

325
10-3-62



MARTIN COMPANY
NUCLEAR DIVISION

MARTIN
MARIETTA 

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

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

MND-P-2483-6
UC-33
TID 4500, 17th edition

SNAP 7 PROGRAM
QUARTERLY PROGRESS REPORT NO. 6
Task 8--Strontium-90 Fueled
Thermoelectric Generator Development

February 1 through April 30, 1962

MASTER

Prepared by
W. A. McDonald
Ass't Program Manager

Approved by
W. L. Kershaw
Program Manager

Printed in U. S. A. Charge: \$0.50. Available from U. S. Atomic
Energy Commission, Office of Technical Information Extension, P. O.
Box 1001, Oak Ridge, Tennessee.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

DISTRIBUTION

This report is to be distributed according to Division of Technical Information Extension Category No. UC-33.

FOREWORD

This quarterly report covers the period from February 1 through April 30, 1962. It has been prepared by the Martin Company according to the requirements of Contract AT(30-3)-217, Task 8, with the U. S. Atomic Energy Commission.

CONTENTS

	Page
Legal Notice	ii
Distribution	ii
Forward.	iii
I. Introduction and Summary	1
II. SNAP 7A and 7C Five-Watt Electric Generation Systems .	3
A. Objectives	3
B. Discussion of Achievements.	3
III. SNAP 7B and 7D Thirty-Watt Electric Generation System .	7
A. Objectives	7
B. Discussion of Achievements.	7
IV. Strontium-90 Fuel Processing	21
A. Objectives	21
B. Discussion of Achievements.	21
V. References	23

I. INTRODUCTION AND SUMMARY

During this quarter the prime effort on this contract has been directed toward:

- (1) Manufacture, assembly and test of the 60-watt thermoelectric generators.
- (2) Procurement of SNAP-7D system components.
- (3) Installation and checkout of the Strontium-90 isotope process equipment.

Some effort was utilized in investigating the unaccounted for decrease in power output from the SNAP 7A system.

Manufacture, assembly and parametric testing of the first 60-watt thermoelectric generator was completed. Some delays were encountered, however, in this phase of the program as a result of manufacturing problems on thermoelectric P-type elements when they were moved from the laboratory to production. Differential thermal expansion damaged some of the thermoelectrics and caused short circuits during the initial attempt to test. Design modifications were made and the first generator now meets design requirements. These problems are not expected to cause a delay in delivering the SNAP 7B and 7D systems.

All of the Sr-90 fuel process equipment was installed and equipment checkout, dry runs and operator training was initiated.

Equipment checkouts and process dry runs have indicated a number of problems that were not anticipated during the design phase. However, subsequent dry runs indicate that satisfactory solutions are being found. The problems will probably cause a slight delay in hot operation; however, it is expected that schedule commitments in completing the fuel for the SNAP-7B system will be met.

The power output of the Coast Guard SNAP-7A system is decreasing much faster than anticipated. A comparison of power output and hot and cold junction temperature readings with parametric operational data leads to the conclusion that the generator is performing as predicted. A plot of the decay in hot junction temperature (cold junction temperature approximately constant) indicates that the isotope fuel had too much Sr-89 (a short-lived isotope) and, therefore, too little Sr-90.

A program for restoring the generator to design performance has been proposed to the AEC. Essentially, restoration would consist of changing the high conducting inert gas, helium, to a mixture of helium and argon which has better thermal insulating properties.

II. SNAP 7A AND 7C FIVE-WATT ELECTRIC GENERATION SYSTEMS

A. OBJECTIVES

The major objectives for the quarter ending April 30, 1962, were the continuation of testing of the SNAP 7A-7C reliability model and the maintenance of liaison with the Navy and the Coast Guard for exchange of data on the SNAP 7A and 7C systems.

B. DISCUSSION OF ACHIEVEMENTS

During the 164 days that the SNAP 7A-7C reliability model has been under test, the water-cooled heat sink has ruptured twice. It is believed that the ruptures were caused by internal residual weld stresses. The cooling plate distorted considerably during the repair welding operation; as a result, the cold junction temperature is running approximately 10° F higher than before the repair. (Since the water jacket is peculiar to the reliability model, this problem cannot affect the actual generator.) A 0.25-watt power output decrease is attributed to the increase in the cold junction temperature and a slight change in the external load resistance.

The unit was operated for 14 days at a hot junction temperature of 800° F, after which it was operated for 150 days at a hot junction temperature of 900° F. The average operating conditions at these two temperatures were:

<u>T_{hj}</u> (°F)	<u>T_{cj}</u> (°F)	<u>E_{oc}</u> (volts)	<u>R_i</u> (ohms)	<u>P_o</u> (watts)
800	120	2.22	0.61	1.99
900	130	2.58	0.69	2.35

The Hydro-Catylators mentioned in the last quarterly report (Ref. 1) have been shown conclusively to be virtually ineffective. These devices were employed to prevent the loss of electrolyte from the batteries. Tests show that the electrolyte loss incurred when these "water savers" are used will be no more than 10% less than the loss incurred without them. The Hydro-Catylators were not included in the delivered equipment and are not recommended for future use.

During this report period, the hot junction temperature and power output of the SNAP 7A system showed an unexpectedly high rate of decrease with time. The system was analyzed completely in an effort to

determine the cause. Since electrical component failure in the generator would effect a stepwise, rather than steady, decrease in power output, it was eliminated from the list of possible causes. Leakage of air into the generator would not account for the decrease in temperature since the thermal conductivity of air is approximately that of argon and certainly, much lower than that of helium, with which the generator is filled. Therefore, air would cause a temperature rise because of thermal conductivity decrease and thermocouple deterioration.

A third possible cause of the power-temperature decrease was then considered: excess Strontium-89 in the fuel. The slope of semilogarithmic plot of hot junction temperature versus time (assuming constant 40° F water temperature) was found to correspond exactly to that of a Strontium-89 decay curve (see Fig. 1). Accordingly, the curves of hot junction temperature and power output were extrapolated further in time, and measurements taken after the initial extrapolation have been found to lie on the curves (Fig. 2). As the extrapolation is extended for periods of more than a few months, the measured hot junction temperature gradually falls below the curve. This is due to the decay of Strontium-90, which was ignored in the extrapolation. The measured power output has tended to rise above the extrapolated curve, partly because of changes in the actual hot junction and cold junction temperatures resulting from the increase in ambient temperature. The power curve extrapolation is less accurate than the hot junction extrapolation since it is based more on design parameters than on performance.

Estimates of the Strontium-89 content were made by extrapolating the decay curve to determine what the hot junction temperature would have been had the fuel contained only Strontium-90. This temperature was found to be approximately 818° F. By examining electrical test data, it was found that, to produce this hot junction temperature with a cold junction roughly corresponding to that of the submerged generator, a heat input of 232 watts would be required. At insertion of the fuel into the generator, the heat input was approximately 270 watts. The Strontium-89 content at the time of fuel insertion was therefore approximately 40 watts; and the content at the time of encapsulation, approximately 80 watts. The contract was understood to call for a Strontium-89 inventory of no more than 40 watts.

Because of the excellent fit with the decay curve and the agreement of later measurements with the extrapolated hot junction temperature and power output curves, it has been definitely concluded that the cause of the aberration in generator performance is an excess of Strontium-89.

The plan for restoring the SNAP 7A performance to normal levels calls for returning the generator to the Martin Company for changing the gas fill from helium to a mixture of helium and argon. The new gas fill would have a lower thermal conductivity, and less heat would flow through the thermally insulated sections of the generator, raising the hot junction temperature and restoring the electrical power output to the desired level. This proposed plan has been submitted to the Commission (Ref. 7).

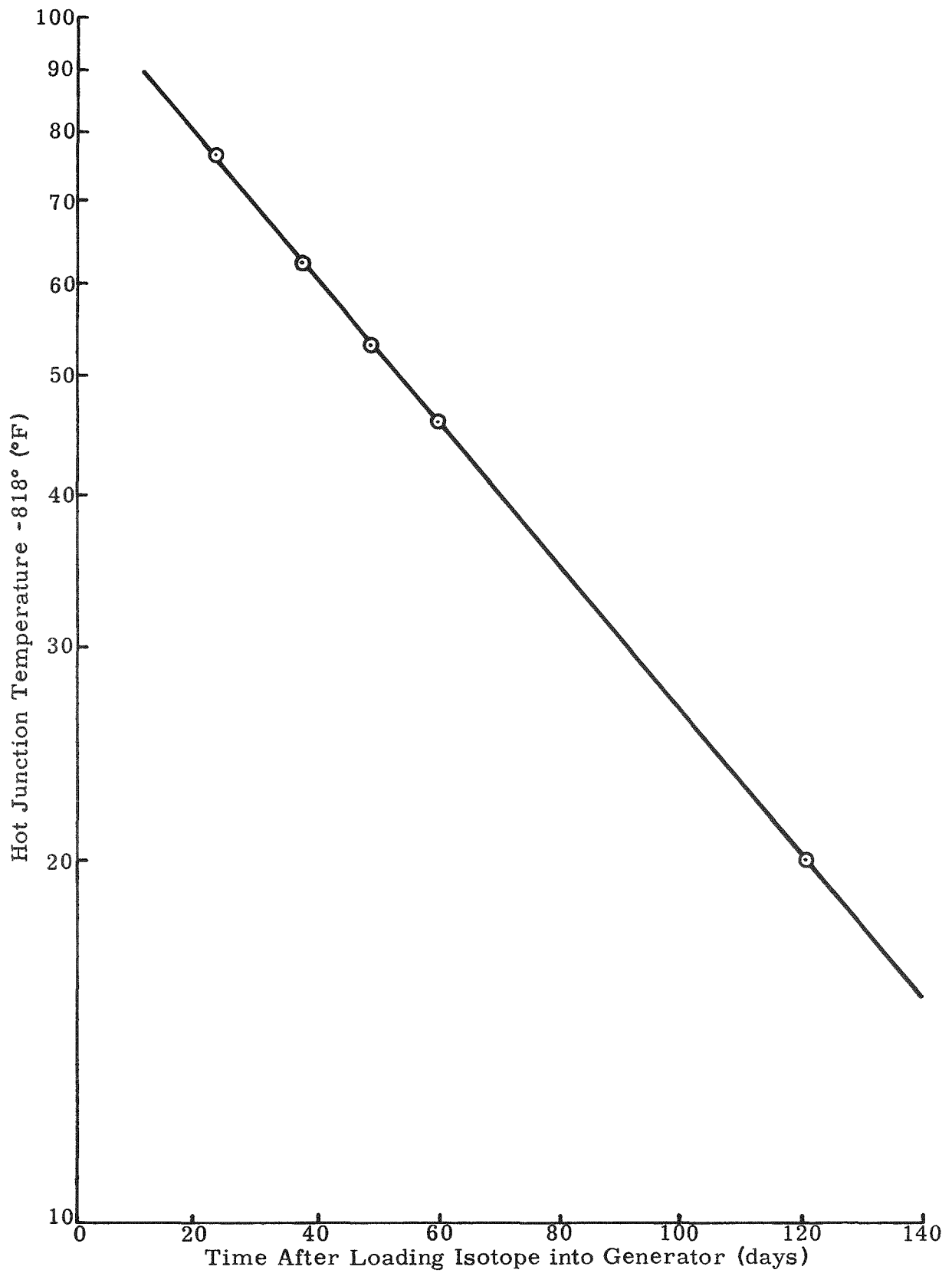


Fig. 1. Determination of Strontium-90 Hot Junction Temperature (ambient water temperature constant)

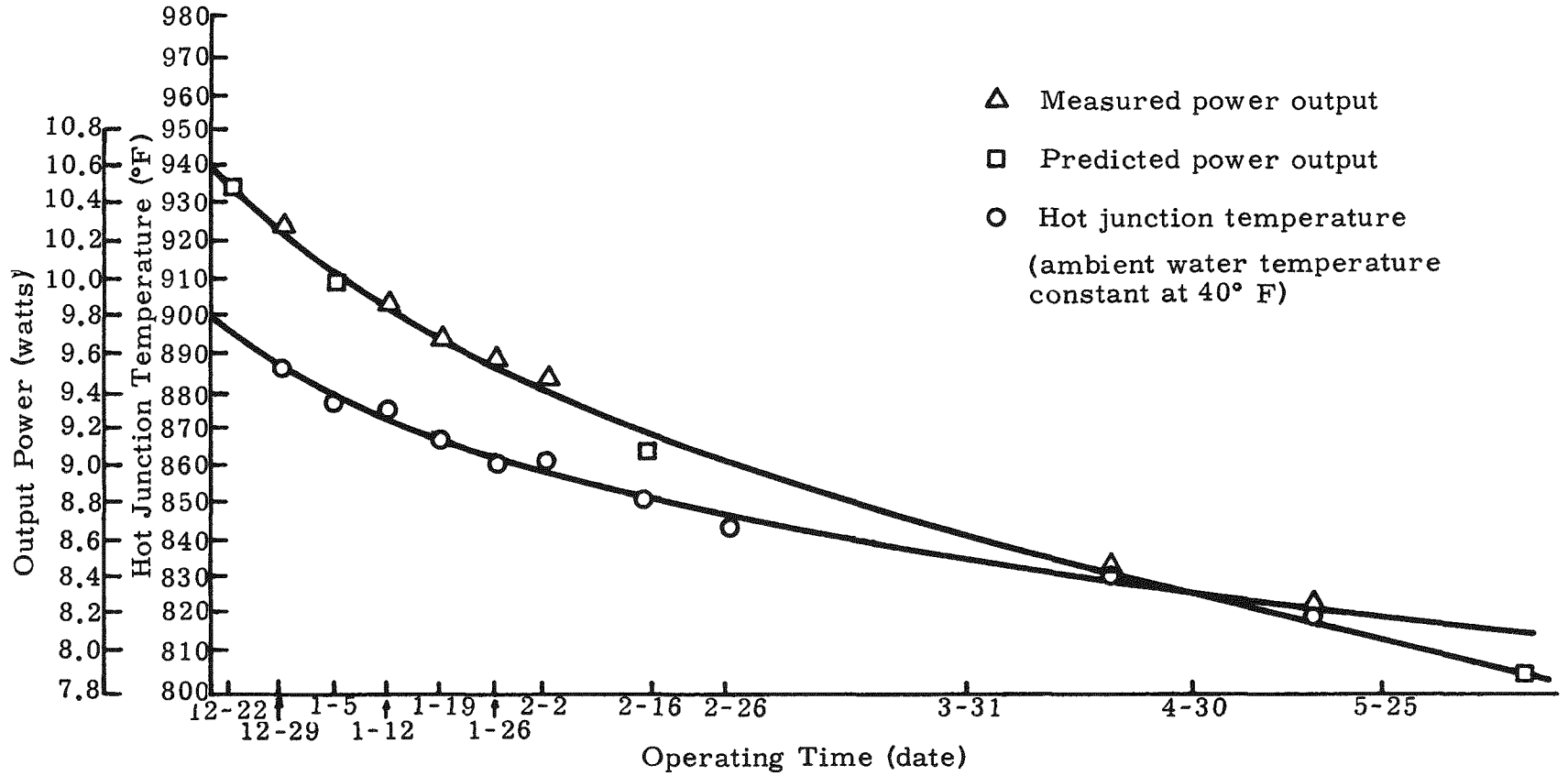


Fig. 2. Operational Data--SNAP 7A Thermoelectric Generator (ambient water temperature constant at 40° F)

III. SNAP 7B AND 7D THIRTY-WATT ELECTRIC GENERATION SYSTEM

A. OBJECTIVES

- (1) To continue reliability model tests.
- (2) To continue evaluation of the all-Martin element couples.
- (3) To assemble the 60-watt operating model.
- (4) To assemble the SNAP 7D generator.
- (5) To procure the SNAP 7D system components.
- (6) To establish SNAP 7B electric system component requirements.

B. DISCUSSION OF ACHIEVEMENTS

The 60-watt reliability model, under test for 110 days, has exhibited no change in electrical properties. The hot junction temperature was increased from 802° to 900° F during the third month of operation. The general performance characteristics obtained during this period were as follows:

T_{hj} (° F)	T_{cj} (° F)	I_{sc} (amps)	E_{oc} (volts)	R_i (ohms)	P_o (watts)
802	132	10.72	2.31	0.224	8.7
900	136	11.48	2.71	--	11.2

The internal resistance at 900° F is not recorded because of erratic readings believed to have resulted from a breakdown of the enamel insulation used on some of the instrument wires. It was decided to continue the test with this condition existing since the remainder of the readings could still be obtained.

In the last quarterly report (Ref. 1), it was stated that the modules in the reliability model would be replaced with Martin P-type elements substituted for Minnesota Mining and Manufacturing Company (3M) P-type elements. It has since been decided not to make this substitution.

In view of the close similarity of materials and fabrication techniques, it is felt that little would be gained. The modules that were fabricated with the 3M elements will be used in the generators.

Three of the 60-watt generator couples with all-Martin elements have been under test for 104 days. There is no apparent change in the electrical characteristics. The elements have been operating with an average hot junction temperature of 970° F.

The first 60-watt generator (Fig. 3) was assembled during the month of March. Thirty modules (Fig. 4) with all-Martin elements were installed. The springs that bear on the elements were inserted as the generator was built up. It was found that the generator cold sink frame (Fig. 5), which retains the module blocks, became barrel-shaped as a result of the force from the compression springs. The deflection was approximately 0.021 inch, and since the design clearance between the generator and container was 0.005 to 0.007 inch, the assembly could not be made. Three special clamping bands were then placed around the generator; the module retaining screws were loosened; and the generator diameter at the midpoint was pulled in. The module retaining screws were retightened, and the clamping bands were removed. The generator could then be readily placed in the container. The generator resistance was monitored during the clamping operation to indicate any damage to the thermoelements.

The generator instrumentation provides for monitoring the temperature of fuel block, hot and cold thermoelectric junctions and heat sink. Other required data can be obtained from the power leads.

Parametric tests of the generator began on March 27, 1962. When the cold generator readings checked out satisfactorily, the generator was evacuated and back filled with an inert gas mixture. As the hot junction temperature approached 350° F, the instrument readings became erratic. When external checks failed to reveal the cause, the generator cover was removed for further examination of the wiring. One of the input power leads was found to have a damaged spot in its insulation. The insulation was repaired and the instrument terminal strip was recleaned to minimize electrical leakage between the terminals. Electrical checks showed the instruments to be normal and revealed no short circuits. The generator was again electrically heated. When the hot junction temperature was a little over 600° F, the troubles reappeared. It was then decided to disassemble the generator for a complete evaluation.

There were signs that the tips of the terminals were shorting against the housing. This condition, caused by insufficient clearance for the radial expansion, was remedied by merely cutting the terminals shorter.

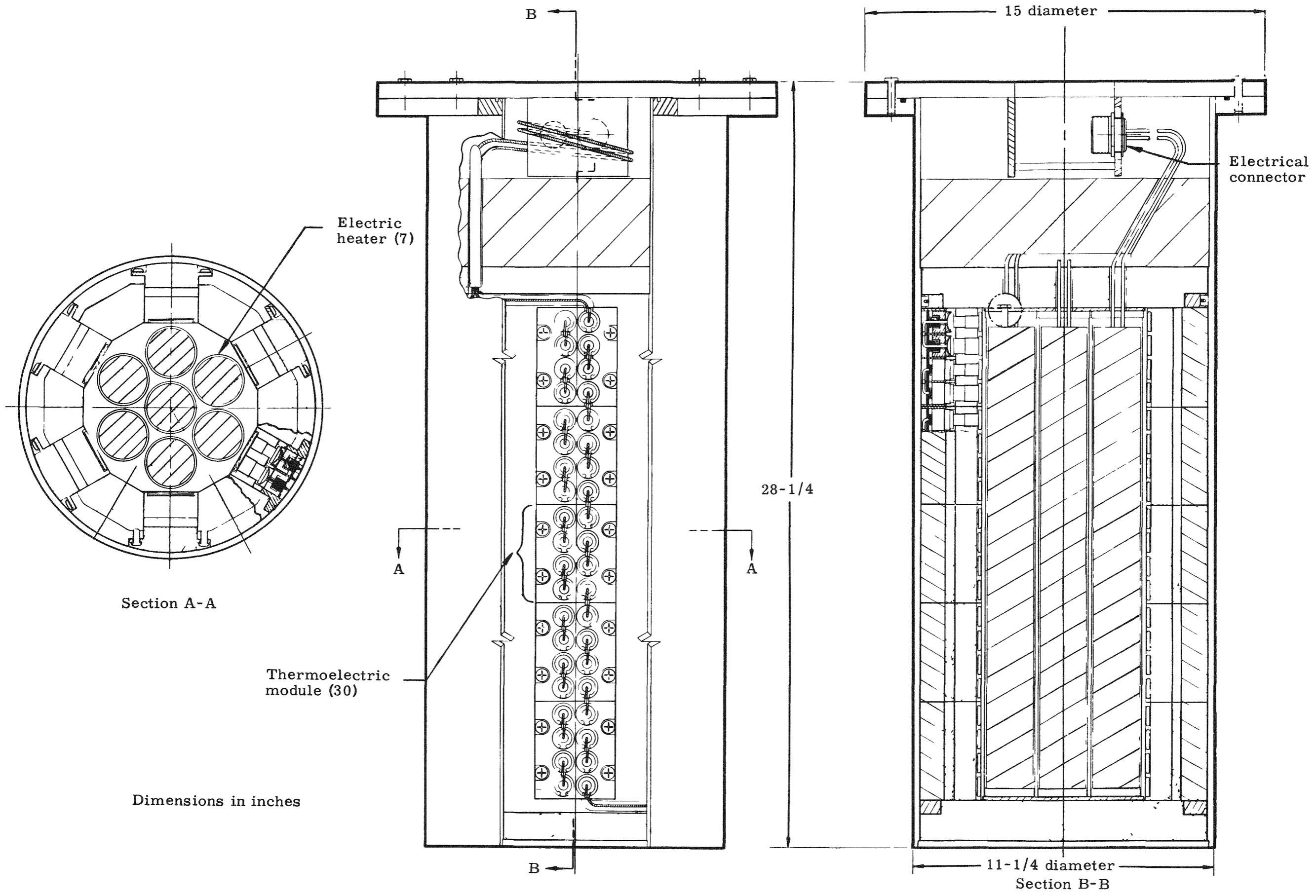


Fig. 3. SNAP 7 60-Watt Operating Model

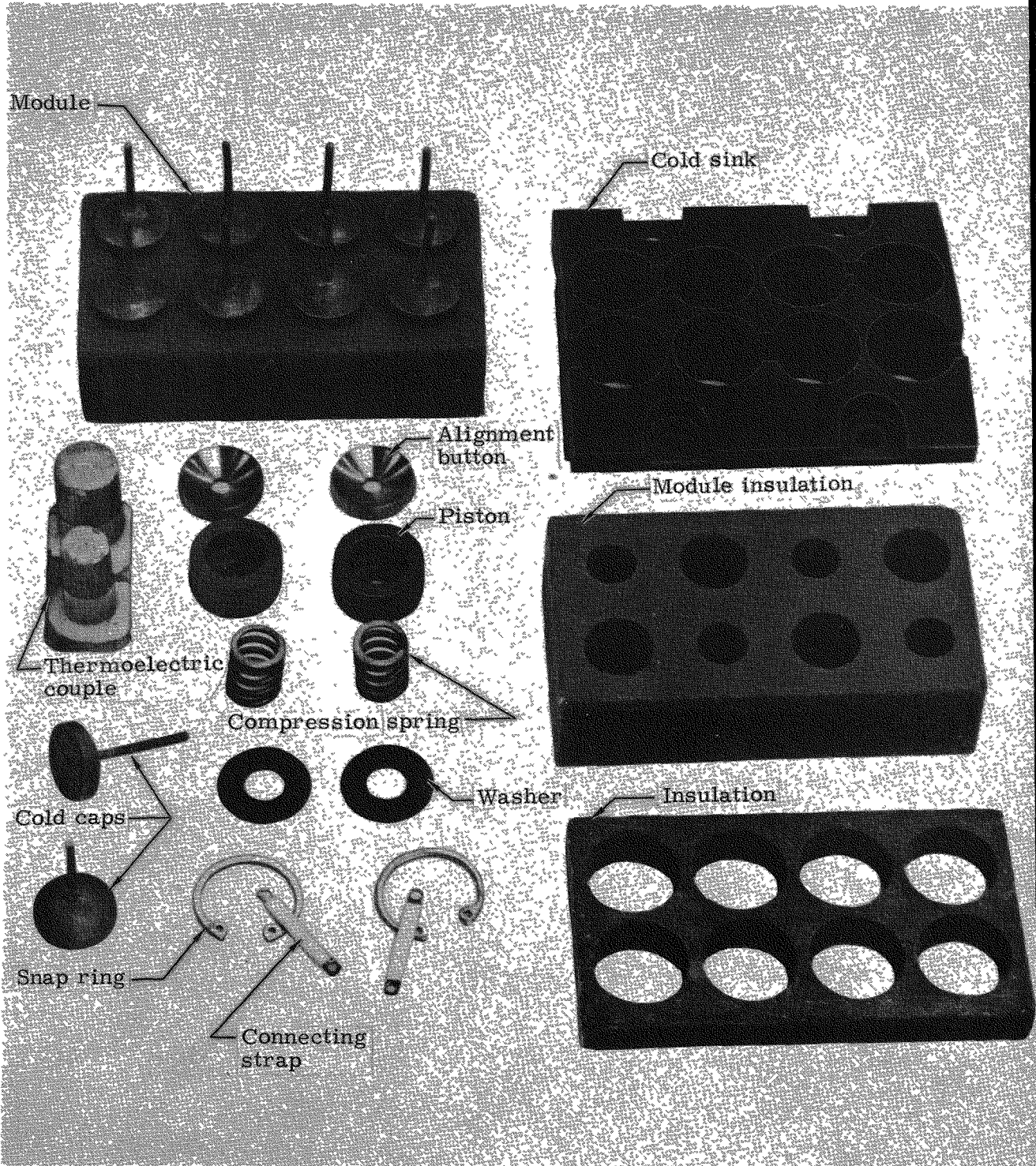


Fig. 4. SNAP 7B-D Thermoelectric Module Components

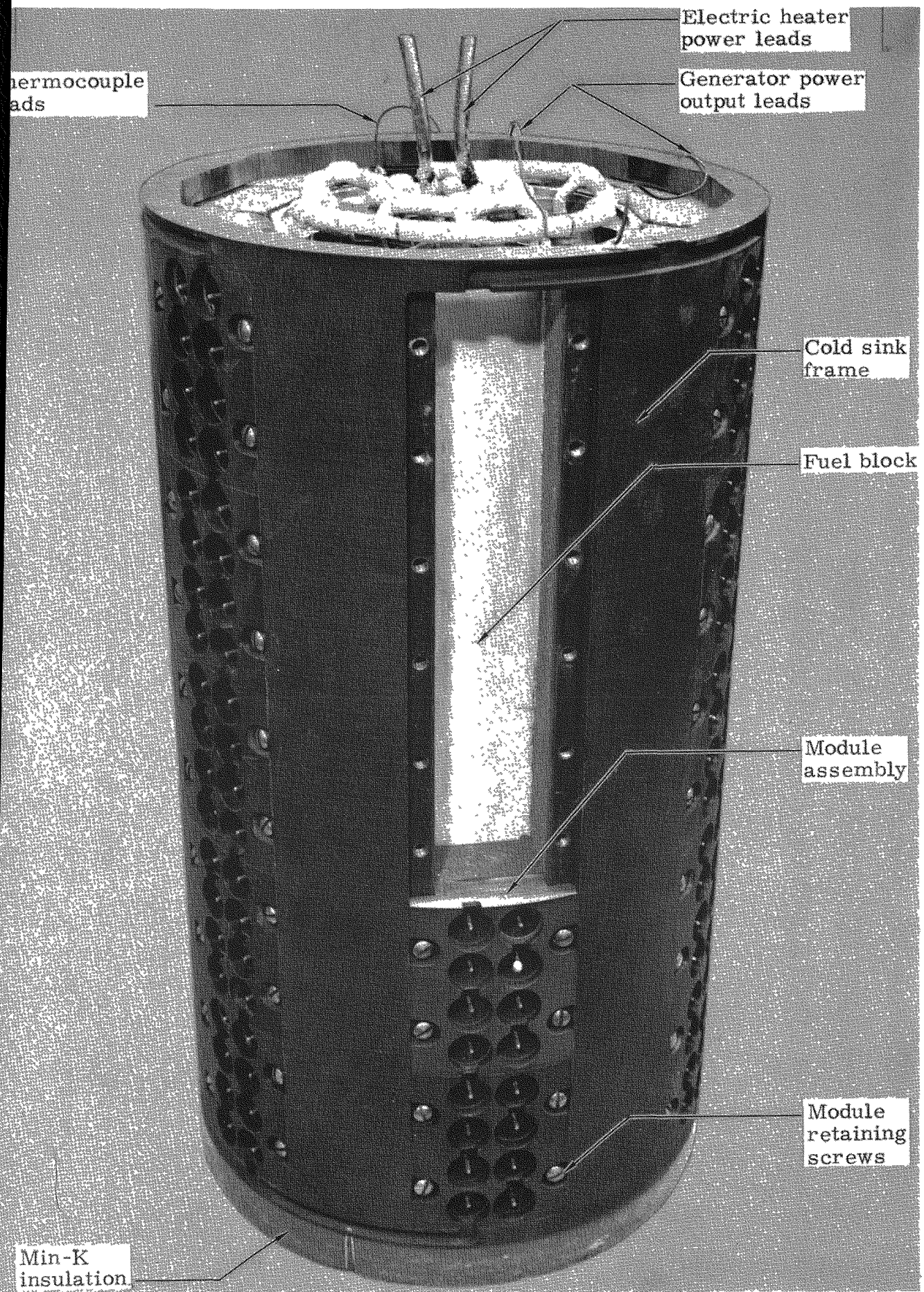


Fig. 5. SNAP 7B-D Thermoelectric Modules Assembled in Electrically Heated Generator (60-watt)

It was found on further examination that there were several broken elements. The breakage was attributed to the difference in longitudinal expansion between the fuel block and the cold sink. Tests with a spring-loaded couple were run to determine the frictional force developed before the couple would slide at either the hot junction or the cold junction. The results were as follows:

Cold junction parts (copper on hardcoated aluminum)	5 lb
Hot junction (aluminum oxide on aluminum oxide)	17 lb

Analysis shows that the 5-pound force acting on the cold junction should not cause the elements to fail. The clearance on the cold junction hardware was increased to allow more relative motion between the elements and the cold sink. In some cases, the combination of assembly tolerances and thermal expansion differential may still exceed the cold junction clearance. Therefore, the sliding forces on the hot junction were evaluated. It was found that the 17-pound force, as in the original assembly, would cause a tensile stress of 1444 psi. The maximum permissible tensile stress in lead telluride is approximately 1000 psi. The broken elements found in the generator after heating confirm this analysis.

The fuel block was removed from the generator, and the aluminum oxide coating was ground off. When the generator was reassembled, a strip of mica sheet was inserted between the fuel block and the hot shoes. The mica sheet provided the second electrical insulation barrier and it reduced the sliding force from 17 to 7 pounds per couple, bringing the stress within acceptable limits.

An alternate recommended solution to the element breakage problem was to change the fuel block from Hastelloy C to graphite, Grade ATJ. The use of graphite, with its low coefficient of expansion, would reduce the differential expansion between the fuel block and the cold sink from 0.101 to 0.022 inch. It is acceptable from both the structural and shielding standpoints, and it is thermally superior. Graphite blocks were ordered as a backup measure, but it was decided to proceed with the solution mentioned previously in order to avoid the three-week delay required for obtaining the graphite.

The generator was completely reassembled by April 21, 1962, with the shorter terminals, the mica strips under the hot shoes, and the increased clearance on the cold junction hardware incorporated in the assembly. The generator was immediately moved to the test area and connected to a vacuum system. On April 23, 1962, the generator was

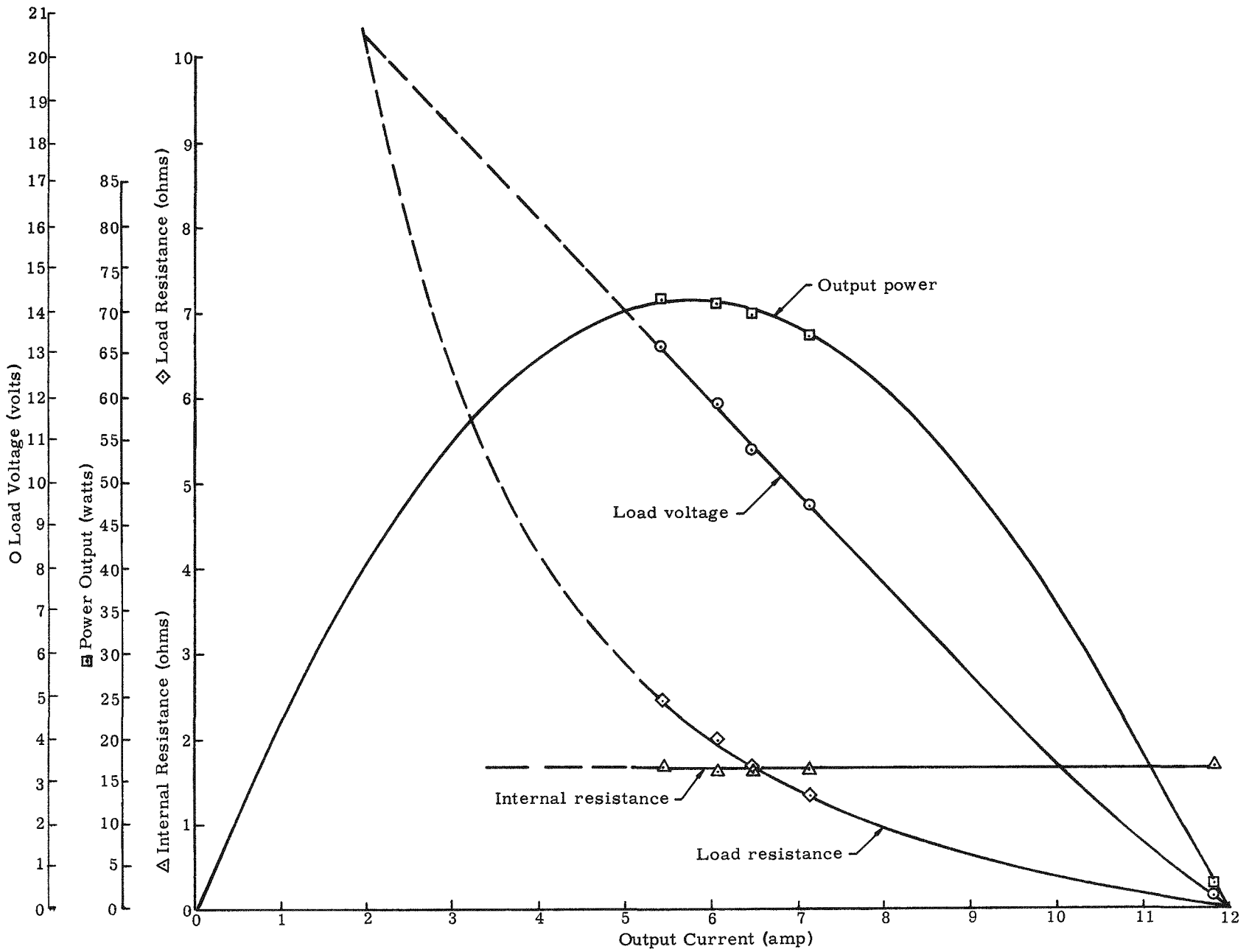
purged with a mixture of helium and argon and then filled with 100% helium. Power was added to the electrical heater at the rate of approximately 100 watts every 2 minutes until the full 1470 watts had been applied. This method simulated the actual fueling conditions and helped determine the time required for the generator to reach a critical temperature. It was found that the generator would reach the 600° F limit (established for the generator prior to sealing and flushing with inert gas) in approximately 4 hours.

Initially, the generator was tested at beginning-of-life conditions. The maximum temperature was established as 960° F at the hot junction, and, with a 100% helium atmosphere, the maximum power input was determined to be 1470 watts. With the power input constant at 1470 watts, the load resistance was varied. The resulting load voltage and output power curves are shown in Fig. 6; the corresponding hot junction and cold junction temperatures are shown in Fig. 7. The maximum measured temperatures are used in each case, rather than the average temperatures.

The fuel is assumed to have a maximum Strontium-89 (plus other contaminants) contribution of 90 watts at the time the generator is fueled (Ref. 2). At the end of life (10 years), the Strontium-89 will have decayed to a point at which it can be disregarded. The remainder of the fuel is assumed to be Strontium-90. At the end of 10 years, the fuel should be producing 1070 watts. This fuel output is just enough to provide the necessary input for the generator, but it does not allow for a loading tolerance. If the power output of the Strontium-89 is below 90 watts, the difference may be used as a loading tolerance.

To allow for losses, the electrical input power was established at 1080 watts for the end-of-life condition. The gas fill in the generator was changed to 100% argon, and the parametric tests were repeated. The results are shown in Fig. 8, and the corresponding hot junction and cold junction temperatures are shown in Fig. 7. In order to achieve the 12 volts required for charging the batteries at the end of 10 years, the system will be operated at 3.5 amperes (42 watts). This is below maximum power but is sufficient to keep the batteries charged, even with only 72% charging efficiency. It is expected that the charging efficiency will be well above 72%.

The second 60-watt generator was approximately 75% complete at the time the first-unit difficulties developed. The assembly was delayed until the results of the modifications on the first generator could be evaluated. The first generator has already passed the electrical tests. The shock and vibration tests will be complete by May 7, 1962. If these are satisfactory, the second unit will be assembled as the first unit was assembled. If there should be a failure (none is expected at this



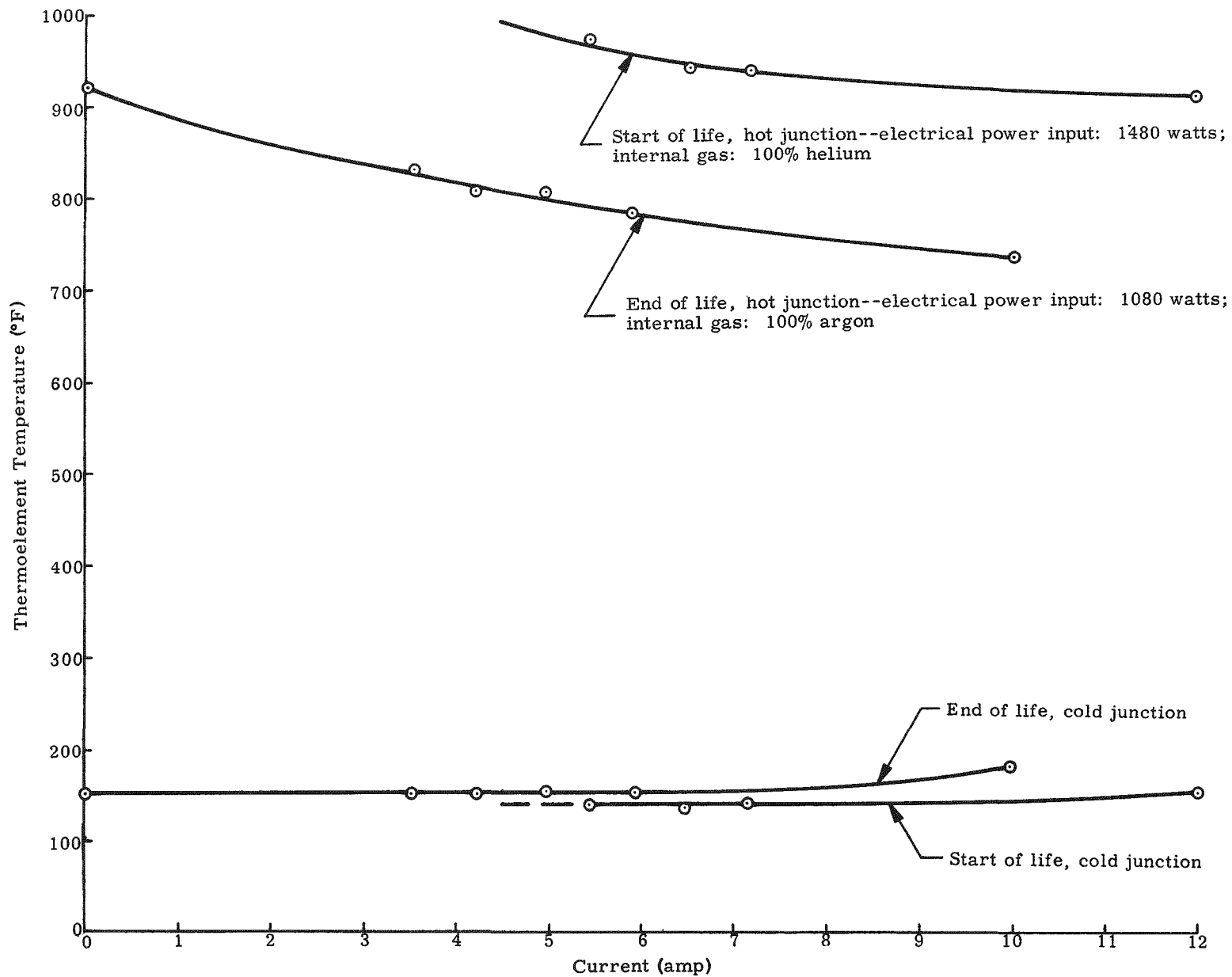
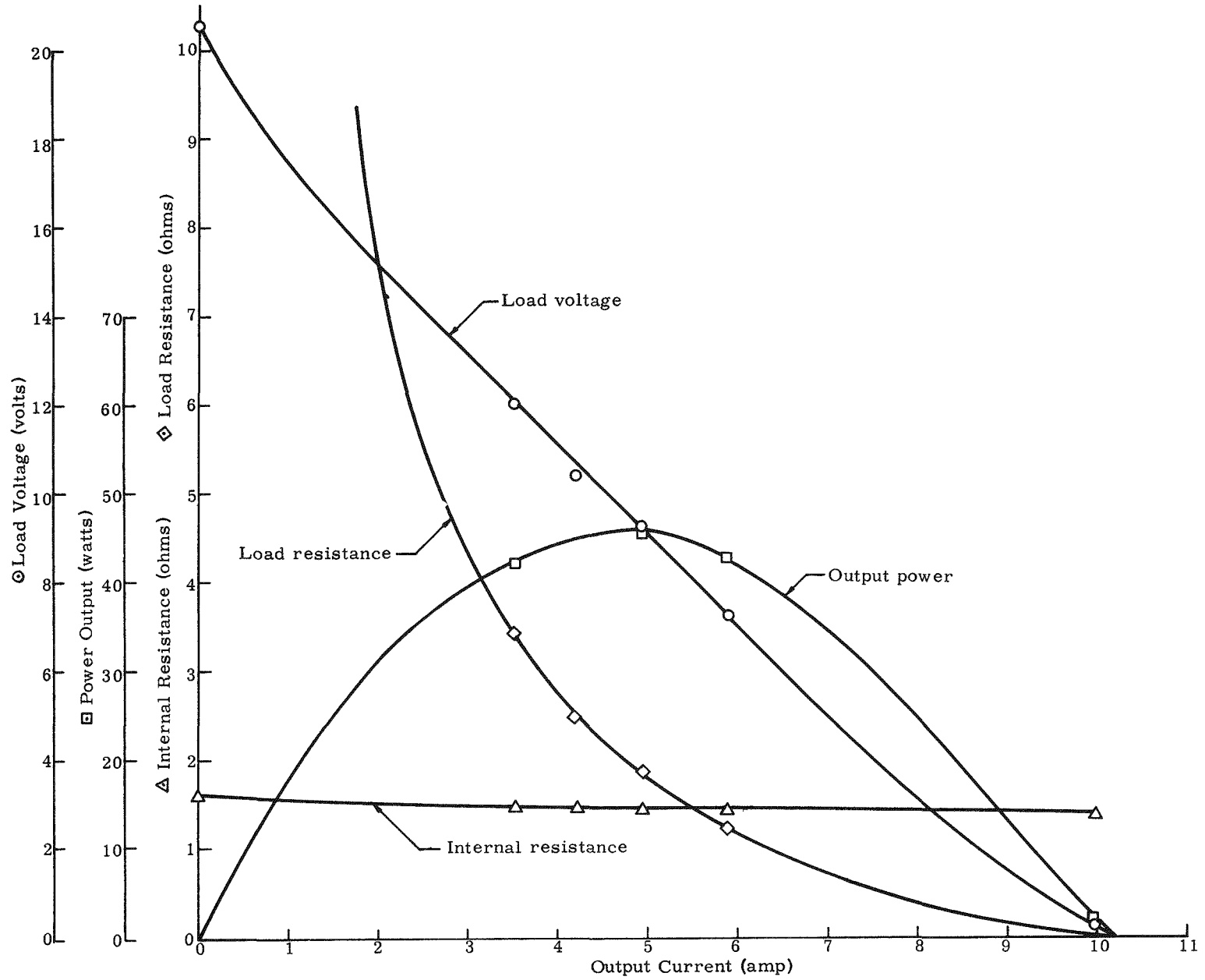


Fig. 7. SNAP 7D 60-Watt Generator--Thermoelement Temperature Versus Current



stage), the alternate plan with the graphite fuel block will be started immediately.

The currently planned SNAP 7D system does not include a converter or voltage regulator although a breadboard converter has been built and tested as a backup, in case the test program should indicate the need for the converter. As shown in Figs. 6 and 8, the output voltage can be kept in excess of the required charging voltage by controlling the load resistance. The battery procured for this system has sealed cells which will withstand 75 psi internal pressure, and 170-amp-hr capacity at a 2-hour rate. Three of these batteries constitute a battery pack. The battery can receive a charging current of 7 amperes continuously (factory tested at 10 amperes). The SNAP 7D wiring diagram is shown in Fig. 9.

All equipment for the SNAP 7D system (Martin components) is available. The 30-day test of the system is being set up with dummy loads since the flashing light and weather station components have not been made available. The results of this test, the shock and vibration tests, the temperature tests, and the fuel loading will be covered in the next quarterly report.

The SNAP 7B electrical system was designed during the last quarter. The output voltage must be stepped up to the 32 volts required for the flashing light. This means that a dc-to-dc converter with a regulated input voltage is required. A breadboard converter has been fabricated. Preliminary tests show that, at full load, the input will be regulated to 12.18 volts and 5.0 amperes and the output will be 33.2 volts and 1.5 amperes. The converter efficiency at full load is 81.8%. The battery for the SNAP 7B system is on order. Its construction is similar to that of the battery used in the SNAP 7D system. The battery pack will consist of four 6-cell units, and it will be rated at 30 amp-hr.

The SNAP 7B and 7D safety reports were reviewed, and additional material was added to the reports. The main changes in the reports include the following:

- (1) Results of capsule pressure tests were added to substantiate the calculations in the section on system integrity.
- (2) The section on fire accidents was expanded to include a "Standard One-Hour Fire" as required by the AEC for shipping containers.
- (3) The section on quality control was expanded to include information on material composition and testing specifications of the capsule and shield material.

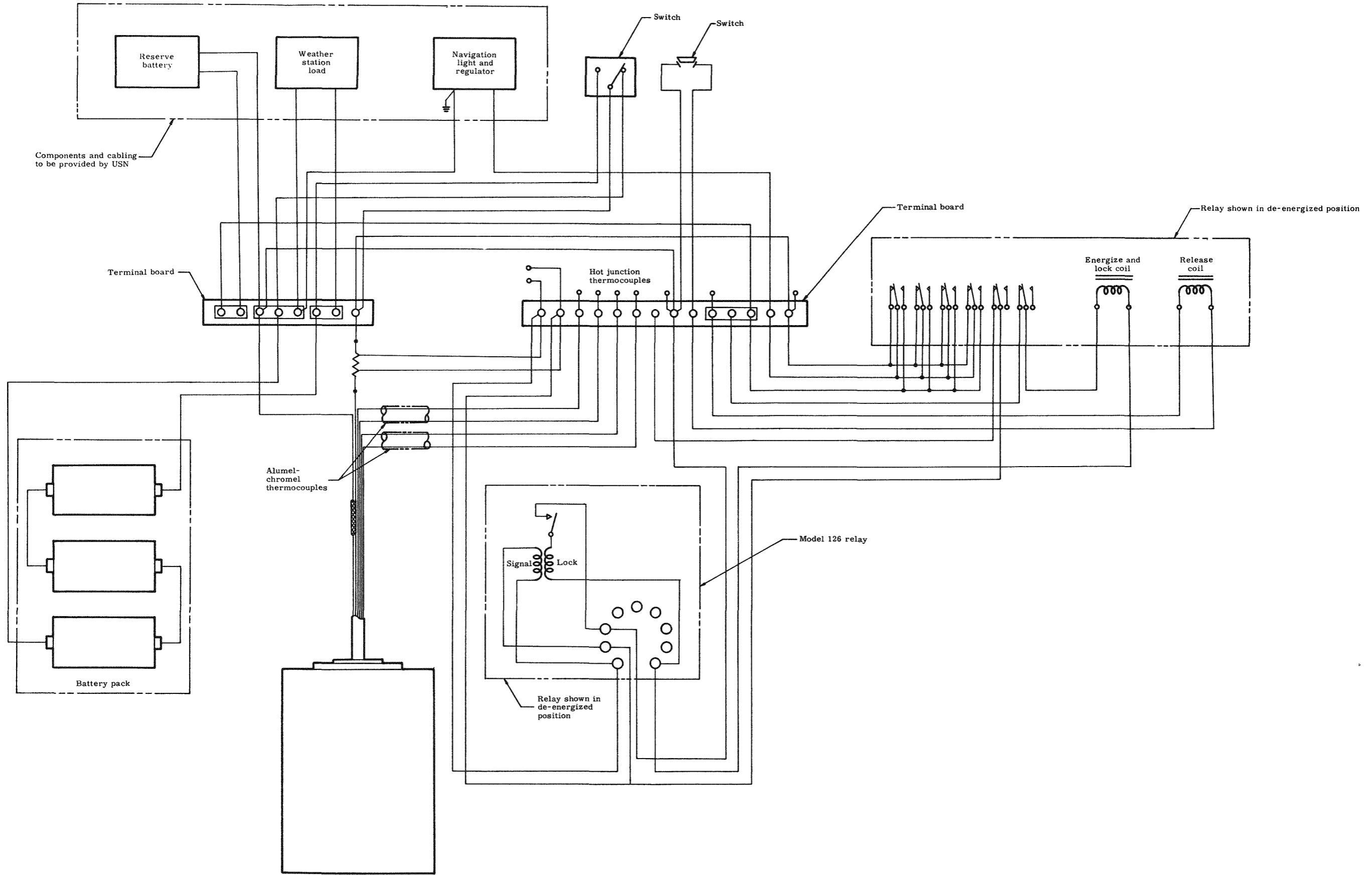


Fig. 9. SNAP 7D System--Schematic Diagram of Electrical System

- (4) An analysis of the results of both corrosion and external hydrostatic pressure was added. The capsule cap thickness was changed to the actual weld penetration thickness as specified. The time to failure was reduced to 510 years.
- (5) Aircraft accidents were deleted from the section on transportation accidents.

The AEC has requested certain other changes to be made prior to their acceptance of the safety reports. These changes will be effected during the next quarter.

IV. STRONTIUM-90 FUEL PROCESSING

A. OBJECTIVES

The objectives for this period were:

- (1) To deliver all remaining fuel processing equipment to the Quehanna Facility.
- (2) To initiate fuel process dry-run demonstrations (inert strontium in place of radioactive strontium) to support equipment and process checkout, and as support when applying for a by-product material license.
- (3) To modify or redesign any fuel process equipment found unsatisfactory during checkout at the Quehanna Facility.

B. DISCUSSION OF ACHIEVEMENTS

All fuel process equipment was delivered to the Quehanna Facility, installed and its checkout initiated. As anticipated, some of the equipment did not function as intended and had to be reworked. In general, the rework has consisted of modifications to facilitate the remote handling capability rather than modification to the chemical process.

The Hanford shipping cask, which is to be used for transporting Sr-90 carbonate from Hanford, Washington, to Quehanna, Pennsylvania, was loaded with inert strontium carbonate and shipped cross-country to check out the routing for the radioactive shipment. Martin procedures and equipment for dissolving the carbonate and removing it from the cask were to also be tested.

The mechanical handling of the cask and dissolving of the carbonate were conducted without problems. Simulation of the cooling down of the cask was not completely checked because the cask heater burned out. The difficulty in raising the temperature of the cask, even with a 2.5-kilowatt power input, may indicate that cooling will not be as much of a problem as anticipated.

Analysis of the dissolved strontium shows that 1595 grams were loaded in the cask. This value has been cross checked by a separate Martin Marietta laboratory and there is good agreement. In addition, a material balance based on analysis for free HNO_3 yields a value for strontium that checks with that of the strontium analysis.

Checkout of the auxiliary equipment used to support the fuel processing indicated changes were needed in the systems for intracell and cell top transfer, manipulator boot changing and ventilation. These changes have all been accomplished.

A demonstration was given to the AEC on transferring an article from the waste disposal box to Cell 2. It was accomplished in approximately 35 minutes. A manipulator boat can be changed without losing containment at any time during the procedure. Emergency activation of the cell ventilation system is now accomplished in about five seconds rather than the two minutes previously required.

Fuel process dry run demonstrations for the AEC Division of Compliance were conducted during the quarter. The ventilation system, waste storage and disposal, Hanford cask unloading procedure and cell transfer systems were successfully demonstrated.

Two deficiencies were noted during the dry run demonstrations. Mechanical deficiencies were present in the powder handling procedures for Sr-90. A new pellet press punch and die set and die charging mechanism now permits the transfer of dry powder from the Waring blender to the pellet press die while it is completely enclosed.

The second deficiency was lack of detailed procedures for some of the operations conducted at the facility. Detailed procedures, with appropriate Health Physics operations, are being written.

Some problems have been encountered in maintaining control of the chemical process. The problems are peculiar to scaled up production equipment and the arrangement of the process piping that allowed some of the reagents to hang up. The modifications made appear to be satisfactory. Fuel pellet densities are now running about 80% of theoretical.

Dry run demonstrations and equipment checkout will continue until hot cell operations begin. After receipt of the by-product material license, but prior to fuel process production, dry runs will be conducted using Sr-89 tracer material.

When the present equipment goes into full-scale operation, the Martin Company should be able to produce approximately 2,000,000 curies of Sr-90 per year.