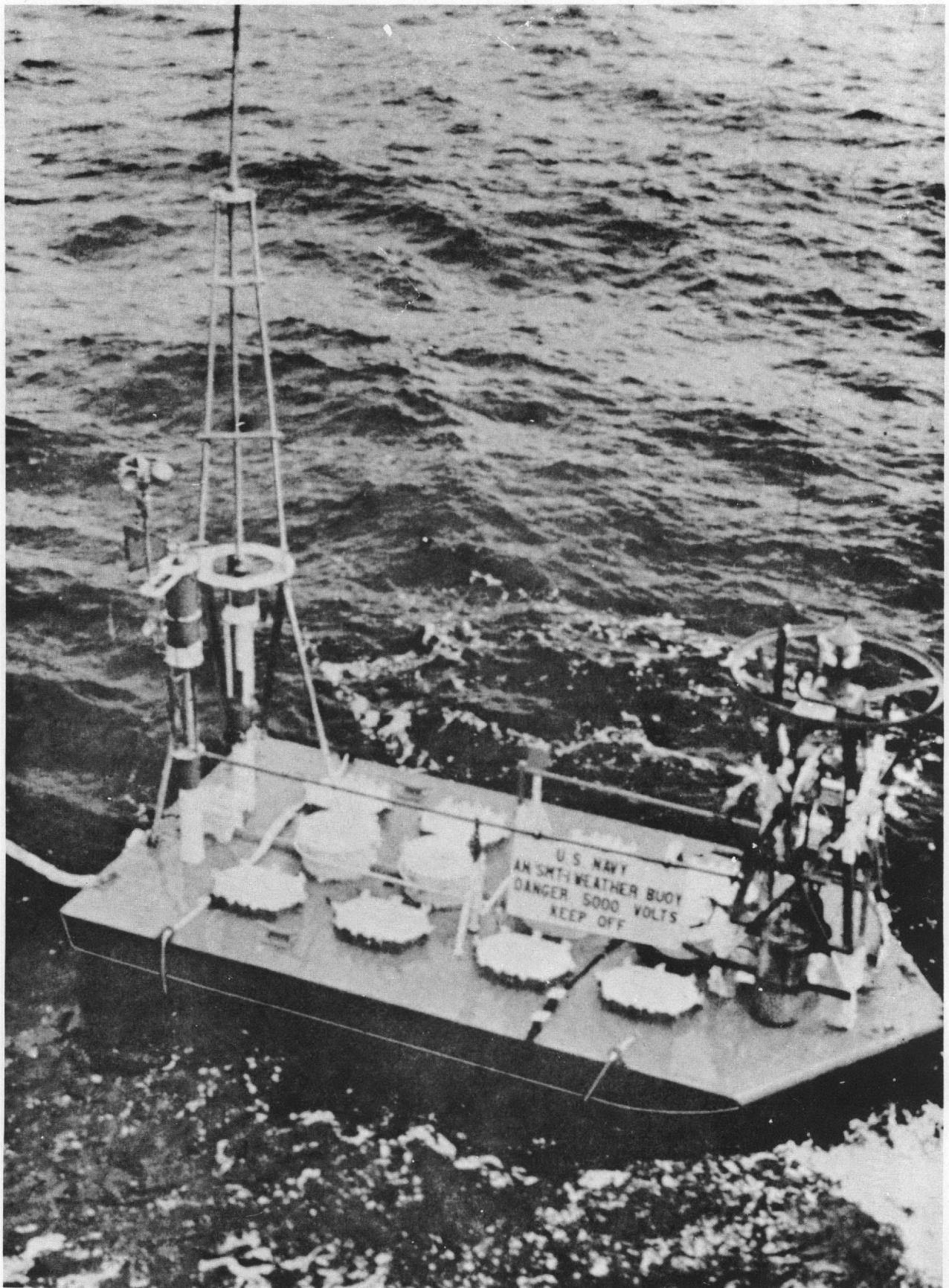


Fig. 13. Boat-Type Weather Station



IV. SNAP 7 FUEL PROCESSING FOR SNAP 7B AND 7D  
GENERATORS--SUBTASK 8.5

A. INTRODUCTION AND SUMMARY OF SIGNIFICANT  
TECHNICAL ACHIEVEMENTS

Fuel process definitions. The fuel process for this program was defined to cover the remote conversion of Strontium-90 feed material to strontium titanate pellets.\*

Investigations also have been carried out relative to decontamination of waste solution generated in the process.

Fuel process engineering. Detailed engineering objectives have been developed for the design of the fuel process equipment. Further, the process flow has been outlined and a performance analysis made to determine thermal, structural and performance compatibility for the process equipment.

Fuel operations. During the period covered by this report no fuel operation, as such, was carried out in the hot cells. Certain functions, such as decontamination, were carried out and are reported here. Also, a description of the hot cells has been inserted. This description is based largely on the information given in Ref. 6.

Strontium-90 fuel specification. Extensive coordination with various AEC sites and offices has been required. The most significant aspect of this effort concerns the specification for the raw fuel materials to be delivered for processing. This is covered in considerable detail in Section E of this chapter.

B. FUEL PROCESS DEFINITIONS\*\*

Several procedures are available for processing strontium to the form required for conversion to the titanate. Two of these procedures have been considered for use at the Quehanna facility.

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\*A brief history of earlier Martin Nuclear work which led to the selection of strontium titanate as the most suitable fuel form for Sr-90 may be found in Appendix B.

\*\*G. Samos and W. Precht

## 1. Process 1

Solution 1. Commercially available titanium tetrachloride is diluted with water to give a solution containing about 75 milligrams of titanium per milliliter of solution. The stock titanium solution must be added dropwise to ice cold water with vigorous stirring; otherwise, a precipitate will form due to hydrolysis of titanium tetrachloride. When the stock solution is prepared, it is analyzed for titanium.

Solution 2. Strontium-90, as received from the Hanford Works of the AEC, will be in the carbonate form, on a filter screen, in a shielded shipping cask. Since the shipping cask is made of stainless steel, nitric acid must be used to dissolve the carbonate (hydrochloric acid will attack stainless steel). The carbonate, after dissolution into a known quantity of solution, is transferred to the storage container. The shipping container is rinsed with a known quantity of water and transferred to the storage vessel. The solution in the storage vessel is thoroughly agitated. The quantity of strontium originally in the shipment and the amount of total solution used during the dissolution of the carbonate are known; therefore, the concentration of the solution in the storage vessel in terms of grams of strontium per liter is accurately known.

A predetermined volume of this solution is transferred to the precipitation vessel. From the known amount of strontium and other reactive cations (principally Ca and Ba) in this volume, a calculation is made to determine how much titanium would be required to react with this quantity of solution. The desired quantity of Solution 1 is measured out and added slowly to Solution 2, which has been cooled to 0° C and rapidly stirred. After these two solutions have been combined, they are then added slowly to a hot (90° C) ammonium oxalate solution sufficiently concentrated to react with the strontium and titanium. A heavy precipitate of hydrated strontium titanyl oxalate ( $\text{SrTiO}(\text{C}_2\text{O}_4)_2 \cdot 4\text{H}_2\text{O}$ ) will form. This precipitate is filtered and washed and is ready for conversion to the titanate.

## 2. Process 2

This procedure is less complex than Process 1 although the final product is not so homogeneous as that produced in Process 1. Dissolution of the material received from Hanford remains the same as outlined above under Solution 2 and will be referred to here as Solution 2.

A predetermined volume of Solution 2 is transferred to the precipitation vessel. A calculation is made to determine the amount of titanium dioxide required to react with the strontium and other cations in

the predetermined volume. The titanium dioxide is weighed out, made into a slurry with water and then transferred into the precipitation vessel. Gaseous ammonia is bubbled rapidly through this mixture to make the solution basic. When the calculated amount of ammonia has passed through the solution, the flow rate is decreased; carbon dioxide is then passed through the same line and through the solution to precipitate the strontium as the carbonate. When a sufficient quantity of carbon dioxide has been passed through the solution, the flow is reduced to keep a slight positive pressure on the line; the precipitate is then filtered and washed. The precipitate is now ready for further physical processing for conversion to the titanate.

These procedures were carefully considered for use at the Quehanna facility. Process 1 was eliminated in favor of Process 2 because the presence of chloride would require equipment fabricated from materials other than stainless steel. Glass equipment would be suitable except for the radiation effects which would cause glass to become brittle and create a hazardous condition. The only other material which could be used is tantalum. Because of its cost and the limited availability of especially fabricated components, tantalum could not be considered for this program. This factor, coupled with the relative simplicity of Process 2, eliminated Process 1 from further consideration.

Ammonia gas and carbon dioxide gas have been substituted in the procedure for ammonium hydroxide and ammonium carbonate to keep liquid volumes to a minimum and thus to reduce the volume of radioactive waste solutions generated.

### 3. Conversion to Titanate

After the titanium dioxide-strontium carbonate mixture has been filtered and washed, it is placed in a ball mill where it is wet-milled for two hours. This ball milling operation helps improve structure and reproducibility of the pellet, as well as the density. After milling, the material is removed from the ball mill and filtered. The precipitate is placed in zirconia crucibles and put into the furnace; the temperature is brought up slowly so that the precipitate will dry. The temperature is finally raised to 1150° C and allowed to remain there for three hours. Tests have shown that calcining at this temperature (or, more specifically, calcining between 1100° and 1200° C) and sintering between 1300° C and 1400° C will give a pellet having maximum density.

After calcining, the required amount of material is placed into a die and a pellet is pressed at five tons per square inch pressure. The pellet is removed from the die and placed into the furnace on a zirconia

plate covered with zirconia sand. The furnace temperature is slowly raised to 1350°C and held for two hours. This results in a hard, dense pellet, approximately 10 to 15% smaller in all dimensions than the green pellet. At the end of the firing period the furnace is allowed to cool slowly to room temperature and the pellet is removed.

During the performance of the chemistry required to give the desired material for conversion to strontium titanate, waste solutions will be generated which will contain trace quantities of radioactive strontium, yttrium and other contaminants which may be occluded in the material received from the Hanford Laboratory. It will be very desirable to remove as much of this activity from solution prior to processing the waste for ultimate disposal.

Several experiments have been run to determine the possibility of removing the radioactive contamination from the liquid waste. Twenty-five-milliliter samples of a Strontium-90 solution containing 2600 counts per minute strontium-yttrium activity per milliliter were pipetted into several centrifuge cones, simulating the waste solution. The samples were set up in groups of two. One sample was acidified, and the second was made basic. The following salts were added, one to each series:

- (1)  $\text{CaCO}_3$
- (2)  $\text{SrTiO}_3$
- (3) Clinoptilolite, a naturally occurring zeolite.

The samples were stirred for 10 minutes and centrifuged. A one-milliliter aliquot from each was evaporated to dryness in a planchet and counted. The following list shows the percent decontamination obtained.

(1) $\text{CaCO}_3$	38%	} Acid (A)	} Basic (B)	
(2) $\text{SrTiO}_3$	0			
(3) Clinoptilolite	~35%			28%



Samples 2(B) and 3(B) were allowed to stand overnight, and another aliquot was taken from each and counted. Sample 2(B) showed 91% decontamination and 3(B) showed 90%.

In another experiment one-milliliter samples of rare-earth fission products were used. To one of these samples was added 100 milliliters of water, thus making 101 milliliters of sample solution, of which 1 milliliter was counted. This sample gave 22,400 counts per minute. One gram of stable strontium titanate was added and the solution was mixed for three hours. A one-milliliter aliquot was taken, evaporated to dryness and counted. This showed a 69% decontamination. The solution was allowed to mix for a total of 25 hours, after which another sample showed 89% decontamination. Several drops of 30% hydrogen peroxide were added to the solution and stirred overnight. Another sample was taken and counted. This sample showed that 99.6% of the original activity had been removed.

Decontamination of waste solutions thus appears very feasible by use of stable strontium titanate or clinoptilolite as scavengers. The effect of the presence or absence of hydrogen peroxide has not as yet been established. However, it is believed that the activity level of waste solutions can be reduced to the point where the solutions can be handled with minimum shielding.

### C. FUEL PROCESS ENGINEERING\*

#### 1. Design

The design of the Strontium-90 process equipment is based on the following objectives:

- (1) Double containment. All open vessels containing radioactive substances will be located in a dry box within a hot cell. If the substance is contained in a completely sealed metal vessel (except for emergency maintenance), it need not be placed in a dry box but will be located within the hot cell.

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\*C. Young

- (2) Liquid transfer. Transfers will be accomplished by vacuum methods, except that gravity transfer of premeasured quantities will be permitted. Quantity indicators will be provided for the critical vessels.
- (3) Valves. When possible, the valves will be located at the highest point in the line, with drainage either way. Where possible, the design will permit emergency replacement by remote methods or bypass of the faulty valve. The installation will be on a fail-safe basis. Manual, solenoid-operated or air-operated valves will be selected for the individual application.
- (4) Plumbing. Stainless steel tubing will be used in all lines to permit the transfer of nitric acid and radioactive solutions. The use of glass will be avoided because of radiation damage and possible breakage.
- (5) Fittings. Swagelok flareless fittings or the equivalent will be used to join the plumbing lines.
- (6) Dry box. The dry box will provide, as nearly as possible, absolute containment. Both inlet and outlet air will go through a prefilter and absolute filter. The box will be kept at a negative pressure in relation to the air in the hot cell.
- (7) Manipulators. The manipulators will be booted by a standard arrangement proposed by Los Alamos Scientific Laboratory (LASL) and commercially available from Central Research Laboratories or American Machine and Foundry (AMF). The manipulator fingers will be modified to provide a connection that is less likely to rupture the booting. In addition to the internal booting, a bellows will be provided over the external arms in such a fashion that the manipulators may be bagged upon removal from the cell.
- (8) Equipment. The various pieces of equipment will be selected to withstand the maximum radiation determined on a conservative basis. The radiation figures allow for 1-1/2 times the expected checkout and processing time, all at maximum levels. When possible, equipment that is subject to failure will be designed to permit maintenance by remote methods. A manned-entry philosophy will be pursued for heavy maintenance.

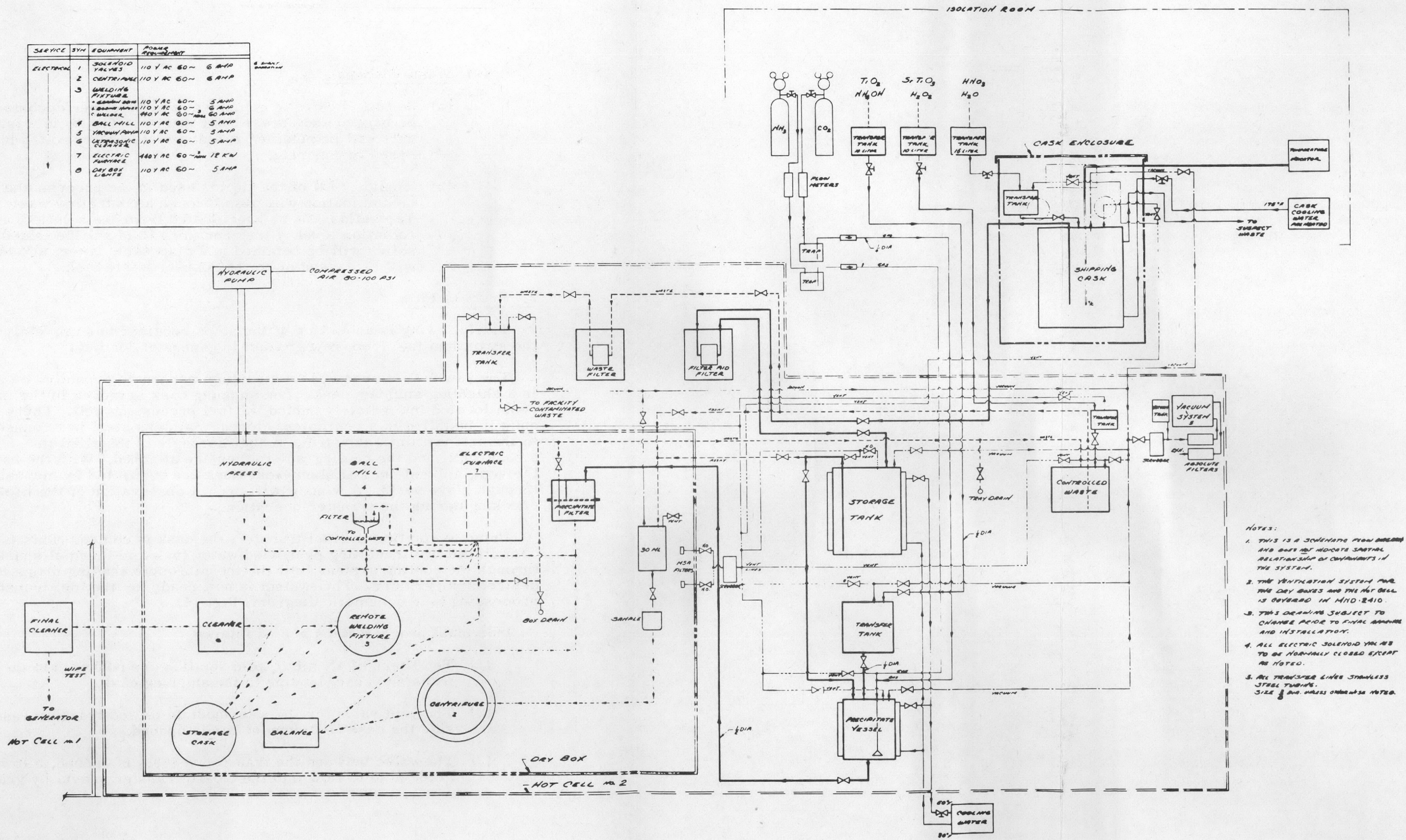


Fig. 14. Preliminary Fuel Process Flow Diagram

(9) Waste disposal.

- (a) Solids. Pieces of equipment will be either decontaminated or bagged and encased in concrete. The solid waste products will be retained in a disposable filter which is encased in concrete.
- (b) Liquids. All of the liquids used in the process and for decontamination will pass through a controlled waste tank. The solids will be precipitated from the solution, and the radiation level of the remaining fluid will be tested. The solids will be retained in a disposable filter, and the fluids will be passed on to the suspect-waste tank.

2. Process Flow

The following is an outline of the steps required to completely process the strontium fuel from raw product to generator loading.

The raw fuel is received as strontium carbonate deposited on a filter in a shielding shipping cask. The shipping cask is moved to the isolation room located immediately behind the fuel processing cell. There the cask is installed in a ventilated, closed, stainless steel box connected to the cell ventilation system. A thermocouple is installed in a cask well, and the cooling water lines are attached. With the box closed, the vent, solvent-in, and slurry-out lines are connected by manual means through glove ports. A window will permit observation of the hookup and checking during the transfer operation.

Prior to starting the fuel transfer, the cask is cooled. This is accomplished by circulating preheated water (to avoid thermal shock) through the cooling system until the thermocouple reading drops to the desired temperature. The system is now ready for the fuel transfer and processing (see schematic diagram, Fig. 14).

Individual process steps are as follows:

- (1) Ten liters of 4N nitric acid solution are poured into the open transfer vessel leading to the shipping cask.
- (2) The vent valve for the intermediate closed transfer vessel inside the cask containment box is opened.
- (3) The valve between the transfer vessels is opened, allowing the nitric acid to flow into the closed transfer vessel by gravity.

- (4) The valve between the transfer vessels is closed.
- (5) The shipping cask vent valve is opened.
- (6) The cask solvent valve is opened, allowing the nitric acid to flow into the cask by gravity.
- (7) The cask solvent valve is closed.
- (8) The nitric acid dissolves the strontium carbonate.
- (9) The vacuum valve on the storage tank and the valve connecting the storage tank to the shipping cask are opened. The strontium slurry is transferred to the storage vessel. The slurry passes through a filter which removes the filter aid prior to storage.
- (10) Steps (1) through (9) are repeated with a second 10-liter quantity of 4N nitric acid.
- (11) Steps (1) through (9) are repeated with a rinse of 5 liters of 4N nitric acid.
- (12) Steps (1) through (9) are repeated with a rinse of 5 liters of distilled water.
- (13) All valves to the shipping cask are closed.

Note: At this point the cask lines can be monitored, and, if the radiation levels are reduced to the specified levels, the shipping cask may be removed from the enclosure.

- (14) With the vacuum line and the lower vent line of the storage tank open, vent air is allowed to bubble through the liquid to provide agitation.
- \*(15) The vacuum line and lower vent line on the storage tank are closed, and the upper vent valve on the storage tank is opened. The vacuum line on the sample vessel is opened and 30 milliliters of the liquid is transferred.
- \*(16) The sample is removed in a 50-milliliter centrifuge tube. The solution is made basic with ammonia and precipitated with carbon dioxide. The sample is centrifuged, and the precipitate is weighed.

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\*These steps may be removed if sufficient analytical data on the feed material are provided by Hanford.

- (17) A quantity of the liquid is transferred from the storage vessel to the precipitation vessel. The exact volume to be transferred is based on the amount of sample precipitated but will be approximately three liters.
- (18) A titanium dioxide slurry is added to the precipitation vessel via the open transfer vessel and intermediate closed transfer vessel.
- (19) Ammonia is bubbled through the mixture in the precipitation vessel to make the solution basic.
- (20) Carbon dioxide is bubbled through the mixture to precipitate the strontium carbonate-titanium dioxide.
- (21) A vacuum is pulled on the controlled waste tank, and the precipitate and liquid are transferred to the precipitate filter. The liquid passes on into the controlled waste tank. The precipitate is washed prior to removal from the filter.
- (22) The precipitate is removed from the filter housing and ball-milled.
- (23) The precipitate is removed from the ball mill and placed in the furnace for calcining.
- (24) The calcined precipitate is removed from the furnace and transferred to the dispensing device.
- (25) The precipitate is loaded in the pressing die and compacted at a pressure of 5 tons per square inch.
- (26) The pellet is removed from the pressing die and transferred to the furnace for sintering.
- (27) The pellet is removed from the furnace and stored.
- (28) Steps (17) through (27) are repeated until enough pellets have been produced to load one capsule.
- (29) The capsule with pellets is placed into the welding fixture. The capsule lid is placed on the capsule. The dome over the welding fixture is closed, and the pressure is raised to two atmospheres by adding helium under pressure (the helium permits reliable leak checking of the weld with commercial equipment). The lid is welded to the capsule.

- (30) The closure weld is checked with helium leak detecting equipment.
- (31) The completed capsule is placed in the storage container.
- (32) Process Steps (17) through (31) are repeated until the required number of capsules to load one generator has been produced.
- (33) The generator is moved into the No. 1 cell and made ready for fuel loading.
- (34) The capsule is removed from storage and placed in the ultrasonic cleaner tank.
- (35) The transfer door in the dry box is opened.
- (36) The manipulator hand in the ultrasonic cleaner is cleaned.
- (37) With the clean manipulator hand the capsule is transferred by the sliding tray to Cell No. 1 and the dry box door is closed. The procedure for transfer is proposed but is being re-evaluated due to concern about "tracking" contamination during transfer. Presently LASL's proposed transfer can is being evaluated as a more positive transfer method.
- (38) With the manipulator in Cell No. 1 the capsule is removed from the transfer tray and placed in a second ultrasonic cleaner tank.
- (39) The capsule is removed from the cleaner and a wipe test is made.
- (40) The capsule is installed in the generator.

### 3. Performance Analysis

The equipment has been evaluated for thermal, structural and performance compatibility. As a result of the thermal analysis, it has been determined that water cooling should be provided for the storage tank, the precipitation vessel, the storage container and the welding fixture. The total heat to be removed has been conservatively estimated at a maximum of 6000 Btu per hour. This is to be handled by a water cooling system which is to be provided as a part of the cell facility.

The vessel designs meet the ASME unfired pressure vessel code. A design pressure of 15 pounds per square inch external (perfect vacuum) has been used for the vessels exposed to vacuum transfer operations to

allow for the possibility of a closed vent or transfer line. Stainless steel construction (AISI 316) has been used throughout to ensure resistance to corrosion and breakage.

In addition to the heat generated by the strontium, there is heat lost from the furnace. Preliminary data received from the vendor sources indicate that it is possible to maintain the furnace surface temperature low enough that it will not become a problem. A design value has been set at 125° F.

#### 4. Safety Analysis

A complete safety-failure analysis will be made on the completed design to determine modes of failure, consequences and the procedure to follow after failure. This analysis is scheduled to start in February.

The system design has been based on a fail-safe system. The use of any fragile materials has been avoided. In the event of a valve failure, the valve can be replaced by remote means or bypassed by means of a parallel system. Spill and leakage possibilities have been held to a minimum, and in case one of these should happen, provisions will be made to swab down the affected area.

#### 5. Shielding Analysis\*

For the purpose of evaluating material damage, it was assumed that the equipment would be subject to maximum operation radiation for a period of 18 months. Actually, the amount of processing scheduled will take less than 12 months, including the checkout and dry-run period.

The total radiation was computed on two different assumptions. First, the computation was made with the assumption that the maximum amount of radioactive material would be in each piece of equipment for the entire 18 months. The radiation level was then checked for an estimated percent of the total time based on the estimated operating time. The following list shows the total radiation dose (roentgens) from the critical items of equipment:

(1) Welder motor	$10^7$	} Full time	$3 \times 10^6$	} Part time
(2) Furnace insulation	$2.4 \times 10^8$		$1.3 \times 10^8$	
(3) Ball mill motor	$2.5 \times 10^7$		$6 \times 10^6$	
(4) Hydraulic press seals	$2.3 \times 10^7$		$6 \times 10^6$	

\* A. Spamer



As stated before, these values are very conservative in terms of time span and the fact that the construction of the equipment provides some shielding which has not been included in these calculations. The doses shown are within the limits of commercially available materials.

Other items of equipment given especially careful attention are the solenoid valves. With no shielding the total dose from a valve will be approximately  $10^6$  roentgens. It is planned to test these valves, prior to installation, to  $2 \times 10^8$  roentgens.

In addition to the natural shielding of the equipment, it is planned to add shielding around the electric motors, capsule storage container and shipping cask. This shielding should further prolong the life of the equipment and make feasible manned entry into the cell for short periods of time.

#### D. FUEL OPERATIONS\*

At the beginning of this report period, decontamination of the Quehanna Facility was well under way. All but two cells were below 750 disintegrations per minute per 0.1 square meter. As of December 8, 1960, about 75% of the contaminated surface was below 100 disintegrations per minute; the remainder (25%), below 300 disintegrations per minute. Weekly scrubbing of the floor area has reduced the level of the service area to the same level as the office area, relieving the staff of having to put on shoe covers and smocks when entering the service area. As a training measure, the staff members are required to dress properly before entering a cell or the decontamination area.

During December and January the emphasis was on cleaning and repairing equipment that was turned over to Martin as a part of the lease agreement. This includes such items as a remote-control welder, 3000-pound fork lift, counting instruments and various machine tools. In addition, considerable effort has been expended in learning the proper operation of the waste disposal facility. The primary components are a steam-vacuum evaporator, receiver, holding tank and various electronic probes. Considerable maintenance and repair were required before the facility would operate over long periods of time without attention. The Instrumentation Laboratory supervisor spent two days in instruction of the proper maintenance of relays and probes in this system.

A general analysis of the cooling requirements for the hot cells was made. This indicated that a water pump supplying 0.8 gallon per minute at 15 pounds per square inch through a 2-ton water chiller and a 300-gallon

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\* W. Stringham

storage vessel would be sufficient to cool all hot sources. The staff has started the installation of this equipment. At this time a 300-gallon tank, available at the site, has been descaled, painted and installed on the mezzanine over the cells. The order for the water chiller and pump has been placed. The copper manifold (1/2-inch OD) will be purchased locally and installed by the staff.

A pair of Model 8 manipulators were transferred to Baltimore so that design personnel could become more familiar with remote operation problems. In addition, more accurate mockup of equipment is assured.

#### E. STRONTIUM-90 FUEL SPECIFICATION\*\*

The Strontium-90 to be provided by the AEC for fueling all four generators in this program will represent the concerted effort of several contractor sites and AEC offices. Since the processes employed are to be used for the first time, and since no recovery of Strontium-90 has been carried out heretofore on such a large scale, it has been necessary to reinvestigate the limits of allowable impurities in terms of process restrictions and effects on the performance and safety of the generators.

Approximately 1 million curies will be separated from Purex fission product cuts at the AEC's Hanford Works, operated by the General Electric Company. Of this amount about 80,000 curies will be processed in a pilot plant especially constructed for this purpose, and the resulting purified product will be transported to the Fission Products Pilot Plant of Oak Ridge National Laboratory. At the latter site further purification will be performed if necessary; and the Strontium-90 obtained will be converted to titanate pellets, whereupon the pellets will be shipped to the Martin Quehanna facility for loading into the SNAP 7A and 7C generators.

Based on the flowsheet developed in the Hanford pilot plant, the remaining Strontium-90 is scheduled to be processed in the Hanford Hot Semi-Works Plant, now being modified for this purpose. The purified product will be shipped either to ORNL for completion of the fueling of 7A and 7C or to Quehanna in the form of strontium carbonate for conversion to the titanate and loading into the SNAP 7B and 7D generators.

Qualitative processing goals are as follows:

- (1) To reduce the concentration of gamma-emitting impurities to as low a figure as is economically feasible so that shielding requirements for the generators are minimized.

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\*\* J. Bloom

- (2) To maintain as high a strontium chemical purity as possible, while at the same time avoiding the addition of stable strontium diluent. The principal contributors to degradation of the Strontium-90, besides the stable strontium isotopes, are the chemically similar elements barium and calcium. Barium is present to a small extent in the strontium because it is a fission product also, while stable strontium isotopes are fission products and are introduced as minor impurities in the reagents employed in the process. Calcium is found in the product because it is employed as a reagent and is present as an impurity.
- (3) By employing gross fission products which are as old as possible (measured from the time of discharge from the reactors) as feed to the final purification steps, to reduce the concentration of short-lived isotopes which perturb the heat generation characteristics of the Strontium-90 fuel. Those isotopes which are chemically different from Strontium-90 are also removed by the separation techniques used, as in Item (1) above, but major difficulties are encountered because of the presence of fission product Strontium-89. This isotope (51-day half life) is formed in substantial activity relative to Strontium-90, and since it cannot be separated chemically, the only recourse is to permit it to decay to the point where its concentration in the product at the time of loading into the generators does not contribute materially to the thermal output of the heat source. If the isotopic ratio of Sr-89 to Sr-90 at the time of discharge from the reactors is assumed to be in the range 0.2 to 0.6 (the ratio is actually a function of the integrated neutron exposure in the reactors), then the relative thermal contribution of Strontium-89 as a function of time can be shown as in Fig. 7. High Strontium-89 concentrations also increase the cooling requirements during titanate preparation and increase the radiation level in the hot cells and from the loaded generators.

Based on the above reasoning, the Statement of Work for this program contains the following fuel specification:

"Fuel. Fuel shall be provided for the generator and shall consist of strontium-90 carbonate in dry-powder form as contained in its in-process filter. The fuel shall have a thermal output of approximately 1,546 watts and shall conform to the following:

- a. Assuming 90 per cent compaction, the fuel shall provide at least 0.50 thermal watts (excluding output of contaminants) per cubic centimeter.

- b. The required thermal output shall be provided by strontium-90. The thermal output of contained contaminants, including strontium-89, shall not be considered in measuring the output of the fuel. However, the thermal output of such contained contaminants shall not exceed 40 watts at the scheduled time of insertion of the fuel capsule in the generator. Stable contaminants shall not exceed 25 per cent of the calculated weight of a pure strontium-90 fuel element.
- c. Radiocontamination shall not exceed the following:

<u>Beta Emitters</u>	<u>Beta Curies/gm Sr-90 (Max.)</u>
Cerium-144	0.15
<u>Gamma Emitter Energy</u>	<u>Gamma Curies/gm Sr-90 (Max.)</u>
0.2 Mev to less than 1 Mev	0.15 curies
1 Mev to less than 2 Mev	0.015 curies
2 Mev or greater	0.0015 curies"

After extensive discussions between Hanford and Martin representatives, the following product definition for Strontium-90 separated at Hanford is close to mutual acceptance:

- (1) Product shall be shipped from Hanford to Martin as the carbonate, codeposited with a filter aid if necessary. The weight of the filter aid shall not exceed the weight of the product (measured as the carbonate). Product shipped from ORNL to Martin shall be in the form of the titanate, sealed in capsules provided by Martin.
- (2) Of the cations present in the product, not more than 25 weight percent shall be stable isotopes other than stable strontium isotopes.
- (3) The Strontium-90 content of the product shall be such that the minimum thermal power density of the titanate mixture produced from it shall be 0.50 watt per cubic centimeter. In

the computation, a material density 90% of the theoretical density of the mixed titanates shall be used, and the thermal output of the Strontium-90 isotope only shall be considered.

- (4) Radiocontamination (except for Sr-89) in the product, computed as of one year from reactor discharge, shall be less than the following:

(a) Beta emitters

Cerium-144--0.15 curie per gram of Strontium-90.

(b) Gamma emitters

0.2 Mev <  $\gamma$  energy < 1.0 Mev--0.15 curie per gram of Strontium-90.

1.0 Mev <  $\gamma$  energy < 2.0 Mev--0.015 curie per gram of Strontium-90.

$\gamma$  energy > 2.0 Mev--0.0015 curie per gram of Strontium-90

- (5) Concentration of Strontium-89 in the product shall be computed as of the time of shipment to Martin and shall be less than the following:

<u>Type of Product Shipped</u>	<u>Use</u>	<u>Sr-89 Concentration at Shipment (curies/gram Sr-90)</u>
SrTiO <sub>3</sub>	SNAP 7C	41
SrTiO <sub>3</sub>	SNAP 7A	41
SrCO <sub>3</sub>	SNAP 7D	34
SrCO <sub>3</sub>	SNAP 7B	34



## V. REFERENCES

1. "Preliminary Safety Report--SNAP 7A, Task 8--10-Watt Radiostrontium Generator for an Unattended Light Buoy," The Martin Company, MND-SR-2474, January 1961.
2. "Preliminary Safety Report--SNAP 7C, Task 8--10-Watt Radiostrontium Generator for a Portable Meteorological Station," The Martin Company, MND-SR-2475, January 1961.
3. Schneider, P. J., "Conduction Heat Transfer," Addison-Wesley, 1955, Equation 4-11.
4. McAdams, W. H., "Heat Transmission," Second Edition, McGraw-Hill, 1942.
5. Giedt, W. H., "Heat Transfer," D. Van Nostrand, 1957.
6. "Radioactive Materials Laboratory Safety Report--Martin Nuclear Facility, Quehanna Site," The Martin Company, MND-2410, September 1960.
7. "Program Plan for SNAP-7 Strontium-90 Fueled Thermoelectric Generator Development," The Martin Company, MND-P-2442 (Rev. 1), November 30, 1960.
8. "Statement of Work for SNAP-7 Systems," The Martin Company, MND-SW-1013 (Rev. A), November 30, 1960.
9. Monthly Status Letters, Task 8--SNAP 7 Program, Nos. 1 through 3, MND-P-2464, November 1960, December 1960 and January 1961.





## APPENDIX A

### HOT CELL FACILITY

A summary report (Ref. A-1) has been prepared by Martin to provide a general evaluation of the potential hazards associated with the Pennsylvania State University Hot Cell Facility located at Quehanna, Pennsylvania. It is an excellent background document; however, certain portions have been reproduced here for ready reference.

#### I. QUEHANNA SITE

The site of the hot cell facility is the former Curtiss-Wright Quehanna Research Facility located 37 miles northwest of State College in north-central Pennsylvania and includes parts of Elk, Cameron and Clearfield Counties. Figure A-1 shows the site location relative to the State of Pennsylvania. The site is perhaps the most remote one for a nuclear facility in the Eastern United States, since it is in a sparsely populated region and has a controlled exclusion area of approximately 80 square miles. The nearest population centers, Sinnemahoning (population 450) and Frenchville (population 558), are nine miles from the site. The site is ideally located for the proposed hot cell operations with respect to its population, meteorologic and hydrologic characteristics, and its general features are representative of those of the Appalachian Plateau physiographic province. The Commission has previously reviewed the Quehanna site for purposes of licensing former nuclear facilities and operations there (Ref. A-1).

#### A. SITE LOCATION

##### 1. Regional

Figure A-2 shows the site location relative to the surrounding region with circles of 5.1-, 10.4- and 25-mile radii superimposed. The highway network circles the site region and serves the closest population centers. This is well beyond the exclusion area of the site. The intervening land between the surrounding road network and the exclusion perimeter is principally State Forest land.

##### 2. Exclusion Area

The exclusion area is shown in Fig. A-3. Access is from Karthaus on a hard surface road running northerly to Quehanna at the edge of the

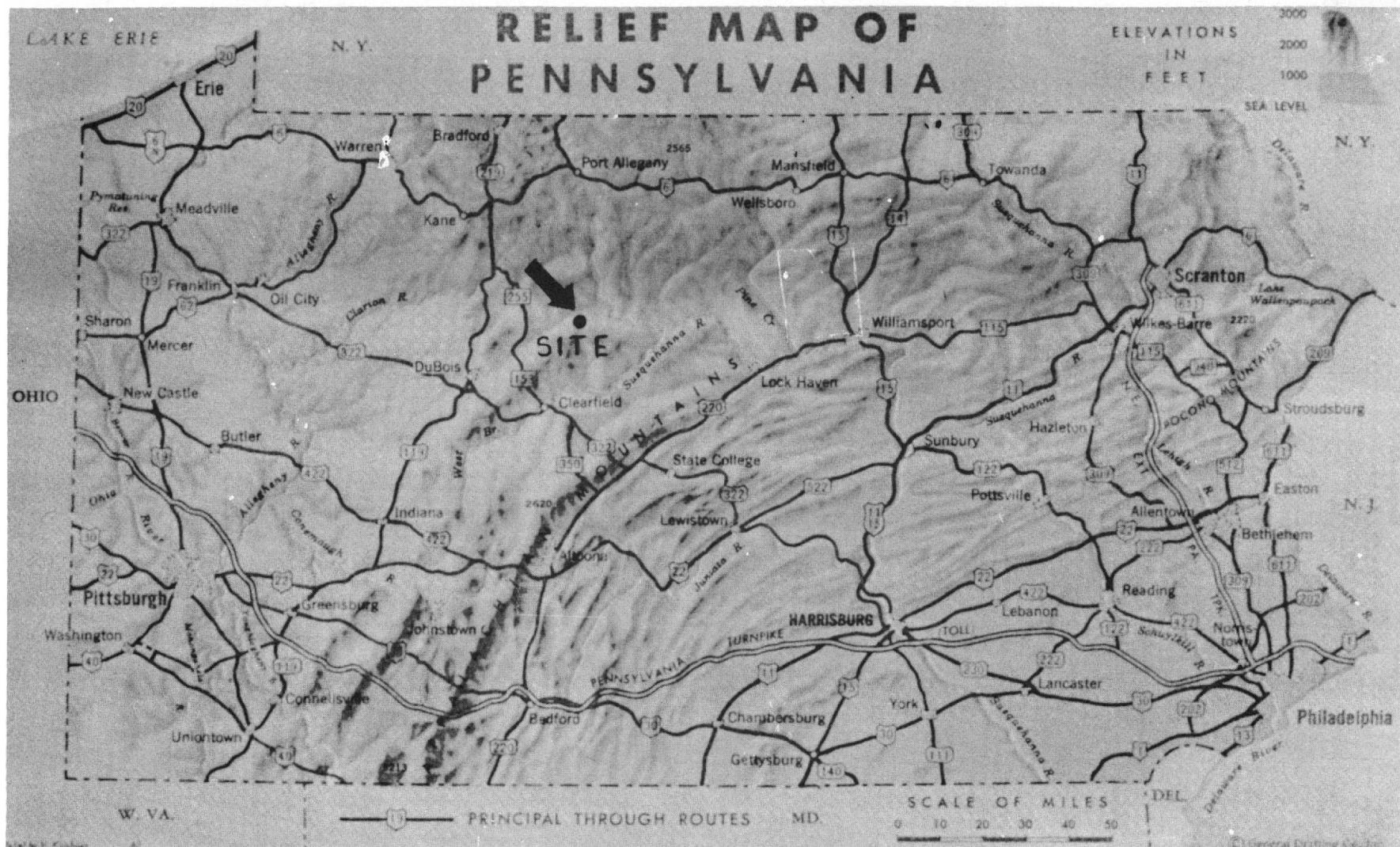


Fig. A-1. Site Location in Pennsylvania

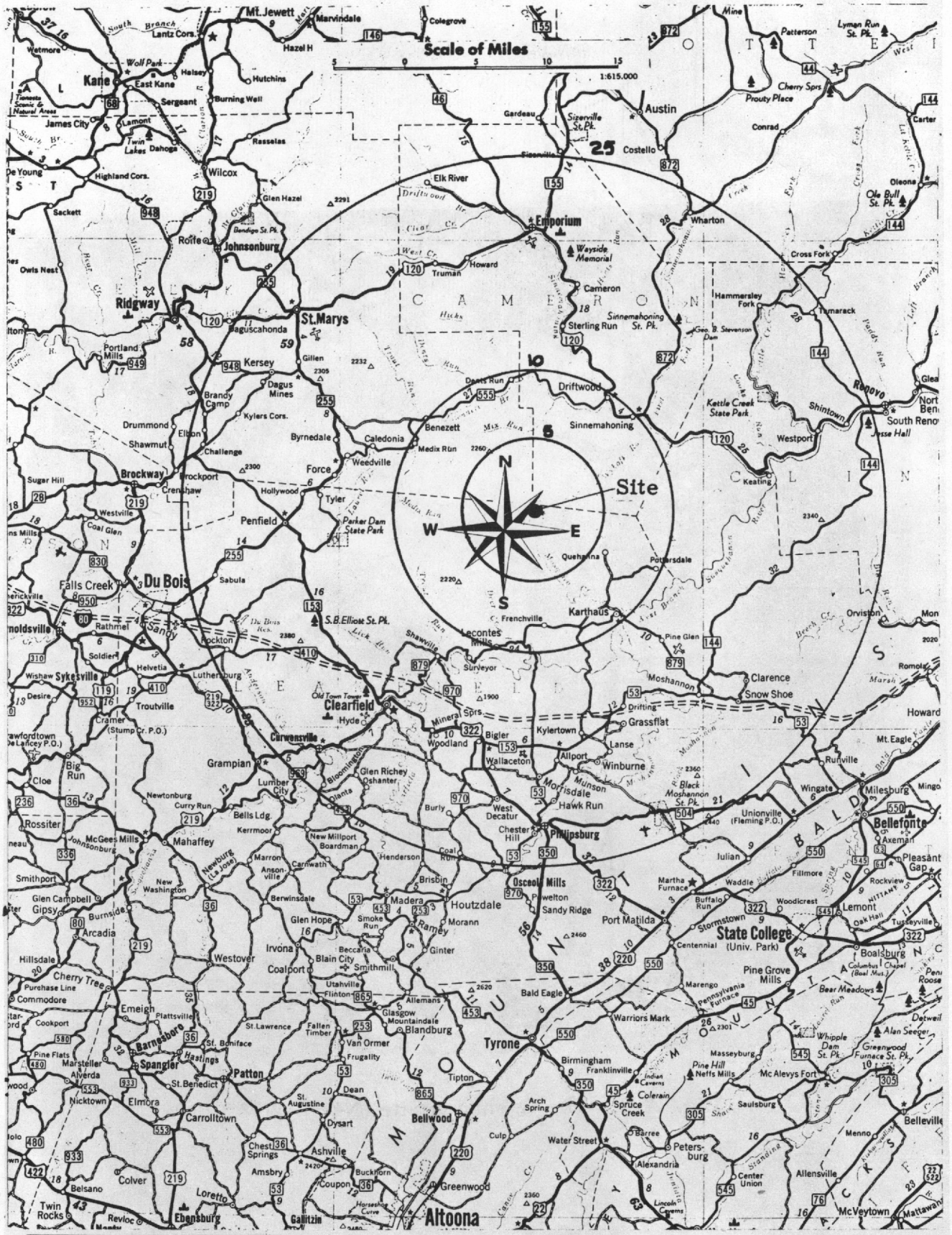


Fig. A-2. Regional Site Location (road map)

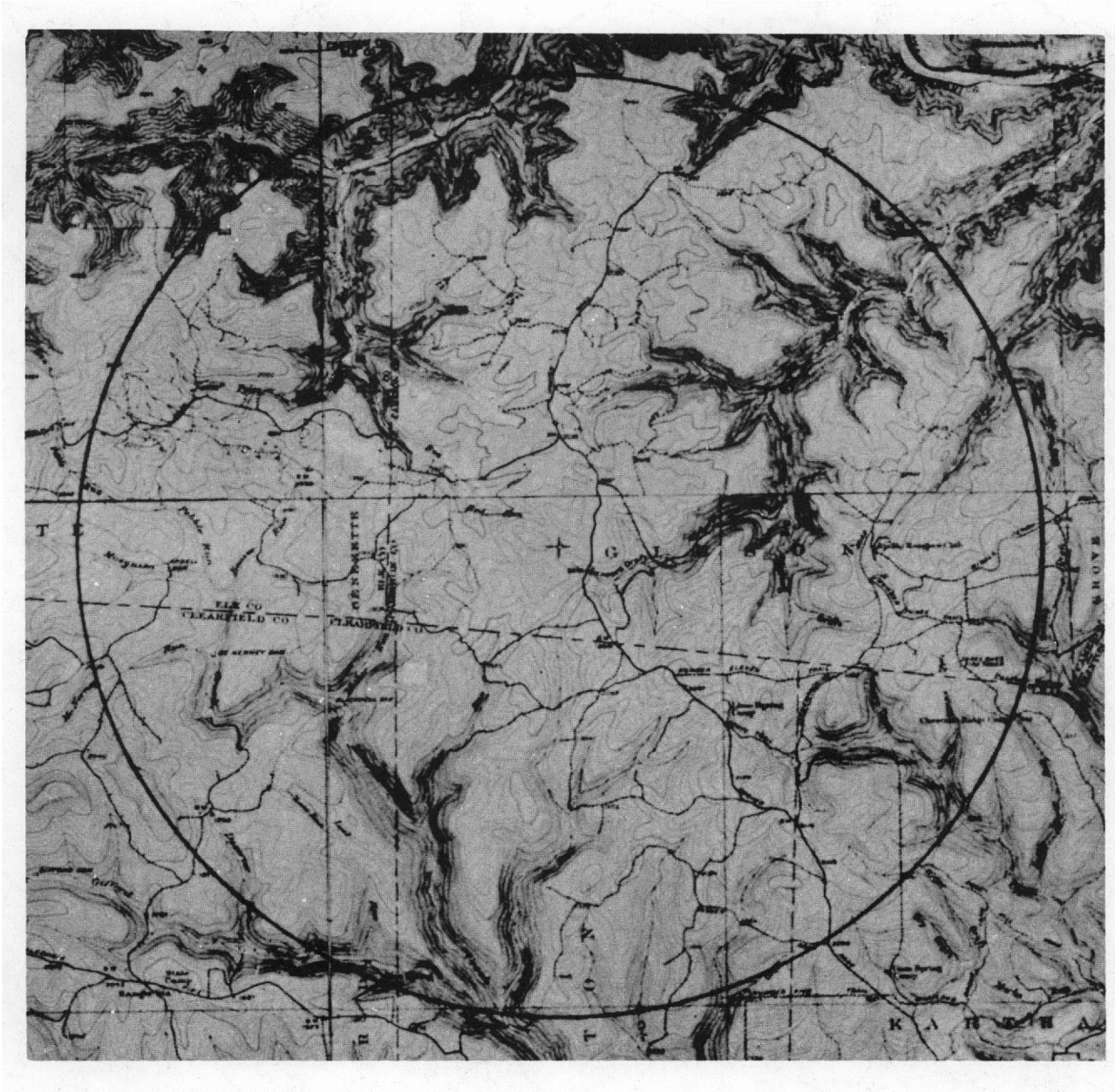


Fig. A-3. The Quehanna Site (N41°--W78°)

exclusion perimeter where entry is controlled by a manned gate. Other roads and trails into the exclusion area are closed. This area consists of a polygonal block of land which is roughly a circle 10 miles in diameter encompassing approximately 80 square miles. The facility site, consisting of the laboratory, attendant structures and a fenced security area, consists of several acres located in the southwest quadrant of the exclusion area.

## II. RADIOACTIVE MATERIALS LABORATORY

A view of the exterior of the building is shown in Fig. A-4. The exterior, of simplified contemporary design, consists largely of insulated aluminum curtain walls applied to the steel frame. The two large bays housing the reactor and the hot cell service area are evident. The reactor area is isolated from the cell service area except for a large truck door and a pedestrian door, both of which will normally be closed and locked.

The 24,700-square foot reactor and hot lab building is basically a one-story structure. Figure A-5 shows the main floor plan. It has been designed to provide as positive separation as possible between contaminated and radioactively clean areas. Those areas which will remain free of contamination include offices, dark room, staff laboratory, cell operating area and the reactor area. As reference to Fig. A-5 will reveal, the only normal access between clean and contaminated areas is via the change area. The large access door between the operating and service areas will normally be closed and locked. Care will be exercised in passing large objects through this door. Because of the relative isolation of the contaminated areas, an intercommunication system to connect all parts of the building has been installed.

### A. HOT CELLS

This facility consists of five cells with the physical configurations as indicated in Table A-1.

TABLE A-1

<u>Cell No.</u>	<u>Floor Size (ft)</u>	<u>Ceiling Height (ft)</u>	<u>Wall Thickness (ft)</u>
1	6 x 7	12	3
2	6 x 12	15	3
3	6 x 7	12	3
4	8 x 12	12	2
5	8 x 8	12	2



Fig. A-4. Exterior View of Laboratory Building

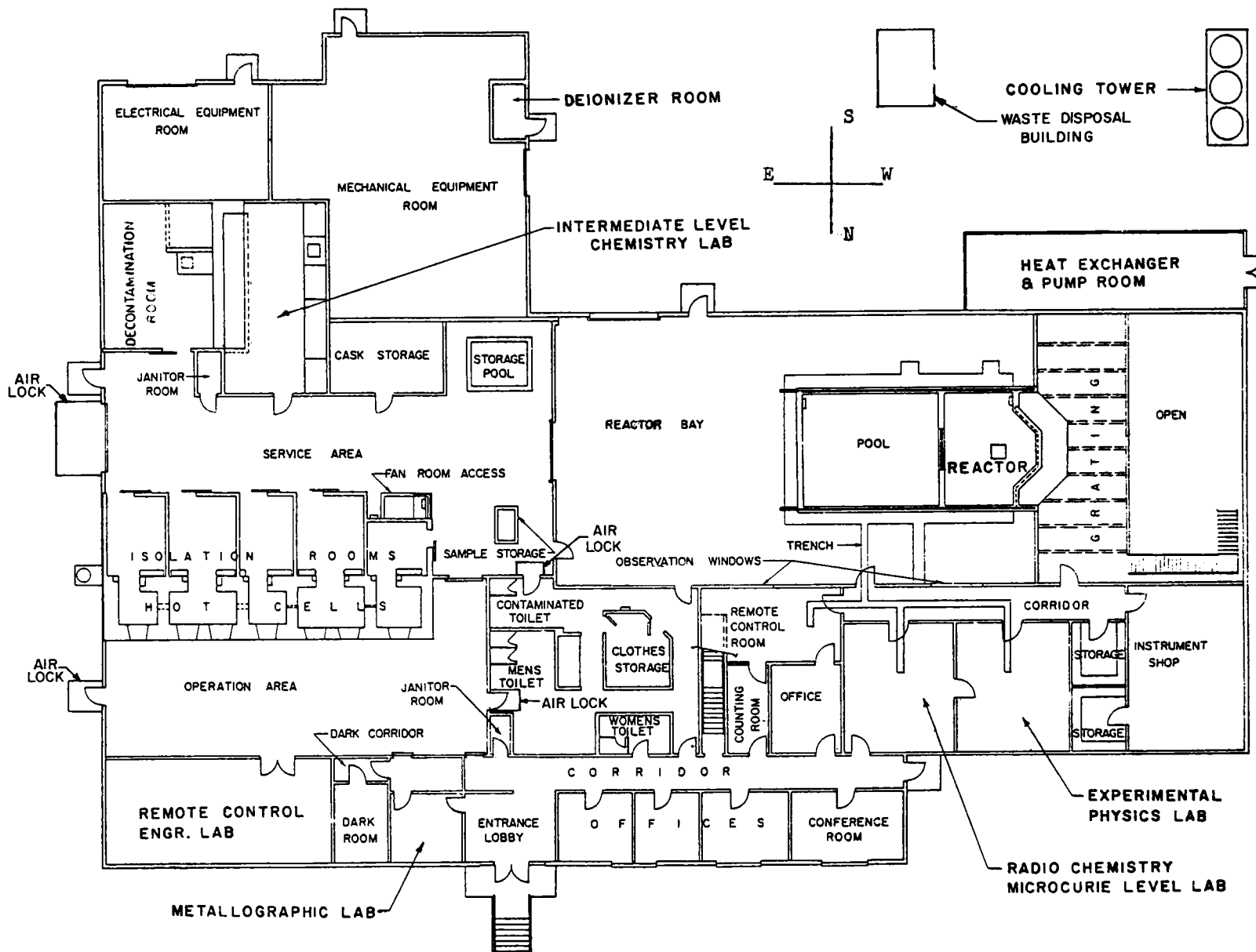


Fig. A-5. Main Floor Plan

Each of these cells is provided with manipulator ports for the use of Argonne Model 8 manipulators. Cells 2 and 4 have two pairs of manipulator port openings so the entire area of the cell can be covered. There is also a General Mills Manipulator, Model 300, in Cell 2. The configuration of the cell with the General Mills Manipulator is shown in Fig. A-6. A detailed plan view of the cell block is shown in Fig. A-7.

The shielding walls of the cells which reach to a height of 9-1/2 feet above the floors are constructed of ferrophosphorous concrete with a minimum weight of 280 pounds per cubic foot. The concrete above 9-1/2 feet is ordinary concrete having a minimum density of approximately 150 pounds per cubic foot. All of the dense concrete was poured with 1/4-inch steel plate forms which remained in place. The steel plates simplify decontamination of the cells, afford a good base for attaching fixtures to the interior or exterior and prevent chipping of the concrete around plugs, drawers and access ports.

The radiation shielding windows are of 3.6-density glass and were received as packaged, oil-filled units ready for insertion into previously installed steel frames. Each window is roughly the same thickness as the 4.5-density wall in which it is installed so that it is undershielded as compared with the wall. This undershielding can be made up by using an additional 4-inch thick slab of 6.2-density glass. There is one 4-inch thick 6.2-density glass block for supplementing the window shielding that can be used any time additional shielding is required. There is a total of seven windows, two in each of the larger cells and one in each of the remaining three cells. The windows are 36 by 48 inches on the hot face, giving a view of all of the cells.

Access to the cells is through doors at the rear which open into the isolation rooms. As reference to Fig. A-7 will reveal, the openings are of sufficient size and have been placed so that modular equipment tables covering one half, or, in the case of the large cells, one third of the floor area, can easily be removed or reshuffled within the cell. Considerable thought was given to the method of operating the doors. Equipment which is too large to pass through the doors may be placed in the cell by removing one or more of the two-foot thick slabs which form the cell roof. By using the 15-ton bridge crane to remove these slabs, the entire cell area may be uncovered if necessary.

Monochromatic lighting is provided in Cell 1 only. However, provision has been made to install sodium vapor lamps in the other cells if this proves desirable. In the meantime, illumination is supplied by incandescent lamps. Stand-by or emergency lighting is supplied in all cells so that lamp failures will not cause shutdown. A propane-fueled motor-generator set supplies sufficient power to continue operation of the cell ventilating and lighting systems in case of an electrical failure.



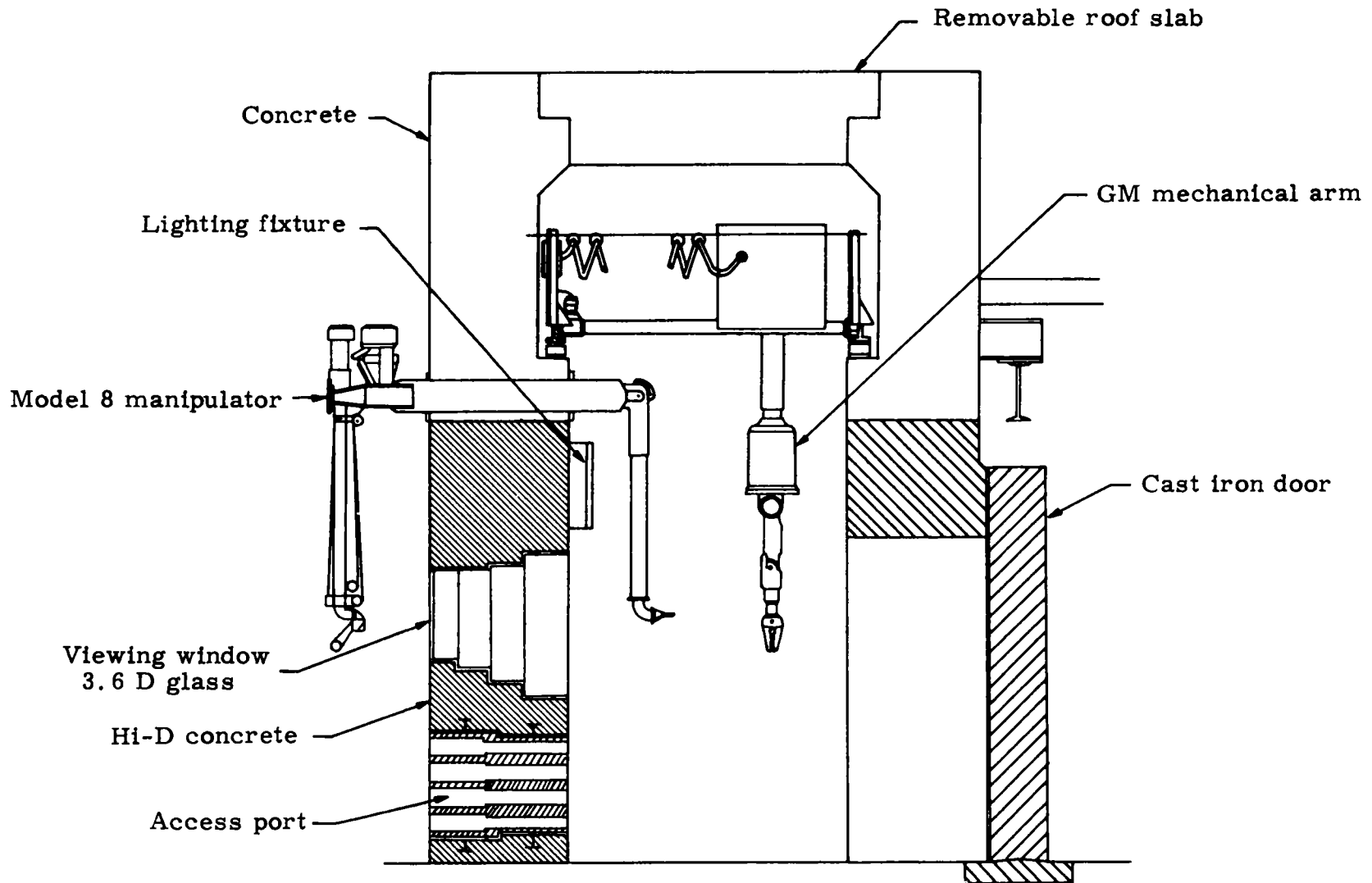


Fig. A-6. Cell Detail

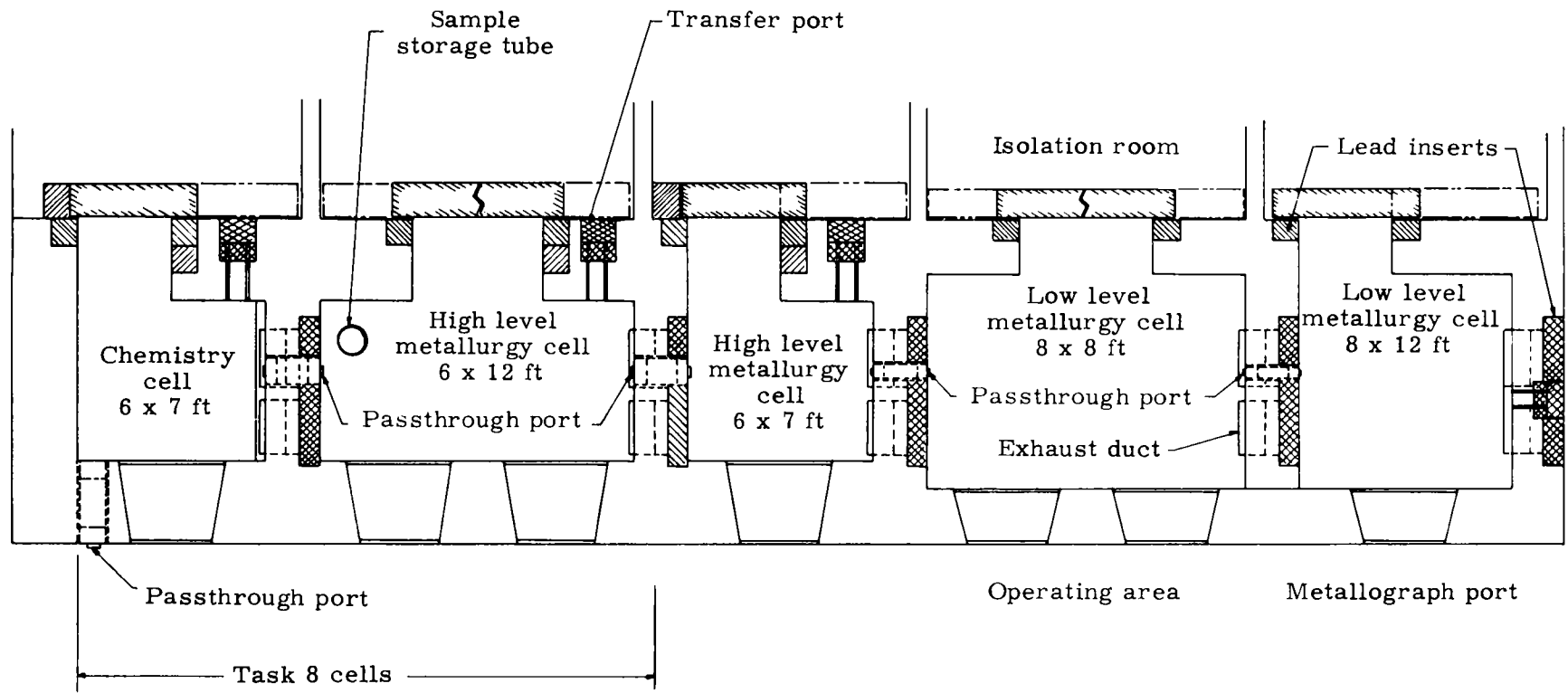


Fig. A-7. Detailed Cell Layout

Transfer of samples between cells is accomplished by using transfer drawers which can accommodate samples up to 12 inches on a side. These drawers are accessible to and may be operated by the manipulators. A pass-through drawer also penetrates the front face of Cell 1 so that small objects can be placed in the cell by the operator.

The operating face of the cells is liberally supplied with access ports. All standard services are available in a trench immediately in front of the cell face so that they may be extended through the wall as needed. Channels through plugs carrying services into the cells are bent so that there is no direct path for radiation from the cell to the operating area. Only a few 110-volt electrical lines are permanently installed within the cells. The arrangement of ports is shown in Fig. A-7. Many of them have been placed in rectangular drawers or inserts which can be removed and replaced with drawers having plugs designed for a specific application. Individual intercommunication sets connect the operating face and interior of each cell.

### III. REFERENCES

- A-1. "Radioactive Materials Laboratory Safety Report," MND-2410, The Martin Company, September 1960.



APPENDIX BHISTORY OF DEVELOPMENT OF STRONTIUM TITANATE  
AS A FUEL FORM

## A. PURE COMPOUND INVESTIGATION

A program was initiated\* whereby chemical and physical properties of the compound forms of Strontium-90 which might be used in a thermo-electric generator could be examined. Compounds originally considered were:

- (1) Strontium silicate.
- (2) Strontium biphosphate.
- (3) Strontium aluminate.
- (4) Strontium fluoride.
- (5) Strontium sulfate.
- (6) Strontium boride.
- (7) Strontium zirconate.
- (8) Strontium titanate.

These compounds were examined competitively in an effort to identify the superior material. The properties desired were:

- (1) Insolubility in hot or cold fresh water or sea water.
- (2) High strontium density.
- (3) High sintered density.
- (4) High melting point.
- (5) Good thermal conductivity.
- (6) Resistance to thermal shock and stress.

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\*AEC Contract AT(30-1) - 2220

During the early phase of this program the silicate, biphosphate, aluminate, fluoride and sulfate were eliminated. In each instance undesirable properties were discovered which seriously challenged the safe containment of Strontium-90 in the desired application.

As testing continued, strontium boride and strontium zirconate were eliminated. Strontium boride was dropped because of:

- (1) Incomplete tieup of strontium as the boride.
- (2) Widely varying rates of solubility in fresh and sea water.
- (3) Difficulty in fabricating by hot or cold pressing.

Strontium zirconate was eliminated because of:

- (1) Undesirably high fabricating temperatures.
- (2) Excessive solubility in fresh and sea water.
- (3) Moderate compound density.

Of all the compounds investigated, the titanate showed the best fabricability and highest degree of insolubility in fresh and sea water. Tracer experiments with Strontium-90 mixed into the strontium titanate pellets showed no activity present in tap water after 2400 hours of testing at 150° F. With natural sea water, after 2448 hours at 150° F, the average fraction of strontium titanate dissolved was 0.0003. At this rate it would take over 900 years to dissolve the pellet tested. Additional experiments were conducted with as much as 25% impurities of calcium and barium without destroying the strontium titanate ceramic linkage throughout the pellet. Samples of strontium titanate have been consistently compacted into various right cylindrical sizes with compact densities greater than 90%.

## B. IMPURITY EFFECT

The original fabrication was performed on pure strontium titanate bodies as it was originally assumed that all impurities would be removed. It has been found, however, that the strontium compound may be contaminated with other alkaline earth materials. Assuming all impurities to be calcium carbonate and/or barium carbonate, a study on the effect of these impurities has been made.

Figures B-1, B-2 and B-3 show the relations and effects of these impurity additions. Figure B-1 shows the relation of titanium dioxide per gram of carbonate blend for 90, 80 and 70 mole percent purity of strontium carbonate. An excess of 0.8 percent titanium dioxide has been used to ensure complete combination of the strontium compound. It should be noted that maintaining stoichiometric relations with titanium dioxide for all component carbonates is of the utmost importance to ensure chemical stability. Figure B-2 shows theoretical densities of the final sintered titanate bodies as a function of impurities present, and Fig. B-3 shows the corresponding strontium density in grams per cubic centimeter per mole percent impurity. These densities represent fully dense bodies, calculated on theoretical data, while the densities of pellets fabricated by normal techniques are expected to reach approximately 90 to 95% of these theoretical values. During the experiments conducted with impurity additions, pellets 90 mole percent pure were prepared using first a deficit of titanium dioxide, and second, an excess of titanium dioxide to determine whether any soluble compound of strontium would result. The basic results are as follows:

- (1) Pellets with approximately an 11% deficit of titanium dioxide show a strontium solubility of 0.002%.
- (2) Pellets containing an excess of 23% titanium dioxide indicate no strontium solubility.
- (3) No soluble barium salts were found.
- (4) Calcium was found to be soluble in bodies having either an excess or a deficit of titanium dioxide, but was more soluble with a deficit of titanium dioxide.

Bodies containing barium and calcium were also found to be much less homogeneous than the pure strontium titanate bodies.

### C. BLENDING

As a result of information obtained at ORNL, the dry blending of powder has not been further developed, as it was indicated that a wet-blend technique could be used. Wet ball-mill operations have produced the best results to date, and normal blending procedures are not effective enough to obtain uniform structures when impurities are present. In an effort to improve structure and reproducibility, a second ball-mill blending operation has been considered after calcining. This has improved reproducibility and density. Densities obtained with this method are plotted in Fig. B-4. It might be expected that the amount of strontium present in the filtrate after the first blending operation is insignificant as strontium is still in the form of the slightly soluble carbonate.

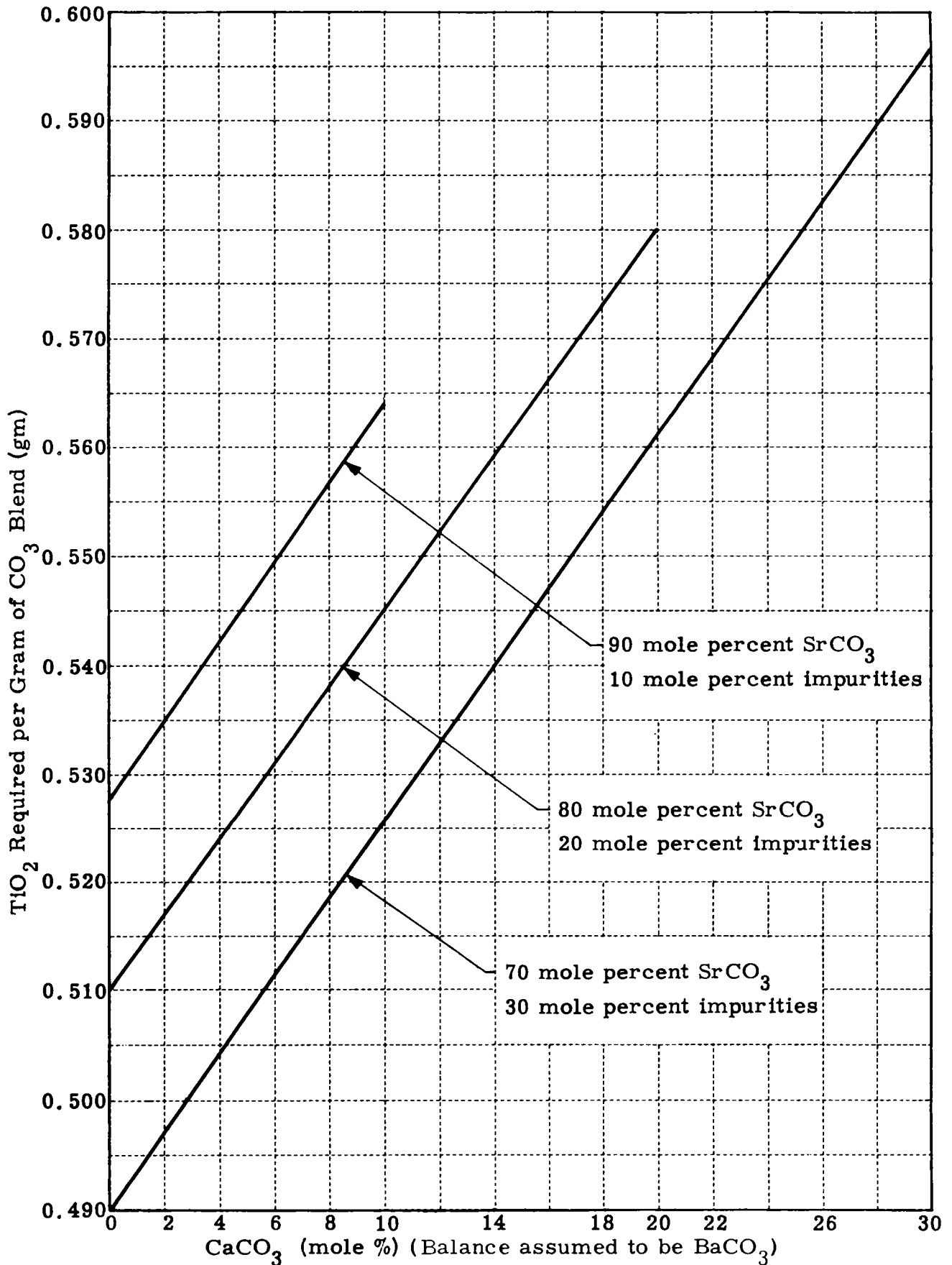


Fig. B-1. Effect of Impurities on the  $\text{TiO}_2$  Balance (Strontium titanate pellets)



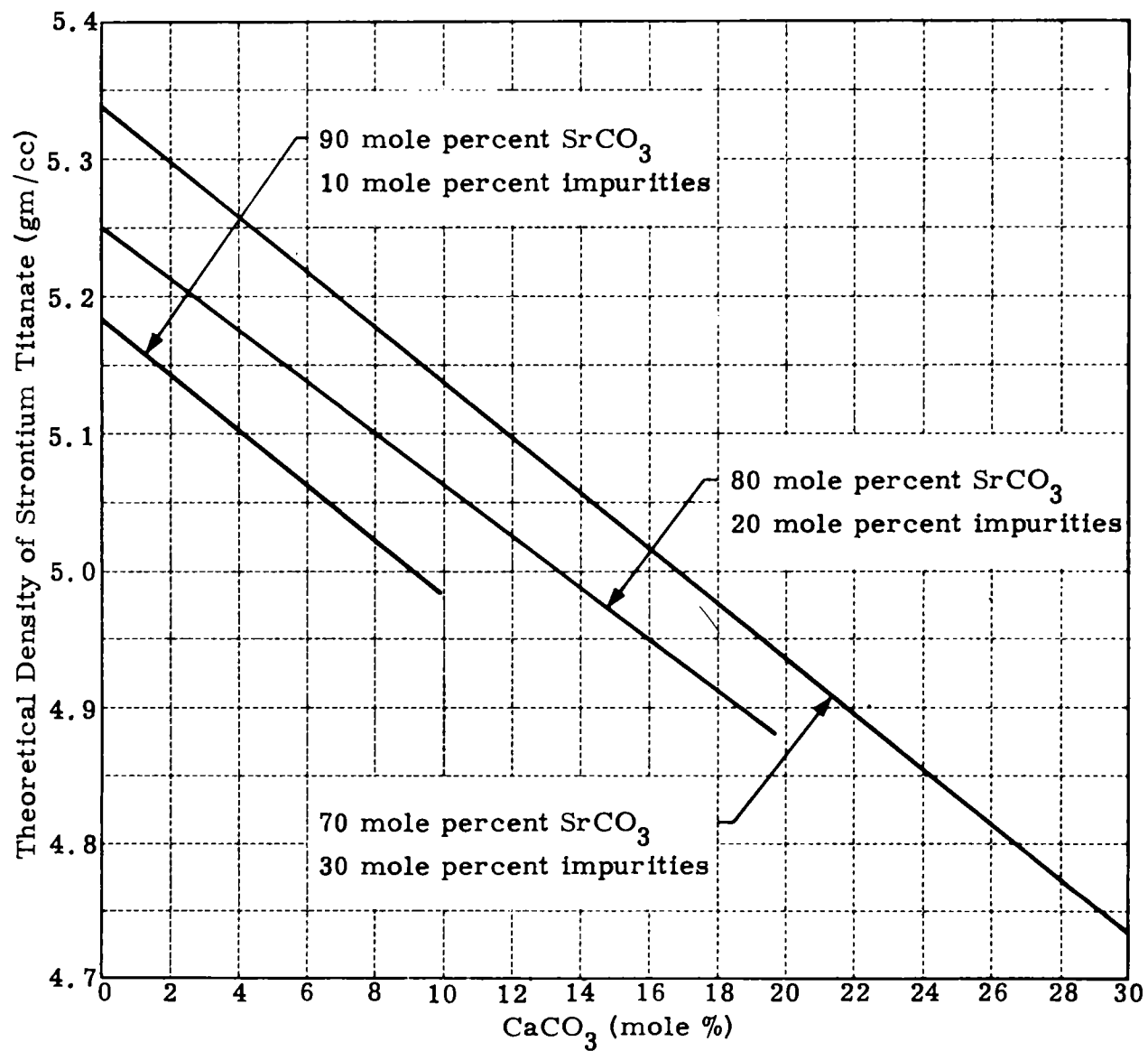


Fig. B-2. Theoretical SrTiO<sub>3</sub> Pellet Density as a Function of Impurity Present

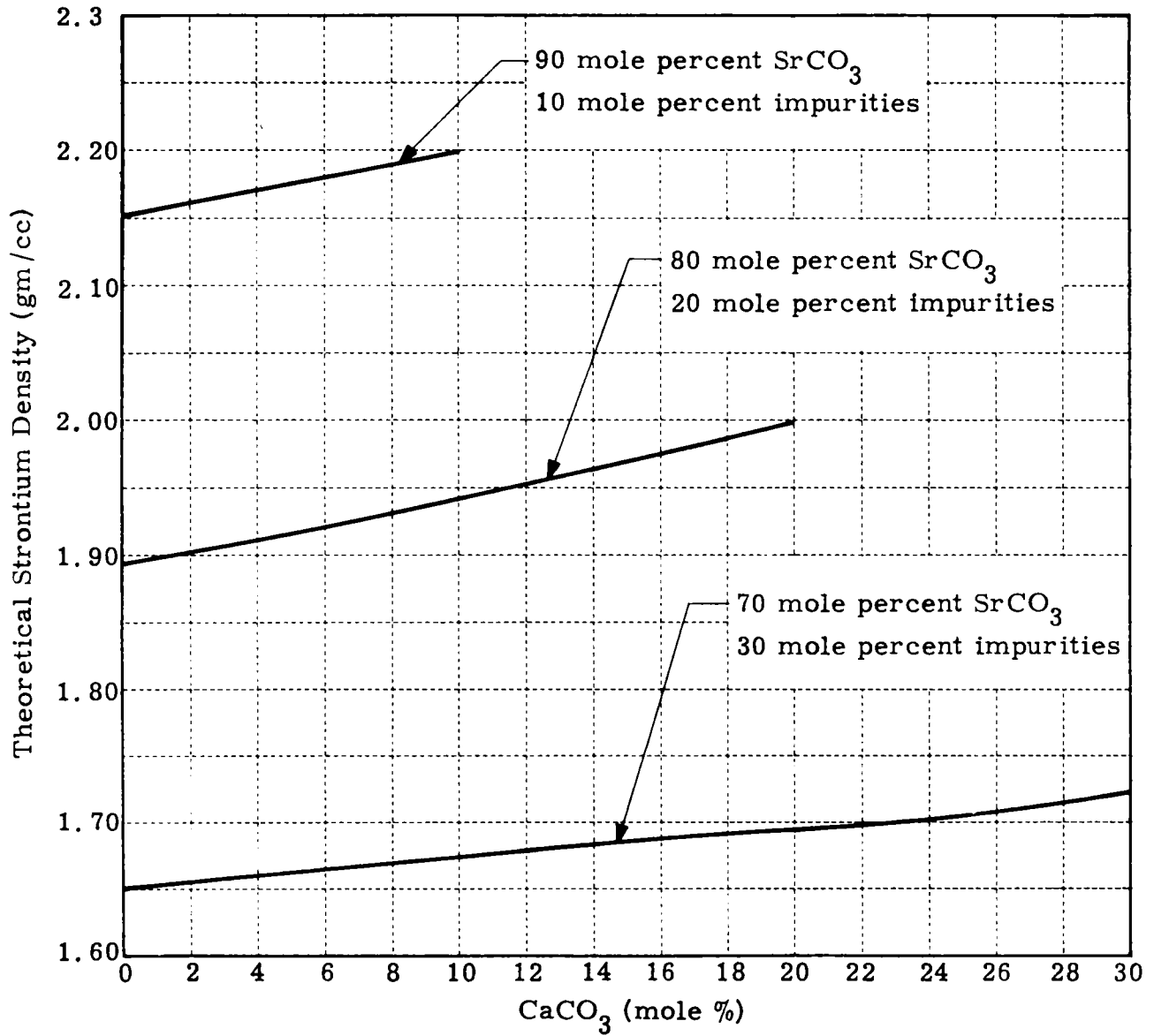


Fig. B-3. Theoretical Strontium Density as a Function of CaCO<sub>3</sub> Impurity

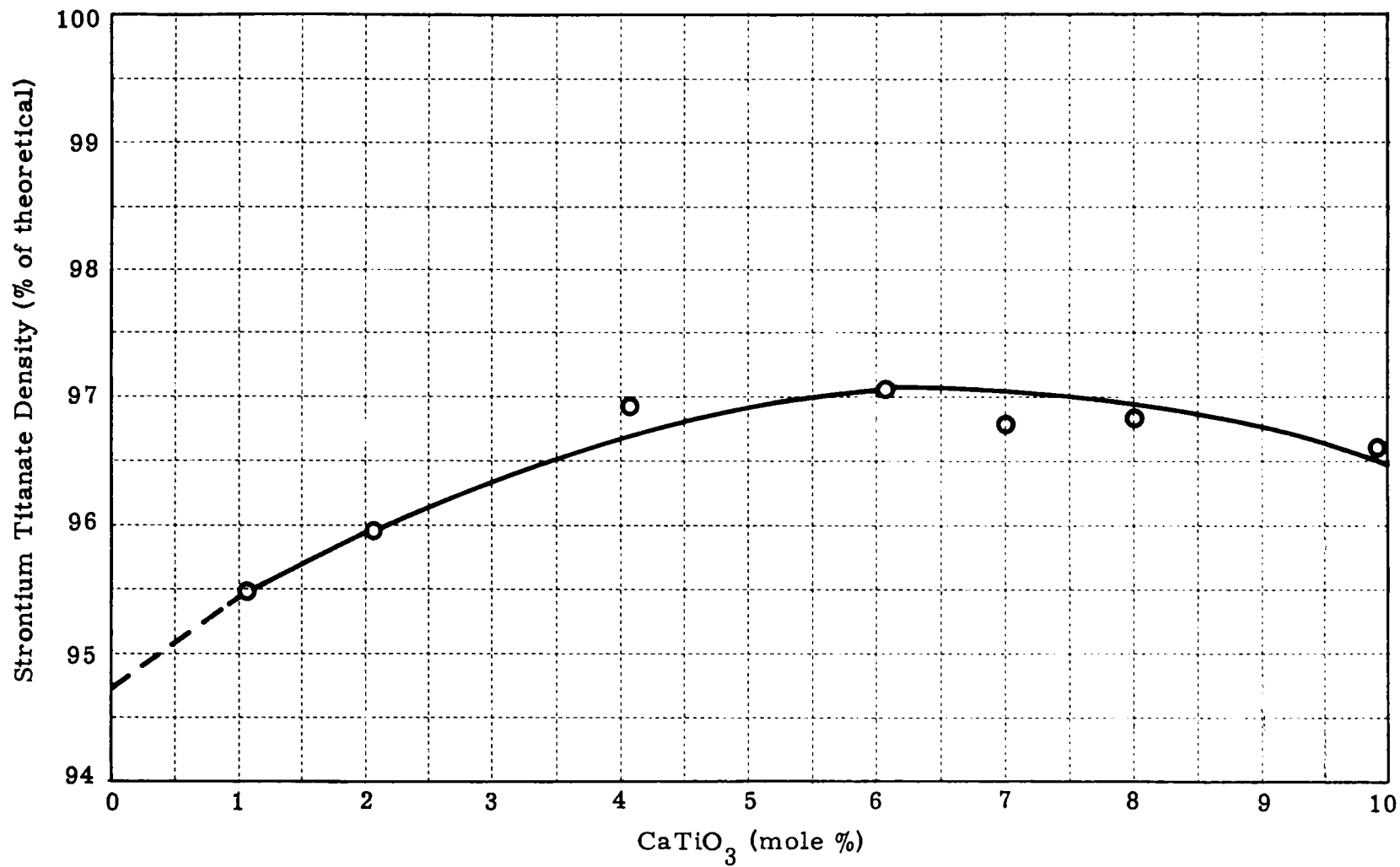


Fig. B-4. Effect of Mole Percent  $\text{CaTiO}_3$  on Pellet Density with Aid of Additional Milling

It was found, however, that wet blending after calcining removed some soluble strontium, now in the form of the oxide SrO; and, although the loss was not detrimental, a different firing schedule has been established to help eliminate this loss. Although a shift in the stoichiometric relation (addition of a 10% excess of titanium dioxide) aids in stabilization for the second blending, the use of a higher calcining temperature proved more satisfactory. Analysis of the filtrate of material calcined at a higher temperature (1200° C) showed a 0.02% solubility of strontium, and material processed with a lower (1100° C) calcining temperature showed a solubility of approximately 0.3%. Further tests have shown that pure strontium titanate, as received, also shows a 0.02% solubility when wet blended and filtered through the same filter as used in the process. This 0.02% of strontium found in the filtrate from the "as received" strontium titanate indicates a carrythrough of material with a 20-micron filter, with perhaps some slight solubility of strontium. Table B-1 compares final pellet densities obtained from powders calcined at 1100° C and 1200° C. Both powders were ball-milled after calcining.

TABLE B-1

Effect of Increased Calcining Temperatures on Final Density

<u>Calcining Temperature (°C)</u>	<u>Sintering Temperature (°C)</u>	<u>Diameter Shrinkage (%)</u>	<u>Theoretical Density (%)</u>
1100	1350	22.75	95.46
	1400	22.9	95.56
1200	1350	19.6	95.31
	1400	19.7	95.52

Table B-1 shows that high densities can be achieved by using a higher calcining temperature with the aid of ball-milling after calcining. Early tests in which a single ball-milling operation was used with a 1200° C calcining temperature gave a lower density. Another point which favors the use of a higher calcining temperature is the change in the calcined material on aging. Material processed with a lower calcining temperature that has been ball-milled after calcining shows a lowering in final pellet density if an excessive time lapse is allowed before sintering. Although X-ray analysis showed no change in structure, pellets pressed and sintered 8 to 10 weeks after calcining suffer a 10 to 15% drop in density. It is fairly reasonable to assume that material calcined at a temperature high enough for a more complete reaction will yield a more stable product.

Because of the difficulty of performing ball-mill operations in a hot cell, future developmental effort will be pointed toward complete elimination of this type of blending operation.

APPENDIX C

## MANUFACTURING DRAWINGS AND WEIGHTS\*

The majority of the manufacturing drawings for the 10-watt thermoelectric generator were completed during this period. For convenience, the complete list of details and assembly drawings is tabulated with their drawing numbers.

<u>Item</u>	<u>Drawing Number</u>
Fuel element cylinder	398-3021001
Fuel element end plates	398-3021002
Fuel element assembly	398-3021003
Fuel block	398-3021004
Fuel block plates	398-3021005
Shield cylinder (uranium)	398-3021006
Shield block (uranium)	398-3021007
Insulation strip	398-3021008
Insulation block	398-3021009
Thermoelectric elements	398-3021010
Hot shoe	398-3021011
Cold shoe and contact	398-3021012
Piston	398-3021013
Spring washer	398-3021014
Heat sink frame	398-3021015
Heat sink bar	398-3021016
Insulation corner strip	398-3021017
Insulation spacer	398-3021018
Insulation block	398-3021019
Wire guide	398-3021020
<u>Assembly</u>	<u>Drawing Number</u>
Generator--biological shield assembly	398-3021000
Biological shield	398-3021041
Generator assembly	398-3021042
Generator cover assembly	398-3021043
Hot shoe assembly	398-3021045
Cold shoe assembly	398-3021046
Thermoelectric module assembly	398-3021047
Heat sink assembly	398-3021048
Generator shell assembly	398-3021049

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\* H. Morton

In addition to the drawings, the estimated weights and dimensions for the 10-watt generator are as follows:

	<u>Weight (lb)</u>
Top uranium shield	114
Biological shield (uranium)	1320
Hastelloy C clad, fins and solder for biological shield	285
Fuel, generator and Hastelloy C generator housing	<u>121</u>
	1840 total

	<u>Dimensions</u>	
	<u>Size (in.)</u>	<u>Height (in.)</u>
Fuel block	4.36 between flats	5.73
Heat sink	8.75 OD	7.00
Generator and housing	9.25 OD	15.38
Biological shield (cylinder)	9.50 ID	19.75
(over fins)	16.25 OD	20.88
	19.25 OD	(including lugs)









