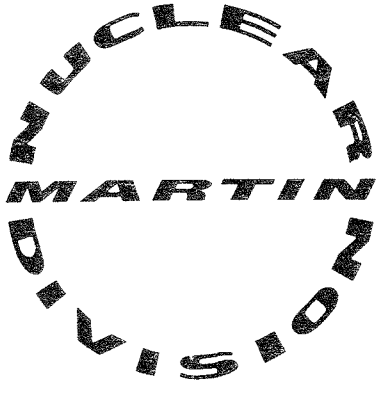

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AUG 361

SNAP Programs

Quarterly Progress Report No. 2

Task 8--Strontium-90 Fueled

Thermoelectric Generator Development

February 1, 1961, through April 30, 1961

MND-P-2483-2

Prepared by

U.S. West

Ass't. Project Engineer

NUCLEAR DIVISION MARTIN

Approved by

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Project Engineer

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FOREWORD

This quarterly report covers the period from February 1, 1961 through April 30, 1961 and has been prepared under the requirements of Contract AT(30-3)-217 with the Atomic Energy Commission.

CONTENTS

CONTENTS (continued)

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 these thermoelectric generators. Fuel process flow and associated equipment requirements for remote conversion of Strontium-90 feed material to strontium titanate pellets are covered. Previously evolved technical standards concerning raw fuel material specification have been coordinated among interested agencies.

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 Subtasks 8.1 and 8.3, one for Subtasks 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 the last report period (Ref. 1). During the period of this report, many components and subassemblies were fabricated; these are shown in Appendix A. Insulation and heat sink items, which are to be tested in the reliability model, were completed.

Engineering and analysis continued during this period. This activity centered about:

(1) Evolution of installation details, including electrical and physical attachments.

(2) Correlation of actual dose rates measured at Oak Ridge National Laboratory with shielding calculations previously made with the IBM 709 computer.

(3) Heat transfer analysis of the proposed SNAP 7C installation in Antarctica.

(4) Ten-watt generator couple tests.

In all activities undertaken during this period, excellent results were obtained and the program progressed as planned.

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 5 watts, direct current, at a nominal output of 12 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 service in Antarctica. Figure 3 shows the installation of the weather station in a circular steel housing. During its first year of operation, the weather station will be located in Antarctica on the Ross Ice Shelf (Fig. 4). The power supply system will consist of a 10-watt thermoelectric generator, a 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 SNAP 7C power supply system is 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 fuel capsules will be loaded in the thermoelectric generators at the Martin-operated Quehanna facility (Ref. 1).

The major effort during this report period was the completion of the design of the 10-watt thermoelectric generator and the integration of the generator into the SNAP 7A and 7C power systems. The succeeding pages of this section will present pertinent details concerning the respective SNAP 7 systems under the following headings:

(1) Generator design.

(2) Shipping cask.

(3) Heat transfer analysis.

(4) Safety and shielding considerations.

(5) Thermoelectric tests.

(6) Manufacturing drawings and weights.

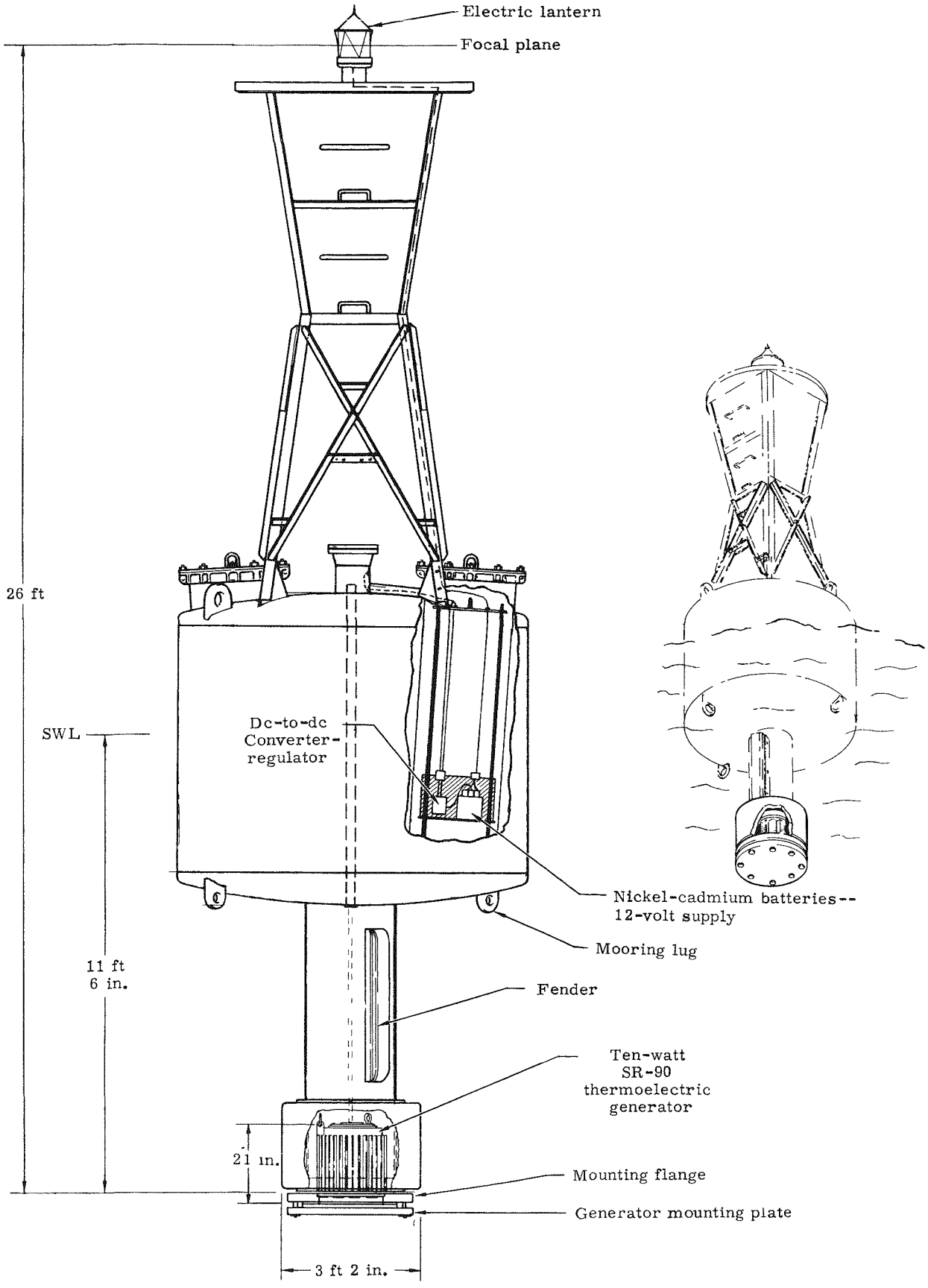
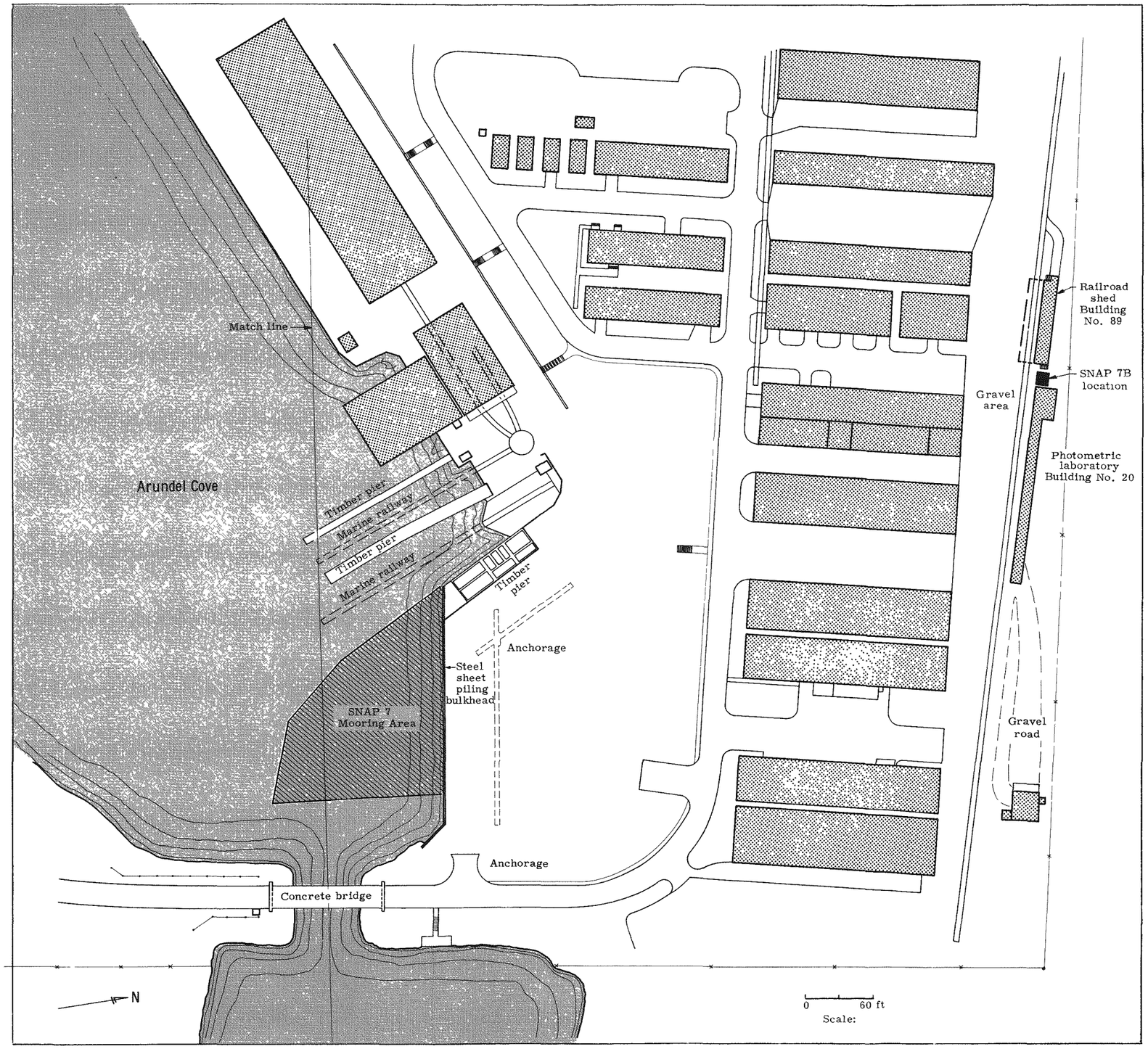


Fig. 1. U.S. Coast Guard Light Buoy



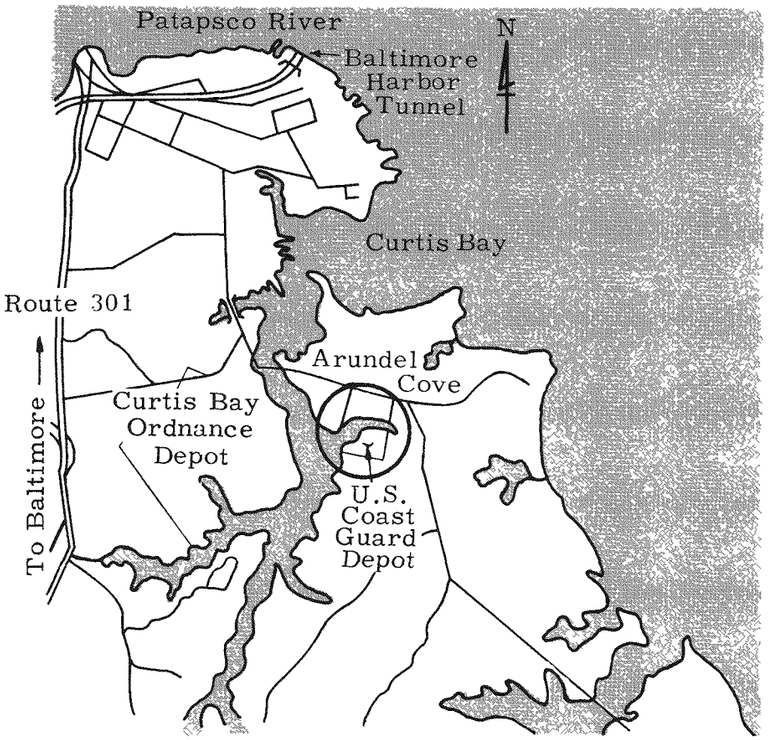
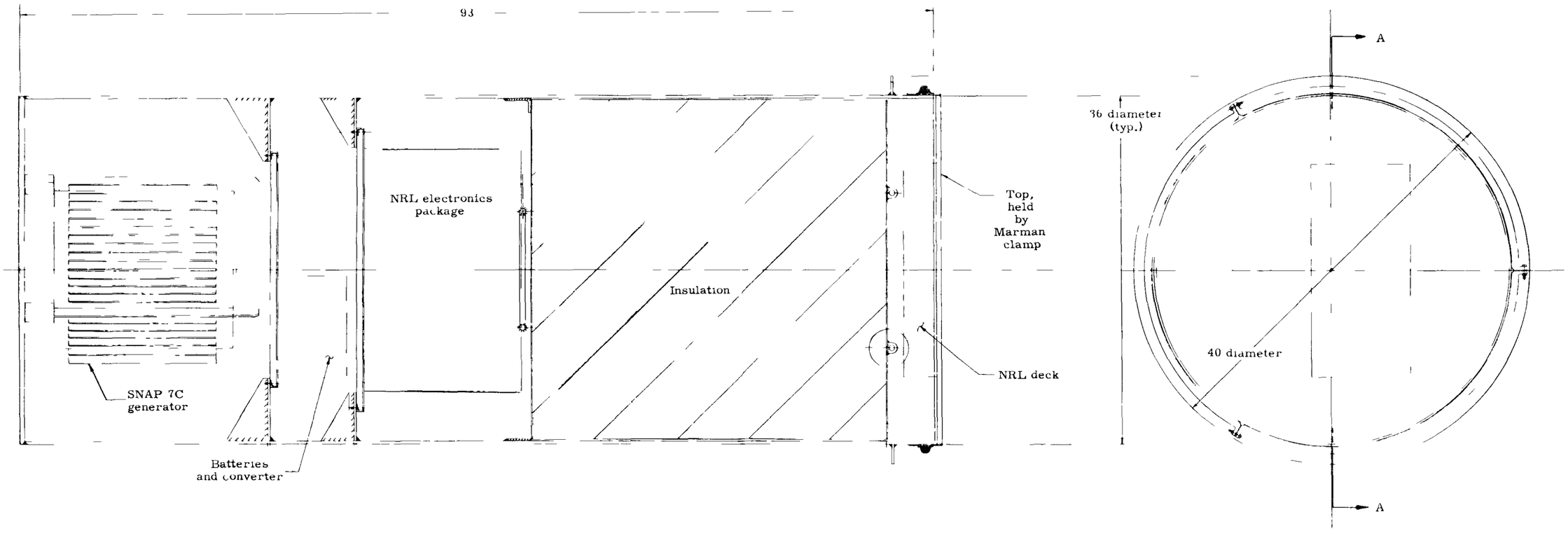


Fig. 2. Site of SNAP 7B Installation



Section AA

Dimensions in inches

Fig. 3. SNAP 7C Weather Station Emplacement Tube

Fig. 4. Antarctica, Showing SNAP 7C Location

sign*

the SNAP 7A and 7C thermoelectric generators was the last period, with the exception of the design of electrical connector. Several designs were considered nies manufacturing these connectors were contacted. mited quantity (4), only two connector suppliers were s special application. One of these later declined to n decided to use a connector similar to the one employed -watt Strontium-90 generator developed by The Martin Com- C Contract AT(30-1)-2519) and described in Ref. 6. This is ification of a special glass seal connector design, pressuretight mixtures of gas containing hydrogen, which can withstand long ls of immersion in water. For purposes of describing the under- hilosophy of the connector design, it is assumed that the generator ivering current to the connector and that the junction box is flooded with sea water, thus creating a short circuit across the outer terminals of the connector. Since the electrical connector case is fabricated of Hastelloy C, there is no greater corrosion at that point than 0.0001 inch per year, the rating of Hastelloy C. Even with current flowing, the platinum electrodes in the connector will not go into solution as other metals would. Therefore, the electrodes will remain intact and maintain the outer Hastelloy C shell as a second corrosion barrier.

The cover and connection from the generator junction box to the batteries and dc-to-dc converter will vary with the system application. The SNAP 7A generator has been designed to withstand being submerged in sea water. Further, using Hastelloy C pipe to protect the extension of the cables to the converter and batteries will give better protection and greater reliability for that specific application. In the case of SNAP 7C, the generator is enclosed in a steel container and buried in ice; therefore, a simple cable connection is suitable for internal wiring.

A preliminary drawing (Fig. 1) for the installation of the SNAP 7A generator in a flashing light buoy (Model 8X26E) was prepared and reviewed with the Coast Guard. From this review, the revisions of the buoy were agreed upon; they will be incorporated during the next quarter.

A preliminary drawing (Fig. 3) was drafted for the Navy installation of the SNAP 7C power system. The drawing was reviewed with Navy representatives, and the installation was found acceptable for use in Antarctica. Only the design of outriggers, which are required to keep the housing from melting through ice, and the attachment of the transmitting antenna have to be resolved to complete installation of the SNAP 7C system.

2. Shipping Cask

Fuel capsules for the SNAP 7A and 7C thermoelectric generators are scheduled to be loaded with $SrTiO_3$ at Oak Ridge National Laboratory. A shipping cask will be needed to transport these capsules from ORNL to Quehanna, Pennsylvania. Rather than build special containers, it was decided to modify the biological shield of the 10-watt generator so that the

*H. Morton

shield would serve as the shipping cask. The inte
was replaced with a bored aluminum block, holding
in the same position, relative to the shield, as whe
generator. The shielding calculations were checke
was found to be within the ICC requirements of 10
and/or 200 mr/hr on the surface, for the new appl
is shown in Fig. 5. The biological shield and the t
later with the 10-watt generator after the fuel has b
Quehanna. Prior to fuel loading, a wooden block will
shield for shipping from Baltimore to Quehanna.

3. Heat Transfer Analysis*

A temperature analysis of the SNAP 7C power supply emp
was performed in order to determine the wall temperature of
weather station housing during operation in Antarctica.

Basically, it is undesirable to expose the generator to the Antarctic
environmental temperature variations because of the effect on generator performance. Further, the weather station batteries and electronic gear are not designed to withstand the Antarctic environment. A suitable environment for the batteries and electronic gear can be provided and temperature excursions minimized by packaging all weather station components, including the generator system, within a single container which will be buried in permanent ice.

At depths below approximately four feet, the ice temperature responds slowly to variations in local air temperature. The maximum variation in ice temperature at a depth below four feet has been measured to be about 50° F when the local air temperature variation is in excess of 100° F.

The generator and equipment will be located at the bottom of a cylindrical container and covered with a plug of styrofoam insulation which completely fills the upper end of the cylinder (see Fig. 3). An exact thermal analysis of this container is difficult because of the geometry and temperature gradient in the ice itself. An approximate analysis* was performed to determine roughly the temperature which may be expected within the buried container. The results of this analysis indicate that the air temperature within the steel container housing will range from a maximum of 60° F to a minimum of 20° F.

4. Safety and Shielding Considerations**

Shielding calculations were performed with an IBM 709 computer to compare calculated dose rates from a 1000-curie source of Strontium-90 with actual values measured at Oak Ridge National Laboratory. A detailed report of the comparison between calculated and measured dose rates was prepared and is included in Appendix C of this report.

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*A. Streb (see Appendix B)

**A. Spamer

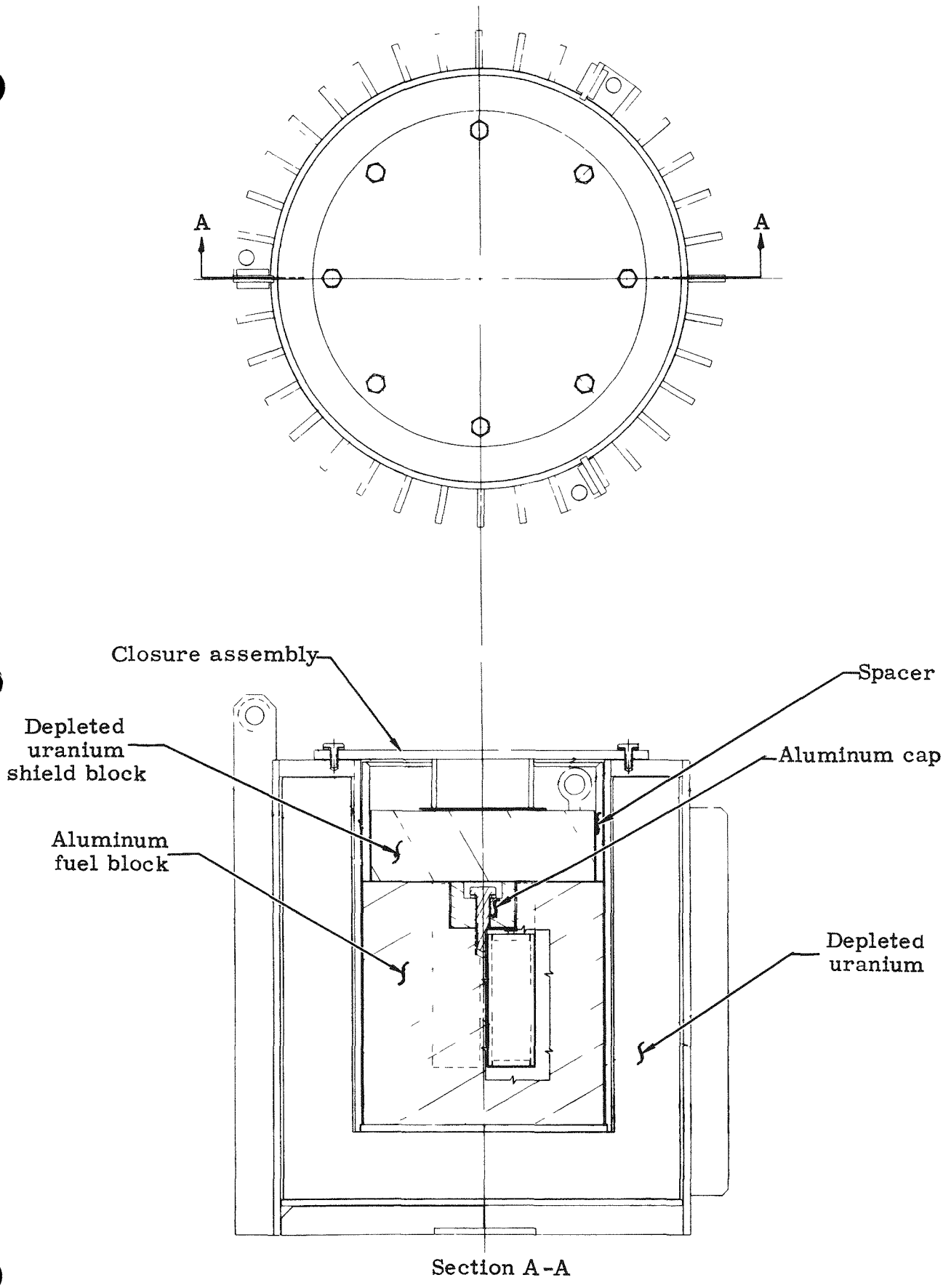

Fig. 5. SNAP 7 Fuel Capsule Shipping Cask

The comparison of calculated dose rates with measured values showed the calculated values were almost uniformly higher than the measured values by a factor of 4 for thicknesses of lead greater than one inch. Below this thickness, the calculated dose rates were lower than experimental values. In terms of shielding thickness, the factor of 4 means that the shields were too large by two half-value thicknesses. Therefore, lead shields could have been reduced by 0.88 inch and the uranium shield by 0.526 inch.

It should be noted that, for two reasons, the shield design was not revised to reflect these data:

(1) These dose rates are the first ever measured from a kilocurie source of Strontium-90; future testing and analysis may introduce additional factors, tending to modify conclusions based on the early measurements.

(2) The test data became available after the shield design had been established.

It is important to note that, in view of the dose rate measurements, the established shield design embodies a considerable safety factor.

A more detailed examination of dose rates in the vicinity of the Hastelloy C container shells of the 10-watt generator was undertaken to determine whether "streaming" will occur in this area. Dose rates at various locations in the vicinity of the double can of the 10-watt generator were calculated by the use of a modified IBM 709 code. Results are given in Table C-1, Appendix C. Figure 6 shows the location of the dose points in relation to the assembled generator. Examination of the dose rates listed failed to show any great discontinuity, indicating that the shield design for the area investigated is quite adequate and no difficulty is to be anticipated with the 10-watt generator.

5. Thermoelectric Tests*

a. Couple life tests

Couples of the 10-watt generator size were placed in the General Engineering Laboratory test fixtures and are presently under test at a hot junction temperature of 980° F. These tests are being conducted to assure continued output of the couples after extended periods of operation at high temperature.

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* H. Miller

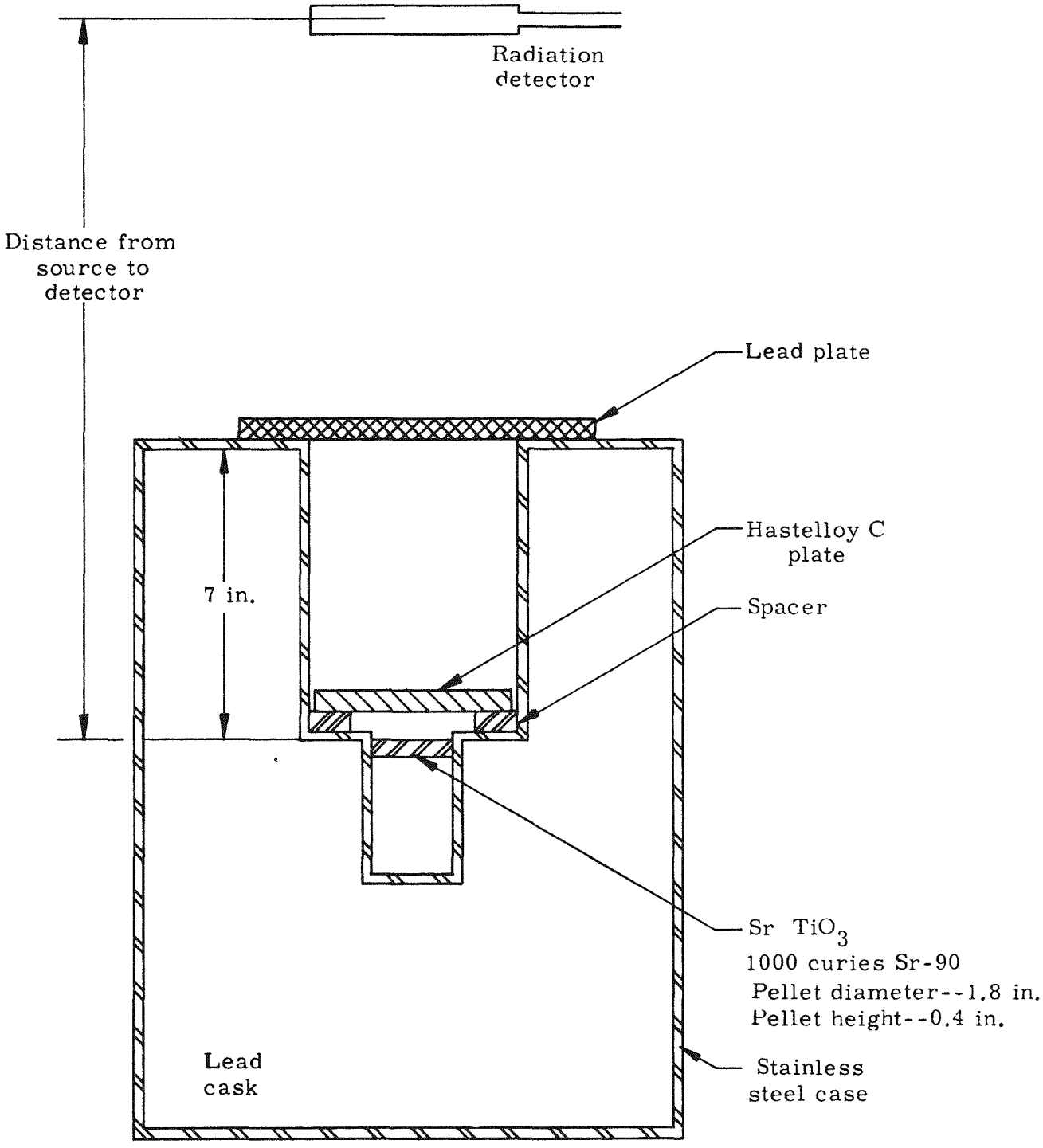

Fig. 6. Physical Arrangement used in Measuring Dose Rates

Three couples of the 10-watt size were installed in the laboratory fixtures. Two of the couples were coated around the hot junctions with No. 1 Sauereisen cement and the third couple was left bare. These couples have been operated at a hot junction temperature of 980° F, in 85% nitrogen-15% hydrogen at one atmosphere, for a period of 52 days.

Test results for the 10-watt-size couples are illustrated in Figs. 7 through 9. The graphs show 8 and 14% power drops for the coated couples and a 19% power drop for the bare couple. The test power drops for the coated couples are considered to be higher than actual since holes were cut into the Sauereisen seals to attach instrumentation.

The No. 1 Sauereisen cement seems to provide a barrier to degradation of contacts on the 10-watt generator couples. The present 10-watt generator couple materials and fabrication techniques, which include cementing around the hot junctions, appear to be satisfactory for use in the SNAP 7A and 7C systems, based upon all tests performed to date.

b. Reliability program

The reliability program was established to demonstrate the feasibility of module assembly techniques, to determine thermal and electrical characteristics of module designs and to obtain an indication of generator reliability through long term operational tests.

A 15-couple reliability test model has been fabricated to simulate one fourth of the 10-watt generator configuration. Instrumentation has been provided to evaluate each couple in the model individually and to determine the temperature of each component in two of the couples. A thermal calibration test was performed on the test container to determine the added heat loss imposed by the couples. A photograph of the complete reliability model and test container is shown in Fig. 10. Figure 11 shows the parts of one 5-couple module in detail. The instrumentation and apparatus are complete and ready for the reliability test.

The model has not yet been assembled because of the lack of enough completed couples; however, the model will be assembled as soon as the necessary couples are received. The test will begin with a parametric study to provide a basis for lifetime operational tests and to provide an estimate of the generator's operating characteristics. Tests will continue for 10-1/2 months at a hot junction temperature of 900° F.

c. Laboratory facilities

The General Engineering Laboratory is assembling a checkout facility for both the 10- and 60-watt generators. This facility includes a fixture to check the performance of each couple installed in the generator at a hot junction temperature of 500° F in an inert atmosphere. Unsatisfactory couples can be replaced easily without removing the generator from the fixture.

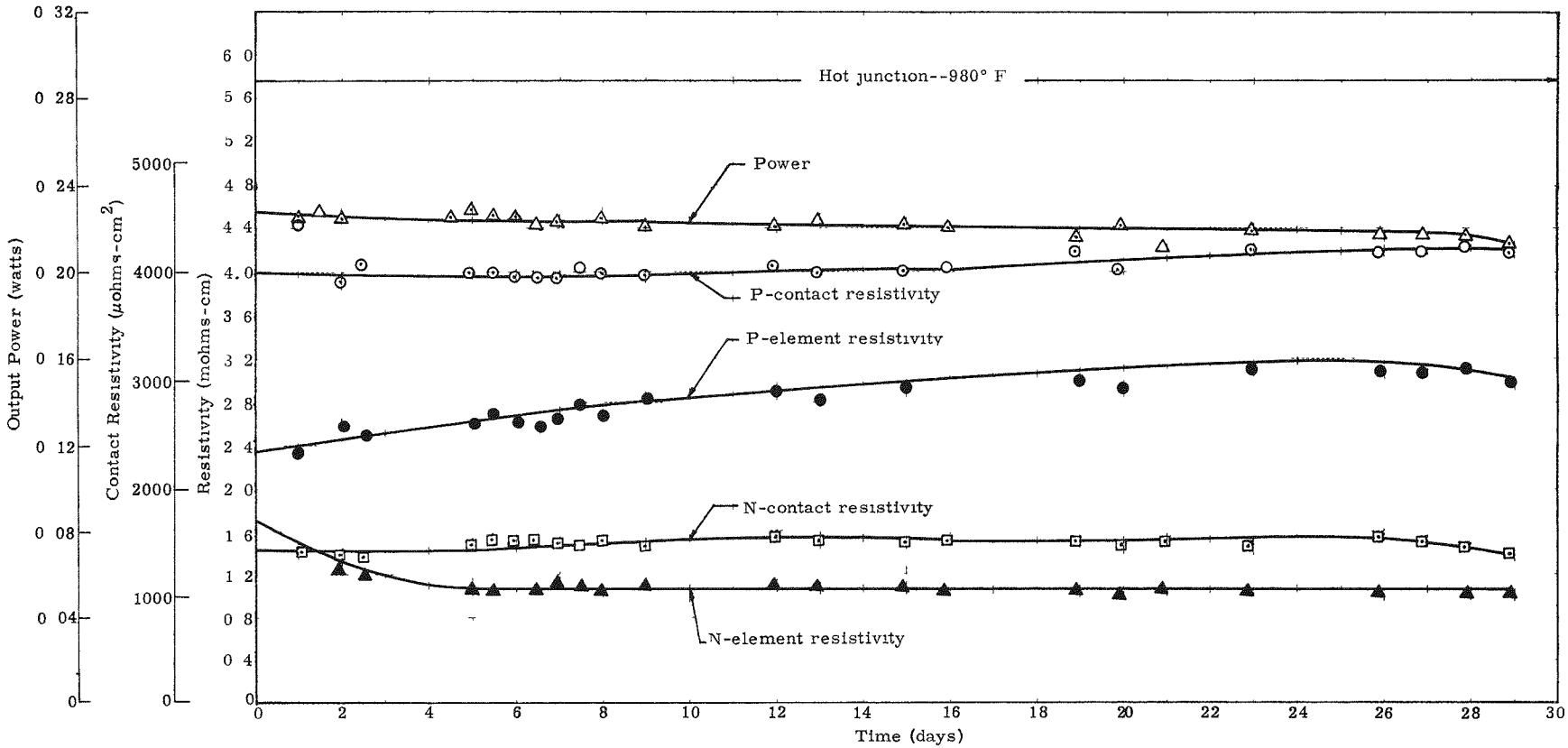

Fig. 7a. SNAP 7 Sublimation Test--Couple No. MAR 6, P diameter = 0.277, N diameter = 0.233--Coated with No. 1 Sauereisen

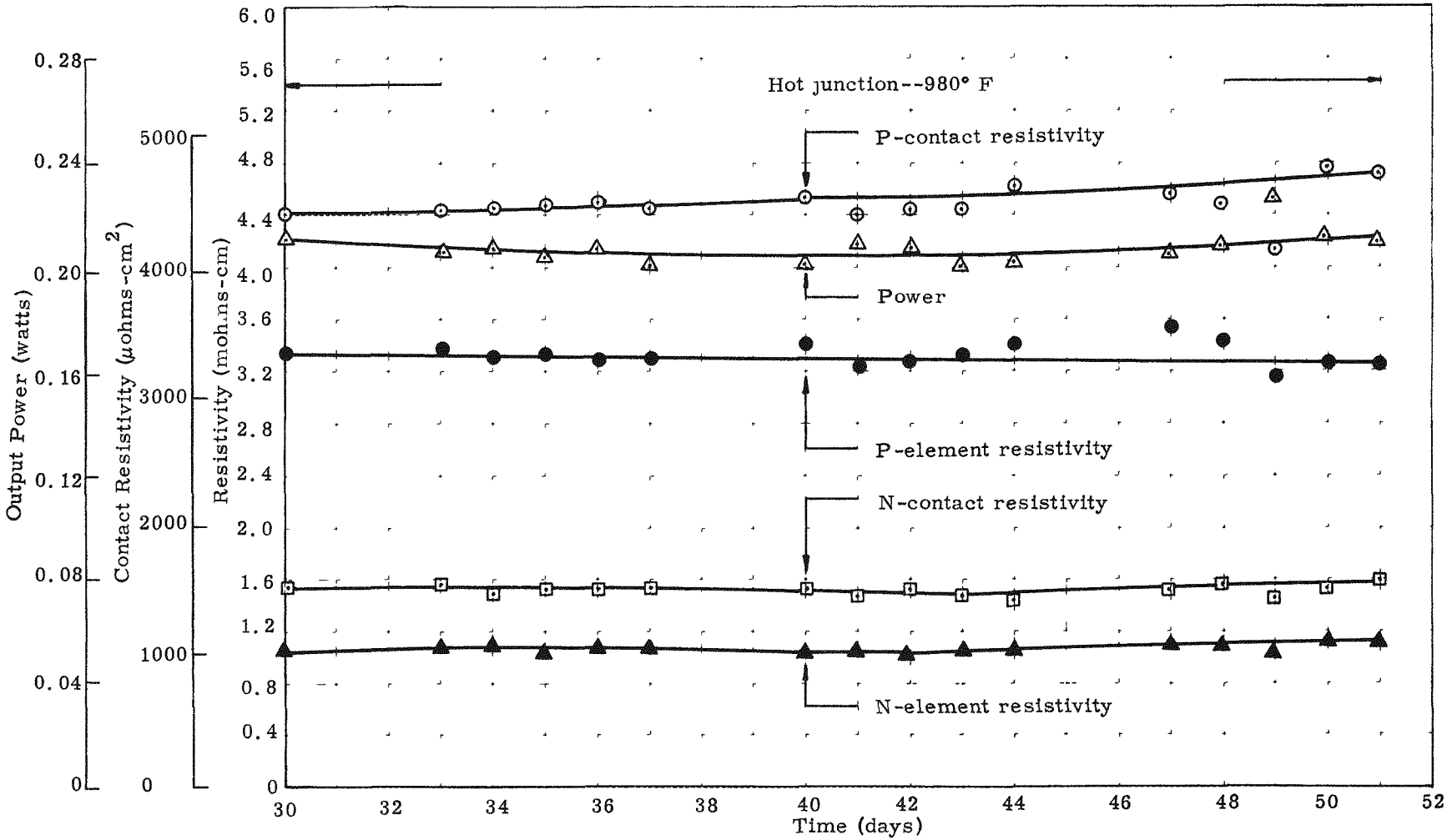

Fig. 7b. SNAP 7 Sublimation Test--Couple No. MAR 10-6, P diameter = 0.277, N diameter = 0.233--Coated with No. 1 Sauereisen

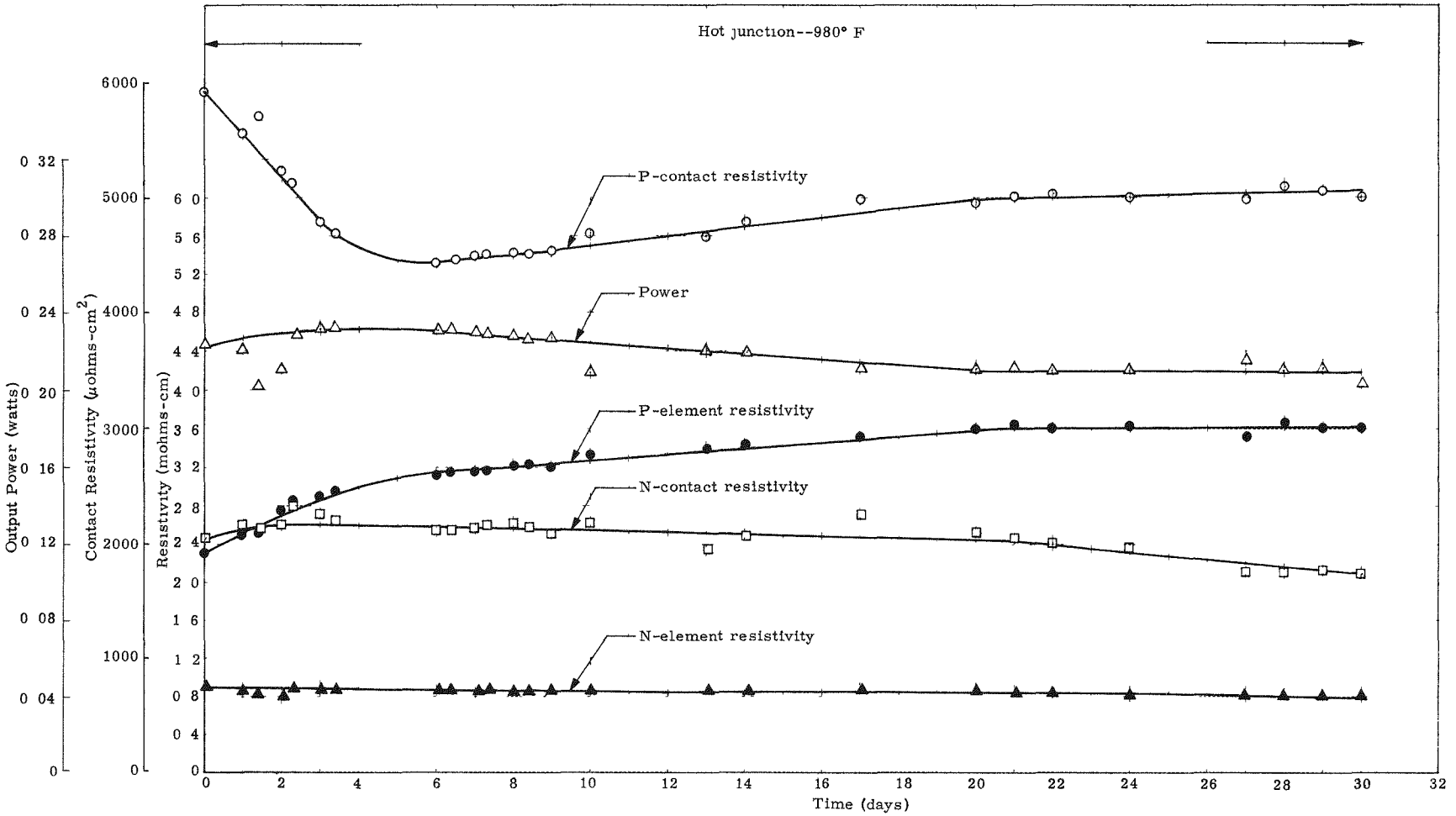

Fig. 8a. SNAP 7 Sublimation Test--Couple No. MAR 10-10, P diameter = 0.277, N diameter = 0.233--Coated with No. 1 Sauereisen

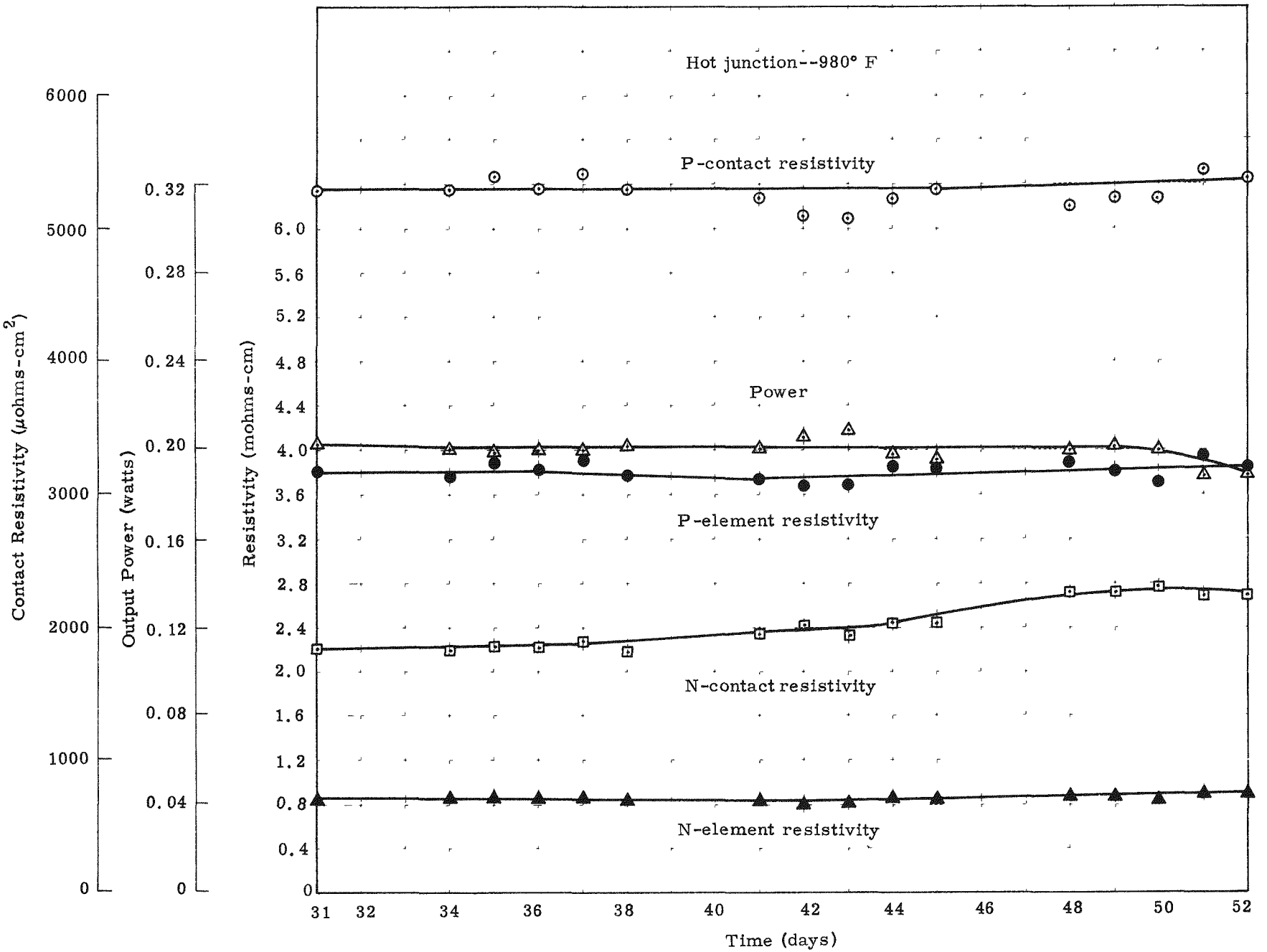

Fig. 8b. SNAP 7 Sublimation Test--Couple No. MAR 10-10, P diameter = 0.277, N diameter = 0.233--Coated with No. 1 Sauereisen

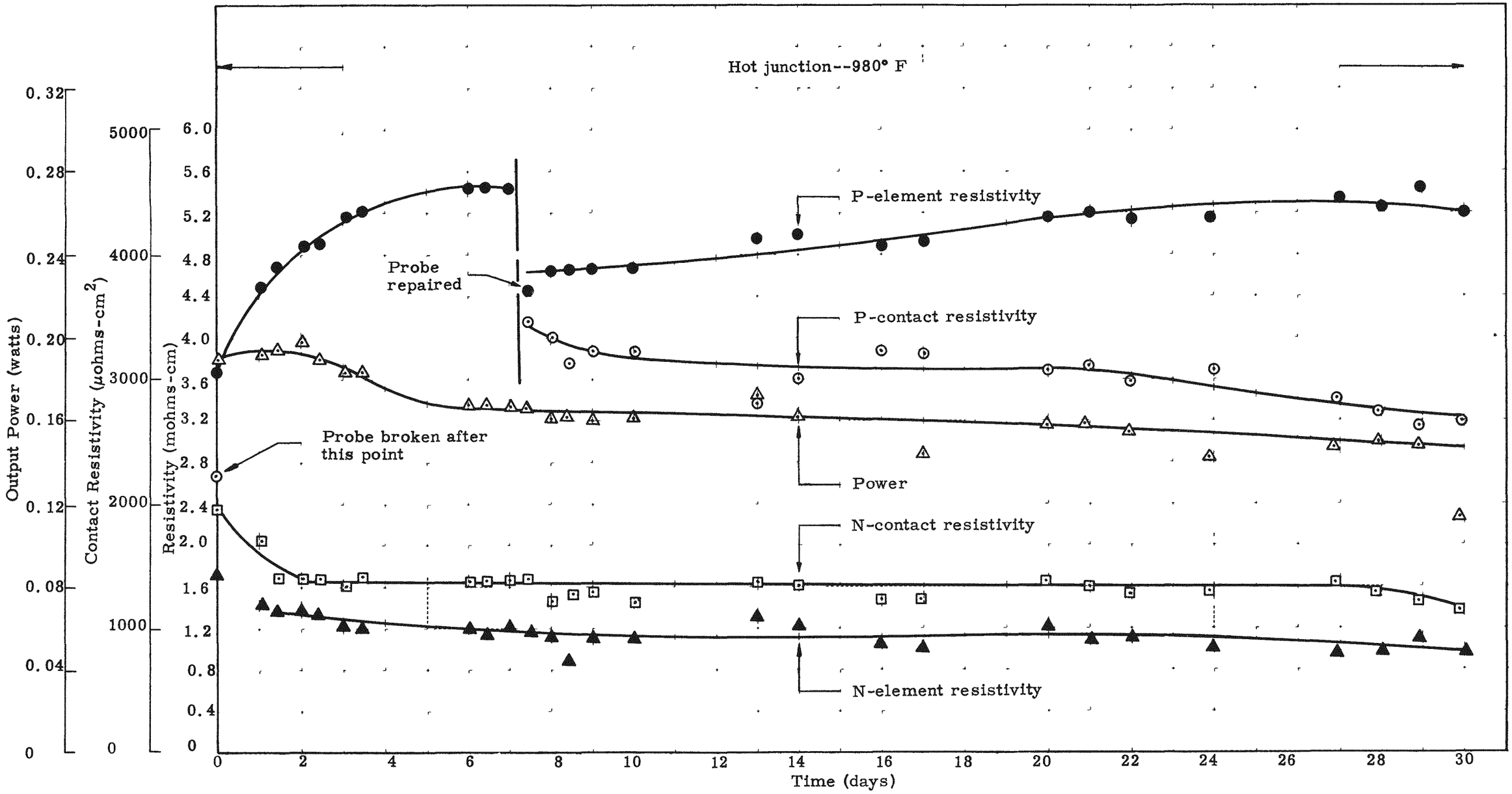

Fig. 9a. SNAP 7 Sublimation Test--Couple No. MAR 10-12, P diameter = 0.277, N diameter = 0.233--Uncoated

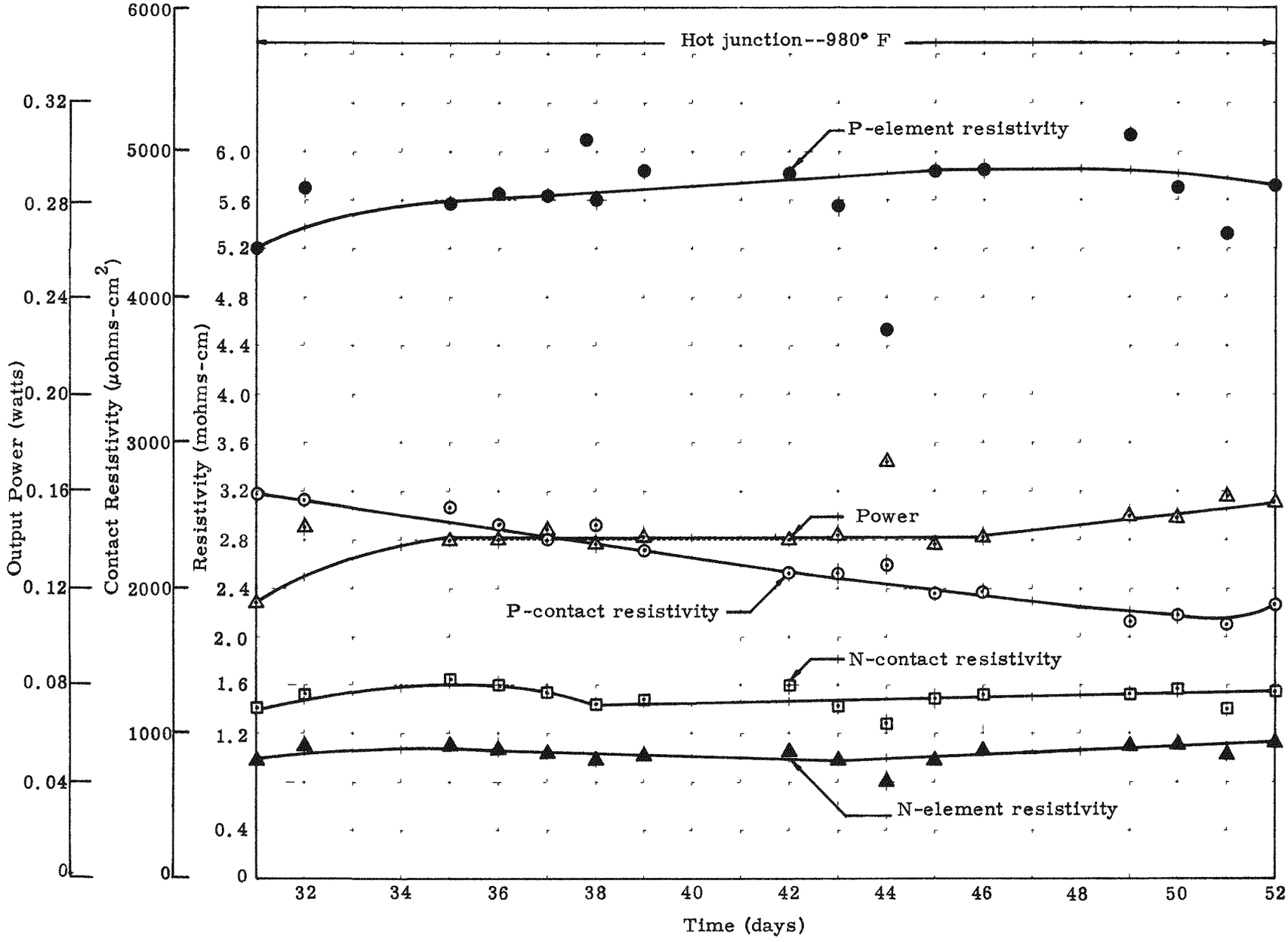

Fig. 9b. SNAP 7 Sublimation Test--Couple No. MAR 10-12, P diameter = 0.277
N diameter = 0.233--Coated

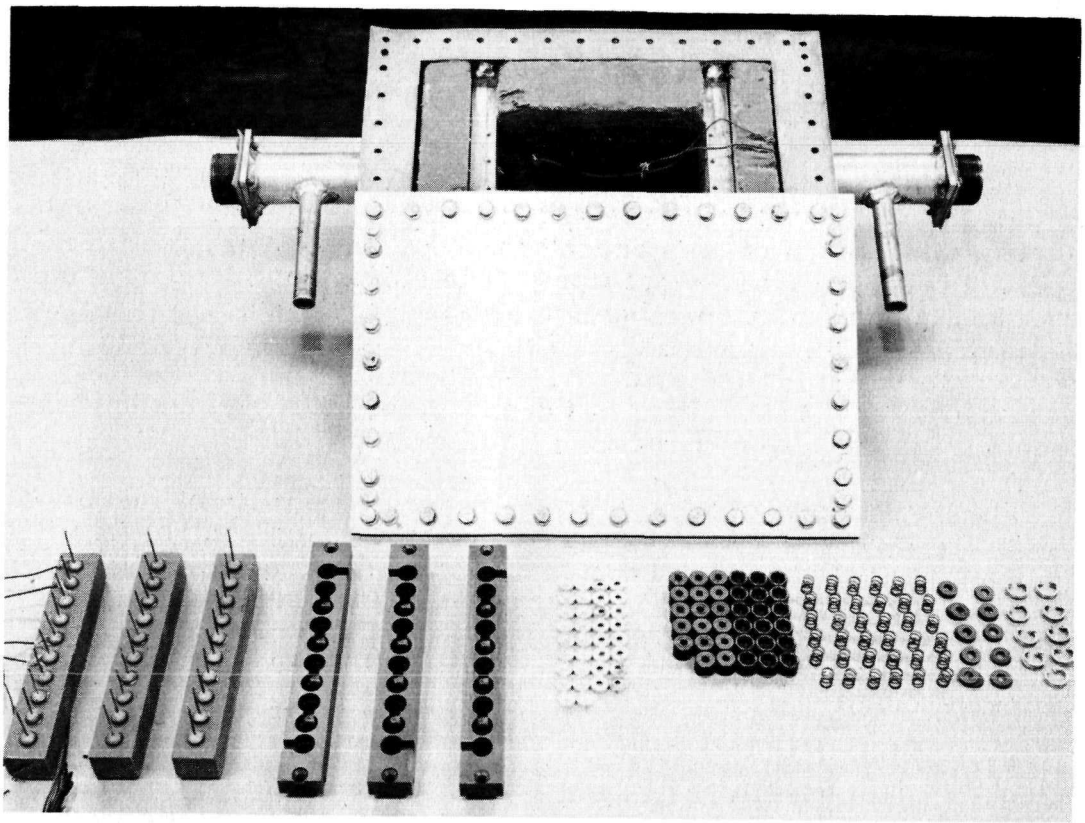

Fig. 10. Reliability Model, Components and Test Container

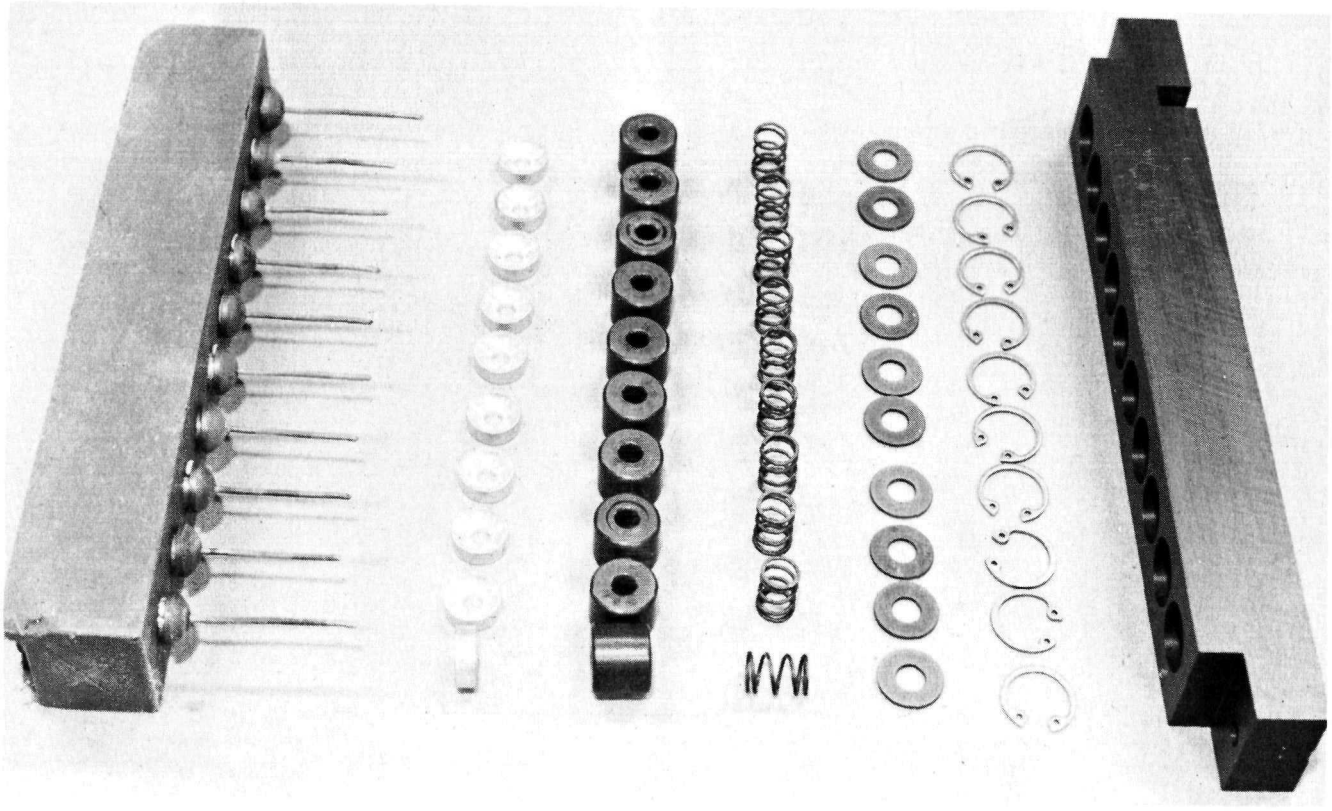

Fig. 11. Parts of Five-Couple Module

The generator-checkout fixture consists of a specially fabricated dry box to accept both the 10- and 60-watt generators. It provides free access to all parts of the generators and all necessary facilities to operate and repair the generators. The box is completely sealed and provides an entrance chamber for passing parts in and out without contaminating the atmosphere. The box and associated equipment will be completed in the next quarter. A parametric test fixture is being provided to check the output of the generator with various anticipated thermal inputs, sink temperatures and external load conditions.

The parametric test fixture is now ready for tests of the 10-watt operating model. This fixture provides power and control facilities for 24-hour operation. An atmosphere conditioning and control system for the generator has also been completed and checked out. Electrical test instrumentation is available for the initiation of tests. High temperature cutout is provided to prevent any over-temperature condition in the generator.

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 60-watt thermoelectric generators were designed, and the associated engineering drawings were released for fabrication and procurement during the period of this report.

In general, the SNAP 7B and 7D generators are patterned after the SNAP 7A and 7C units. However, because of different design requirements, such as maximum overall diameter limitation, the 60-watt generators cannot be said to be an exact scaleup of their 10-watt counterparts. The SNAP 7B and 7D generators, as designed, feature many interesting aspects, such as:

(1) Employment of integral thermoelectric modules, each of which consists of several pretested thermoelectric couples.

(2) Use of a multicapsule fuel block which accommodates 14 fuel capsules.

(3) Use of depleted uranium as shielding material in the biological shield.

(4) Control of thermal-to-electrical conversion efficiency by adjustment of the thermal conductivity of the insulation.

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

The objectives of Subtasks 8.2 and 8.4 are to analyze, design and fabricate SNAP 7B and 7D thermoelectric systems.

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

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*J. Keenan

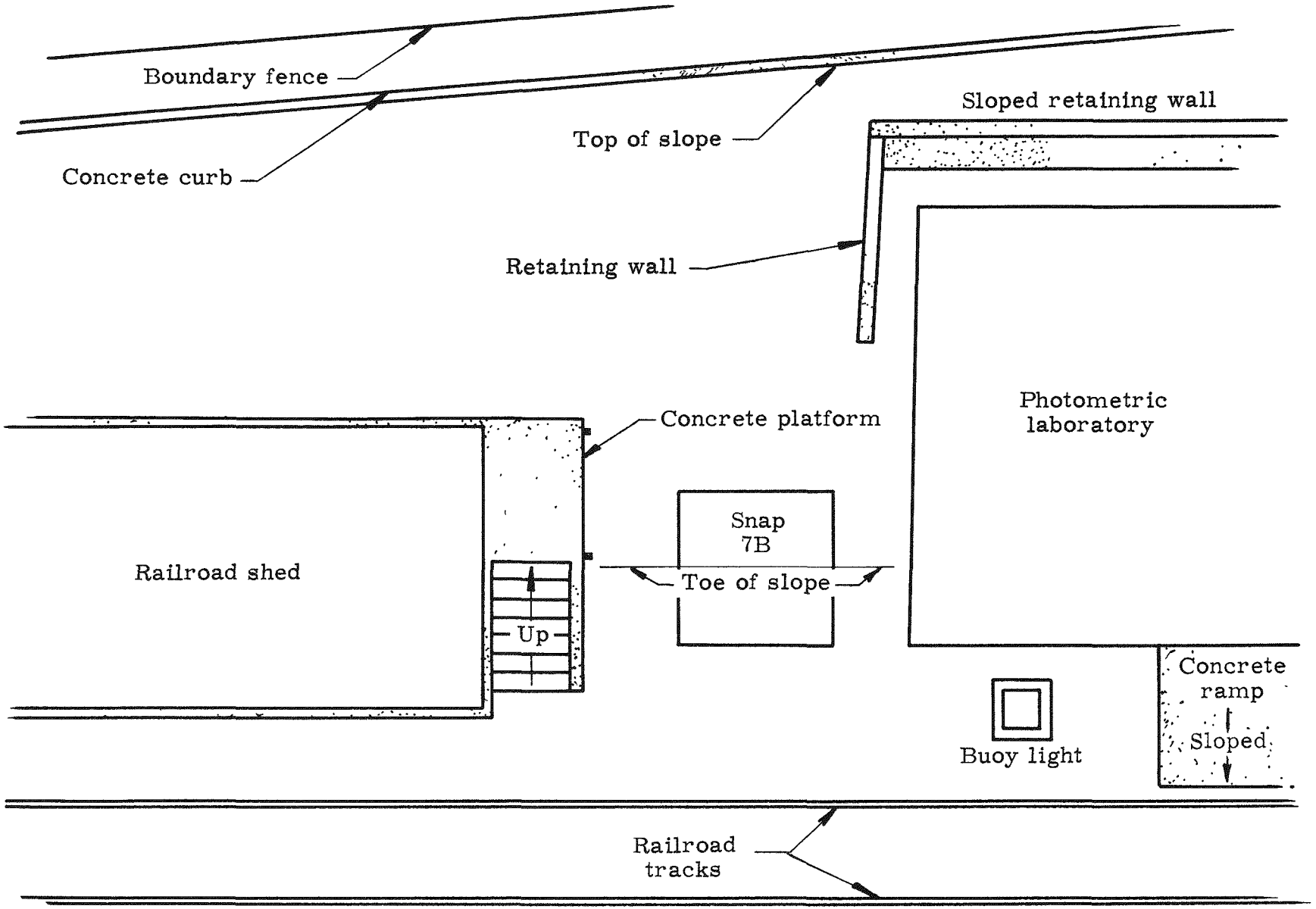


Fig. 12. Site of Snap 7B Installation

The SNAP 7D system will be used with a U. S. Navy boat-type floating weather station. The system will consist of a 60-watt thermoelectric generator, a dc-to-dc converter and nickel-cadmium batteries to produce 30 watts 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.

During this period, the 60-watt thermoelectric generator was designed and detailed for manufacturing. Considerable analysis was required in the heat transfer, shielding and thermoelectric areas. More specific details of these studies, as well as the actual design, will be discussed in the succeeding pages of text.

1. Generator Design

The major effort during this period was the design of the 60-watt thermoelectric generators. The Coast Guard system (SNAP 7B) and the Navy system (SNAP 7D) are designed to use identical generators except for possible differences in deliverable electrical power. Figure 14 shows the design of the 60-watt generator and identifies the major components in the final assembly. Basically, the generator consists of 14 isotope capsules, a thermoelectric direct conversion circuit, a heat rejection system and a biological shield. Each phase serves an essential function for the successful operation of the generator and the safety of personnel in the immediate vicinity.

The preliminary design of the 60-watt generator was essentially similar to that of the 10-watt generator. The thermal input from the Strontium-90 fuel was calculated to be 1546 thermal watts for each generator. Seven fuel capsules were to contain the strontium titanate and were to be arranged as follows: one in the center and six equally spaced around the perimeter. To reduce cost and delivery time, cast 2-inch pipe of Hastelloy C was selected as the cladding for the fuel capsule. The module design was considered in relation to this generator design; and from among various arrangements which used thermoelectric couples in line and in groups, a module of four pairs was selected. The use of an inner shell, as in the 10-watt generator, was eliminated as a result of a thermal analysis which indicated a potential problem in heat dissipation. In addition, there was no requirement for fuel capsule shipping casks. Upon completion of this preliminary design, the diameter over the fins was 23-7/8 inches and the height at the cover was 25-3/4 inches. Since the container tube for the boat-type weather station is 24 inches in diameter, this design, although conforming to the Statement of Work, was discarded in favor of another design of smaller diameter. The design dose rate was also revised from the original 100 mr/hr at 4 feet, as given in the Statement of Work, to 10 mr/hr at 1 meter from the center of the source, as required by ICC regulations. AEC personnel agreed to accept the weight increase in order to meet the dose rate specifications of 10 mr/hr at 1 meter or 200 mr/hr at the surface.

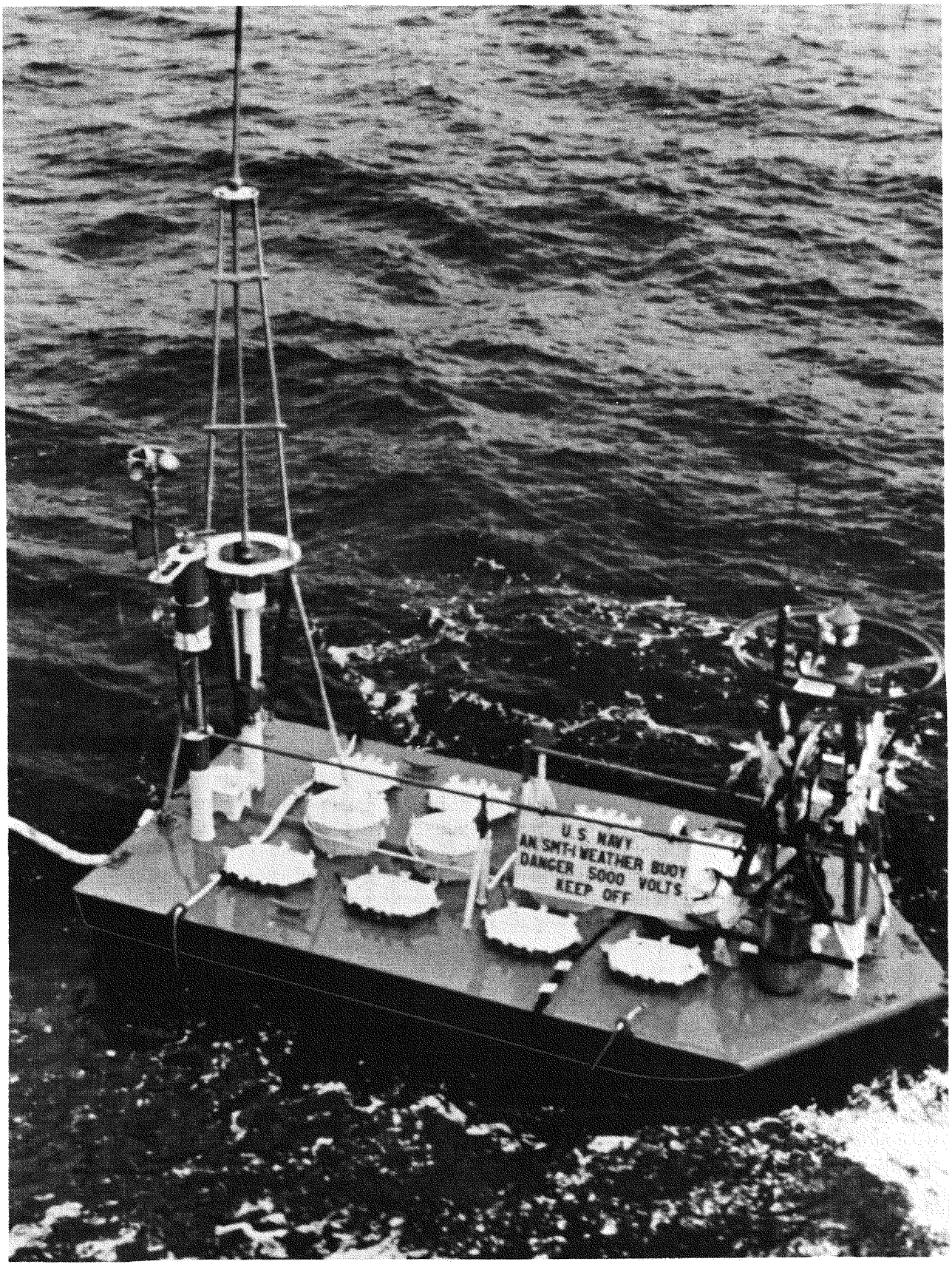


Fig. 13. Boat-Type Weather Station

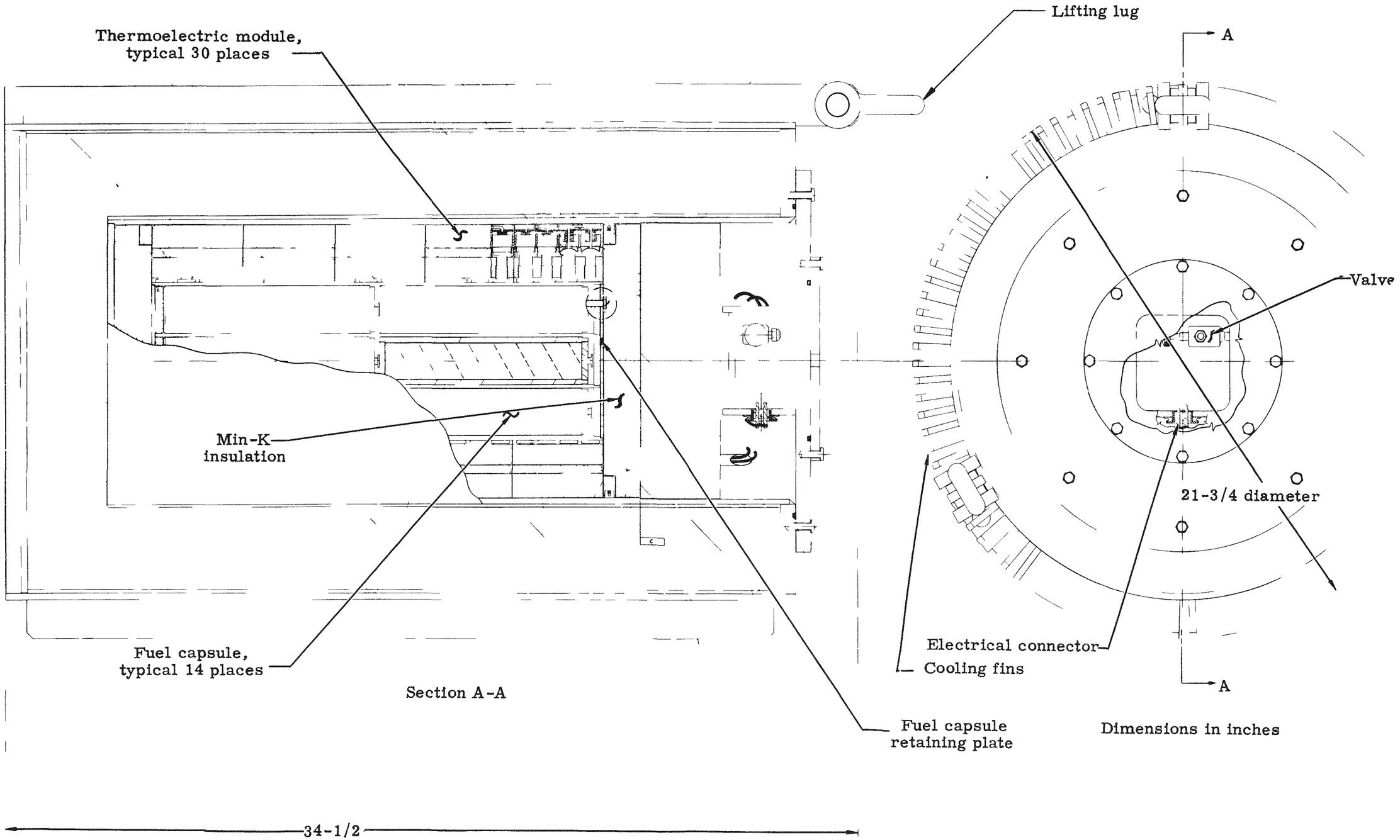


Fig. 14. SNAP 7B and 7D 60-watt Generator

The final design for the 60-watt generator was also based on a 1546-thermal watt input for each generator. The same diameter of fuel pellet and capsule as used in the 10-watt generators was selected for use in the 60-watt generators. Because of the greater quantity of fuel, the fuel block was designed with one hole in the center and six equally spaced holes around the perimeter. The height of the fuel capsules with the smaller diameter was excessive, necessitating a major revision of the welder and making handling more difficult. The fuel capsule height was therefore reduced by a factor of 2; two fuel capsules are to be loaded into each hole. The lateral surface of the fuel block is composed of 12 machined flats arranged as sketched in Fig. 15. The six flats for the modules will be precoated with aluminum oxide and ground. The strips of insulation compressed between the fuel block and the heat sink will position the fuel block.

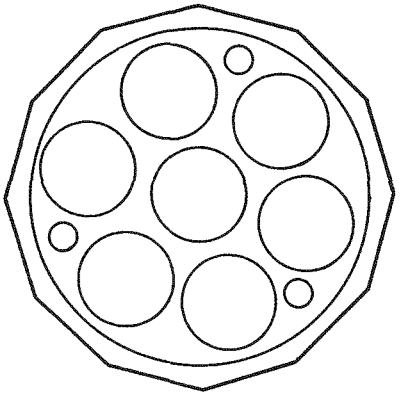
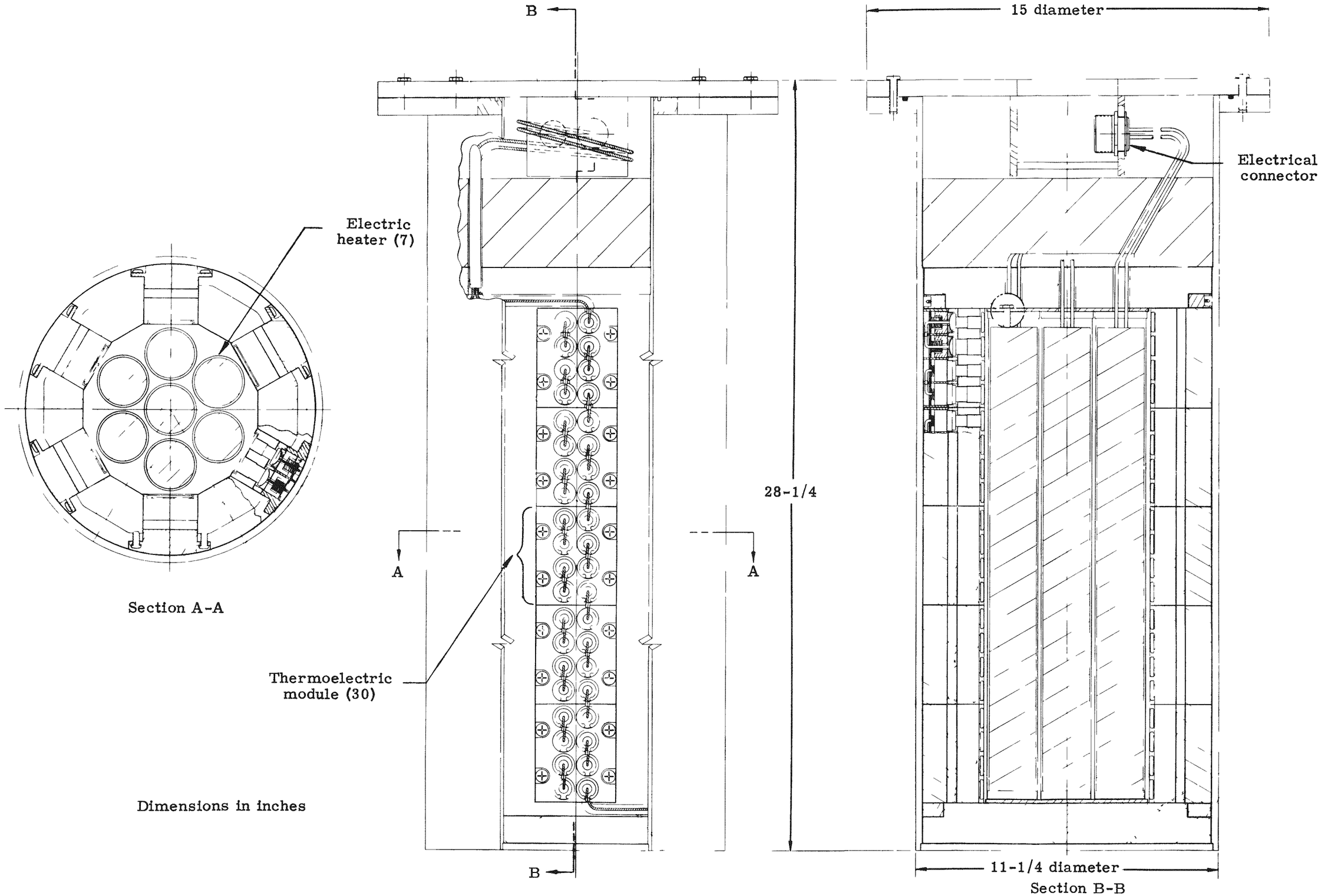


Fig. 15. Fuel Block Arrangement

The heat sink is aluminum and consists of a frame upon which the module bars are mounted and held in place. The connecting insulated wires are in recesses in the heat sink so that the heat sink can be slid into the biological shield after the unit has been assembled. There are four pairs of thermoelectric elements per module, with five modules per group and six groups per generator, or a total of 120 pairs of thermoelectric elements. Figure 16 shows the assembly of a thermoelectric element pair, indicating hot shoe, cold shoe, cold shoe cap, piston and spring. The electrical circuit is a series arrangement, as shown; the thermal path is from the fuel block, through the hot shoe to the thermoelectric elements and through the cold shoe, the cold shoe cap, the piston, the module bar and the heat sink to the biological shield.

The biological shield is similar to the 10-watt generator in basic design. The shield is depleted uranium with Hastelloy C cladding both inside and outside. The use of Hastelloy C gives long-period protection in sea water--not only protecting the shield, but also providing a second corrosion barrier for the fuel. The walls of the cladding, inside and



Dimensions in inches

Fig. 16. SNAP 7 60-watt Operating Module

outside, are bonded to the uranium to give a good thermal path to keep the temperature drop as low as possible. The outer shell is finned, so as to dissipate rejected heat to the atmosphere. The electrical connection is made through a junction box in the cover. The details of this will be covered in the individual installations of SNAP 7B and 7D systems.

2. Heat Transfer Analysis

Thermal analyses were performed to establish the basic configuration of the SNAP 7B and 7D systems. Some of the considerations are reviewed in the following.

a. Hot junction temperature

Due to input power decay, it is impossible to maintain a constant hot junction temperature over a 10-year interval. It would be desirable to operate the hot junction at some temperature in the neighborhood of 900° F at the end of life in order to achieve high conversion efficiency. However, the hot junction temperature at the beginning of life would then be in excess of the maximum temperature tolerated by the thermoelements. Experience with available materials has shown that the hot junction temperature should not exceed 980° F. The thermoelectric elements have been sized, therefore, to produce 800° F hot junction temperature at the end of life. This size gives a maximum hot junction temperature of approximately 980° F at the beginning of life.

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 between the generator shell and cold cap assemblies. Water at 80° F was selected as the design environment. The temperature rise in the film has been calculated to be approximately 8.6° F, and the total temperature rise between the generator surface and the cold end of the elements has been estimated to be 71.4° F. On the basis of these estimates, the cold junction temperature should be 160° F at the beginning of life. At the end of life, the heat output will have decreased to 78% of the original value and the cold junction temperature, correspondingly, will have decreased to 143° 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, and it was concluded that, for the 60-watt generator, the shielding should be located entirely on the outside. This conclusion was due primarily to the significant size and weight of the fuel block.

The outer shell, therefore, must be designed to contain the shield material (depleted uranium) and must 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 (the generator terminals may be short-circuited during shipment, resulting in a significant reduction in temperature drop across the elements).

(2) During operation, submerged in water not exceeding 80° F.

Condition (2) poses no problem relative to cooling the surface; the generator will be designed for operation in temperatures resulting from these conditions. Condition (1), however, could result in elevated generator temperatures. The surface must be designed to operate at temperatures no higher than 200° F in order to maintain internal temperatures at a reasonably low level during shipment. The maximum design heat input is 1546 watts. The heat lost from the surface is given by

$$q_{total} = q_{conv} + q_{rad} = 1546 \text{ watts}$$

where

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

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

h_c = convection film coefficient

h_r = radiation heat transfer coefficient

T_s = surface temperature of container

T_r = environmental temperature

A_c = convective heat transfer area

A_r = radiant heat transfer area

ΔT = = temperature difference between generator shell surface and environment = $T_s - T_r$.

The generator surface is considered to consist of a cylinder 19.00 inches in diameter and 32.75 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 200° F.

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

$$A_r = 17.80 \text{ ft}^2$$

The approximate convective heat transfer area is 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. 2):

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

where

e_s = fin efficiency

ϵ = $W_c^{3/2} \sqrt{h_c/kA}$

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

W_c = $W + \delta$

δ = one-half fin thickness = 0.125 inch

W = length of fin = 1.5 inches

k = thermal conductivity of fin material
= 6.5 Btu/hr-ft^2-°F

h_c = convective heat transfer coefficient

$\simeq 0.27\,(\Delta T)^{0.25}$ (Ref. 3)

$= 0.27\,(75)^{0.25} = 0.790$ Btu/hr-ft^2-°F.

Then

$$\epsilon = \left(\frac{1.5 + 0.125}{12}\right)^{3/2} \sqrt{\frac{0.790}{(6.5)\,(0.00283)}} = 0.319$$

and

$$e_s = \frac{1}{\sqrt{0.638}} \tanh \sqrt{0.638} = 1.260\,(0.659) = 0.830 = 83\%.$$

Therefore,

the total convective heat transfer area A_c is

$A_c = 15.50 + 0.519\,N$

where N is the number of fins.

The radiant heat transfer coefficient h_r may be determined from Eq 13.29, Ref. 4:

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

$= 1.34$

where

ϵ_s = surface emissivity = 0.80.

The total heat transferred is then given by

$q_{total} = (0.790)\,(15.50 + 0.519\,N)\,(75) + (1.34)\,(17.80)\,(75)$

which must equal the total design input of 5270 Btu/hr.

$q_{total} = 915 + 30.6\,N + 1780 + 5270$

gives

$N = 84$ fins.

So a minimum of 84 fins will be required on the shell surface.

In order to provide some degree of conservatism, the number of fins actually incorporated in the 60-watt generator design has been set at 99.

d. 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. Because of its transient nature, the Strontium-89 thermal power cannot be considered part of the total thermal power requirement.

Figure 17 illustrates the combined thermal output of Strontium-89 and -90 for 245,400 curies of the heavier isotope as a function of time 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 60-watt generators may have some significant thermal contribution from the lighter isotope which would require [illegible]tion 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 7B and 7D fuel requirement which permits an overload of 90 watts due to Strontium-89 content.

e. 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 forms a snug fit between the fuel block and the cold sink, providing support for the fuel block. It has a mean depth of 1.5 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 of 0.0190 Btu/ft-hr-°F was selected as a nominal value for the generator design. It is valid for Min-K 1301 with a gas fill mixture of approximately 90% argon and 10% hydrogen.

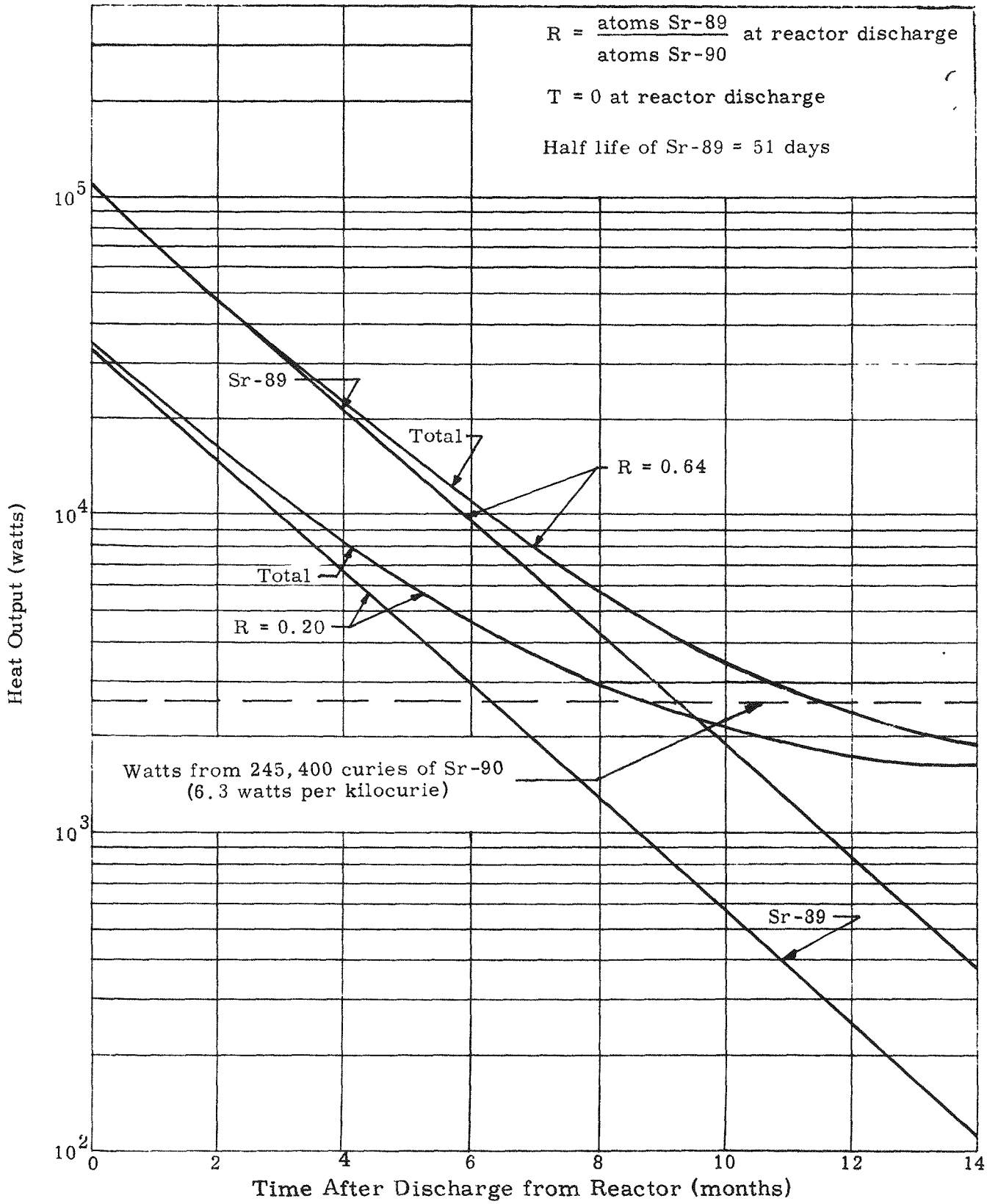


Fig. 17. Effect of Strontium-89 on Heat Output of SNAP 7B 60-Watt Generator

The calculated total heat loss at the end of life is 154 watts, including an estimated 18 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.

3. 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

ρ = resistivity.

The figures of merit for various available materials were compared on the basis of published information. From this comparison, it was apparent that the Martin N-type thermoelectric material and Minnesota Mining and Manufacturing 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 3M material. Samples of each of the materials considered were procured, bonded to suitably simulate the hot and cold shoe installation in the generator (Figs. 18 and 19) and operated at temperatures betwen the limits expected in the generator (143° 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 3M P-type. Long range testing has shown these two materials to be durable, displaying little degradation in performance with time.

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

The gross thermal output at the end of life is 1200 watts; the net available heat input to the thermoelements is 1046 watts (154 watts comprises insulation and other losses).

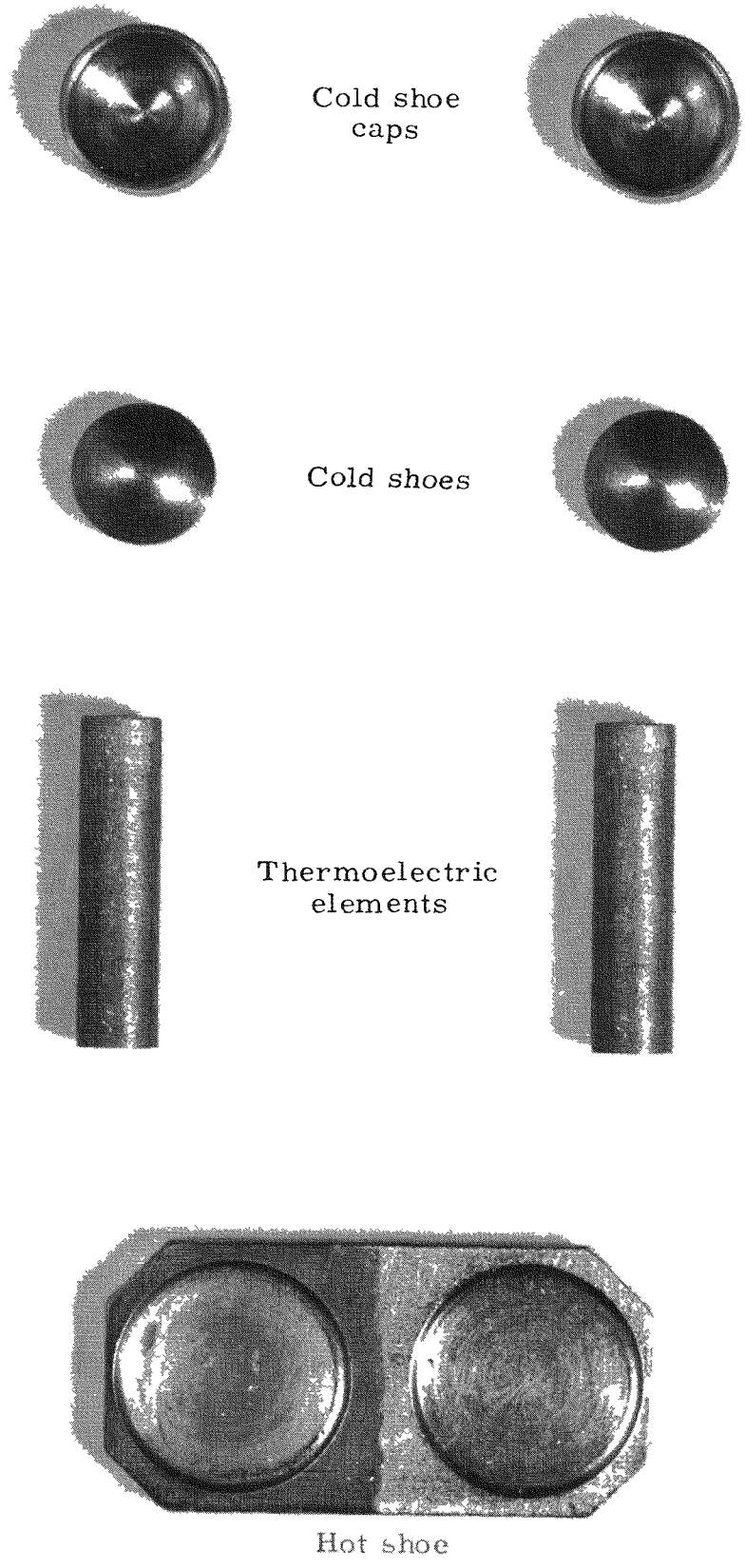


Fig. 18. Components of Thermoelectric Couple

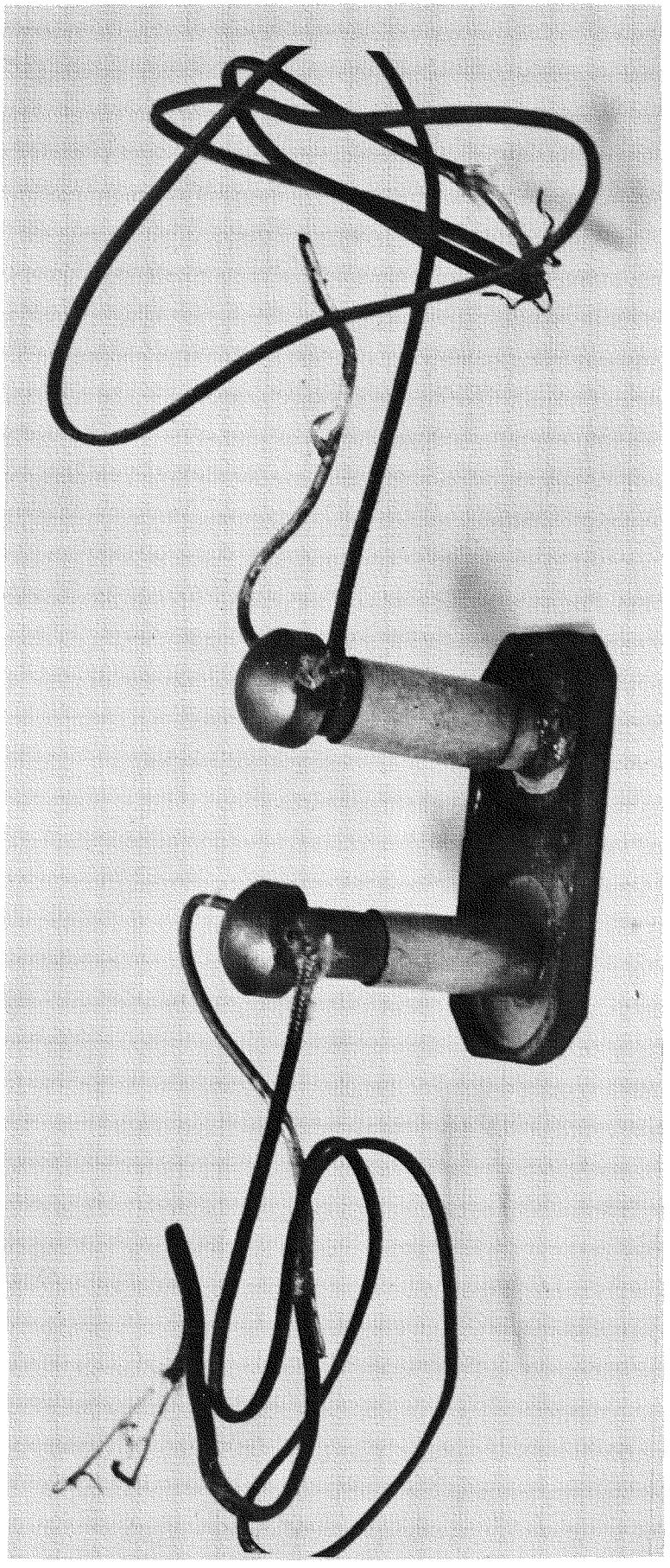


Fig. 19. Bonded Thermoelectric Pair

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 143° F. Figure 20 shows the test fixture for thermoelectric couple tests.

TABLE 1

Summary of Module Test Thermoelectric Properties

Average Properties	Martin N	3M N	Martin P	3M P
Seebeck voltage (μ v/°C)	196	249	217	171
Resistivity (μohm-cm)	1131	2227	5962	3532
Contact resistivity (μohm-cm^2)	464	752	1037	1299
Thermal conductivity (watts/cm-°C)	0.0194	0.0191	0.0208	0.0200
Figure of merit ($s^2/k\rho \times 10^6$)	1533	1463	394	416

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

	N-type	P-type
Seebeck coefficient (μv/°C)	$s_N = 196.2$	$s_P = 171.4$
Resistivity (μohm-cm)	$\rho_N = 1131$	$\rho_P = 3532$
Thermal conductivity (watts/cm-°C)	$k_N = 0.0194$	$k_P = 0.0200$
Contact resistivity (μohm-cm^2)	$c_N = 464$	$c_P = 1299$

The following system parameters used in the calculation are defined:

ℓ = element length = 2.54 cm

T_I = hot junction temperature = 700° K

ΔT_t = thermoelement temperature difference = 365° K

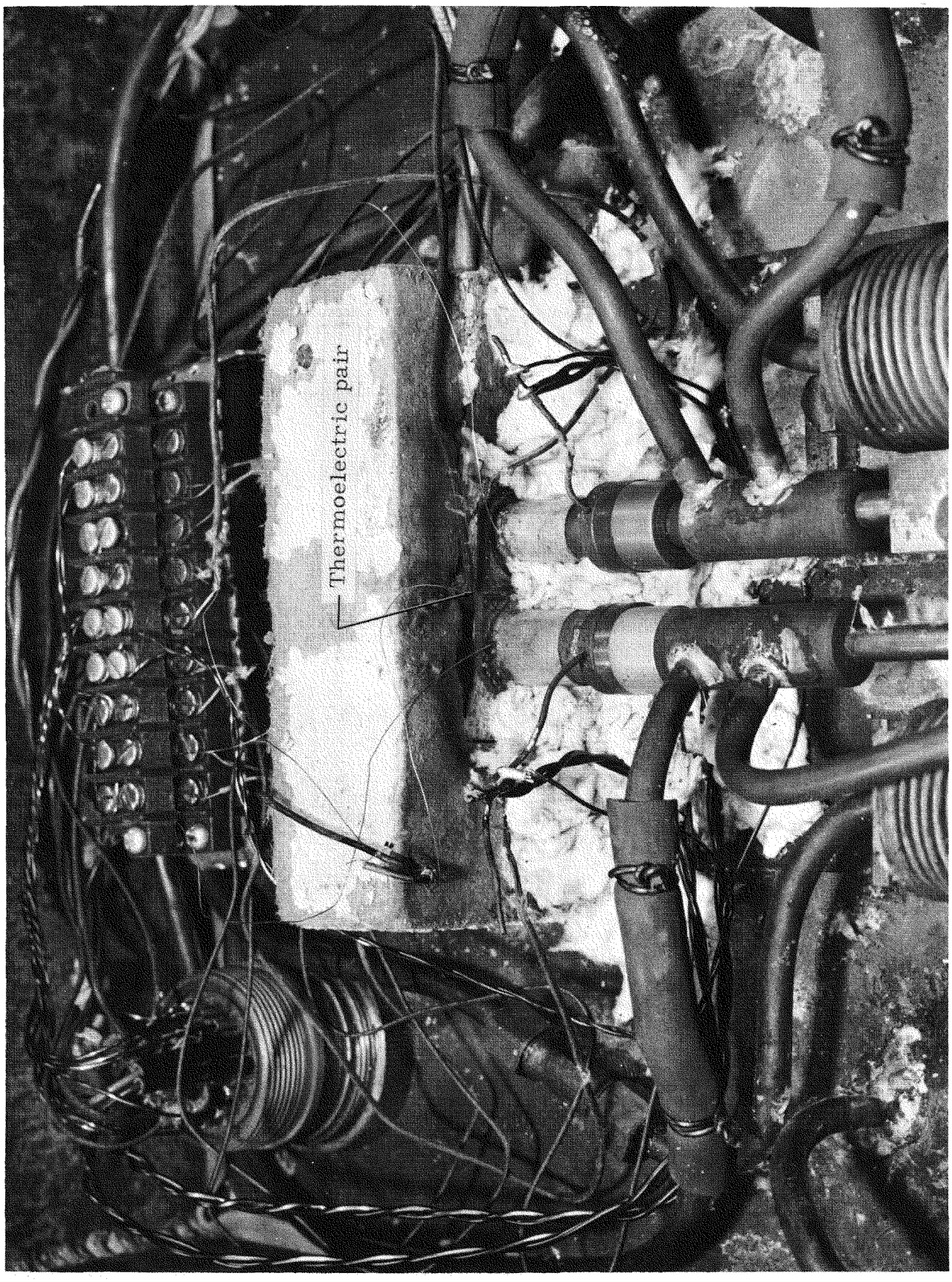


Fig. 20. Test Fixture for Individual Couple Tests

R_i = thermoelement internal resistance

R_c = thermoelement hot junction contact resistance

R_{load} = load resistance

x = ratio of load resistance to internal resistance for maximum thermoelectric efficiency

A_P = cross-sectional area of P element

A_N = cross-sectional area of N element

K = adjusted total thermal conductivity

α = total Seebeck coefficient for element pair

$$= s_N + s_P = 367.6 \times 10^{-6} \text{ v/°C}$$

$$B = \frac{S_N}{S_P} = \left[\frac{k_P}{k_N}\frac{\left(\rho_N + c_N/\ell\right)}{\left(\rho_P + c_P/\ell\right)}\right]^{1/2} = 0.5788$$

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

$$K = (k_P + k_N B)\ S_P = 0.03123\ S_P \text{ watts/°C}$$

$$R_i = \left(\rho_P + \frac{\rho_N}{B}\right)\frac{1}{S_P} = \frac{0.005486}{S_P} \text{ ohms}$$

$$R_c = \left(\frac{c_P}{\ell} + \frac{c_N}{\ell B}\right)\frac{1}{S_P} = \frac{0.00827}{S_P} \text{ ohms}$$

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

$$= 1.157.$$

The thermoelectric efficiency is given by

$$\eta = \frac{\Delta T}{T_I}\ \frac{x}{(x+1) + \dfrac{K(R_c + R_i)(x+1)^2}{\alpha^2 T_I} - \dfrac{\Delta T (R_i/2 + R_c)}{T_I R_i + R_C}} = 0.0522$$

The net power output is given by

$$P = (P_{in})(\eta) = 54.58$$

The open circuit voltage is

$$e = N\alpha\Delta T = 16.10 \text{ volts}$$

and the load voltage is

$$v = \frac{x}{x+1} e = 8.636 \text{ volts.}$$

The resistances may be determined as follows:

$$P = \frac{v^2}{R_{load}}$$

$$54.58 = \frac{74.58}{R_{load}}$$

$$R_{load} = 1.366 \text{ ohms}$$

$$x = \frac{R_{load}}{R_i + R_c}$$

$$= 1.157$$

$$R_i + R_c = 1.18 \text{ ohms}$$

$$(R_i + R_c)/\text{pair} = \frac{1.18}{120} = 0.00983 = \frac{0.006313}{S_P}.$$

The shape factor for the P element is then

$$S_P = \frac{A_P}{\ell_P} = 0.642 \text{ cm}$$

and the cross-sectional area is

$$A_P = 1.631 \text{ cm}^2.$$

The P- and N-element diameters are then

D_P = 1.441 cm = 0.567 in.

D_N = 1.096 cm = 0.431 in.

These diameters have been incorporated in the SNAP 7B and 7D generator design.

4. Safety and Shielding Analysis

Radiation dose rates from unshielded capsules of the 60-watt thermoelectric generator were computed. Based on a capsule containing 17,500 curies of Strontium-90, the results are as follows:

	Dose at Side (r/hr)	Dose at Top and Bottom (r/hr)
One meter from single fuel capsule	25.3	9.4
One meter from 14 fuel capsules	130	67.4

Radiation dose rates are reduced by the application of a 3.25-inch (thickness) uranium shield. The dose rates with the shield in place are as follows:

	Dose at Side (mr/hr)	Dose at Top and Bottom (mr/hr)
One meter from center of generator	2.5	2.5

All values stated above reflect the corrections in dose rate indicated by experiments performed at ORNL.

Preliminary safety reports for the SNAP 7B and 7D power supply systems are presently being completed (Refs. 5 and 6).

5. Manufacturing Drawings and Weights

The majority of the manufacturing drawings for the 60-watt generator were completed during this period. The complete list of component and assembly drawings is tabulated in Appendix E.

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.*

The wet chemical process has been established and is given in Appendix F. The variables in the pelletizing process have been investigated, with only one or two operational parameters still to be confirmed before completion of the operational fuel preparation procedure.

Fuel process engineering. During the last period, detailed engineering objectives were developed for the design of the fuel process equipment. The process flow was outlined, and a performance analysis was made to determine thermal, structural and performance compatibility for the process equipment. During this period all of these items were under constant and critical review. Further, information from others involved in hot cell operations has been received. As a result, engineering objectives, as well as process flow, have been modified. Section C of this chapter expands upon and clarifies various aspects originally presented. This section presents a detailed description of the design philosophy, scaleup and process flow, metallurgical steps and process equipment.

Near the end of the quarter, it became apparent that consideration should be given to employing an additional cell; too many tanks were being installed in the isolation room associated with Cell 2 (Ref. 1, p 44 and 63). Such an innovation, however, necessitates a re-evaluation of equipment locations within all Strontium-90 processing cells. Currently under study is the possible rearrangement of the equipment in Cell 2 to allow for shorter fuel process pipes to be compatible with tank relocations.

Fuel operations. During the period covered by this report, no fuel operation, as such, was carried out in the hot cells. Equipment and systems required to support Task 8 fuel processing were established. This work covered the cooling loop and decontamination dry box. Also, maintenance and repair of existing equipment continued.

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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, Ref. 1.

B. FUEL PROCESS DEFINITIONS**

1. Wet Chemical Processing

During this period a series of experiments were run to establish the wet chemical processing procedures. These experiments were:

(1) To determine the efficiency of the use of carbon dioxide gas and ammonia gas, versus sodium carbonate and sodium hydroxide, in order to minimize liquid waste.

(2) To determine the amount of strontium adsorbed by the filter aid.

(3) To determine the feasibility of using strontium titanate as an adsorbent for fission products from waste solutions.

The basic chemical process previously reported (Ref. 1) utilizes:

(1) Dissolution of the carbonate, as received, with nitric acid.

(2) Introduction of titanium dioxide as a slurry.

(3) Precipitation of the strontium carbonate in the presence of the titanium dioxide slurry.

In an attempt to minimize liquid waste, it was planned to introduce ammonia gas prior to precipitation to make the solution baic; carbon dioxide gas would then be used to precipitate the strontium carbonate. However, experiments showed this approach to be impractical. Ammonia gas and carbon dioxide gas reacted in the feed lines to form ammonium carbonate. This reaction clogged the flow meter as well as the glass frit through which the gases enter the solution. It was found necessary to pass ammonia gas into the hot solution continuously to keep the solution basic. Further, after a three-day period of bubbling of carbon dioxide at approximately 1 ft^3/hr through the solution, precipitation was still not complete. The solution was maintained at about 90° C since material precipitated at this temperature has been shown to possess better compacting and sintering properties than material precipitated at room temperature.

**G. Samos and W. Precht

For these reasons and because ammonium salts inhibit precipitation of strontium carbonate, it was decided that the base and carbonate would be added as solutions in the form of sodium hydroxide and sodium carbonate. Chemical analysis of material prepared in this manner shows the sodium retention after washing to be from 0.2 to 0.5%. Since there is no obvious effect on the pellet fabrication of materials having this level of sodium content, it is considered of little consequence. Therefore, the use of liquids instead of gases has been incorporated into the chemical processing procedure (see Appendix F for wet chemical process).

To show the amount of strontium adsorbed by the filter aid, the following investigation was made. A five-gram sample of strontium carbonate was dissolved with nitric acid. A 1-ml solution of Strontium-90 containing 10^6 counts/min was added to the solution. This solution was then made basic with sodium hydroxide, and strontium was precipitated as the carbonate by the addition of sodium carbonate. Five grams of filter aid (Celite) were added and the slurry thoroughly mixed. After filtration, the filtrate was diluted to 200-ml and a 1-ml aliquot was removed, dried and counted. This sample gave 0 ± 4 counts/min above background. The precipitate was then dissolved with 3M nitric acid and filtered to separate the filter aid. The filter aid was washed with 3M nitric acid and dried. One-half gram of this filter aid was put into a planchet and counted. The count was 4 counts/min ± 4 above background. The strontium was reprecipitated as carbonate and filtered without a filter aid. A sample of the filtrate gave a count of 7 ± 4 counts/min. This investigation showed the amount of strontium adsorbed by the filter aid to be of little significance and showed that $SrCO_3$ could be filtered without filter aids.

The use of strontium titanate as an adsorbent for fission products from waste solutions was also investigated. A 100-ml solution of mixed fission products was prepared, containing 124,100 counts/min-ml. The solution was made basic with sodium hydroxide, and one gram of strontium titanate was added. This was stirred overnight for approximately 16 hours. A portion of this solution was centrifuged, and a 1-ml aliquot of the supernate was evaporated to dryness and counted. The evaporated sample gave 3680 counts/min, equivalent to 97% decontamination.

A second sample, similar to the first, in which several milliliters of 30% peroxide were added with the strontium titanate, was prepared. It, too, was allowed to stir overnight. It was found, however, that a colloid formed, due to the presence of peroxide, preventing the complete removal of the strontium titanate from solution. The material would not settle upon centrifugation nor would filter paper contain it. Counting 1 ml of this sample showed a decontamination of 92.8%. It is believed, however, that this was due to activity carried in the colloid. The addition of strontium titanate in a basic solution of mixed fission products will therefore decontaminate the solution to approximately 97% of its original activity.

2. Pelletizing

a. Pellet fabrication

The process for preparing high density strontium titanate pellets from a powder blend of precipitated strontium carbonate with titanium dioxide involves calcining, ball milling, compacting and sintering. Experiments were performed to optimize some of the processing variables. The first experiment was to establish the necessity of ball mill blending.

A quantity of material was prepared to determine the effects of ball milling before calcining, ball milling after calcining and no ball milling. The quantity of material used for each batch contained a quantity of strontium substitute equivalent to 30 kilocuries of activity. This quantity is also equivalent to a triple batch; a triple batch is three times the size of the batches to be ball milled at Quehanna during the processing of Strontium-90.

An additional variation was tried between batches to include calcium impurities of 11 and 25% for two batches each. The following list indicates the starting weights of materials used to precipitate a triple batch containing 11% calcium contamination:

576 gm $SrCO_3$	660 ml conc HNO_3
96 gm $CaCO_3$	60 ml 50% NaOH
395 gm TiO_2	519 gm Na_2CO_3 in 30% solution

After weighing, the carbonates were placed in a large vessel, where a slurry was prepared by the addition of 700 ml of water. Nitric acid was added dropwise to the slurry to dissolve the carbonate. The resulting solution was made basic with sodium hydroxide, after which the TiO_2 powder was added. The solution was then heated to about 90° C with constant stirring. Sodium carbonate was added at 10 ml/min until the desired amount had been reached. After cooling, the slurry was filtered through a Whatman No. 42 filter paper. One liter of water was used to wash the precipitate, which was then placed in the oven to dry. After drying, the various powder blends were prepared and processed as shown in Tables 2, 3 and 4. Figures 21, 22 and 23 show sintered pellets of the material indicated in Tables 2, 3 and 4, respectively.

All powder blends were calcined at 1125° C for two hours in air. This temperature is slightly higher than previously used as a higher calcining temperature seems to yield a better product. The apparent

TABLE 2

Dimension and Weight Changes During Processing of $SrTiO_3$ Pellets with 11 Atomic Percent Ca^{++}

Material	Lot No.	Weight of Lot at Start (gm)	Weight After Ball Milling (gm)	Weight After Calcining (gm)	Weight After Ball Milling (gm)	Compact Load (tsi)	Pellet Number	Initial Weight (gm)	Pressed Weight (gm)	Pressed Dimensions (in.) Diameter	Pressed Dimensions (in.) Height	Sintered Weight (gm)	Final Dimensions (in.) Diameter	Final Dimensions (in.) Height	Density (% of theoretical) Pressed	Density (% of theoretical) Final**
Precipitate of $SrCO_3$	1	300		220.5	216.3	5	1A	72.1	72.1	1.766	0.622	71.84	1.542	0.546	60.2	88.4
(~ 90°C) + 11% Ca^{++} in slurry of TiO_2						5	1B	72.1	72.05	1.766	0.619	71.75	1.542	0.549	59.8	87.4
Batch A						5	1C	72.4	72.35	1.766	0.625	72.10	1.542	0.547	60	87.9
	2	300		233		5*-10	2A	77.6	77.03	1.766	0.582	76.20	1.723	0.573	68.9	71.2
						10	2B	77.6	77.6	1.766	0.608	76.60	1.728	0.594	66.6	68.6
						20	2C	77.98	77.9	1.766	0.580	77.25	1.728	0.559	69.8	73
	3	300	296.5	233.9		5*	3A	79.9	79.88	1.766	0.759	79.60	1.690	0.719	54.5	61
						10	3B	79.9	79.83	1.766	0.689	79.60	1.701	0.683	60.2	63
						20	3C	74.3	74.22	1.766	0.606	73.90	1.703	0.596	63.7	67.9

* Sample crushed and repressed.

** Based on theoretical density of 4.9 gm/cm^3.

TABLE 3

Dimension and Weight Changes During Processing of $SrTiO_3$ Pellets with 25 Atomic Percent Ca^{++}

Material	Lot No.	Weight of Lot at Start (gm)	Weight After Ball Milling (gm)	Weight After Calcining (gm)	Weight After Ball Milling (gm)	Compact Load (tsi)	Pellet Number	Initial Weight (gm)	Pressed Weight (gm)	Pressed Dimensions (in.)		Sintered Weight (gm)	Final Dimensions (in.)		Density (% of theoretical)	
										Diameter	Height		Diameter	Height	Pressed	Final**
Precipitate of $SrCO_3$ (~ 90°C) + 25% Ca^{++} in slurry of TiO_2 Batch B	4	300		193.0		5*	4A	64.3	64.25	1.766	0.614	64.05	1.606	0.556	56.1	72.5
						5	4B	64.3	64.3	1.766	0.633	64.10	1.622	0.579	54.29	68.9
						5	4C	64.6	64.65	1.766	0.638	64.45	1.633	0.585	53.9	67.7
	5	300		205.0	203.0	5	5A	67.6	67.52	1.766	0.643	67.25	1.490	0.543	56.1	91
						5*	5B	67.1	67.05	1.766	0.625	66.70	1.491	0.535	57.4	91.6
						5	5C	68.3	68.26	1.766	0.650	68.00	1.493	0.563	56.1	88.7
	6	300	298.3	234.3		5	6A	78.1	78.05	1.766	0.781	77.60	1.625	0.720	53.4	66.2
						5*	6B	78.1	77.9	1.766	0.743	77.80	1.636	0.697	56.1	67.7
						5*	6C	78.3	77.98	1.766	0.758	77.90	1.638	0.703	55.1	67.2

* Sample crushed and repressed.

** Based on theoretical density of 4.76 gm/cm^3.

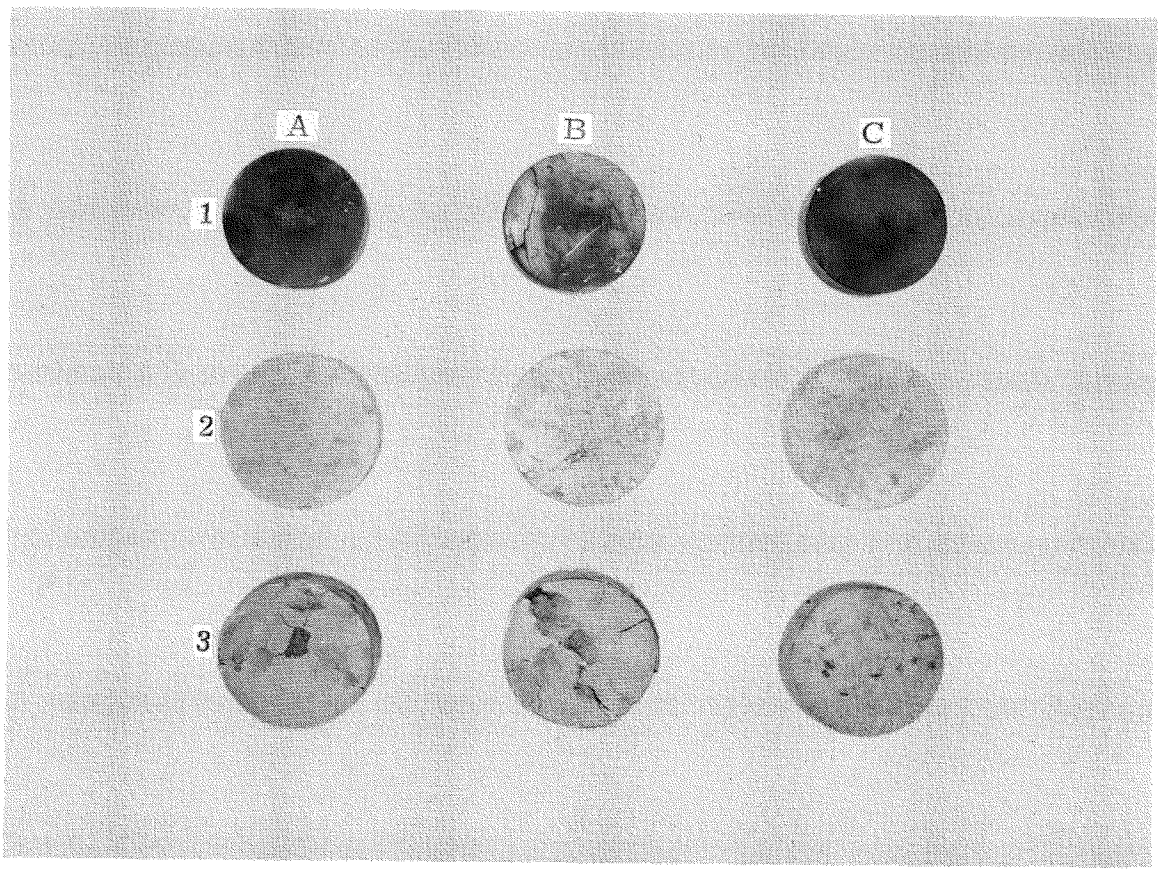
TABLE 4

Dimension and Weight Changes During Processing
$SrTiO_3$ Pellets with Impurities

Material	Lot No.	Weight of Lot at Start (gm)	Weight After Ball Milling (gm)	Weight After Calcining (gm)	Weight After Ball Milling (gm)	Compact Load (tsi)	Pellet Number	Initial Weight (gm)	Pressed Weight (gm)	Pressed Dimensions (in.)		Sintered Weight (gm)	Final Dimensions (in.)		Density (% of theoretical)	
										Diameter	Height		Diameter	Height	Pressed	Final **
Precipitate of $SrCO_3$						5	7A	79.0	78.88	1.766	0,816	78.85	1.603	0.735	51	66.3
(~ 90°C) + 10% Ca^{++} ball mill blended with TiO_2	7	300	298	237.2		5*	7B	79.0	78.97	1.766	0.817	78.85	1.613	0.739	50	65.3
						5*	7C	79.4	79.28	1.766	0.790	79.15	1.602	0.711	52	68.9
$SrCO_3$ + 25% Ca (~ 90°C) ball mill blended with TiO_2	8	300	297.6	234.2		5	8A	78.0	77.9	1.766	0.815	77.75	1.619	0.744	51	64.9
						5	8B	78.6	77.9	1.766	0.791	77.75	1.617	0.724	52	66.8
						5*	8C	78.6	78.5	1.766	0.795	78.40	1.602	0.728	52	68.7
Chemically pure $SrTiO_3$						5	9B	80	79.85	1.766	0.747	79.60	1.533	0.645	53.3	80

* Sample crushed and repressed.

** Calculated density--lot 7 = 4.9 gm/cm^3.
Calculated density--lot 8 = 4.76 gm/cm^3.

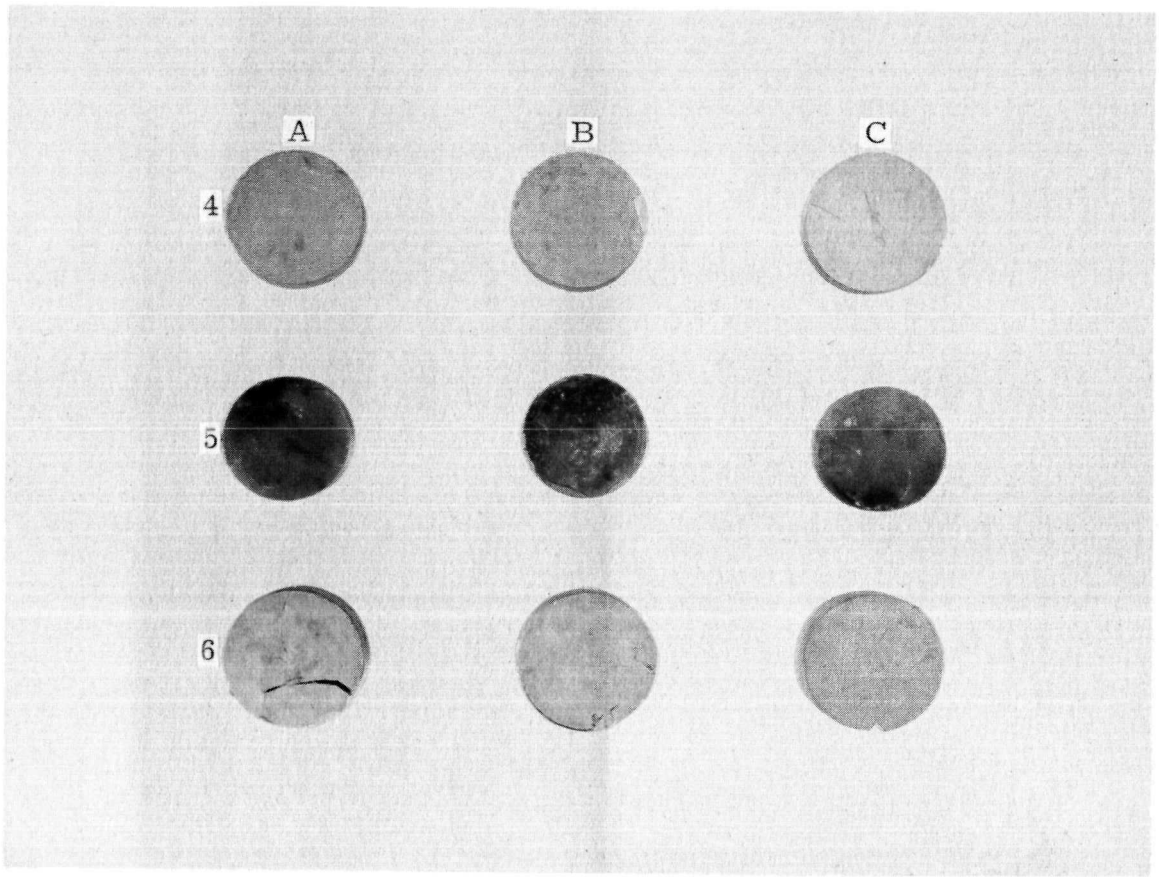


Row 1--calcined, ball milled,
pressed, sinistered.

Row 2--calcined, pressed
sinistered.

Row 3--ball milled, calcined,
pressed, sintered.

Fig. 21. Sintered Pellets--$SrTiO_3$ + 11% Ca (precipitate blend)

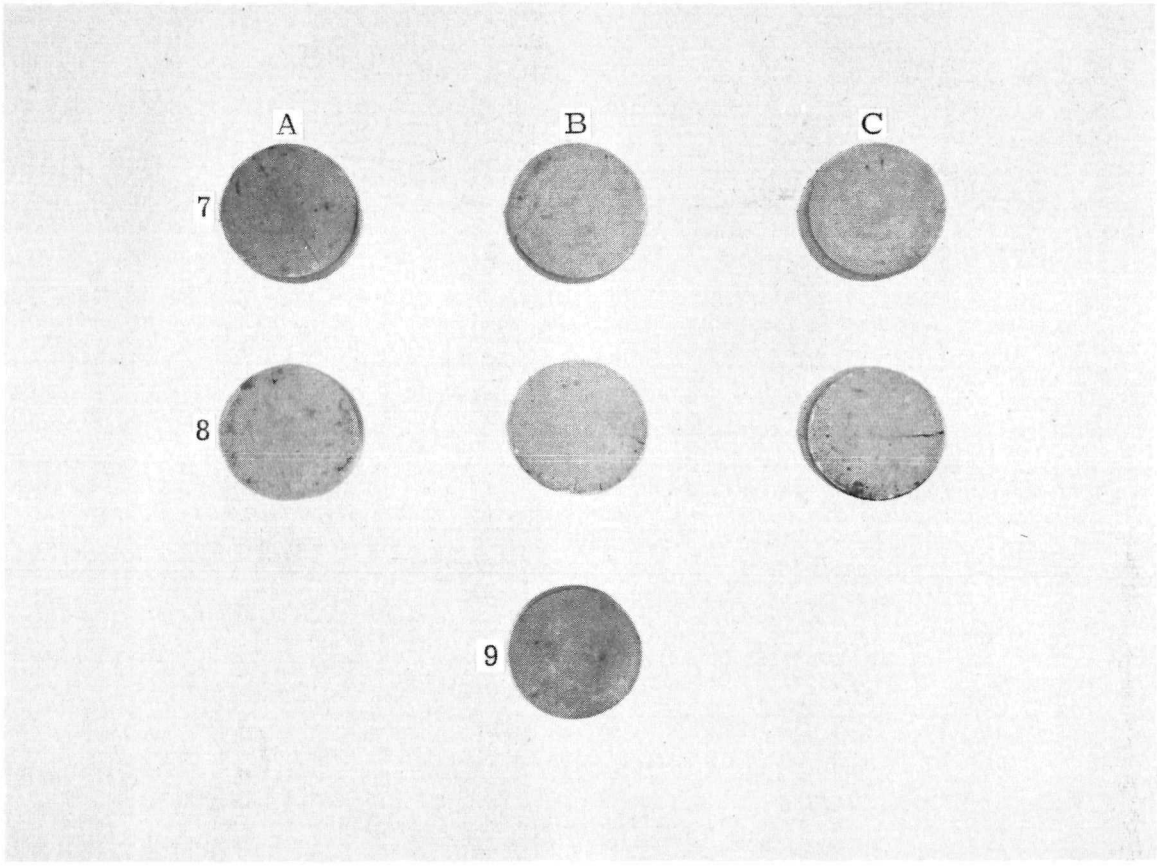


Row 4--calcined, pressed,
sinitered.

Row 5--calcined, ball milled,
pressed, sintered.

Row 6--ball milled, calcined,
pressed, sintered.

Fig. 22. Sintered Pellets--$SrTiO_3$ + 25% Ca (precipitate blend)



Row 7--10% Ca impurity: ball milled, blended with TiO_2, calcined, pressed and sintered.

Row 8--25% Ca impurity: ball milled, blended with TiO_2, calcined, pressed and sintered.

Row 9--Standard $SrTiO_3$

Fig. 23. Sintered Pellets--Plain Carbonate Precipitate and Standard $SrTiO_3$

discrepancies between various lots in weight loss after calcining is probably due to variation in the moisture content of the starting lots. Compacting was accomplished at a pressure of 5 tsi whenever possible. Higher compacting pressures were used only when 5 tsi was not sufficient to yield a sound compact. Although calcining at higher temperature is advantageous in reducing the compression ratio of the powder, it also results in partial sintering of powder during calcining. Unless the partially sintered pieces are properly crushed prior to compacting, the final pellets show poor sintered properties.

The use of higher compacting loads in attempting to crush these presintered pieces results in excessive powder buildup in the die and decreases die life. It has been necessary to polish the die after compacting powders with loads equivalent to 10 or 20 tsi. However, the use of powders which are ball milled after calcining yields satisfactory compacts with loads of 5 tsi. Wiping of the die after compacting at 5 tsi proved sufficient for the removal of any residual powders. Laminations appeared after sintering in some of the pellets which were ball milled after calcining; however, it is believed that this effect can be eliminated by proper particle size control of the calcined and ball milled powders.

In view of the low density obtained with the standard sample (No. 9B, Fig. 23), it appears likely that two hours at temperature was not sufficient for sintering a charge of this size. The location of pellets in the furnace was varied between batches so that pellets processed in a similar manner were not in the same location during sintering. The results show there was no effect in the sintering of pellets processed in the same manner but located in different furnace positions during sintering. As shown in Tables 2 and 3, the processing of the two batches is identical, except for the variation in calcium content. The effect of increased calcium content seems to be of little consequence during pellet fabrication.

The results of these experiments conclusively show the necessity of ball mill blending after calcining in order to yield sound, uniform pellets of high density.

In addition to the investigation mentioned, several experiments are presently being conducted to evaluate the effect of calcining time and temperature, plus additional ball milling time. Results of these experiments will help finalize the pelletizing process.

b. Strontium loss and impurity pickup during ball milling

Several other experiments were also conducted to establish strontium loss and impurity pickup during ball milling.

Strontium loss. Chemical analyses were performed on the filtrate from material that had been calcined at 1200° C for one hour and wet ball milled. The filter used was stainless steel with 20-micron pores. The average amount of strontium found in the filtrate was 6 ppm. A calcining temperature of about 1200° C for one hour is therefore sufficient to prevent the presence of water-soluble forms of strontium during wet ball milling after calcining. The amount of strontium found in the filtrate is probably due to carrythrough of the material through the 20-micron pore size filter. The extremely small amounts of strontium found in both the filter aid and filtrates indicates that the loss of strontium through adsorbtion or solubility will be negligible.

Impurity pickup. Since the fabrication process to be used includes wet ball milling, chemical analyses have also been made to show the amount of iron introduced during wet milling in a steel ball mill. The analyses show an iron content before milling of 0.002%; after 2-hour milling, 0.012%; and after 4-hour milling, 0.038%. As in the case of small sodium addition, these iron additions have proved to be of no obvious consequence in effecting pellet processing or chemical stability of the titanate body.

C. FUEL PROCESS ENGINEERING

1. Design

The design criteria and mode of operation for the Strontium-90 fuel processing are as follows:

(1) Because of space requirements and safety considerations, Cells 1, 2 and 5 were made available for strontium process work at Quehanna. An enclosure box will be provided in Cell 2 to serve as primary containment for the radioisotope during the processes in which radioactivity may be released to the immediate environment. Such processes include the strontium precipitating, drying, calcining, sintering, pressing and welding. Model 8 manipulators will be installed through the enclosure face to perform normal light duty operations. In addition, a small electric hoist will be provided for equipment movement inside the enclosure. Bag-out for waste disposal may be constructed in the back of the enclosure. In transferring large weights in and out of the enclosure, the crane outside the cell will be employed. The containment box has an inlet and outlet air system that must pass through a prefilter and absolute filter.

(2) Emergency manned entry inside any cell will be permitted. This entails:

(a) The shielding of selected hot containers.

(b) Reversing the flow of hot liquids in lines and unshielded vessels back into the shielded containers.

(c) Proper housing of solid materials.

The solid materials may be either pellets, cake, or calcined material; each should be kept in a separate container specifically designed for storage. A progressive check on the radiation level in the cell will be instituted during the shutdown operation prior to manned entry. Provisions will be made for routine entry into Cell 1 to handle casks, waste drums, hooks, etc.

(3) Transfer ports will be maintained between the enclosure in Cell 2 and Cell 1 and out of Cells 1, 2 and 5.

(4) The air flow will be toward the area of highest contamination (i.e., the containment box). All contaminated air will

be exhausted through the containment box atmosphere whereas the uncontaminated air may be passed directly through the absolute filter of the containment box. Contaminated air is defined as any gas that has passed over radioactively contaminated surfaces or evolved from a substance containing radioactive nuclides.

(5) All equipment--parts, insulation, windows, etc.--inside the cell should be able to withstand a total dose of 10^8 roentgen equivalent physical (rep).

(6) Storage and slurry tanks. Cooling capacity for each will be:

(a) 10,000 Btu/hr, based on 168 kilocuries of Strontium-90 and Strontium-89/Strontium-90 ratio = 3.

(b) 5000 Btu/hr, based on 168 kilocuries of Strontium-90 and Strontium-89/Strontium-90 ratio = 0.25.

(7) Provisions will be made for sampling techniques from the following:

(a) Final wash from cask.

(b) Feed solution in storage tank.

(c) Wash from precipitates.

(d) Ball mill wash.

(e) Waste tanks.

(8) The transfer of materials from a relatively high radiation area, such as the containment box, to a low radiation area, such as Cell 1, will be conducted through intermediate confinement and decontamination. A swipe check will be made immediately after the final decontamination of the samples and their removal from the cell.

(9) 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.

(10) Valves. Where 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 applications.

(11) Plumbing. Stainless steel tubing will be used in all lines to permit the transfer of nitric acid and radioactive solutions. A double containment technique will be employed to deal with line rupture or leaks of hot material.

(12) Fittings. Swagelok flareless fittings or the equivalent will be used to join the plumbing lines.

(13) Manipulators. The manipulators will be booted by a standard arrangement, commercially available. 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 manipulator may be bagged upon removal from the cell.

(14) A clean cask surface must be maintained throughout the hot material transfer in order to avoid large areas of decontamination and consequent delays in returning casks to the AEC. Cleanliness may be maintained through the use of a shielded enclosure to house all the connectors leading in and out of the cask. The enclosure must be completely sealed to provide positive containment of any spillage and prevent leakage to the outside surface.

(15) Waste disposal. Pieces of equipment will be either decontaminated or bagged and encased in concrete. The solid products will be retained in a disposable filter encased in concrete. All of the liquids used in the process, and for decontamination, will pass through a controlled waste tank. The solids will be removed from solution by physical means. The activity of the remaining fluid will be checked prior to disposal. The solids will be retained in a disposable filter, and the fluids will be passed on to a waste tank containing vermiculite.

2. Scaleup and Process Flow

The following describes the requirements for processing the supplied Strontium-90 feed material into strontium titanate pellets. Detailed analysis of the process is also given.

The scaleup of the process for Quehanna is based on the handling of a maximum production capacity of 168 kilocuries* of Strontium-90 per shipment, and 100 kilocuries processing per month, in the form of titanate pellets. The scaleup is defined in the sequence of the operations involved together with auxiliary equipment, check points and safety measures.

The raw fuel is received as strontium carbonate deposited on a filter in a shielded shipping cask. The shipping cask is installed in Hot Cell No. 1. A thermocouple is installed in the cask well, and the cooling lines are attached. With the box closed, the vent, the solvent-in line and the slurry-out line are connected by manual means through glove ports. A window will permit observation of the hookup and checking during the transfer operation.

Prior to the start of the fuel transfer, the cask is cooled. The system is now ready for the fuel transfer and processing (see schematic diagram Fig. 24).

Individual process steps are described in the succeeding section.

a. Carbonate cake dissolution and transfer from cask to slurry tank

The cake, together with the filter aid in the cask, will be slowly purged with nitric acid, slowly enough to prevent gas pressure buildup. The exhaust system will meet this basic criterion in order to eliminate back pressure in the lines.** During this operation, most of the carbonates will be dissolved, the carbonate aggregates will be broken loose and the filter aid will fall off the screen. The slurry thus formed will be transferred by vacuum to a receiving vessel called the slurry tank.

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* This figure corresponds to the maximum activity expected to be shipped by Hanford. This is actually the limit of cask capacity at maximum carbonate cake density, 0.75 gm/cc. Scaleups for storage and operation are to 168 kilocuries and 20 kilocuries, respectively

** An isolation tank will be installed between the water-nitric acid reservoirs and the cask in order to contain radioactive materials that may be pushed out of the cask by carbon dioxide back pressure into the acid lines.

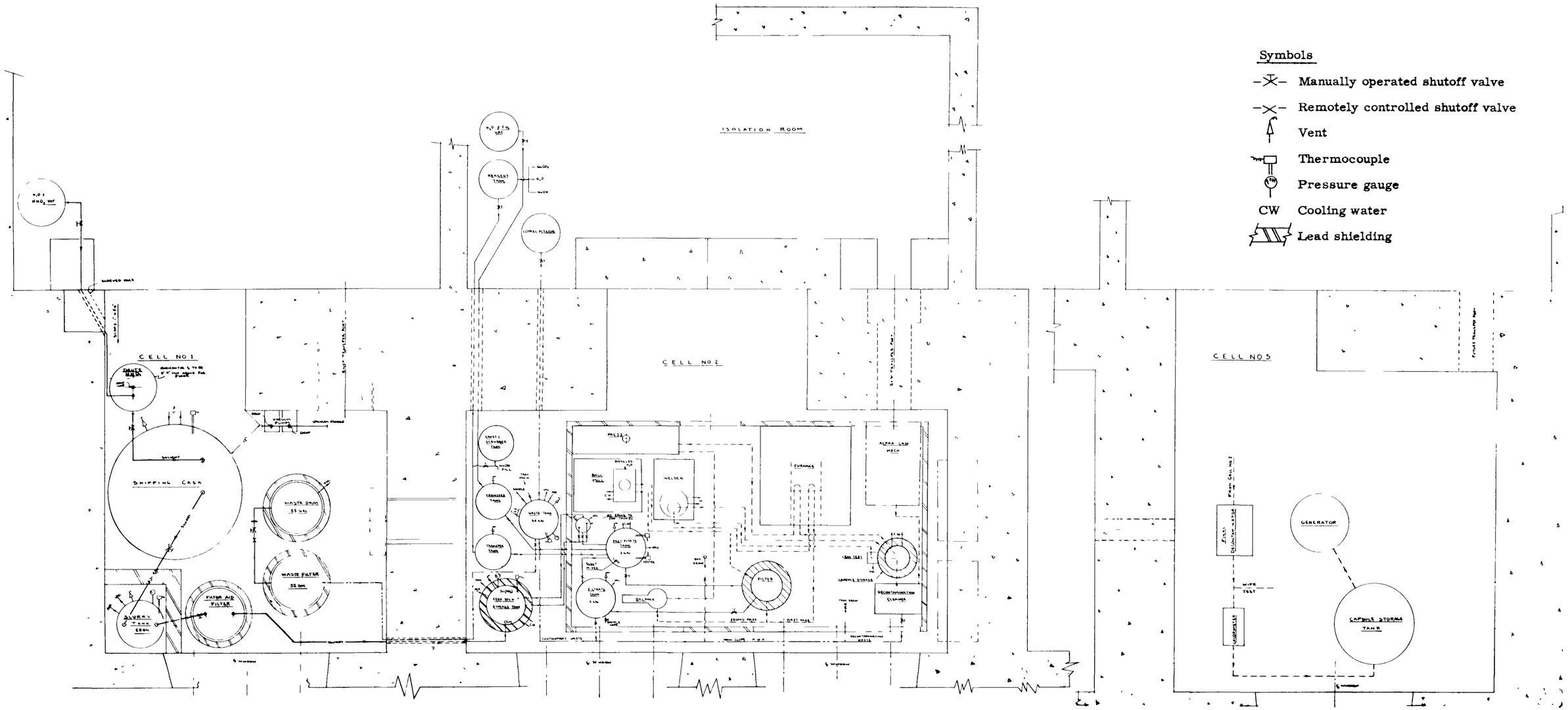

Fig. 24a. SNAP 7 Strontium-90 Fuel Processing Schematic Diagram, Showing Transfer and Waste Lines

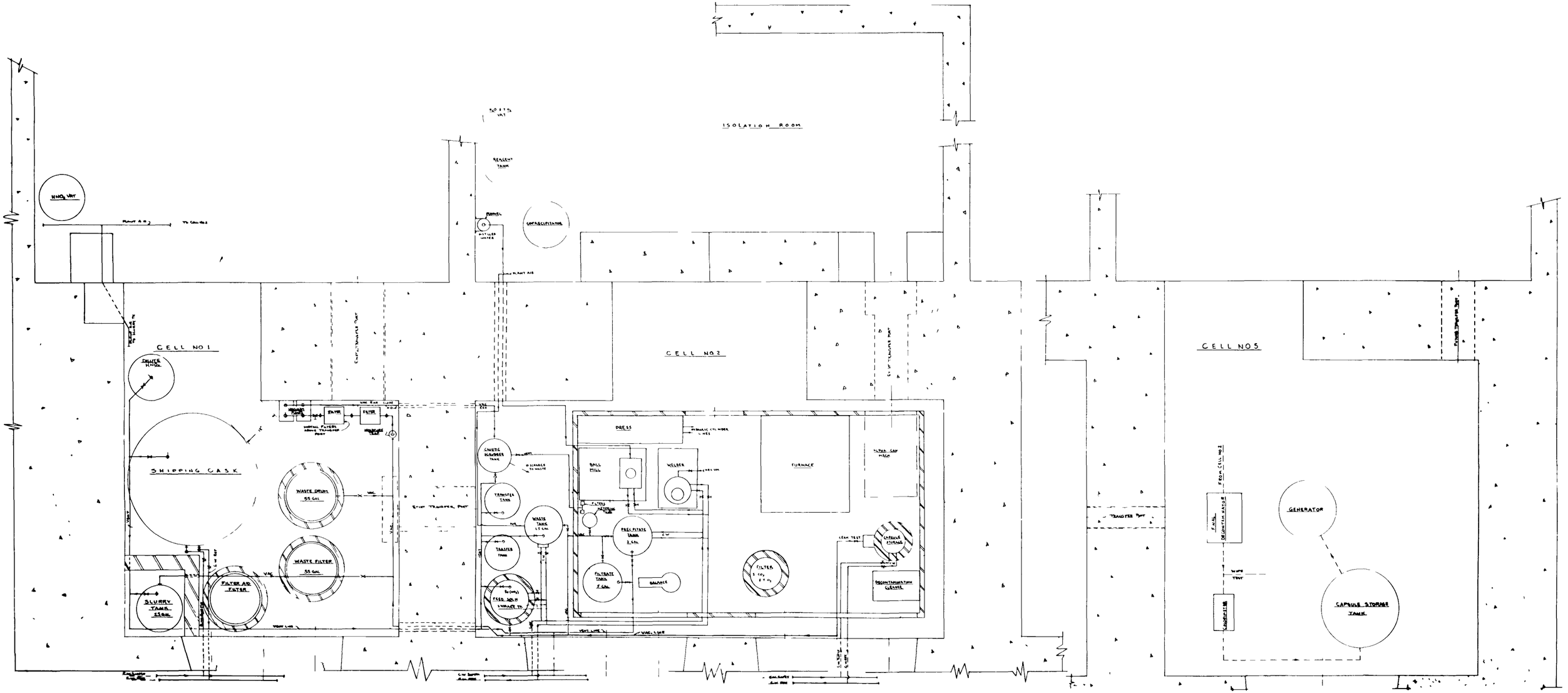

Fig. 24b. SNAP 7 Strontium-90 Fuel Processing Schematic Diagram, Showing Vacuum, Vent and Utility Lines

The nitric acid requirement to dissolve a carbonate cake weighing 4920 grams, with a calcium content of 25 wt % of the cations present, is 77 moles. The total amount of carbon dioxide evolved in the dissolution process is 38.2 moles, or 855 liters at STP. Apportionments of the amounts of nitric acid required and carbon dioxide evolved from the cake components are given as follows:

Compound	Weight (gm)	HNO_3 Required (moles)	CO_2 Generated (moles)
$SrCO_3$	3280	44.7	21.8
$CaCO_3$	1640	32.8	16.4
Total	4920	76.5	38.2

Additional acid will be added for rinsing and digestion of precipitates adsorbed on the inside cask walls and lines leading to the slurry tank, as well as undissolved carbonate in the slurry tank. The following tentative program will be used:

	Volume (liters)	Reagent	Concentration	Digestion (min)
Dissolution	10	HNO_3	3M	5 to 10
	10	HNO_3	3M	5 to 10
	6	HNO_3	3M	5
Rinsing	5	HNO_3	0.5	5
	5	HNO_3	0.5	5
	6	HNO_3	1	10
	5	H_2O	--	5
Total volume	47			
Total acid	89 moles			

b. Check on cake transfer from cask

(1) Cask surface dose rate measurements

The dose rate at the surface of the shipping cask is directly proportional to the amount of fuel inside the cask. As the fuel is removed, the dose rate at the surface decreases. Therefore, the radiation of the surface of the cask will be monitored by a device outside the cask, in Cell 1, for the fuel transfer operation. If, for any reason, the cask surface becomes contaminated, or if the background radiation level is too high, this monitoring technique will give an inaccurate indication of cask content. Other counting methods will be used in support of the main monitor.

(2) Radiometric analysis

Directional shielded probe. Before the hot operation, a directional shielded detector, looking into an unshielded section of the feed line leading to the slurry tank, will be installed in Cell 1. The detector will monitor automatically the activity level reaching the tank as a result of the transfer operation.

Wash activity. After completion of the primary transfer operation, the cask will be filled with 1M nitric acid and any reaction will be allowed to continue for five minutes. The system will be purged with air, and a sample from the stream will be obtained at a point before the stream reaches the slurry tank. A radiometric analysis, at this point, will determine the concentration of Strontium-90 (and associated activities). The amount of activity thus measured will be indicative of the degree of residual internal contamination in the cask.

(3) Chemical and/or radiometric analysis

An aliquot of concentrated solution will be obtained from the storage tank. Following the addition of sodium hydroxide to neutralize the solution, sodium carbonate will be added to the solution to precipitate strontium as $SrCO_3$. The carbonate precipitate will be dried and weighed. If an average shipment is assumed to be 10^5 curies dissolved in 50 liters of acid solution, the weight of the carbonate precipitated from a 1-ml sample will be approximately 0.05 gm, having

a Strontium-90 activity of about two curies. This activity level can be handled and measured adequately at Quehanna. A radioanalysis of the sample may be used to support or replace the above technique.

c. Digestion of slurry solution

In order to speed up dissolution and to leach out all the strontium from the filter aid surfaces, a digestion period of 15 minutes in the slurry tank, with constant agitation, will be allowed. The stirring will prevent settling of the filter aid. By vacuum, the slurry will be transferred through a filter into a storage vessel, the filter aid being retained on the filter while the solution passes through. The shipping cask, slurry tank and filter will be washed in sequence by a slightly concentrated acid, which will be withheld in the slurry tank for final removal of adsorbed solutions or particles. Two dilute nitric acid rinses follow. A final water rinse will be used to clean the acid from the lines and vessels. The acid wash volume must be kept at a minimum to reduce cation contamination in the neutralization process, solubility of Sr^{++} and Ca^{++} and volume of waste solution.

(1) Equipment

(a) Filter

The filter, which must be fine enough to retain the filter aid, will consist of 400-mesh material. It must have the capacity to retain the solids from six shipping casks (approximately five gallons).* The metal parts of the filter must be stainless steel which can withstand 6N nitric acid. The filter will be encased in a shielded container, and the container will be shielded so that the maximum radiation dose rates will not exceed 200 mr/hr at the surface of the shield or 10 mr/hr at a distance of 1 meter from the center of the container. It has been demonstrated by tracer experiment, duplicating as nearly as possible the coprecipitation of strontium carbonate with filter aid, that more than 99% of the strontium was recovered. Actually, the filter aid in

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* If it is assumed that the volume of cake per shipment occupies approximately 1/2 the cask's volume, then the total capacity of the filter should be 6 x 3 = 18 liters.

the shipping cask will be subjected to continuous heating from the time of cask loading to cake dissolution. This may alter significantly the extent to which strontium is adsorbed on the filter aid surfaces. Hanford, on the other hand, is duplicating cask conditions, including the heating. Experiments are in the planning stages. The filter and its container will be located in an enclosure with Cell 1.

(b) Storage vessel

The storage vessel will have a capacity of 25 gallons to store strontium in the form of the nitrate $Sr(NO_3)_2$ after it is transferred from the slurry tank. The vessel will be constructed of stainless steel to be resistant to 6N nitric acid. The vessel must be shielded and must have provisions for connections to the system vent and vacuum systems. In addition, provisions must be made for a feed and discharge line. The discharge line will be a 3/8-inch tube entering through the top of the vessel. A perforated ring, located near the base of the vessel, must be provided for connection to the compressed air system to effect agitation for stored solutions. A thermocouple well will be installed to measure solution temperatures. Adequate cooling capacity will be provided to remove nearly 10,000 Btu/hr of heat from the Strontium-90 solution.

(c) Slurry tank.

The slurry tank will have the same characteristics as the storage vessel. This choice was made on the basis that the wash and digestion will take an appreciable time during which the heat generated, gases evolved, etc., should be removed.

d. Thermal output

The total heat output was calculated as follows:

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* In order to check on the precise total amount of the $Sr(NO_3)_2$ solution available in the storage tank, a complete record of the total mass transferred will be kept. Further, the amount of retention in the lines, if any, will be determined in advance through the dry run operations.

Sr-90 = 168 kc x 6.3 watts/kc = 1060 watts

Sr-89 = (168 x 3) x 3.4 watts/kc = 1714 watts

Total 2774 watts

2774 watts x 3.41 Btu/hr-watt = 9450 Btu/hr

e. Precipitation

A measured volume of the strontium storage solution containing approximately 10^4 curies will be transferred to the precipitation vessel. There it will be mixed with two liters of a titanium dioxide-water slurry containing sufficient excess titanium dioxide to react with all the metal ions in the solution during the sintering process.* The mixture will be mechanically agitated. A sufficient amount of a 50% sodium hydroxide solution containing a 10% excess will be added to neutralize the solution. Sodium carbonate dissolved in 1000 ml of hot water (90°C) will be added to precipitate the carbonate.

(1) Equipment

(a) Transfer vessel.

The stainless steel transfer vessel will have the capacity for metering out 500 to 1000 ml of strontium nitrate solution for the precipitation reaction. It will be provided with openings to connect to vent and vacuum and for liquid inlet and discharge. Its total capacity will be approximately 0.5 gallon.

(b) Precipitation vessel.

The precipitation vessel, also constructed of stainless steel, will have a 3-gallon capacity. A source of heat will be provided to maintain a temperature of 90° C for 10 liters of slurry. A thermocouple well must be installed to measure the temperature of the slurry. The vessel must be equipped with connectors to vent and vacuum lines. Among other equipment are an inlet line and a bottom discharge line and a mechanical stirrer near the base of the vessel for agitation. (A reflux condenser is required to retain condensible vapors in the precipitator.) A spray ring will be provided to rise off precipitate from the vessel walls.

* Exact amounts will depend on shipment size and the extent of dilution.

f. Strontium filtration

The slurry from the precipitator will be cooled to 20°C and filtered through a medium porosity filter, after which the filtrate will be passed to a hold tank and recycled through the filter cake. The filter cake will be washed with one liter of a dilute carbonate solution. Liquid transfer is accomplished by applying a vacuum to the receiving vessel.

(1) Solubilities of alkaline earths.

The carbonates of calcium, strontium and barium have a slight solubility in pure water. The following list indicates their solubilities (Ref. 5):

(1) At 25°C, 0.015 mg of $CaCO_3$ dissolves in 1000 mg of water.

(2) At 18°C, 0.011 mg of $SrCO_3$ dissolves in 1000 mg of water.

(3) At 18°C, 0.02 mg of $BaCO_3$ dissolves in 1000 mg of water.

Further, higher solubilities are expected at elevated temperatures. Since the precipitation is being carried out at 90°C, the solubilities of the carbonates might present, first, an appreciable loss of Sr-90 and second, a high-activity waste. However, estimates based on the above solubility data for strontium indicate that the loss is only a few curies in 10 kilocuries. The large concentration of soluble electrolytes present at the time of precipitation may tend to depress the solubilities of the precipitated salts of alkaline earths. This point has been verified by laboratory investigation. In addition, the precipitate will be washed by a dilute carbonate solution in order to take advantage of a mass action effect which favors the precipitate formation.

(2) Equipment

(a) Filter

The filter has a capacity of two liters. The filter medium is tentatively defined to be a fine, porous stainless steel filter. ORNL is currently using a stainless steel filter and a ceramic alumina filter manufactured by Norton. It is provided with a removable head that is in place during operation. For removal and transfer of the solid cake, the head is taken off.

(b) Holdup tank

A 5-gallon holdup tank is provided to receive the filtrate from the strontium precipitation vessel. It is a stainless steel vessel equipped with a feed line, a discharge line and a connection to the vent and vacuum system.

g. Waste disposal system

All liquid drains from process equipment in the strontium system will be drawn into the tank either by gravity drain or by vacuum applied to the storage tank. Inert strontium titanate powder (1 gm/100 ml of filtrate) or clinoptilolite will be added to the waste solutions. The slurry will be drawn through a filter, and the filtrate will be mixed with vermiculite for disposal as solid waste.

(1) Equipment

(a) Waste storage tank

A 25-gallon stainless steel vessel will be provided to store liquid wastes from the strontium processing. The tank will be equipped with a liquid level indicator system to measure volumes from 0 to 25 gallons. A manifold will connect all equipment discharge lines with the waste storage tank; an air line will be attached to a ring in the base of the tank for agitation. A 1/2-inch discharge line and connections to the sampling line, vent and vacuum system will be included in the system.

(b) Filter

The filter must be fine enough to retain 400-mesh solids, and it must have a 5-gallon capacity. After it has been filled, the filter is to be removed from the system. For that purpose it will be encased in a shielded container such that the maximum dose rate at the surface will not exceed 200 mr/hr or the dose rate at 1 meter from the center of the vessel will not exceed 10 mr/hr.

(c) Vermiculite absorber

A shielded container containing a charge of vermiculite will be employed to absorb about 15 gallons of water solution. The container will be stainless steel, and the shield will be adequate to reduce the maximum dose rate to below 200 mr/hr at its surface and below 10 mr/hr at 1 meter.

h. Air vent system

An air vent system will be provided. The vents from the individual pieces of equipment will be manifolded and discharged through an MS absolute filter. All vent lines will be 1/4-inch stainless steel resistant to nitric acid vapors. Prior to discharge, appropriate vent gases will be passed through a caustic scrubber to prevent damage to the absolute filters by acid fumes.

3. Metallurgical Steps

a. Calcination

The cake will be introduced in the furnace for calcination. The calcination process will bring about the thermal decomposition of the carbonates and a partial conversion of the oxides thus formed to the titanates:

$$X\,CO_3 \xrightarrow{1300^\circ C} X\,O + CO_2$$

and

$$X\,O + TiO_2 \xrightarrow{1300^\circ C} X\,TiO_3$$

where X represents the cation of alkaline earth. The following firing schedule for an 80-gram charge has been defined by the Nuclear Components Department:

Temperature (°C)	Time (hr)
100	1.5 to 2
300	1
500	1
700	1
900	1
1100	1
1300	2

The sample will be allowed to cool to ambient cell temperature before transfer out of the furnace.

To simplify the calcination procedure, the ORNL technique will be considered.

4. Process Equipment*

The following descriptions define the size, material, and pertinent features of the various pieces of equipment used to process Strontium-90 into a usable fuel form.

Containment box. The approximate dimensions of the containment box are width, 9 feet; depth, 5 feet and height 9 feet. The box is constructed of stainless steel with No. 2 Plexiglas windows. Large windows are provided in the front face of the box to match with the cell windows for observation. Above these, in line with the cell lights, are smaller windows for auxiliary lighting, and in the top surface of the box there are additional windows. The main box lighting system rests on top of the box in such a manner that the lights may shine down into the box. The overhead lights provide 60,000 to 66,000 lumens and are supplemented by the auxiliary lighting to eliminate shadow problems. The lighting plan includes dimmer controls to permit operator selection of the lighting intensity.

The box contains a bridge-type hoist in order that the heavy pieces of equipment may be moved by remote methods. This is especially vital in case of a failure during processing where replacement or repair of contaminated equipment becomes necessary. A large hinged door ($3\ ft^2$) is located in the top of the box for installation and bag-out removal of the large pieces of equipment. In the rear of the box an 18-by 24-inch bag-out door is provided. This door is supplemented by an observation window, and plans include a gloved diaphragm installation to fit over this opening to assist in box maintenance.

Four supporting systems are to be installed: ventilation, transfer, plumbing and electrical. These systems are presently in the design stage. The ventilation system will maintain a reduced pressure of 0.5 inches H_2O in the box, with filtered inlet and outlet air. The system will be installed in such a manner that contamination spreading will be minimized.

The transfer system will provide for the movement of capsules and samples in and out of the box and also the maintenance equipment, wipes, replacement parts, etc. Two general transfer systems are being evaluated, with variations of each system. The first is a lock-type system

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* C. Young.

between cells for transferring materials immediately after they come from an ultrasonic cleaning tank, and it will include provisions to prevent tracking of contamination from one cell to the other. The second type of system is supplemental to the first, and consists of a closed vessel which can receive and discharge material without contaminating the exterior surfaces. In principal, it is similar to the alpha cans used at Los Alamos.

The plumbing is to be installed by development in the box. An installation drawing will be provided to show the locations for all equipment and to show the approximate plumbing runs. The development is to be supervised and approved by the Engineering Department. The plumbing will be stainless steel tubing with stainless steel Swagelok fittings.

The electrical system is to provide individual control over each piece of electrical equipment used from the control panel. The temperature of all critical containments and processes will be controlled and monitored. This electrical system is to consist of an electric furnace, a heater for the precipitation vessel, thermocouples, motors, bridge-type hoist, solenoid valves and lights.

The box structure will be sufficiently strong to permit the installation and removal of the box with an overhead crane with all of the process equipment within the box in place. The box will be braced by external members of structural steel which have been primed and painted with an acid resistant paint. In order to keep the pieces of equipment at a minimum size, the box design permits separating the box from the stand and lights.

Feed solution tank. The tank is to be stainless steel with a 25-gallon capacity; it is currently under procurement. Provisions have been made for solution, vacuum and vent lines. A thermocouple and a liquid level indicator will be installed. All penetrations into the tank will be through the top to minimize the possibility of a leak. A cooling water jacket will surround the tank, which will be base supported. The tank is constructed according to the Unfired Pressure Vessel Code.

This tank is used to store the strontium nitrate solution after the filter aid has been removed.

Slurry tank. This tank is the same as the feed solution tank. It will hold the slurry from the shipping cask. The tank order was placed during April.

Liquid waste tank. This tank, too, is the same as the feed solution tank. It will be used to hold the liquid waste prior to final disposition. The tank has been ordered.

Reagent vessels. The reagent vessels are 5-gallon open top stainless steel tanks with dust covers. These vessels are used to introduce the various solutions into the process system. Wall mounting brackets are provided. These vessels have been ordered.

Transfer tank--large. The transfer tank is a 5-gallon stainless steel tank which is to be placed in the line between the reagent vessel and the reaction vessel with valves on either side. Venting provisions are to be included. The purpose of the transfer tank is to prevent blow-back into the area where the reagent vessels are located. Wall mounting brackets will be provided. The tank is designed to satisfy the Unfired Pressure Vessel Code.

Transfer tank--small. This tank is to serve the same general purpose as the larger transfer tank, but it will be used to handle smaller volumes and naturally, it will occupy less space. The capacity is 0.5 gallon, and the construction is of stainless steel according to the Unfired Pressure Vessel Code. These tanks are on order.

Shipping cask. This is the shielded stainless steel cask used to ship the fuel from Hanford to Martin's facility at Quehanna, and it will be furnished by Hanford. In addition to meeting the requirements for safe shipment, provisions will be made for venting, adding a solvent and transferring the strontium carbonate as a slurry. Quick disconnect fittings are to be provided to facilitate the transfer line connections. Connections will also be provided for cooling prior to introducing the solvents into the cask.

Metering tank. The metering tank will have a total capacity of 0.5 gallon. Fittings are to be provided in the bottom of the tank to install standpipes which can be used selectively to establish desired fluid levels in the tank, with the excess liquid returning to the storage tank by gravity. This device will be used to transfer strontium nitrate from the feed solution storage tank to the precipitation vessel in given quantities. Brackets will be attached for wall mounting. The construction is of stainless steel according to the Unfired Pressure Vessel Code. The tank has been ordered.

Precipitation vessel. The precipitation vessel has a three-gallon capacity and provisions for vacuum, vent, feed solution in, precipitate out, heating (during addition of reagents and precipitation), water cooling (prior to transfer of precipitate), and electric motor driven stirrer and a thermocouple installation. Brackets for wall mounting are included. The construction is of stainless steel according to the Unfired Pressure Vessel Code. The vessel has been ordered.

Filter aid filter. The purpose of this filter is to remove the filter aid from the strontium nitrate after dissolution of the strontium carbonate

cake with nitric acid. The type of filter proposed is a dynel fiber-wound unit. The plan is to enclose the filter in a stainless steel container of sufficient volume to contain all of the filter aid used in processing the total 600,000 curies since tests indicate an almost negligible retention of radioactive material. This steel container, in turn, will be placed in a steel drum with concrete shielding surrounding the inner container. This arrangement will be monitored during the process to ensure that the radiation is well within acceptable limits.

Waste filter. This is the same as the filter aid filter. The liquid waste will be treated to precipitate particulate matter which will be removed by the waste filter.

Waste drum. The waste drum will consist of a measured charge of vermiculite placed in a steel drum, with shielding provided as dictated by analysis. Predetermined quantities of the liquid waste will be added to the vermiculite after passing through the waste filter. The drum will be monitored to ensure that the radiation level does not exceed the specified limits.

Process filter. The process filter is used to separate the precipitate from the process fluids after their removal from the precipitation vessel. It will consist of a modified medium porosity alumina extraction thimble located in a stainless steel housing. The unit is to be arranged in such a manner that the thimble may be installed and removed from the housing without breaking any of the process lines. During use, the cover of the housing can be set to one side and an inner retainer and seal ring removed. The filter can then be lifted out and may be transferred directly to the furnace for calcining. A lid will be provided for the filter during calcining to retain dust particles. These operations can be handled directly with the manipulator. Lead shielding will be provided for the filter housing to permit emergency bag-out operations and reduce the general radiation level in the box.

Filtrate tank. This tank is identical to the large transfer tank and is on order.

Furnace. The furnace will be an especially designed unit which will maintain the heat loss to a minimum. The specifications call for an oven temperature of 1450°C for maximum continuous operation. The external oven temperature is not to exceed 125°F in air at 80°F, ambient. The oven is a nine-inch cube. The furnace will be equipped with a program-type controller. Glow bar heating elements have been selected since they are the most adaptable to replacement by remote methods. The disposal plan for the furnace is to remove a side and then remove the individual pieces (ceramic bricks, etc.) for individual handling. Since the furnace is so bulky, it is felt that the knockdown feature is imperative for easy disposal. Vendor bids have been requested.

Ball mill. The ball mill will be a unit especially designed to facilitate hot cell operation. It will consist of a motor and gear box turning a one-gallon stainless steel mill. The mill is to be equipped at the top with a plug-valve arrangement which swings to one side for loading, and a ball discharge valve with a screen arrangement located opposite the loading valve. The mill drive is equipped with an overrunning clutch to permit the jar to be positioned by use of the manipulators for loading and discharging. A locking pin is to be included to lock the mill in position for loading and discharging, and it is to be equipped with a microswitch to prevent accidental application of power with the mill in the locked condition. The mill will turn at 62 rpm. The order has been placed for the mill.

Balance. The balance selected is a standard, low height, stainless steel, triple-beam balance. It has already been delivered to Quehanna. The maximum rated error for any measurement is 0.1 gram.

Welding fixture. The welding fixture is designed to weld the fuel capsule. A pressure dome enclosing the torch and capsule will permit evacuation of the chamber and backfilling with helium (to 30 psig). The helium environment will produce a better weld, and in addition, the helium that is trapped in the capsule will permit a helium leak test on the weld. The torch will be either located in the welding position or pivoted to the loading position, as determined by spring-loaded detents. The electrode may be adjusted vertically or changed by manipulators. The capsule is to be rotated under the torch by a Graham variable speed drive, which can be set for speeds ranging from 0 to 41 rpm. Water cooling is provided for both the torch and the fixture that holds the fuel capsule. Design and analysis are complete, and bids are being solicited.

Capsule storage and leak test fixture. The capsule storage and leak test fixture is a dual purpose device. First, it will hold the capsule while the pellets are being fabricated. A cooling water jacket will maintain low storage temperatures. After the capsule top has been welded in place, the fixture may be evacuated and used as a helium leak test chamber. A lead shield will surround the fixture to reduce radiation during maintenance operations.

Decontamination cleaner. A 300-watt ultrasonic generator with two cleaning tanks and a tank selector switch has been procured for the decontamination of the fuel capsules. The generator includes a self-tuning feature which assures maximum cleaning action. A green soap and water solution has been selected as the cleaning medium.

Reflux condenser. A reflux condenser is used on the vent from the precipitation vessel to control carryover since the precipitation vessel will be held at 90° to 95°C during the reaction period. It consists

of a simple four-turn coil surrounded by a cooling water jacket. The engineering is complete except for checking and release.

Hydraulic press. The press consists of a frame built from standard structural steel sections, a hydraulic jack, hydraulic power supply, die block, floating punches and ejection system. The jack sits in the bottom of the press with the lower punch protruding through the bottom shelf. The die block is placed over the end of the lower punch, and a measured amount of powder is loaded in the die. The upper punch is inserted and a spacer block is slipped into position for pressing at 5 tsi. To remove the pellet from the die, a holddown device is positioned over the die block and the lower punch is raised, pushing the pellet out of the die. The green pellet is then slipped off onto a tray covered with zirconia sand suitable for sintering. The power supply will be located in front of the cell where the operator will have full control over the pressing operation. Stearic acid is used to lubricate the die and punches before each compaction.

Vacuum pumps. Two vacuum pumps will be used in parallel to supply the required vacuum for liquid transfer, to evacuate the pressure dome on the welding fixture, and to make the roughing evacuation for the helium leak check. The parallel system affords greater reliability.

Vacuum tank. The vacuum tank is identical to the large transfer tank. A moisture drain will be provided in the bottom. The vacuum settings will be determined during the dry runs prior to the start of operations.

Oil trap. A trap will be used to remove the oil and moisture from the vacuum lines prior to their being exhausted through absolute filters.

Caustic scrubber. The vent lines from the various process lines are to be manifolded and they will pass through a caustic scrubber before discharge. The caustic scrubber tank will be constructed of stainless steel. The vent lines are to be directed under the caustic solution. The vent discharge port will be baffled to help stop vapor carryover. Drain and fill provisions are provided for changing the caustic solution by remote methods. The caustic scrubber tank has been ordered.

Moisture trap. The moisture carried by the vent system will be evaluated, and if the need is indicated, a moisture trap will be provided. The reason for keeping the moisture content low is to prevent damage to the filters used on the vent discharge into the containment box.

Valves. All valves will be stainless steel. The plan is to use manual valves where they are readily accessible to the operator. Where the valve is not conveniently located but can be readily serviced (areas

with minimum radiation) solenoid valves will be used. A combination of solenoid valves backed up by manual valves is being considered for the high radiation areas where the valve can be operated by manipulators, but it is felt that the solenoid valve can be used during operation.

Plumbing. All plumbing lines will be stainless steel tubing.

Fittings. The fittings are to be the all-stainless steel Swagelok type. Threaded joints will not be used for sealing.

D. FUEL OPERATIONS*

During this report period the emphasis at Quehanna has been directed toward completion of the cooling loop, construction of a decontamination dry box, repair of various electronic equipment and continued maintenance and repair of the Facility.

A cooling loop for delivering 50° F water, which will remove 10,000 Btu from all the cells, was completed. Figure 25 is a view of the two water chillers which are connected in parallel to the 300-gallon storage tank and the loop itself. Figure 26 shows a typical cell installation. In addition to the inlet and outlet lines for the cold water, this view also shows the emergency connection to the industrial water line. In case both coolers should break down, it will be possible to cool the system with industrial water. There is an air break in this line to prevent contamination from entering the industrial water system.

Figure 27 shows the decontamination dry box at about 95% completion. This box is portable; all solutions will be fed into the box by gravity from solution bottles located on the top shelf. This box was built primarily to decontaminate and repair the slave end of a Model 8 manipulator. An ultrasonic cleaner has been installed in the floor of the box to decontaminate small parts such as manipulator tongs, tools, etc. A monorail and hoist will be installed in the decontamination room to support the manipulator during this operation. The box is of plywood construction with three coats of Amercoat on the inside surface.

Considerable effort was expended on the maintenance of various electronic apparatus. The following list is an example of the type of equipment repaired:

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* W. Stringham.

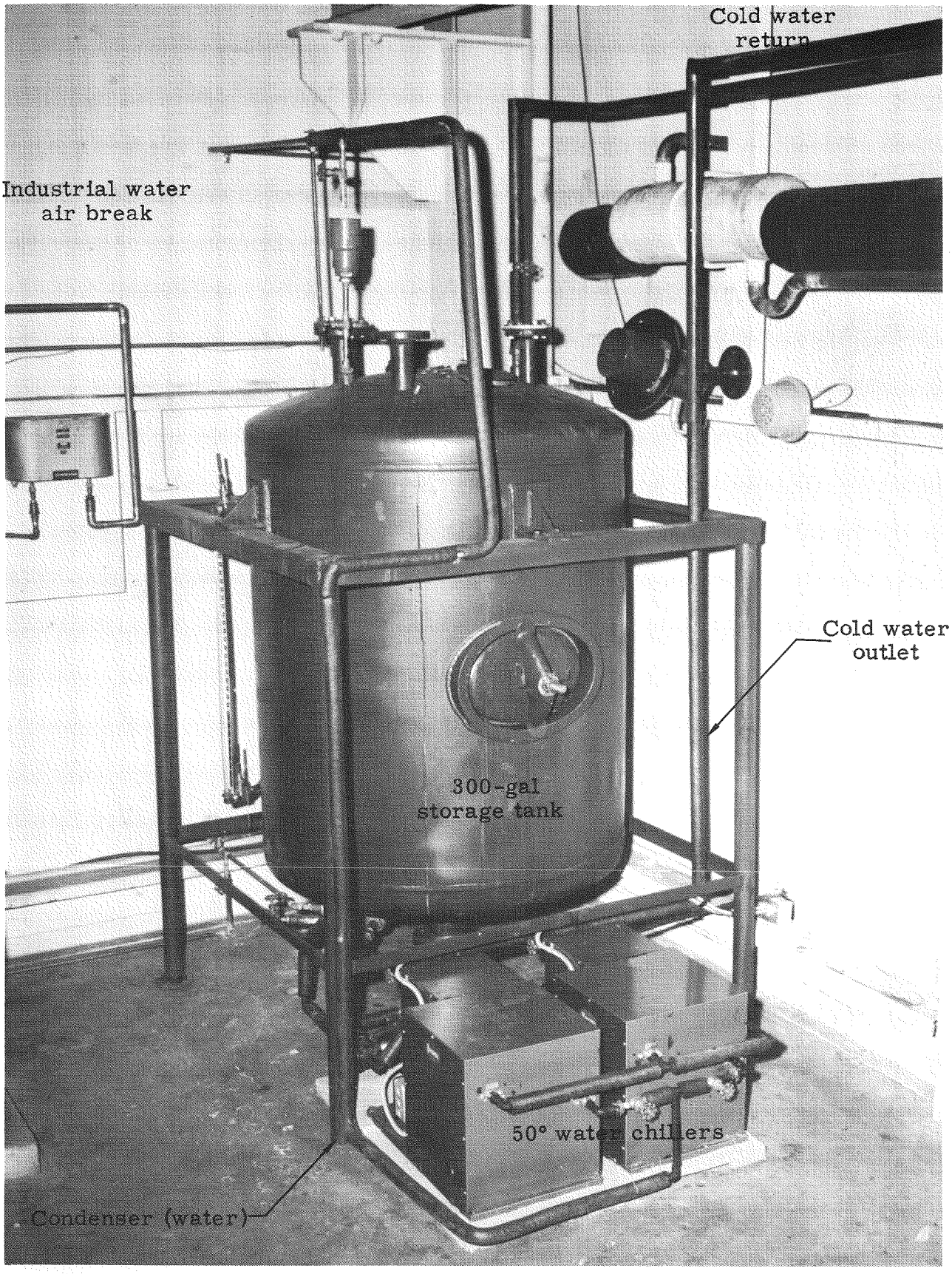

Fig. 25. Cold Water Supply to Hot Cell

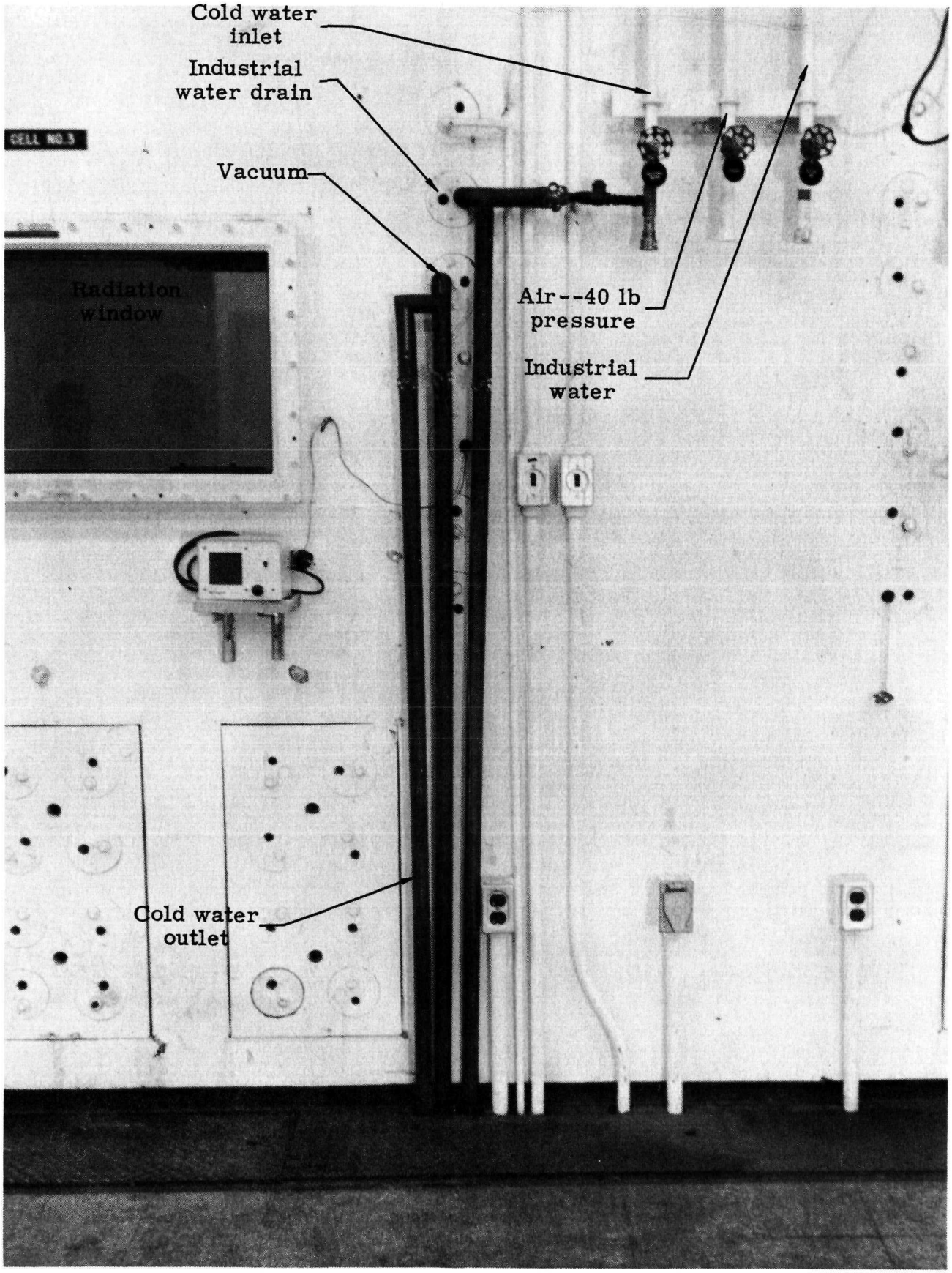

Fig. 26. Typical Cold Water Loop for Cells

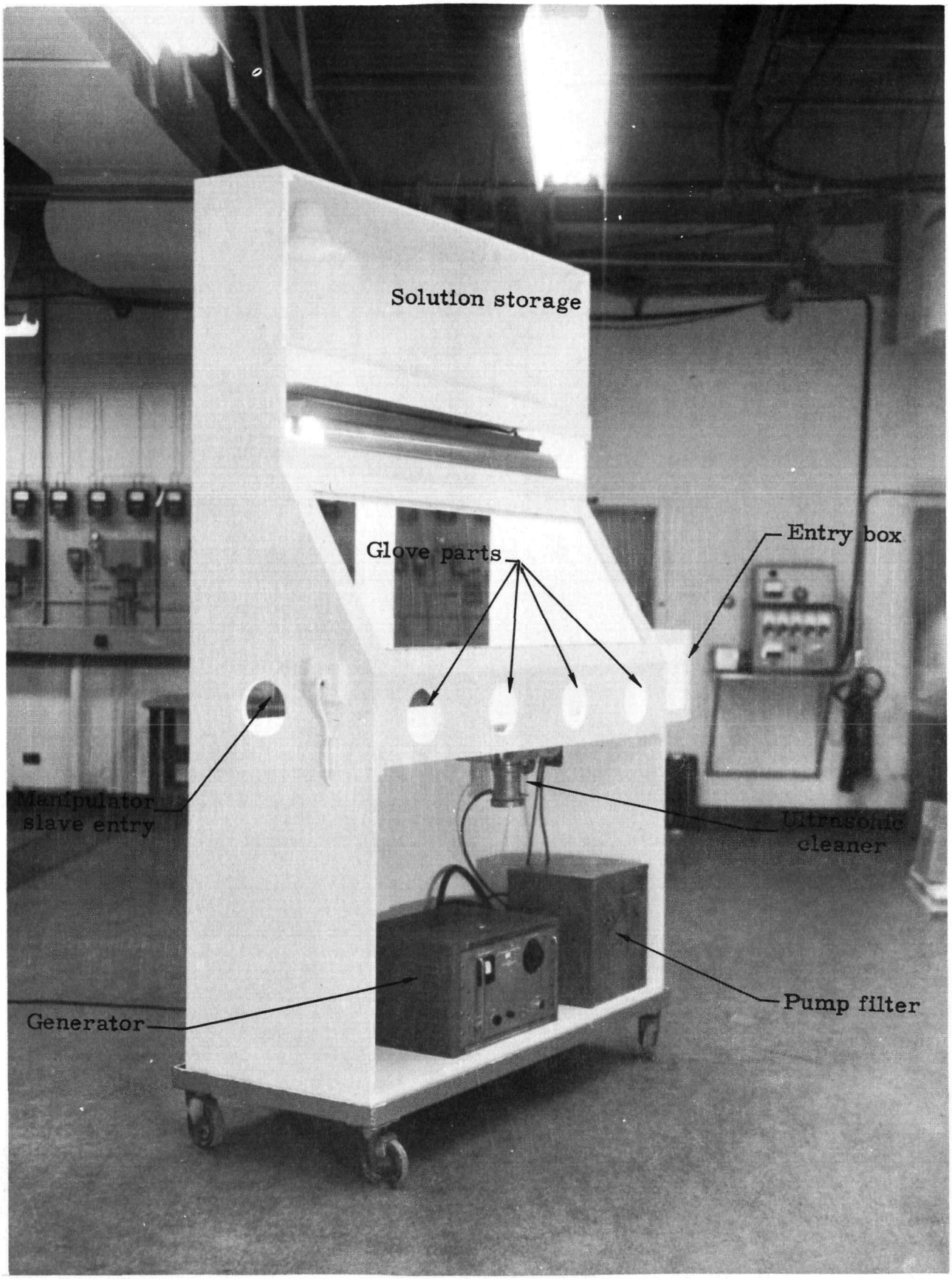

Fig. 27. Decontamination Dry Box

(1) Remote area monitoring system, Jordan Ram II.

(2) Baird Atomic scalers--6 units.

(3) Baird Atomic timers--4 units.

(4) Jordan hand counters.

(5) Tracerlab air monitors--2 units.

(6) Tracerlab hand counters.

Several of the radiation windows in the cells have a precipitate inside the nonbrowning cover plate on the side of the window. This is caused by the degradation of the gasket that is in contact with the oil. New gaskets were ordered, and an elaborate standard operating procedure has been written in preparation for changing the gaskets in all the windows.

The radiochemistry laboratory is equipped with three partially completed dry boxes suitable for analytical determinations in the millicurie range. Parts have been ordered to complete the installation of the boxes.

V. REFERENCES

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4. Giedt, W. H., "Heat Transfer," D. Van Nostrand, 1957, Equation 13.29.

5. Perry, J. H., Editor-in-Chief, "Chemical Engineer's Handbook," McGraw-Hill Book Company, 1950, New York.

6. "Data Telemetry Package Powered by a Strontium-90 Generator," The Martin Company, MND-SR-2428, September 30, 1960.

APPENDIX A

PHOTOGRAPHS OF COMPONENTS AND SUBASSEMBLIES SNAP 7A AND 7C

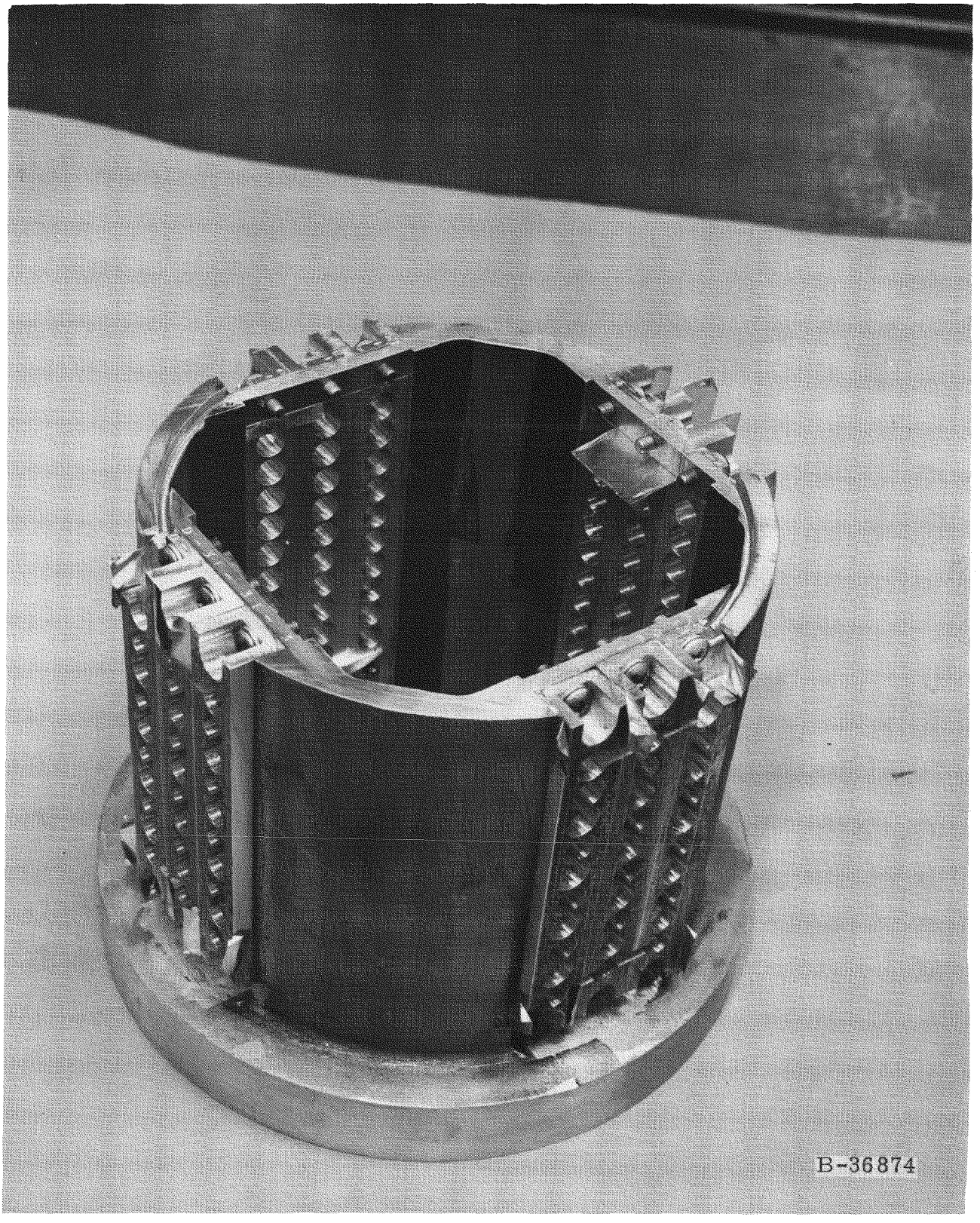

Fig. A-1. SNAP 7A and 7C Prior to Turning (shims compensate for space required for subsequent hardcoat)

Fig. A-2. SNAP 7A and 7C Heat Sink--In-Process Dimensional Check

Fig. A-3. SNAP 7A and 7C Heat Sink After Turning

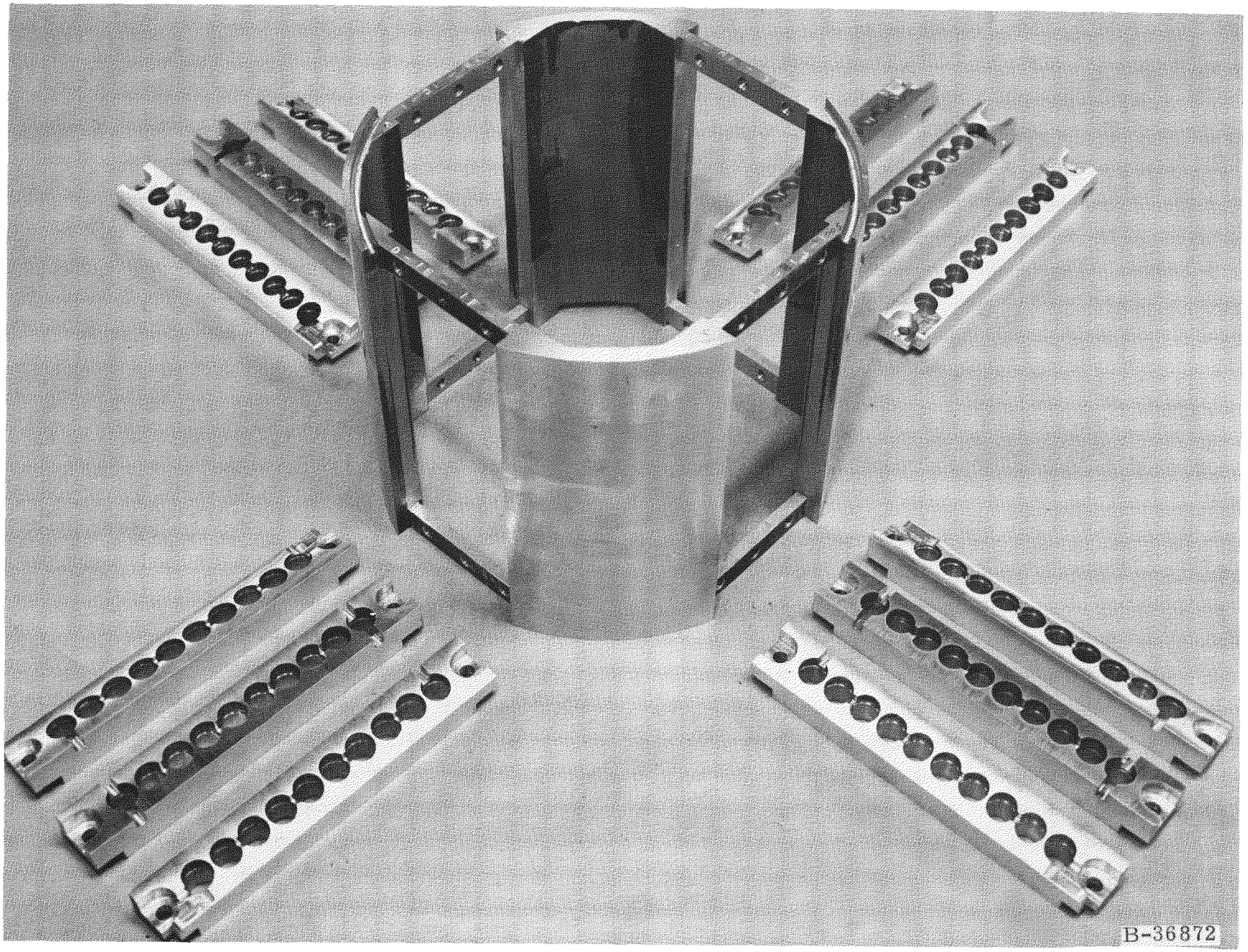

Fig. A-4. SNAP 7A and 7C Heat Sink Disassembled

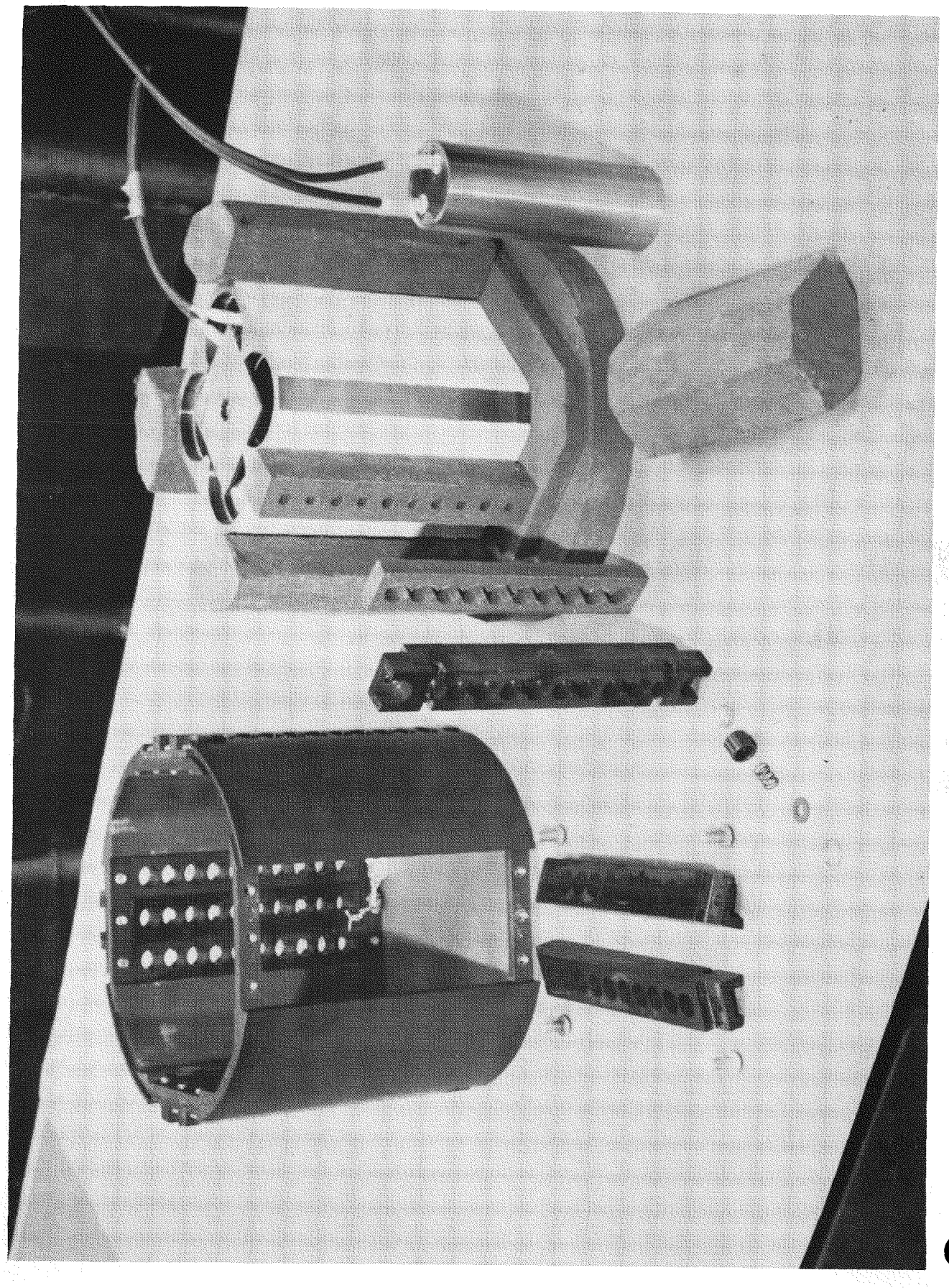

Fig. A-5. SNAP 7--10-w[illegible] Operating Model Components

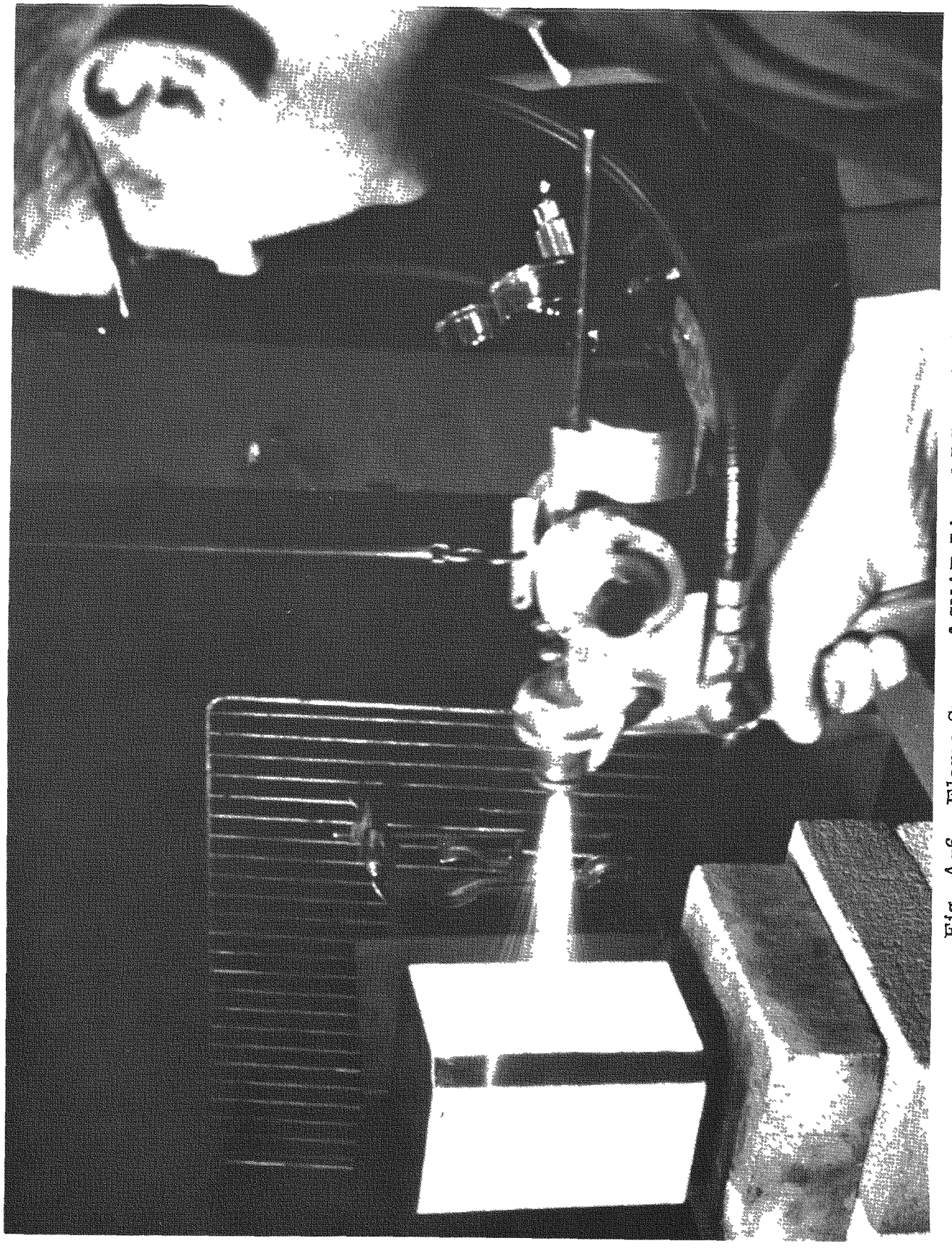

Fig. A-6. Flame Spray of SNAP 7A and 7C Fuel Block

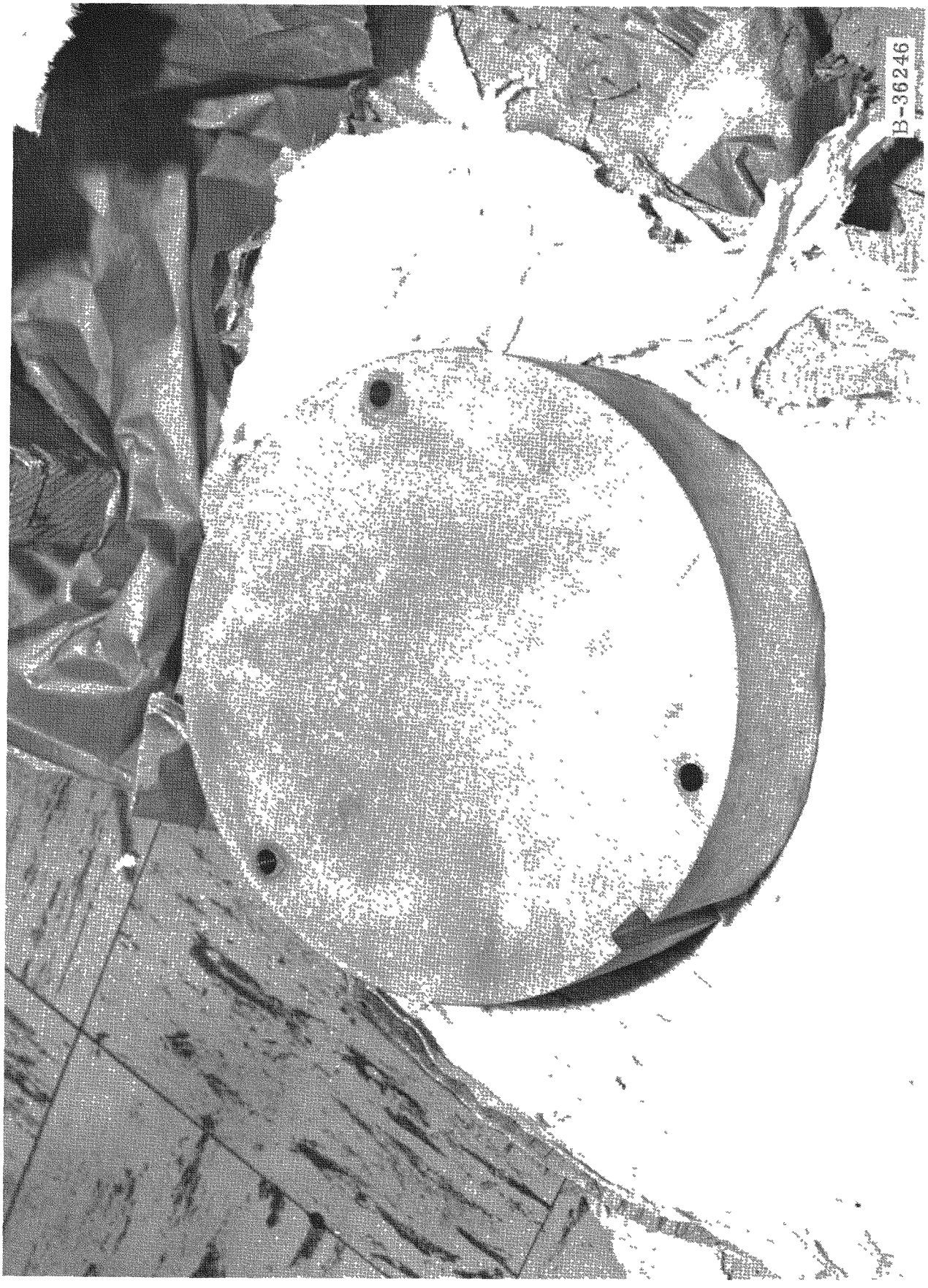

Fig. A-7. SNAP 7A and 7C Depleted Uranium Top Shield Block

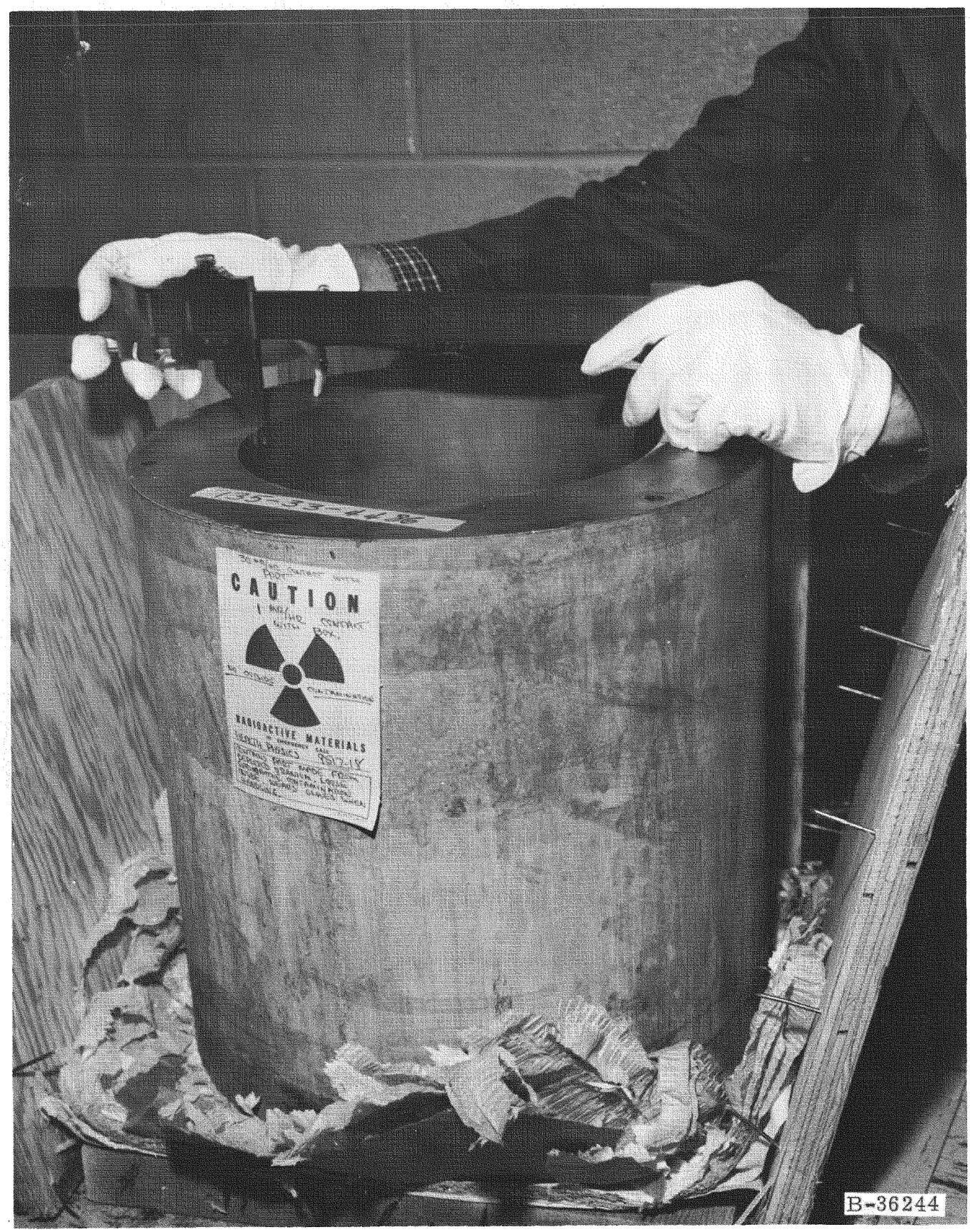

Fig. A-8. SNAP 7A and 7C--Depleted Uranium for Use in Biological Shield

APPENDIX B

THERMAL ANALYSIS OF BURIED CONTAINER SNAP 7C EMPLACEMENT

Figure B-1 is a sketch of the model to be analyzed.

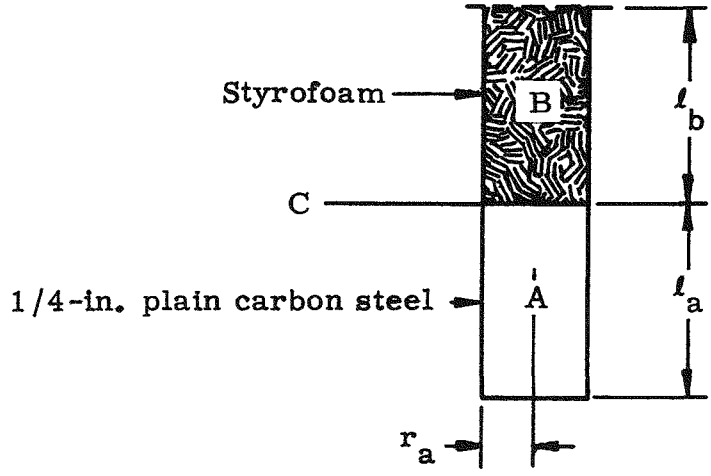

Fig. B-1. Buried Container Model

Section A will be treated as a constant-temperature (T_a) surface, losing heat in all directions below plane C by conduction through the ice. The ice temperature at infinite distance will be taken as a constant (T_∞) at any particular time.

The steady-state heat loss from Section A by this mechanism is then given by the equation

$$Q_a = -k_e A(r) \frac{dT(r)}{dr} \qquad (1)$$

where

Q_a = heat loss from Section A

k_e = thermal conductivity of the ice

$A(r)$ = area of heat flow path at a distance r from equivalent center of heat source.

$T(r)$ = temperature at distance r.

The shape of a constant-temperature surface at distance r will be taken as a hemisphere with center on the intersection of Plane C and the cylinder centerline. This implies that, for the purpose of analysis, Surface A will be treated as a hemisphere with area equal to the cylindrical Surface A.* The variable area A(r) of the surface will then be given by

$$A(r) = 2\pi r^2 \tag{2}$$

Equation (1) then becomes

$$Q_a = -2\pi k_e r^2 \frac{dT(r)}{dr}$$

which becomes, when integrated,

$$\frac{1}{r_2} - \frac{1}{r_1} = \frac{-2\pi k_e}{Q_a} (T_2 - T_1) \tag{3}$$

It is desired to determine the temperature at the surface (T_a) relative to the temperature T_∞ at $r_1 = \infty$. The radius r_2 is chosen equal to the radius of a hemisphere whose surface area is equal to that of the cylindrical Section A, having a radius of r_a and length ℓ_a.

$$2\pi r_2^2 = 2\pi r_a \ell_a + \pi r_a^2$$

$$r_2 = \sqrt{r_a \ell_a + \frac{r_a^2}{2}} \tag{4}$$

Substituting Eq (4) for r_2, $r_1 = \infty$, T_a for T_2 and T_∞ for T_1 gives

$$\frac{1}{\sqrt{r_a \ell_a + \frac{r_a^2}{2}}} = \frac{2\pi k_e}{Q_a} (T_a - T_\infty)$$

$$T_a - T_\infty = \frac{Q_a}{2\pi k_e \sqrt{r_a \ell_a + \frac{r_a^2}{2}}} = \Delta T$$

*This will become apparent later in the analysis.

$$Q_a = 2\pi k_e \sqrt{r_a \ell_a + \frac{r_a^2}{2}} \; (\Delta T) \qquad (5)$$

In Section B of the cylinder, the side wall will act as a fin losing heat to the surroundings according to the relation derived in Ref. B-1.

$$Q_b = \sqrt{UPkA} \; (T_b - T_\infty) \; \frac{(1 - pe^{-2m\ell_b})}{(1 + pe^{-2m\ell_b})} \qquad (6)$$

where

$$p = \frac{km - U}{km + U}$$

$$m = \sqrt{\frac{UP}{kA}}$$

U = heat transfer coefficient from surface

P = perimeter = $2\pi r_b$

A = $2\pi r_b t$

t = thickness of material

k = thermal conductivity

T_b = temperature on root of fin at Plane C.

According to the definition of the problem, $T_b = T_a$. Equation (6) may be rewritten

$$Q_b = \sqrt{UPkA} \; \frac{(1 - pe^{-2m\ell_b})}{(1 + pe^{-2m\ell_b})} \; \Delta T \qquad (7)$$

All quantities in Eq (7) are defined and determinable except the effective surface heat transfer coefficient U. Such a coefficient would be expected to be same in Sections B and A. U is defined in the heat conduction equation as follows.

$$Q = UA\Delta T \tag{8}$$

Substituting into Eq (8) the heat loss for Section A given by Eq (5) and the area A for Section A given by

$$A = 2\pi r_a \ell_a + \pi r_a^2 \tag{8a}$$

and solving for U give

$$U = \frac{k_e}{\sqrt{r_a \ell_a + \frac{r_a^2}{2}}} \tag{9}$$

The total heat production Q_p must equal the heat loss $Q_a + Q_b$

$$Q_p = \left[2\pi k_e \sqrt{r_a \ell_a + \frac{r_a^2}{2}} + \sqrt{UPkA}\ \frac{(1 - pe^{-2m\ell_b})}{(1 + pe^{-2m\ell_b})} \right] \Delta T \tag{10}$$

In order to determine the container surface temperature the temperature rise (ΔT) in the ice pack must be evaluated from Eq (10).

The following geometry is assumed:

r_a = 1.5 ft
ℓ_a = 4.1 ft
ℓ_b = 3.33 ft
k_e = 1.3 Btu/hr-ft-°F
k = 28 Btu/hr-ft-°F.

The heat input to the system at the beginning of life is 256 watts, and the calculated temperature rise in the ice is 33° F. At the end of life the heat input decays to 200 watts and the temperature rise becomes 25.5° F. Reference B-2 indicates a maximum ice temperature for Antarctica as follows:

	Temperature (°F)	
	Maximum	Minimum
At a 1-meter depth	20	-30

Based on these values and the above estimates, the minimum container wall temperature will be -4.5° F. The maximum wall temperature is 53° F, but since this is above the ice freezing point, the above analysis does not hold. In this condition the problem reduces to that of a cylinder submerged in water at 32° F. In summary, the container wall temperature will range from -4.5° F to +36° F, depending on time of year and fuel age.

The generator will transfer heat to the container by both radiation and natural air convection.

$$G_{total} = G_{rad} + G_{conv}$$

$$G_{rad} = h_r A_r \Delta T$$

$$G_{conv} = h_{c_1} A_{gen} \Delta T_1 = h_{c_2} A_{con} \Delta T_2$$

$$\Delta T_1 + \Delta T_2 = \Delta T$$

where

h_r = radiation heat transfer coefficient

h_{c_1} = convection heat transfer coefficient for generator surface

h_{c_2} = convection heat transfer coefficient for inside container walls

A_r = effective radiation area, generator to container

A_{con} = area of inside container wall

A_{gen} = convective surface area of generator

ΔT = temperature difference between generator and container wall

ΔT_1 = temperature difference between generator and surrounding air

ΔT_2 = temperature difference between container wall and enclosed air.

$$G_{conv} = \frac{h_{c_1} h_{c_2} A_{gen} A_{con}}{h_{c_1} A_{gen} + h_{c_2} A_{con}} \Delta T$$

$$G_{total} = \left(h_r A_r + \frac{h_{c_1} h_{c_2} A_{gen} A_{con}}{h_{c_1} A_{gen} + h_{c_2} A_{con}} \right) \Delta T$$

Evaluating this equation for G = 874 Btu/hr (maximum heat input) and for the appropriate geometry results in

$$\Delta T = 73.5^\circ F$$

The maximum surface temperature of the generator is then 73.5 + 36 = 109.5° F. At the end of life the total temperature difference will be 57.3° F and the minimum generator surface temperature will be approximately 53° F.

Air temperature within the steel container housing will range from a maximum of 60° F to a minimum of 20° F.

APPENDIX C

SHIELDING KILOCURIE AMOUNTS OF STRONTIUM-90*

I. INTRODUCTION

Strontium-90 is one of the radioactive isotopes used to generate heat for small auxiliary power systems. Kilocurie amounts of this isotope are required to produce several watts of heat. Since the decay sequence of Strontium-90 contains no nuclear gamma radiation, it would be easy to believe that no shielding is required. However, bremsstrahlung X-rays are present, and shielding must be provided for them. Most of the bremsstrahlung are generated when the beta rays are slowed down in the compound or mixture of which the fuel pellet is made. A smaller number are generated by the betas which escape the pellet and are slowed down in the cladding material.

Bremsstrahlung from Strontium-Yttrium-90 are usually measured by using small sources in the microcurie and millicurie range. These results invariably stress the large quantity of low energy gamma rays produced but do not give adequate distributions for the high energy end of the spectrum. These results are wholly inadequate for use in designing shields for high kilocurie amounts of Strontium-90. Calculated bremsstrahlung distributions indicate that heavier shielding is required than is indicated by experimental results obtained in measuring microcurie and millicurie amounts of Strontium-90.

To obtain a confirmation of the amount of shielding required for large Strontium-90 sources, Oak Ridge National Laboratory was requested to measure the attenuation by lead absorbers of the radiation from a 1000-curie source of strontium titanate. The purpose of this report is to compare the experimental results with calculated values.

II. BREMSSTRAHLUNG

The total intensity (number of photons times the photon energy) of bremsstrahlung from monoenergetic beta rays in thick targets is given by

$$I = kZE^2$$

---

*A. M. Spamer

where

I = bremsstrahlung intensity

k = constant

Z = atomic number of absorber

E = beta energy (mev).

The spectral distribution of photons is a straight line function with the maximum photon energy equal to the beta ray energy (Ref. C-1). The number of photons at the maximum energy is, however, zero. By equating the total intensity to the area of the triangle formed by the distribution curve and the coordinate axes, the number of photons at zero energy is easily found to be equivalent to 2EkZ. If the photon distribution is divided into energy groups, the average number of photons in each group is equal to the area under the distribution curve bounded by the energy limits of the range, divided by the energy increment.

Beta rays from isotope decay are not emitted monoenergetically but in spectral distributions which vary greatly for different isotopes (Ref. C-2).

If the distribution of betas is known for a particular isotope, it may be broken into energy groups and the photon production for each group found.

By use of the curves in Ref. C-2, the beta distribution of the nominal 2.2-mev beta from the disintegration of Yttrium-90 was found. The results were normalized to have the area under the curve represent the distribution from one Yttrium-90 disintegration. As a check of these results, the average energy of the betas calculated from this curve was found to be 0.876 mev, which compares favorably with the value 0.90 mev given in Ref. C-3.

The beta distribution was divided into 10 equal energy groups; the number of betas in each group per Yttrium-90 disintegration is given in Table C-1. This grouping of betas was then used to calculate bremsstrahlung distribution. The energy grouping for the bremsstrahlung was chosen to be the same as that for the betas. The number of gammas for each group and the number of gammas divided by kZ are listed in Table C-1.

TABLE C-1

Grouped Spectral Distribution of Betas and Bremsstrahlung from Yttrium-90

Energy Group	Number of Betas per Yttrium-90 Disintegration	Number of Gammas ÷ kZ	Number of Gammas per Yttrium-90 Disintegration: As Used k = 0.0007 Z = 26	Number of Gammas per Yttrium-90 Disintegration: Adjusted k = 0.000175 Z = 26
2.2-1.98	0.0068	0.000157	2.97×10^{-6}	7.42×10^{-7}
1.98-1.76	0.0349	0.00143	2.70×10^{-5}	6.75×10^{-6}
1.76-1.54	0.0696	0.00611	1.155×10^{-4}	2.89×10^{-5}
1.54-1.32	0.1013	0.0180	3.404×10^{-4}	8.51×10^{-5}
1.32-1.10	0.1231	0.0425	8.035×10^{-4}	2.09×10^{-4}
1.10-0.88	0.1389	0.0924	1.747×10^{-3}	4.37×10^{-4}
0.88-0.66	0.1482	0.188	3.550×10^{-3}	8.87×10^{-4}
0.66-0.44	0.1469	0.385	7.286×10^{-3}	1.82×10^{-3}
0.44-0.22	0.1308	0.887	1.677×10^{-2}	4.19×10^{-3}
0.22-0	0.0993	3.50	6.612×10^{-2}	1.65×10^{-2}

Published values for the constant k vary widely from 0.4×10^{-3} to 1.1×10^{-3} (Ref. C-1). One theoretical determination gives values one order of magnitude lower. The value 0.7×10^{-3} was used in the calculations presented here.

An effective value of Z = 26 was obtained for strontium titanate by using the following relation, found in Ref. C-1:

$$Z_{eff} = \frac{N_1 Z_1^{\,2} + N_2 Z_2^{\,2} + N_3 Z_3^{\,2} + \ldots}{N_1 Z_1 + N_2 Z_2 + N_3 Z_3 + \ldots}$$

where N_1, N_2, N_3, . . . are the atoms per cm^3 of the mixture having atomic numbers Z_1, Z_2, Z_3, . . .

The total intensity of bremsstrahlung from a distribution of beta energies is expressed by

$$I = kZ(E_{rms})^2 \doteq \sum_{\substack{\text{all} \\ \text{groups}}} N_i \bar{E}_i$$

or

$$(E_{rms})^2 \doteq \sum_{\substack{\text{all} \\ \text{groups}}} \frac{N_i}{kZ} \bar{E}_i$$

E_{rms} is the root mean square energy of the beta distribution; $\bar{E}_i$ is the average group energy of the bremsstrahlung, and N_i is the number of bremsstrahlung in the group. The root mean square energy of the betas was calculated and found to be 1.015 mev. The sum of the number of gammas, divided by kZ, multiplied by the average energy, was calculated and found to have a value of 1.217 mev. This is about 18% higher than the calculated $(E_{rms})^2$.

Reference C-4 states that the dose rate from one curie of Strontium-90 is about the same as that from 12 mgm of radium, and the average energy of the bremsstrahlung is about 300 kev. The dose rate at one meter from 12 mgm of radium is 12 mr/hr. Using the distribution of bremsstrahlung given in Table C-1, the bare (without self-absorption and shielding) dose rate at one meter from one curie of Strontium-Yttrium-90 was found to be 12.53 mr/hr, and the average energy of the bremsstrahlung was calculated to be 236 kev.

The bremsstrahlung from the 0.5-mev beta of Strontium-90 were not included since they will be in the low kilovolt range and will be attenuated rapidly in the first few mils of shielding.

III. SHIELDING PROGRAM

Dose rates were calculated by means of a generalized shielding program coded for the IBM 709. The source is divided into a number of point sources, and the program calculates the dose rate from each of these points. The program was coded to accomodate up to 400 source points and a maximum of 10 initial source energies. Path lengths through the various materials, along a line joining a point source and the dose point, are found and used to calculate relaxation lengths and buildup for each of the materials between these two points. The individual relaxation lengths are added to obtain the total relaxation length. Buildup along the individual path segments is defined as the infinite medium buildup factor minus one. The total buildup factor along the path from the source to the dose point is assumed to be one plus the sum of the individual buildups. Infinite media buildup factors are approximated by the sum of two exponentials.

The direct energy flux at the dose point is evaluated for each source energy and source position and converted to dose rates by the appropriate flux-to-dose conversion factor. The total dose rate is, of course, equal to the sum of the dose rates from each individual source point.

IV. DESCRIPTION OF EXPERIMENTS*

Dose rates from a 1000-curie source of Strontium-Yttrium-90 were measured by ORNL personnel. The kilocurie of Strontium-90 was contained in 65 grams of titanate powder which had been compacted and sintered to a density of 4.5 grams per cubic centimeter. The only radioactive contamination in the pellet was 305 millicuries of Cerium-144 at the time the measurements were made.

Measurements were made with a Cutie Pie Model 740 (Victoreen Instrument Company) and a survey meter No. 2610A (Nuclear Instrument and Chemical Company) which had been calibrated by the ORNL Health Physics Department, using standard radium gamma sources.

The physical arrangement used when the measurements were made is shown in Fig. C-1. Two sets of measurements were made. Case I was made with the detector located 16-3/8 inches above the pellet when the pellet was shielded with a 1/8-inch Hastelloy C plate and lead plates which varied in increments of 1/2 inch up to 6-1/2 inches. The measurements for Case II were made with the detector 19-1/4 inches above the

*This information was drawn freely from an advance copy of Ref. C-5.

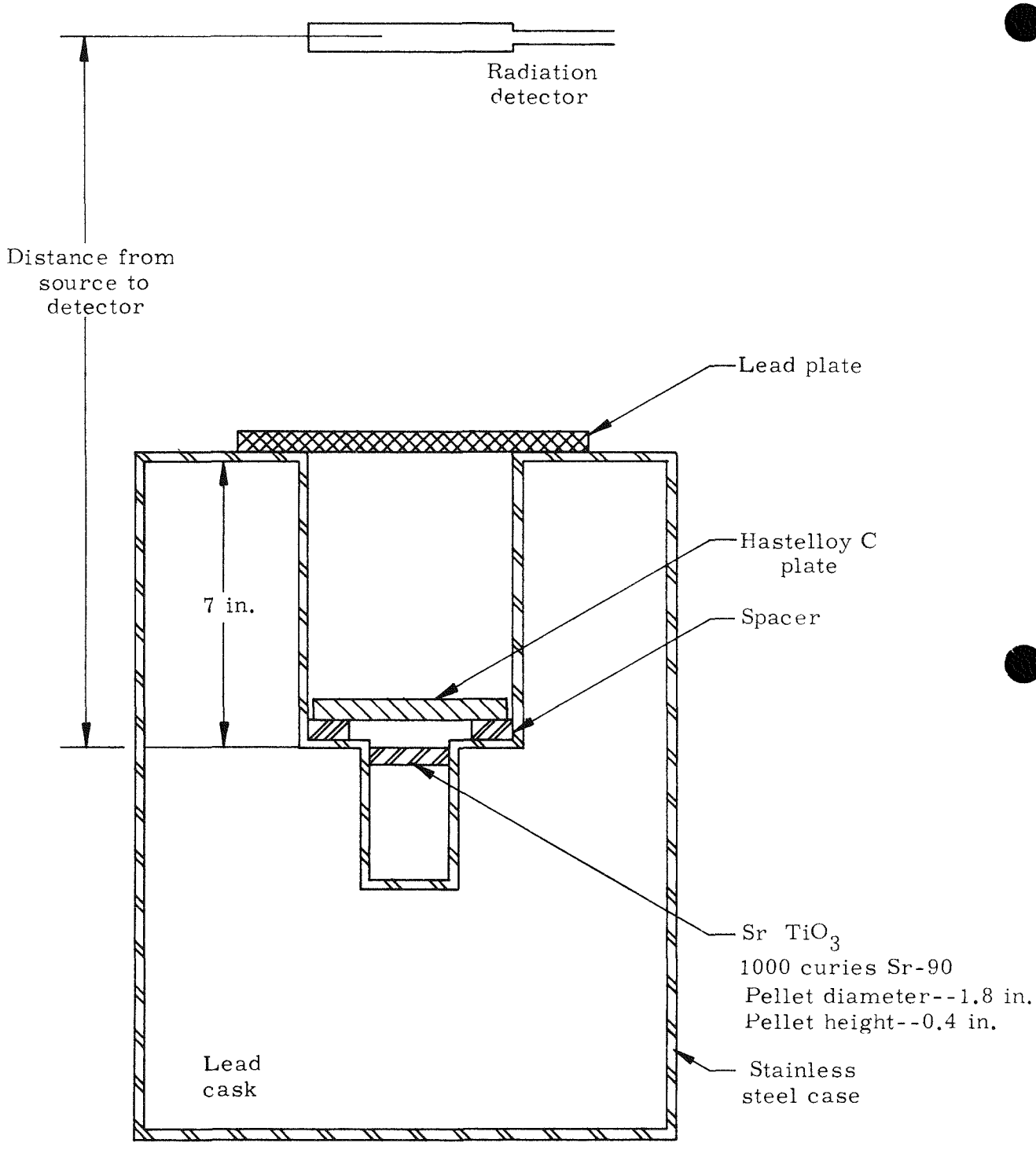

Fig. C-1. Physical Arrangement used in Measuring Dose Rates

pellet when it was shielded with 1/2 inch of Hastelloy C and various thicknesses of lead. The results of these experiments are plotted in Fig. C-5 and C-6.

V. RESULTS OF CALCULATIONS

The accuracy of point-by-point approximations to integrals depends upon the number of points used. To determine the number of points to use to obtain reasonably accuracte results, several problems are run with different numbers of points. Experience has shown that comparatively few points are required to have the third and fourth significant figures in agreement with the results from a greater number of points. To determine the number of source points to use for the present problem, the source was divided into 5, 10, 20 and 40 points. The circular cross section of the fuel pellet was divided into 5 and 10 approximately equal areas, as shown in Fig. C-2 and C-3, and the height was divided into 1, 2 and 4 equal divisions. Calculated results are given in Table C-2. After examining these results, it was decided to run the remainder of the problems with 20 source points. Dose rates versus thickness of lead shielding are tabulated in Table C-3 and are plotted in Figs. C-5 and C-6. Table C-3 also contains the contribution to total dose rate from the 305 millicuries of Cerium-144 present in the strontium pellet. As with Strontium-90, most of the gamma radiation associated with Cerium-144 is bremsstrahlung radiation. The bremsstrahlung radiation from Cerium-144 results from the 2.97-mev beta of Praseodymium-144. The higher energy beta of Praseodymium-144 causes the bremsstrahlung to be more penetrating than the bremsstrahlung of Yttrium-90. If the contribution from Cerium-144 to Strontium-90 dose rates were limited to a percentage of the Strontium-90 dose rate, the allowable amount of cerium would vary with the shield thickness. To illustrate this point, it was assumed that a 10% increase in dose rate would be acceptable. The curies of Cerium-144 per curie of Strontium-90 were calculated for various thicknesses of lead shielding and are plotted in Fig. C-4.

VI. COMPARISON OF RESULTS AND CONCLUSIONS

Examination of Figs. C-5 and C-6 shows that the calculated results are higher than the experimental results by an almost constant amount. Analysis of the data shows the calculated results are higher than the experimental results by a factor of about 4.

This could be caused by either of two items used in the calculations. One of these could be the curie strength, and the second, the product kZ which enters into the bremsstrahlung calculation. The curie strength used should be close to the stated amount since care was exercised to determine the amount of Strontium-90 present in the pellet. When the spread of values given for k is considered (Ref. C-1) it would be safe to assume that the factor kZ used in the calculations is incorrect.

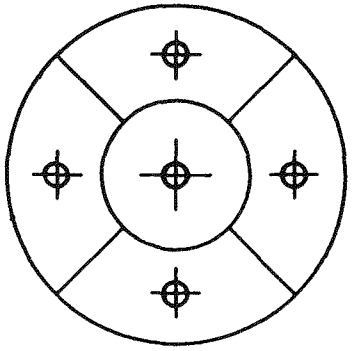

Fig. C-2.
Source Point Configuration--
Five Divisions

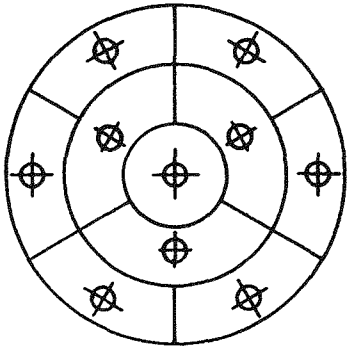

Fig. C-3.
Source Point Configuration--
10 Divisions

TABLE C-2

Comparison of Dose Rates for Various Numbers of Source Points

Number of Divisions in Circular Area	Number of Divisions in Height	Total Number of Source Points	Dose Rate* No Lead (mr/hr)	1/2-in. Lead ($mr/hr \times 10^4$)	5-in. Lead (mr/hr)
5	1	5	38.181	1.0144	6.6837
	2	10	38.318	1.0146	6.6937
	4	20	38.354	1.0146	6.6963
10	1	10	38.175	1.0142	6.6787
	2	20	38.311	1.0143	6.6890
	4	40	38.347	1.0144	6.6914

* Detector--16-3/8 in. from pellet;
Self absorption in pellet and 1/8-in. Hastelloy C.

TABLE C-3

Calculated Dose Rates Versus Thickness of Lead
for Comparison with ORNL Data--

Self Absorption + Hastelloy C + Lead

Thickness of Lead (in.)	Dose Rates (mr/hr) Sr-Y-90* 1000 Curies	Ce-144* 305 Millicuries	Sr-Y-90** 1000 Curies
1/2	1.015×10^{4}	23.6	5.61×10^{3}
1	3.828×10^{3}	12.0	2.08×10^{3}
1-1/2	1.575×10^{3}	6.4	8.47×10^{2}
2	6.81×10^{2}	3.5	3.62×10^{2}
2-1/2	3.03×10^{2}	1.9	1.60×10^{2}
3	1.38×10^{2}	1.045	7.26×10
3-1/2	6.38×10	0.575	3.34×10
4	2.98×10	0.316	1.55×10
5	6.70	0.0952	3.47
6	1.54	0.0285	8.00×10^{-1}
7	3.64×10^{-1}	0.00851	1.87×10^{-1}

* Distance, source to detector = 16-3/8 in.
Hastelloy C thickness = 1/8 in.

** Distance, source to detector = 19-1/4 in.
Hastelloy C thickness = 1/2 in.

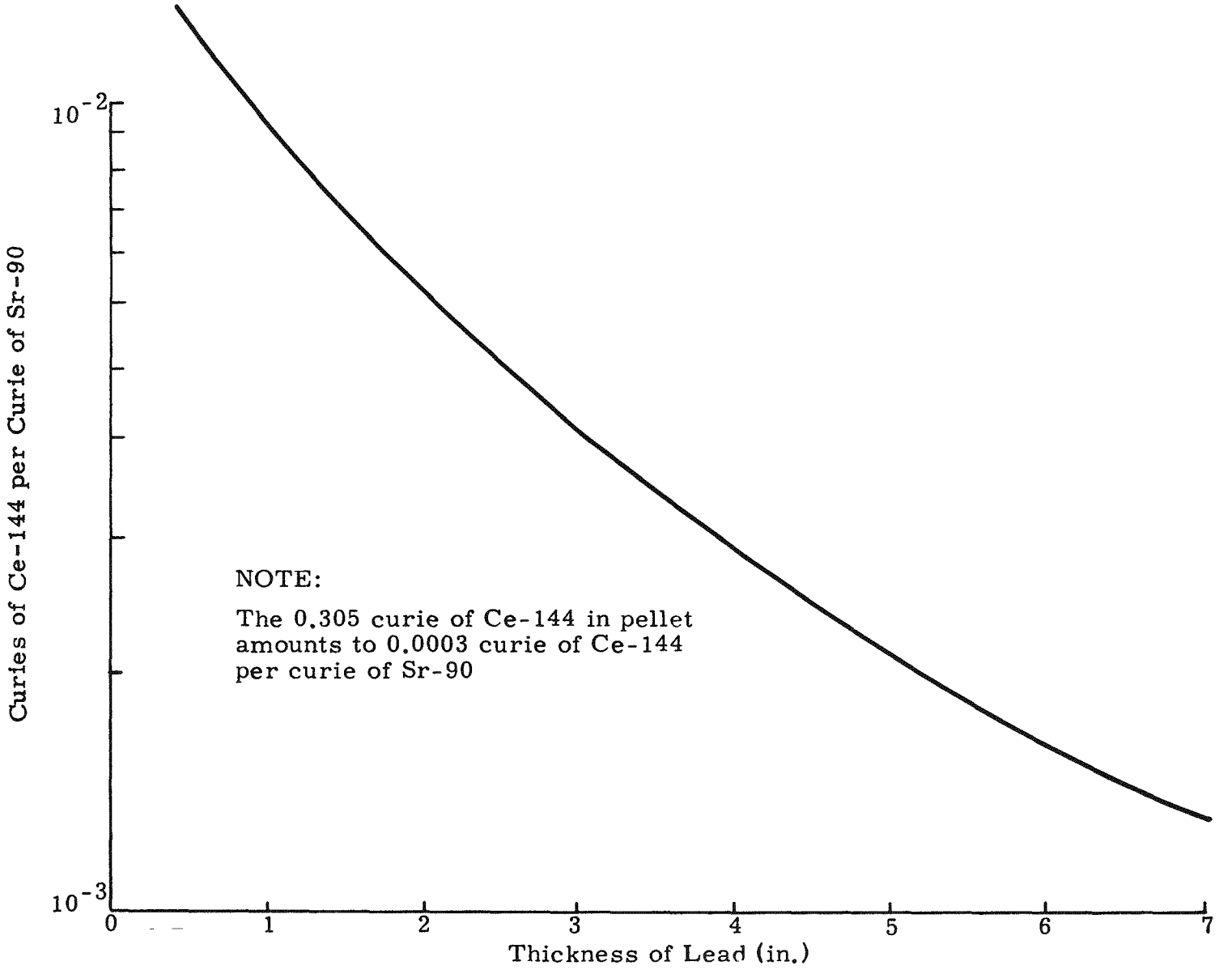

Fig. C-4. Curies of Ce-144 per Curie of Sr-90 Required to Increase Strontium-90 Dose Rate by 10%

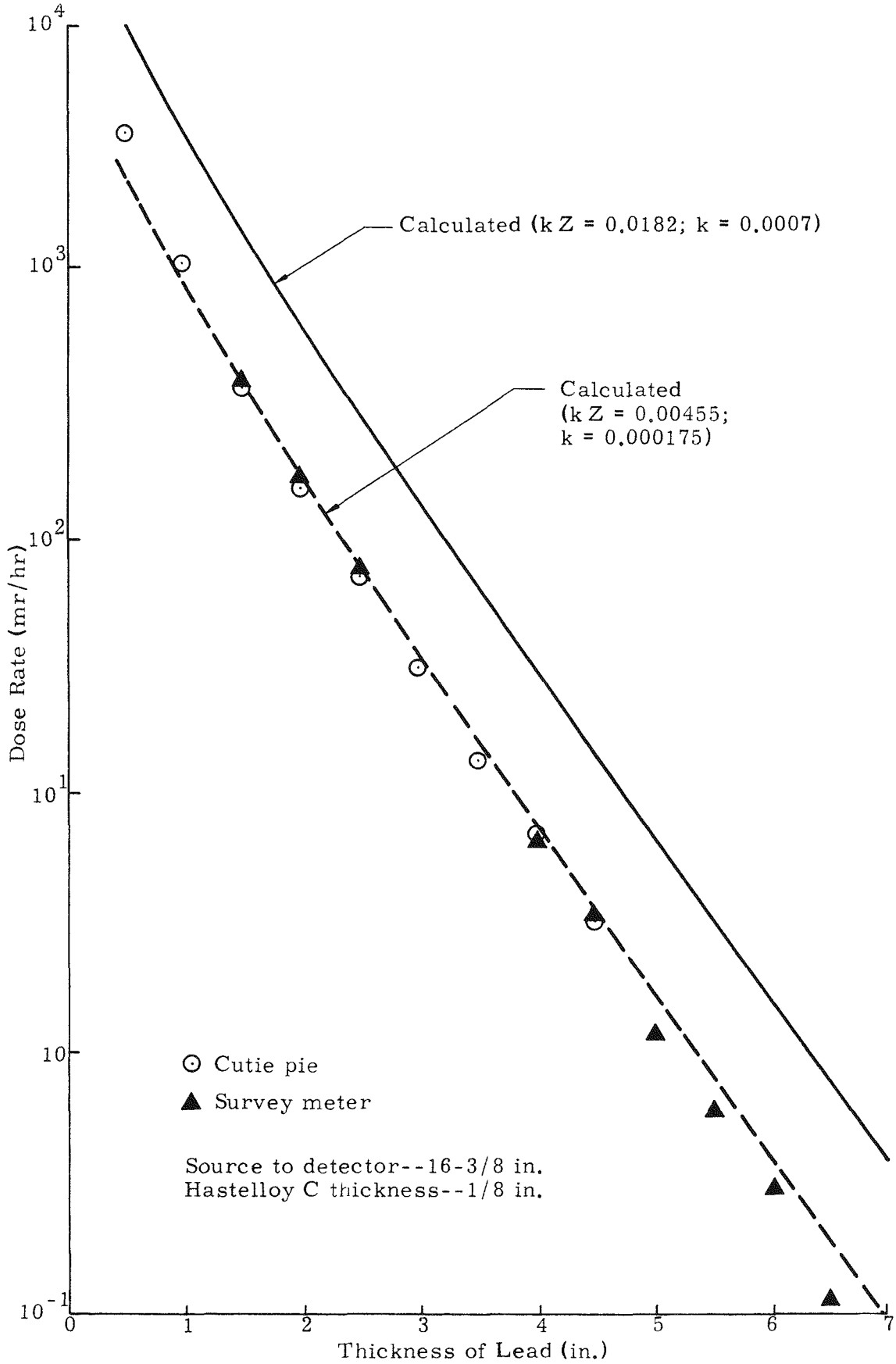

Fig. C-5. Measured and Calculated Dose Rates for Case 1

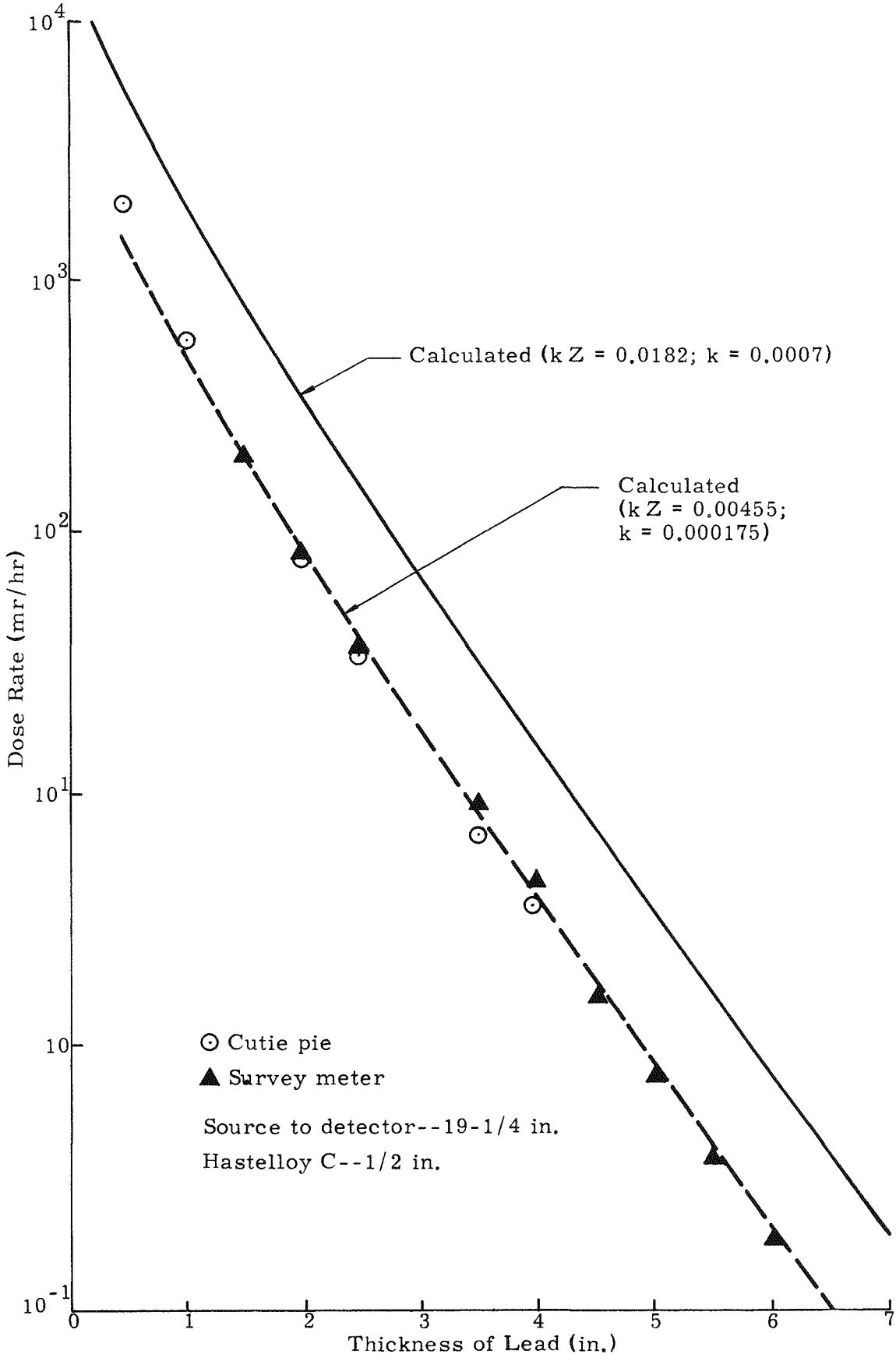

Fig. C-6. Measured and Calculated Dose Rates for Case 2

The originally calculated values were divided by 4 and plotted as the dashed curves in Figs. C-5 and C-6. These curves are in close agreement with the experimental points.

The value of kZ used to make the original calculations is 0.0007 x 26 = 0.0182, and the value which gave the dashed curve is 0.00455. If it is assumed that the calculated effective Z of 26 is correct, the value of k would become 1.75×10^{-4}. The number of gammas per disintegration of Yttrium-90, using the adjusted value of kZ, are tabulated in Table C-1.

The Cerium-144 present in the pellet while the experiments were being performed did not contribute appreciably to the Strontium-90 dose rates. Depending upon the shield thickness, the amount of cerium present in the pellet could be increased by factors between 4 and 40 to have a 10% increase in dose rate.

VII. REFERENCES

C-1. Evans, R. D., "The Atomic Nucleus," McGraw-Hill Book Co., Inc., New York, 1955.

C-2. Marshall, J. H., "How to Figure Shapes of Beta Ray Spectra," Nucleonics, Vol 13, No. 8,p 34, August 1955.

C-3. Marinelli, L. O., Quimby, E. H. and Hine, G. J., Nucleonics, Vol 2, No. 4, p 60, April 1948.

C-4. Haybittle, Phys. in Med. Biol 1, 3:270, 1956

C-5. ORNL-CF-61-1-25 (to be published).

APPENDIX D

SNAP 7A AND 7C ENGINEERING DRAWINGS

Detail	Drawing Number
Fuel capsule cylinder	398-3021001
Fuel capsule end plates	398-3021002
Fuel capsule 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
Plug for flange screw holes	398-3021021

Connector (Hastelloy C and platinum)	398-3021022
Connector mount	398-3021023

Assembly	Drawing Number
Generator--biological shield assembly	398-3021000
Fuel shipping cask assembly	398-3021040
Biological shield	398-3021041
Generator assembly	398-3021042
Generator cover assembly	398-3021043
Fuel shipping cask subassemblies	398-3021044
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

SNAP 7A and 7C Calculated Weights

Section	Weight (lb)
Biological shield (uranium only)	1320
Top shield (uranium)	114
Fuel capsule (one)	3.21
Generator (including top uranium shield)	235
Biological shield assembly	1584
Mercury	21
Total	1840

SNAP 7A and 7C Dimensions

Item	Size (in.)	Height (in.)
Fuel capsule	1.87 OD	5.39
Fuel block	4.36 between flats	5.73
Heat sink	8.75 OD	7.00
Generator shell	9.25 OD	15.38
Biological shield	9.50 ID	--
(cylinder)	16.25 OD	19.75
(over fins)	19.25 OD	20.88 (including lugs)

APPENDIX E

SNAP 7B AND 7D ENGINEERING DRAWINGS

Detail	Drawing Number
Fuel capsule cylinder	398-3021101
Fuel capsule end plates	398-3021102
Fuel capsule assembly	398-3021103
Fuel block	398-3021104
Fuel block plate	398-3021105
Shield cylinder (uranium)	398-3021106
Shield block (uranium)	398-3021107
Insulation strip	398-3021108
Insulation block	398-3021109
Thermoelectric elements	398-3021110
Hot shoe	398-3021111
Cold shoe and contact	398-3021112
Piston	398-3021113
Spring washer	398-3021114
Heat sink frame	398-3021115
Heat sink bar	398-3021116
Insulation corner strip	398-3021117
Insulation block	398-3021118 398-3021119
Wire guide	398-3021120
Plug for flange screw holes	398-3021121

Connector (Hastelloy C and platinum)	398-3021122
Connector mount	398-3021123

Assembly	Drawing Number
Generator--biological shield assembly	398-3021100
Biological shield	398-3021141
Operating model assembly	398-3021142
Generator cover assembly	398-3021143
Hot shoe assembly	398-3021145
Cold shoe assembly	398-3021146
Thermoelectric module assembly	398-3021147
Heat sink assembly	398-3021148
Operating model container assembly	398-3021149

SNAP 7B and 7D Calculated Weights

Section	Weight (lb)
Biological shield (uranium only)	3310
Top shield (uranium)	202
Fuel capsule	5.20
Generator (internal parts)	238
Biological shield and cover	4110
Top shield	202
Total	4550

SNAP 7B and 7D Dimensions

Item	Size (in.)	Height (in.)
Fuel capsule	1.87 OD	8.64
Fuel block	6.19 between flats	18.13
Heat sink	10.75 OD	19.13
Biological shield	10.75 OD	--
(cylinder)	18.75 OD	32.25
(over fins)	21.75 OD	34.50 (including lugs)

APPENDIX F

WET CHEMICAL PROCESS DIRECTIONS

1. Slowly add 10 liters of 2M nitric acid to cask; wait 10 minutes.
2. Transfer to receiving tank.
3. Repeat Steps 1 and 2. Agitate solution in receiving tank for at least 15 minutes.
4. Transfer solution from receiving tank to cooled storage vessel, through the filter, to remove filter aid.
5. Rinse cask with two successive five-liter portions of 5M nitric acid, running each portion through the cask, through the receiving tank and through the filter into the storage vessel.
6. Repeat the rinse using one 10-liter portion of water.
7. Agitate solution in storage vessel for 15 minutes.
8. Remove 1-ml sample for analysis.
9. Transfer four liters of the strontium solution to the precipitation vessel.
10. Prepare a slurry of 165 grams of titanium dioxide in 500 ml of water.
11. Transfer this slurry into the precipitation vessel, rinsing all vessels thoroughly with water.
12. Add 200 ml of 50% sodium hydroxide to the slurry in the precipitation vessel.
13. Heat this solution to at least 90° C.
14. Add 700 ml of 30% sodium carbonate at the rate of 10 ml/min to the hot, rapidly stirred solution.
15. Turn heater off and allow solution to cool to 50° C without external cooling; cool to room temperature with external cooling.

16. Filter and wash precipitate with five 200-ml volumes of 0.1M sodium carbonate and three 200-ml volumes of water.

17. Take 25-ml sample of waste for analysis.

18. Transfer the precipitate into aluminum oxide combustion crucibles and place in furnace.