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MASTER

SNAP PROGRAMS

Quarterly Progress Report No. 5

Subtask 5.3 and Task 6

October 1 through December 31, 1960
MND-P-3013-II

Approved by:

Director
Advanced Development Section

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MARTIN-NUCLEAR HEAVY ELEMENTS PROCESSING FACILITY

FOREWORD

This is the fifth unclassified SNAP Quarterly Progress Report prepared by The Martin Company. It covers the work performed during the period October 1, 1960 through December 31, 1960 under Contract AT(30-3)-217 (Subtask 5.3 and Task 6) for the U.S. Atomic Energy Commission.

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SUMMARY

The Martin Company is currently under AEC contract to develop suitable isotopic power sources for use in thermionic and thermoelectric power generators for space applications.

In the thermionic generators (SNAP-TIP Systems) Curium-242 will be used for the short missions (up to approximately 180 days) while Plutonium-238 is being considered for missions of longer duration. As availability, cost and processing procedures are improved for other potential isotopes, additional investigations will be included in the program to meet expanded Government requirements.

A. TASK 5, SUBTASK 5.3--THERMIONIC ISOTOPIC POWER SYSTEM

The procedure for fueling a thermionic generator with a Curium-242 heat source, which was established last quarter, was evaluated by Thermo Electron Engineering Corporation (TEEC) for its effect on the thermionic converter. The procedure then became part of the design criteria for Generator 2B.

The aspects which were stressed in the development of Generator 2B were:

- (1) The ability of the generator to withstand the dynamic loads specified in the Statement of Work.
- (2) Reducing the interelectrode spacing.
- (3) Reducing the collector work function.
- (4) Reducing heat losses.

For a second activation test unit which was fabricated and tested, a collector work function of 2.0 volts was achieved. The temperature dependence of the collector work function was also checked, and was found to occur at 900° K. The second purpose of the unit was to check out the radiation heat transfer between the heater and the emitter. The actual temperatures were in excellent agreement with the analytically determined temperatures. The design temperature distribution with Generator 2B, using the same analytical techniques, should be quite accurate.

The final design of Generator 2B was completed, except for details concerning the brazing of the emitters to the emitter support and the method of spacing the electrodes. The leadthroughs used on Generator 2A were revised and thoroughly tested. Tests were in progress at the close of the quarter on other components such as the heater and collector springs and on methods such as techniques of spacing and the brazing of emitters. A detailed dynamic analysis of the design of Generator 2B was undertaken and was 90% complete at the close of the quarter.

B. TASK 6--FUEL TECHNOLOGY DEVELOPMENT

The Task 6 fuel development program is being conducted for the purpose of supplying suitable radioisotope heat sources for direct conversion to electrical power by means of thermoelectric and thermionic SNAP generators. Various processes and methods for preparing optimum isotope compounds on a large-scale, pilot-stage basis are being investigated and evaluated with the aid of information from the literature and from laboratory-scale experiments conducted at The Martin Company and a few national laboratories.

The current effort is centered on processes involving Curium-242 and Plutonium-238 for medium- and long-lived applications, respectively. However, new isotopes which show promise are continually being investigated, along with new concepts such as the blending of different isotopes in a single capsule to achieve power flattening throughout the entire spectrum of short- to long-lived missions.

Containment materials for various isotope compounds and alloys are also undergoing intensive testing and evaluation. Test programs currently in progress include compressive-drop impact, high-velocity impact (bazooka), sea-water corrosion and air oxidation at elevated temperatures and compatibility with isotope compounds. Safety analysis of the overall Task 6 program is also under way.

I. INTRODUCTION

This is the second Quarterly Progress Report required under Contract AT(30-3)-217 for Fiscal Year 1960, including an appraisal of work under Task 5.3, Thermionic Isotopic Power Systems (SNAP-TIP Systems), and Task 6, Fuel Technology Development Program.

The SNAP programs covered in this report are divided into the respective tasks and subtasks. Each task is separately administered. Subtask 5.3 is directed by Mr. R. J. Harvey, and Task 6, by Mr. W. M. Bowes. This report covers the effort for the period October 1, 1960, through December 31, 1960. This report was prepared by R. J. Harvey and W. M. Bowes, based on principal contributions by Thermo Electron Engineering Corporation and G. H. Storrs, J. B. Zorn, G. Samos, J. Kane, P. Hess, S. Zwickler, E. Smith and P. Burkhalter of The Martin Company.

II. TASK 5--SUBTASK 5.3

1. Objectives

The objectives of this subtask are to design, develop, fabricate and test an operational power system coupling a thermionic converter device to a radioisotopic heat source. The thermionic development is being performed by Thermo Electron Engineering Corporation (TEECo) of Cambridge, Massachusetts, as a subcontractor to The Martin Company. The work on the isotopic heat source, environmental testing and other systems work is being carried out by The Martin Company.

2. Scope of Work

Subtask 5.3 included in its scope during the quarter work on the following sub-subtasks:

- (1) Evaluation of effects of fueling procedure on generator performance (TEECo) (5.3.2).
- (2) Dynamic analysis of Generator 2B (TEECo) (5.3.3).
- (3) Design and development of Generator 2B (TEECo) (5.3.4).
- (4) Testing of components and life testings of Generators 1A and 2B (Martin) (5.3.11).

Work on all four sub-subtasks continued into the following quarter.

3. Technical Discussion

a. Evaluation of fueling procedure (5.3.2)

The procedure for fueling a thermionic generator, prototype Unit 2A, with Curium-242 (described in Ref. II-1) is in general agreement with the procedure selected for fueling test unit 2B. However, many of the details must be revised so as to reflect the design differences between units 2B and 2A. Most phases of the fueling, listed in Chapter II of Ref. II-1, would apply to both units. However, the method of inserting the heat source in the thermionic generator for Unit 2B must be changed since the isotope will include the multishield heater section. The geometry of the fixture for holding the isotope, as well as the cooling system for keeping the temperature of the isotope below 700° C, must be adjusted.

The procedure for brazing the generator should include the possibility of inserting a seal plug for evacuation of the fueled unit, a facility not incorporated in the present fueling method.

The unsolved problems in the development of the fueling procedure, listed in Chapter IV of Ref. II-1, have been answered as follows:

- (1) Problem: Can the preactivated thermionic diodes be exposed to air, later outgassed, and regain full power?
Solution: It has been found that activated diodes can regain full power after exposure.
- (2) Problem: Can the electrical leadthroughs tolerate the temperature gradients during the additional outgassing encountered at the time of fueling?
Solution: With the new leadthrough design, experimental tests have shown, the leadthroughs to be used in the fueled unit can tolerate the temperature gradients.
- (3) Problem: Will a more rigid support of the emitter cause prohibitively high heat losses?
Solution: The rigid support designed for Unit 2B will cause minimum heat losses.
- (4) Problem: Will the normal contact between isotope container and emitter holders be sufficient to achieve the proper temperature distribution?
Solution: The design of Unit 2B, as compared with that of Unit 2A, transfers heat from the isotope to the emitter by radiation; there is, therefore, no problem of thermal contact resistance between the isotope and the emitter.
- (5) Problem: Can the final brazing of the thermionic generator be perfected to the point where a leak-free closure can be achieved with a high degree of reliability?
Solution: A low temperature braze (BT) has proved to be reliable with regard to leaks. It is therefore recommended that it be used in the final braze in the fueled unit.

b. Dynamic analysis of Generator 2B (5.3.3)

Generator 2B has been designed in such a way that the diode assemblies and the heater section are independent entities. This permits the two sections to be analyzed separately.

(1) Vibration and acceleration

Isotope supporting structure. The isotope (heater) supporting structure was analyzed for vibrational and accelerational effects in three orthogonal directions. The analysis showed that the various natural frequencies of the isotope structure subassembly ranged from 1300 to 2350 cycles per second. All the natural frequencies were higher than the 1500 cycle per second maximum driving frequency except for one torsional mode. Since no torsional vibration was specified and no apparent coupling effect exists, the isotope supporting structure is satisfactory from the standpoint of vibrational stability.

Stresses induced in the structure by vibration and acceleration loads were all below the working stresses of the materials at the operating temperature.

Diode assembly. The emitter-collector subassembly was designed to prevent separation of the emitter and collector during constant acceleration up to 23 g's and during vibration as specified in the environmental specification.

The spring load was designed to be 1500 grams. The design of the spring and interelectrode spacers is discussed in Section C.4.

Since separation of the emitter and collector does not occur under the specified environmental conditions, no vibrational analysis is required. Stresses in the sapphire spacers are well within working limits.

(2) Impact

Isotope supporting structure. Impact analysis of the isotope supporting structure is in progress. No definite results have been achieved.

Diode assembly. Impact analysis of the diode assembly is nearing completion. During 50-g impact, the emitter and collector will separate by no more than 0.001 inch. Resultant stress on the sapphire rods when contact is re-established is approximately 3000 pounds per square inch. Unless stress concentrations occur, resulting in chipping of the rods, this is within the working stress limits of sapphire at operating temperature.

c. Design of Generator 2B (5.3.4)

(1) Configuration of Generator 2B

Generator 2B is shown in Fig. II-1. Two identical diode assemblies are mounted at the opposite ends of a cylindrical housing. They are heated by a radiant heater suspended centrally in the split housing by a supporting structure made of multiple concentric cylinders.

Each diode assembly consists of a disc-shaped emitter, one inch in diameter, spring-loaded against the collector. The spacing is maintained by three sapphire rods embedded in the collector. The emitter is barium-impregnated tungsten brazed to a dense tungsten pellet to prevent warpage. The spring load, produced by a leaf spring mounted behind the collector, is applied to the emitter along a thin-walled tube which passes through the collector, locates the emitter axially and acts as the emitter lead. The tube is electrically insulated from both the collector and the leaf spring, and the collector is in turn insulated from the housing.

The radiant heater is a low-current electrical heater. A ribbon filament is coiled inside a cylinder of the same size as the isotope it simulates. The individual turns of the filament coil are held in place by ceramic spacers. The heater is mounted inside a heater sleeve which extends beyond the flat ends of the heater to heat both emitters from the sides in addition to heating the circular bases of the emitters. One heater lead plugs into the emitter lead of one diode assembly; the other lead is provided by ground.

The heater supporting structure is made of six concentric tantalum cylinders each 0.005 inch thick. They are brazed together alternately at each end with molybdenum fitting rings. The inside cylinder is spot-welded to the heater sleeve, and the outside cylinder is brazed to a ring which fits between the two halves of the housing. This arrangement provides a relatively rigid structure of low thermal conductivity and little thermal expansion. Radiation losses are kept low by spiral shielding made of 0.0005-inch tantalum foil inserted between the structural cylinders.

Isotope fueling can be accomplished by electron-beam welding the isotope casing into the finished heater sleeve-supporting structure assembly. The whole assembly can then be stored until subsequent incorporation into the unit. During storage, the assembly can be kept adequately cool by clamping the isotope at the flat faces. For final assembly, the whole structure is simply lowered into one half of the housing.

Fig. II-1. Generator 2B

Heat is transferred from the heater to the emitter by radiation. Activation Unit 2 was constructed to confirm the radiant heat transfer analysis and, in addition, to confirm the relation between the work function and the temperature of the collector. The complete report on the testing of Activation Unit 2 is included in this report as Appendix A. Both the temperature of the heater and the work function of the collector relate to temperature characteristics approximately as predicted.

(2) Heat transfer

The heat flow pattern inside the prototype is shown schematically in Fig. II-2. The individual calculations are presented in Appendix B. The results for the anticipated specifications are as follows.

Generator specifications.

$$T_E = 1500^\circ \text{ K}$$

$$T_C = 900^\circ \text{ K}$$

$$T_H = 600^\circ \text{ K}$$

$$1.90 \text{ electron volts} \leq \phi_C \leq 2.00 \text{ electron volts}$$

$$10 \text{ microns} \leq w \leq 15 \text{ microns}$$

$$A = 5 \text{ square centimeters per diode}$$

$$0.25 \text{ watt per square centimeter} \leq P_O \leq 0.45 \text{ watt per square centimeter.}$$

Heat input to both diodes. (The input equals the sum of the losses and the output.)

Radiation loss (watts)	38	± 6
Electron cooling loss	41	± 5
Sapphire losses	12	± 2
Lead losses	12	± 2
Power output	3.5	± 1
Total (watts)	106.5	± 16

Fig. II-2. Heat Flow Diagram

Heat losses from heater to housing.

Conduction (watts)	5 ± 0.2
Radiation	4 ± 0.5
End losses	<u>5 ± 1.0</u>
Total (watts)	14 ± 1.7

The total heat input is the sum of losses and output, and is approximately 120 ± 18 watts. The anticipated efficiency is

$$\eta = \frac{P_o}{\Sigma (P_o + \text{losses})} = \frac{3.5 \pm 1}{120 \pm 18} = 3.5 \pm 1\%.$$

(3) Interelectrode spacing

The present design of Generator 2B utilizes sapphire rods to maintain interelectrode spacing. During vibrational testing, a spring load of 3.3 pounds had to be used to prevent widening of the space. To keep contact stresses at the emitter below 500 pounds per square inch (a stress level which in the past has not caused creep), the diameter of the sapphires was enlarged to 0.060 inch at the hot end. To compensate for the increased heat losses, the diameter at the cold end was reduced to 0.030 inch. This design should yield a net loss of 2 watts per spacer. The total length is reduced to give a differential thermal expansion of less than 0.0003 inch (Appendix B).

A difficulty arising in the use of sapphire rods is the necessity for even higher spring loads to prevent separation during impact. This would imply sturdier sapphire geometries with increased heat losses if differential expansion is to be held below 0.0003 inch. Also, the lead tube which transmits the spring load and locates the emitter axially would have to be rigid to withstand environmental testing, leading to further heat losses. The present design appears marginal.

Because of these inherent difficulties of sapphire rods as spacers, TEECo is currently investigating two new ways of maintaining interelectrode spacing, under this project and under other vacuum converter projects. One new method uses wires instead of the sapphire; the other uses thin metal sheets around the periphery of the diode (see Fig. II-3). In either method the wire or the sheet is brazed to an electrically insulated member in the collector and brazed or electron-beam welded to the emitter and serves both as spacer and as emitter lead. Both methods, now in their developmental stage, present the

Fig. II-3. Spacer Arrangements: (a) Wire Spacer, (b) Sheet Spacer

advantage of matched thermal expansion of spacer and electrodes, lower conduction losses, reduced creep, simpler structure and higher reliability.

Thermal expansion of the metal spacer can be matched to that of the collector and emitter to result, ideally, in zero differential thermal expansion. Lower conduction losses can be achieved because the length of the spacers is no longer limited by differential thermal expansion (only by rigidity) and the functions of spacer and lead are combined. Creep is greatly reduced because the stresses due to spring loading are eliminated. Spring loading with the leaf spring and tension tube are eliminated, resulting in a simpler structure. Greater reliability is attained because the interelectrode spacing is maintained positively during shock and vibration.

(4) Springs

A spring is used to ensure that the emitter remains in contact with the sapphire rod spacers, regardless of relative thermal expansions and acceleration forces. Based on the specified acceleration and vibrational environment, a steady-state spring loading of 1500 grams was chosen.

Because the spring will be operating at a temperature of approximately 700° C, a material that retains its elastic properties at this temperature is required. Any creep in the spring is undesirable. To simplify assembly and permit accurate spring loading, a spring deflection of over 0.020 inch at a load of 1500 grams is desirable.

The material chosen was Tantalloy (an alloy of tantalum and tungsten) produced by Fansteel Metallurgical Corporation. Because the only available sheet stock was 0.011 inch thick, the spring was designed for this thickness. Various shapes and sizes were analyzed before a design was chosen.

The tests showed that the spring will perform satisfactorily. The deflection for a 1500-gram load is approximately 0.045 inch. No inelastic deformation was observed during operation of the spring over 120 hours at 700° C under a 1500-gram load.

(5) Emitter braze

It has been observed that emitters sometimes warp. This warping is independent of any external load and increases with time, as the emitter is maintained at operating temperature. Emitters of the type used in the present test diodes are least susceptible to warpage, but some deformation occurs which may limit the closeness of spacing

attainable. To eliminate any warpage and ensure a tolerable flatness of the emitter surface during the life of the device, it was decided to braze the thin (0.040-inch) emitter to a heavy tungsten backing plate. Two different brazes have been tried and a third method of brazing is almost completed.

The first braze utilized the 1505° C tungsten-nickel eutectic. Suitable ratios of NiO and W powder were mixed with a binder (amyl acetate) and the braze material applied with a spray gun. The brazed emitter was mounted in a test diode, and the tube was processed and activated in a bell jar-oil diffusion pump vacuum system. Following activation, a life test was run. The collector work function increased from 1.9 to 2.2, as shown in Fig. II-4, indicating poisoning of the collector surface. Emission capability of the emitter was determined at the end of the life test, but no emitter poisoning was detected. The tube was disassembled, and the electrode surfaces were inspected. The emitter face was clean, but the collector showed a heavy brown-black deposit. A spectrographic analysis of the deposit showed it to be largely barium, with traces of nickel, titanium, and magnesium.

The second brazing method investigated used a nickel-molybdenum eutectic (1350°C). Binder, braze application and brazing procedure were identical to the Ni-W braze, except for the lower brazing temperature (1350°C). This emitter was mounted and processed in a sealed glass envelope on a mercury diffusion pump. Following activation, a life test was run. The collector work function increased with time (Fig. II-5), rising to about 2.2 volts. Inspection of the tube at the end of the test showed the emitter to be clean, but the collector surface was covered by a deposit similar to that in the previous tube.

The third brazing method also was a nickel-molybdenum eutectic. The braze material in this case was applied by evaporating a layer of molybdenum on the tungsten backing piece, followed by a thin layer of nickel, also deposited by evaporation. The apparatus used is shown in Fig. II-6. The piece from which the material was evaporated was electron-bombardment heated to 2100°C for molybdenum and 1250°C for nickel. The required amount was deposited in about ten minutes. It is hoped that this method will eliminate any possibility that NiO or binder will remain after completion of the braze to cause contamination. This method also allows better control over the thickness of the metal layer and permits a smaller amount of nickel to be used.

d. Testing of components and life testing of Generators 1A and 2B (5.3.11)

A development effort was undertaken during this reporting period to refine the design of the leadthrough developed for Generator 2A.

Fig. II-4. Collector Work Function Versus Time for Test Diode No. 1110 (Ni-W Braze)

Fig. II-5. Collector Work Function Versus Time for Test Diode No. 1120 (Ni-Mo Braze)

Fig. II-6. Evaporation Deposition Apparatus

As described in the last quarterly report (Ref. II-2), the necessary change involved increasing the flexibility of the metallic sections. Since some parts were already on hand, the first change involved only increasing the flexibility of the upper cap. This is shown in Figs. II-7 and II-8. Metallographic examination revealed that cracking of the ceramic near the interface still occurred, but to a lesser degree. The design was again revised as shown in Fig. II-9. The dimensions of the ceramic were held constant while the wall thickness of the titanium was reduced.

A number of leadthroughs were made, thermally cycled, and leak-tested. They proved to be vacuum-tight even after severe thermal cycling. The temperature was raised to 800° C within 45 minutes, maintained for one hour, and reduced within 45 minutes. No leakage was observed after five applications of this cycle to a representative leadthrough. This result is very encouraging because no previous design would withstand such severe cycling. This leadthrough design will be used in Generator 2B.

4. Conclusions

- (1) The present design of Generator 2B will withstand vibration tests and have a minimum of heat losses.
- (2) The collector work function is expected to be between 1.9 and 2.0 volts.
- (3) The heater is expected to last at least 3000 hours.
- (4) The spring holding the electrodes together will withstand a load of 1500 grams for long periods at high temperature with negligible setting.
- (5) The creep rate of the sapphire rods into the electrodes, particularly the emitter, will be minimized by increasing the contact area of the rod.
- (6) Previous leadthrough difficulties will be eliminated by use of the new design.
- (7) The design of Generator 2B is such as to permit fueling the generator with a Curium-242 heat source.

Fig. II-7. Cross Section of Primary Leadthrough for Generator 2A

Fig. II-8. First Revision of Leadthrough, Showing Thick Wall

Fig. II-9. Second Revision of Leadthrough, Showing Thin Wall

5. References

- II-1. "Procedure for Fueling a Thermionic Generator with a Curium-242 Heat Source," The Martin Company, MND-P-2401, August 1960. Also published as Appendix C of MND-P-3012-II.
- II-2. "SNAP Programs Quarterly Progress Report No. 4--Subtask 5.3 and Task 6," The Martin Company, MND-P-3012-II, July through September 1960.

III. TASK 6--FUEL TECHNOLOGY DEVELOPMENT

A. SUBTASK 6.1--GENERAL DEVELOPMENT AND MATERIAL REQUIREMENTS

A literature survey on americium, curium and plutonium was completed during the second quarter of 1960 in preparation for the final evaluation of the americium and curium processing methods. It was followed up last quarter with progress on the compilation of a compendium of technical information pertaining to actinides and other isotopes for power device heat sources.

1. Objectives

The objectives of this subtask were:

- (1) To compile data pertaining to the properties of Americium-241, Curium-242, Plutonium-238 and other fuel sources.
- (2) To survey published information concerning processes for making various compounds of Curium-242 and Plutonium-238.
- (3) To direct and coordinate subtasks under Task 6 and to serve as liaison between the project and the AEC.

2. Scope of Work--Evaluation of Literature

The scope of the search on which the compendium is based was defined in a draft statement of work dated October 6, 1960. The 18 nuclides on the final list are given in Appendix D. On the bases of the preliminary search completed in June 1960 and the eclectic studies during the third quarter, a subject index was prepared for final search of the Abstracts of Classified Reports (ACR) and Nuclear Science Abstracts (NSA). The boundaries of interest were also defined by a list of topics excluded from the search (Appendix E).

A thorough search was made through the ACR (Volumes 1 through 16) and NSA (Volumes 1 through 14). This search yielded over 1500 items of prime interest plus an additional 600 of secondary or marginal interest.

In order to cut the list of references requiring detailed evaluation to a more manageable size, the following divisions have been made:

- (1) All classified reports.
- (2) All microcards.

(3) All patents.

(4) All reports not available in The Martin Company.

It is intended to examine every available original document and to prepare notes and comments for as many of the better publications as time allows. This work is now under way; work on the reviews has been deferred.

3. Technical Discussion and Evaluation

The effectiveness of the compendium will depend greatly upon the thoroughness and care with which the source material is selected and examined. Hence, as far as practical, maximum attention has been directed to these factors.

Thoroughness is stressed most for the transuranium isotopes on the list. Data on the others are being chosen selectively, with most general information about the elements and their compounds omitted to avoid the inclusion of a large number of unnecessary references. The most pertinent information about Cesium-137 and Cerium-144 is usually noted because of their occurrence as fission products. Limited information is available on Radium-228, Neptunium-237, Actinium-227, Polonium-210 and Promethium-147. Strontium-90 is a special case, because over 95% of the publications are concerned with biological matters and, hence, are of no interest for this compendium.

4. Conclusions

During the quarter, the preparation of the compendium was divided into two phases:

- (1) Information research--scientific evaluation and preparation of the manuscript.
- (2) Information processing--the manipulation of documents and delivery of information in finished form.

B. SUBTASK 6.2--AMERICIUM-CURIUM RADIOISOTOPES PREPARATION PROCESSES

Laboratory experiments were conducted on the ion-exchange purification of Americium-241 which will be subsequently encapsulated and irradiated to produce Curium-242. The thiocyanate purification procedure is being explored and the various process conditions are undergoing refinement. An alpha-counting system is under construction to aid in the analysis of solutions containing americium, curium and plutonium.

The americium-encapsulation process was modified and is currently operating satisfactorily, as evidenced by prepared dummy specimens which met all the design specifications. The capsule-decontamination process has been established for removing any traces of radioactive material prior to shipment of the capsules to an irradiation facility.

Investigations were made into the pressure buildup which may occur inside the capsule as a result of oxygen and helium accumulation. It was concluded that slightly reducing the compacted core density, in addition to incorporating a void in the core, would prevent any excessive pressure. Under these conditions the capsule would be seal welded at a one-atmosphere helium pressure and subsequent leak detection would require merely "sniffing" the capsule with a helium-leak detector. Also, the etching procedure for final seal welding of the capsule was established.

Several methods were explored for achieving an improved separation of curium from aluminum during the chlorination-volatilization of irradiated capsules. Results with simulated materials were not satisfactory; so effort is now under way on a wet-chemistry method in which the capsules are dissolved in a solution of sodium hydroxide and sodium nitrate.

A set of Argonne Model VIII manipulators was procured and will be installed in a hot-cell mockup at Baltimore.

Design analysis of a shipping cask for seven irradiated capsules was completed, and cask specifications were established. A vendor has been chosen and final negotiations are under way for fabrication of the cask.

1. Objectives

During the quarter, the objectives of Subtask 6.2 were as follows.

a. Americium processing

This phase of the subtask had as its objectives:

- (1) To assemble the dry-box line for americium encapsulation.
- (2) To check out and modify equipment for proper operation.
- (3) To run the americium-purification procedure to establish the chemistry and operating conditions.

- (4) To determine the handling equipment and procedures needed for americium encapsulation.
- (5) To initiate work on the building of a vacuum volatilizer for use in preparing samples for counting analysis.
- (6) To design and have fabricated a sample-holding chamber for alpha counting under vacuum.

b. Curium purification and processing

The objectives of this program were:

- (1) To assist in the preparation of a hot-cell mockup for design of the Quehanna processing equipment.
- (2) To continue work on the purification of curium by ion exchange according to methods found in the literature (Refs. III-1, -2, -3 and -4).
- (3) Curium volatilization--to determine whether filter frits would prevent loss of rare earths from the reaction chamber.
- (4) To determine whether vacuum sublimation would prevent the loss of rare-earth chlorides.

c. Design, fabrication and equipment installation

The following objectives were established for the americium- and curium-processing system:

- (1) To coordinate and inspect the installation of equipment in the Heavy Elements Processing Facility (HEPF).
- (2) To review the design of the disposal system for radioactive liquid wastes and recommend procedures for safe handling of the wastes.
- (3) To incorporate revisions to Martin drawings.
- (4) To review and revise as required the specifications for the curium-shipping cask.

2. Scope of Work

a. Americium processing

Purification. Tracer-level experiments on the procedures to be followed in the large scale purification of americium were carried out during the quarter. The experiments were run in accordance with the step-by-step procedure outlined for processing americium (Ref. III-5):

- (1) Removing plutonium by passing the americium feed solution in 8M hydrochloric acid plus 0.1M nitric acid through a Dowex 1 anion-exchange column.
- (2) Loading that solution on a Dowex 50 cation column.
- (3) Eluting with 5M ammonium thiocyanate to prepare a solution for a final Dowex 1 separation. Americium remains on the column while the rare earths are eluted with more thiocyanate.
- (4) Finally, eluting purified americium with 6M hydrochloric acid.

Design drawings of a vacuum evaporator used at Lawrence Radiation Laboratories, Livermore, California, have been made available to Martin. The fabrication of this equipment for use in Martin laboratories will begin as soon as the necessary materials are available.

A unit has been designed for holding and counting an alpha sample under vacuum, with silicon diodes as detectors.

Encapsulation. A major part of the work of this subtask was spent in modifying the hydraulic press, die and capsule shells to effect satisfactorily filled capsules. Work continued during the quarter in these areas:

- (1) Checking the powder mixer and leak-detection equipment.
- (2) Removing sources of leaks in the dry-box system.
- (3) Decontaminating capsules.
- (4) Evaluating etching procedures in support of the welding effort.

b. Curium purification and processing

A set of Argonne Model VIII manipulators was borrowed from Martin's Quehanna facility for installation in the hot-cell mockup. The mockup is designed for developing hot-cell procedures and techniques for the various processes which will be conducted at Quehanna.

Work continues on the purification of curium, with americium as a substitute. One of its objectives is the accurate determination of the number of column volumes of solution required to remove all contamination as well as to elute all curium from the various resin columns used in the procedure.

Curium volatilization. Tests were performed with a filter frit incorporated at the end of the reaction chamber of the volatilization apparatus. Vacuum sublimation experiments were also performed with aluminum chloride and a mixture of aluminum chloride and gadolinium chloride.

c. Design, fabrication and equipment installation

Much of the design effort during the quarter was devoted to inspecting and coordinating the installation of the processing equipment in the HEPF.

Solenoid valves supplied with the americium-purification boxes were found to be inoperable and failed to pass quality control. A Valcor valve, Model 51P18N24, was tested to the satisfaction of the Radioisotopes Laboratory, and a purchase order was released to Valcor for the development of a similar valve made of irradiated polyethylene. The valve is to be constructed according to Martin Specification NPS47270001. After acceptable performance tests have been completed, a sufficient number of valves to satisfy the requirements will be purchased.

Following a design revision for proper operation of the liquid-waste disposal system at the HEPF, purchase orders were released to secure the necessary equipment. A liquid-level monitor will indicate liquid levels together with a high-level alarm which will sound when the tank is 75% full. A liquid-discharge system was designed to make possible the assay of the tank contents and the discharge flow of the liquids to the local sewage pumphouse servicing the HEPF. A recording meter in the pumphouse will monitor the dilution achieved during discharge into the sewer mains.

Schematic flow diagrams were prepared during the quarter to show the purification and encapsulation of americium, plutonium and curium.

Specifications for the curium-shipping cask were re-evaluated on the basis of the higher thermal requirements per capsule. Revised specifications, made more stringent to include performance requirements in accordance with anticipated federal shipping regulations (ICC), were released to vendors.

The americium-box exhaust systems were modified to house the thermal indicators for fire detection.

Removable metal covers were designed for the glove ports. These will be installed as an additional safety feature when the box is not in operation.

The layout of the containment boxes in the HEPF has been initiated.

Early work dealt with the location of equipment required in the chemical purification procedure. Work has been initiated to design dry boxes for processing the irradiated americium capsules at the Quehanna facility.

The alarm system for the HEPF has been discussed with Health Physics and Security representatives. It was concluded that the available telephone lines would be used to connect the American District Telegraph alarm for any of the following situations:

- (1) Fire.
- (2) Radiation.
- (3) High liquid level in waste tank.
- (4) Loss of vacuum.

A panel of lights to indicate the source of the trouble will be mounted outside the facility.

3. Technical Discussion and Evaluation

a. Americium processing

Purification. Bench-scale experiments were conducted on the americium-purification process, involving the use of three separate ion-exchange columns. The procedure followed is outlined in Ref. III-6.

Experimental results showed that the eluate from the first Dowex 1 ion-exchange column could be concentrated by a factor of only 10 rather than a factor of 50 due to the large quantity of salts present. This concentration represented a volume equal to one half of a column volume. After the 10-to-1 concentration of the solution, the eluate was diluted with an equal volume of water and the pH was adjusted to 1 with ammonium hydroxide. This adjusted solution was then passed through a column of Dowex 50. The rare earths and tracers (americium and fission products) were retained on the column. After washing with two column volumes of 0.1N ammonium chloride, an eluant solution of 5M ammonium thiocyanate was passed through the column to remove the rare earths and tracers as SCN^- complexes. This eluant, with the SCN^- complexes, was subsequently passed through a second anion column to retain the Americium-241 while the rare earths and fission products passed through. Figure III-1 represents a flow sheet of the experiment and shows that nine column volumes of 5M ammonium thiocyanate washings were used to separate the fission products from the americium. However, the decontamination factor was only about 30. A second experiment was run in an attempt to improve the decontamination factor. In this run, 26 column volumes of 5M ammonium thiocyanate were used and decontamination was greatly improved. Future work will emphasize decreasing the volumes of eluant in the final separation of fission products from Americium-241 while maintaining good decontamination.

A set of drawings of the alpha-sample volatilizer has been received from Lawrence Radiation Laboratories. Fabrication of this piece of equipment is expected to start soon. The vacuum and diffusion pumps necessary for its operation are being ordered.

An apparatus for holding and counting alpha samples under vacuum with silicon diode detectors has been designed and is being fabricated. Researchers at Brookhaven and Oak Ridge National Laboratories have found that by removing the air between the alpha sample and diode, better resolution can be obtained. A power supply and a preamplifier for the diode system have also been ordered. The rest of the required electronic equipment is available at the critical facility on a temporary basis (i.e., Tracer Laboratory Model 213 256--Channel Analyzer, Franklin Amplifiers, etc.).

The original valves of the americium-processing equipment were found to leak and were returned to the vendor for repair before the start of the quarter. Another valve manufacturer has been contacted, and two valves containing different types of core material are being sent to The Martin Company for test purposes. If they are found satisfactory, similar valves will be ordered to replace the original valves, as well as additional valves for future needs.

* Radioactive rare earths

Fig. III-1

Encapsulation. The blender for mixing americium oxide and aluminum powder was modified from the original design and installed in the glove box. With the new design the powder can be mixed in much less space. Tests of blend homogeneity were performed with ceramic grade uranium dioxide as a substitute for americium oxide. A mixture of aluminum powder, uranium dioxide and fluorescein powder (to check for contamination) was blended for one hour. Three capsules were then processed by the method to be used with the americium oxide. These capsules were first radiographed, after which the aluminum shell was removed. The individual charges are being checked for uranium dioxide content. During the test, the equipment was sprayed with alcohol to activate the fluorescein powder and check for contamination. The fluorescent die check indicated that there was some contamination at the top of the molding die; however, the amount was very small, and periodic decontamination should keep the activity within tolerable limits. Figure III-2 is a radiograph showing the uranium dioxide distribution obtained when the blended powder (Al with UO_2) was pressed into capsules. Many of the apparently large agglomerates are a buildup of many particles aligned behind one another. This distribution was obtained after one hour of mixing. Further tests will be necessary to attain the optimum mixing schedule.

Oxygen-pressure buildup during the irradiation of americium dioxide (AmO_2) to obtain curium oxide (Cm_2O_3) was calculated and found to be significant. It was decided to provide a void in the capsules and/or reduce the core density to alleviate the pressure effect, rather than to use a getter for the oxygen since any foreign material would increase the problem of separation of curium and americium from the aluminum.

A recommendation was made that the capsules be fabricated from the higher strength Type 5052 aluminum rather than Type 1100 aluminum to withstand the oxygen- and helium-pressure buildup. It was found that the magnesium and chromium present in the Type 5052 aluminum would not be detrimental to reactor operation, nor would they seriously increase the activity of the capsule.

The detail pieces of the compacting press were received and the die was assembled on the press. When the stepwise compacting procedure was attempted, it was found that the press was not functioning as required. The electrical wiring did not conform to the wiring diagram. The vendor was notified and necessary modifications were made for correct operation. During a runoff check of the press, it was found that the aluminum capsules exhibited a tendency toward galling of the inner walls and decreasing length. These problems were rectified by compacting smaller charges of aluminum powder, enlarging the inside diameter of the capsule and changing the capsule material to Type 5052 aluminum.

Fig. III-2. Radiograph of Americium Capsules Containing UO_2

When capsules of 0.996- to 0.997-inch outside diameter were compacted, it was very difficult to remove them from the die. Changing the outside diameter to 1.000 inch eliminated this difficulty.

In view of these and earlier findings, the following specifications have been established for the americium capsules:

(1) Type of material:

- (a) Shells--Type 5052 aluminum, H38.
- (b) End plugs--Type 5052 aluminum, H14.
- (c) Weld filler metal--Type 5052 aluminum or 43 alloy.

(2) Dimensions:

- (a) Outside diameter--1.000 $\begin{matrix} +0.000 \\ -0.001 \end{matrix}$
- (b) Inside diameter --0.888 $\begin{matrix} +0.000 \\ -0.001 \end{matrix}$
- (c) Wall thickness --0.055 to 0.056 inch.

(3) Welding procedure:

- (a) Bottom end capsule--welded on side of capsule with filler metal; chill block used to prevent excessive annealing.
- (b) Top end capsule--milk-can type with void, end welded.

A pressure of six tons per square inch has been found optimum for compacting the core.

Loaded capsules were sectioned to show the effect of pressing. Figure III-3 shows a section of one of the first capsules made in the automatic press. The individual pressed charges were only mechanically bonded to each other and separated easily. However, aluminum powder is still stuck to the inner wall of the shell, indicating that some bonding occurred between the clad and core, which will tend to aid in containment of internal gas accumulations. Present fabrication of the capsules employs an increased number of smaller charges, further improving bonding characteristics between clad and core.

Fig. III-3. Section of Aluminum-Filled Capsule

MND-P-3013-II

Since temperature calculations on the irradiated capsules will be influenced to some extent by the core density, it was decided to evaluate the thermal conductivity of specimens compacted to 75, 85, 95 and 100% of theoretical density. Tests for relative values of thermal conductivity were initiated, but it was found that the instrumentation was not sufficiently sensitive to provide the desired degree of accuracy. Consequently, this effort has been postponed until more sensitive equipment becomes available.

Capsule decontamination and etching. Prior to shipment of the americium capsules to an irradiation facility, any radioactive contamination on the outer surfaces will be removed with an ultrasonic cleaner. Tests were performed in which fluorescein powder and Dye-check (Magneflux Corporation) were applied to the surface of a capsule to simulate contamination; the capsule was subsequently decontaminated in the ultrasonic cleaner. It was found that a mixture of green soap and water (5 cc soap to 1000 cc water) completely cleans the surface within a period of 30 seconds.

Investigations were made into the etching procedure required to prepare the capsules for the final seal-welding operation. It was found that a 1N solution of sodium hydroxide, when applied for 30 seconds, will provide a satisfactory etch. After the 30-second etching period, the end of the capsule shell is rinsed in nitric acid, followed by a water rinse.

Figure III-4 shows a weld typical of those obtained in aluminum capsules. The crevice which is readily apparent in the weld is caused by an aluminum oxide layer on the aluminum surfaces. The weld thickness obtained at this point by the best cleaning method is 0.034 inch, minimum. Determination of the bursting strength of a weld with this thickness indicated that failure should not occur as a result of the pressure buildup anticipated.

Helium-leak checking of capsules. Prior to recent modifications, the procedure for leak testing the americium capsules involved:

- (1) Placing capsules in evacuation chamber.
- (2) Pressurizing with ten atmospheres of helium.
- (3) Placing in a second chamber, partially evacuating, then sniffing for helium with a helium-leak detector.

Since a void volume filled with helium will be incorporated in the newer capsules, the evacuation and pressurization steps in the leak-checking procedure may be replaced by the simplified operation of merely sniffing the completed capsules for helium.

Fig. III-4. Typical Weld in Top of Aluminum Capsule Showing Crevice

The present plan for encapsulating americium is to incorporate a void in the end cap along with a center hole for the admission of helium. Following compaction of the core, the end cap will be welded to the capsule shell (milk-can seal); a pipe, with an O-ring at the end to effect a seal, will then be placed against the cap over the center hole (Fig. III-5)

Fig. III-5

and the void evacuated to 25 microns of mercury. After the proper vacuum has been attained, helium will be bled into the void until a one-atmosphere pressure has been attained and the center hole will then be seal welded. After the sealing operation is complete, the capsule will be placed in an evacuation chamber and subsequently checked for leaks.

Another method of checking for leaks is currently under investigation. It involves placing the capsules in water which is slightly below its boiling point and detecting the formation of bubbles. It has been found by this method that a leak of 1.1×10^{-4} cubic centimeter per second produces a bubble every 20 to 30 seconds. The effectiveness of this method may be further improved by pressurizing to increase the internal pressure of the capsules prior to checking for leaks. In pressurizing the capsules, a problem exists in determining the effect of hole size (leak) and pressure on the rate of pressure buildup in the capsule. Further testing of this method will continue.

Dry-box line. Considerable effort was expended in sealing leaks in the dry-box system. It was found necessary to employ 1/4-inch gasket material to prevent leaks. All bolts through the boxes and all lead-throughs were sealed with Glyptol or Permatex. The mixing, pressing and welding boxes were sealed so that only one liter per minute would be lost at a pressure differential of two inches of water.

A small vacuum cleaner will be installed in the mixing box and will be interconnected with the pressing and welding boxes to aid in decontamination of these boxes.

b. Curium processing

Curium volatilization. In an effort to reduce the loss of curium chloride formed during the separation of aluminum by chlorination sublimation, filter frits of various porosities were incorporated in the exhaust line from the volatilization chamber. These frits did trap a portion of the entrained product, but overall results were not sufficiently satisfactory to warrant their use in the system.

It was postulated that the fine particles of rare earth (used to simulate curium) were lost as a result of entrainment in the helium stream passing through the system to sweep the aluminum chloride vapor into the condensate receptacle and that elimination of this gas stream might reduce entrainment to within tolerable levels. Consequently, several laboratory experiments were conducted in which aluminum chloride and rare-earth chlorides were volatilized under a 25- to 50-micron vacuum. Even though the only physical flow in the system was that provided by the aluminum chloride vapor, it was found that carryover of rare-earth chloride particles still occurred to the extent that the separation was not considered satisfactory. Based on these results, further effort on the sublimation process will be discontinued until vapor-pressure data can be established for curium chloride. Work will now be centered on a method in which aluminum in the irradiated capsules is separated from the curium product by wet-chemistry techniques employing a sodium hydroxide-sodium nitrate solution.

Curium purification. A series of experiments have been conducted to determine optimum conditions for the ion-exchange purification of curium on Dowex resin columns and also to determine the number of column volumes of eluant which can be passed through the column for fission product removal before curium begins to elute. Since curium is not available for bench experiments, americium is being used to simulate the curium.

With 10.5M lithium chloride solution as the eluant, lanthanides and actinides will elute chromatographically from the column, fission products and lanthanides eluting first; therefore, curium and the actinides will be decontaminated of lanthanides (Refs. III-7 and -8).

Experiments were run by preparing a solution containing rare earths, fission product activity and Americium-241 in 11.5M lithium chloride plus 0.1N hydrochloric acid. The solution was then loaded on a Dowex 1

column and the column subsequently eluted with 10.5M lithium chloride. Results of the experiments are shown in Figs. III-6, -7, -8 and -9. Figures III-6 and -7 show that, with proper column length, nine column volumes of 10.5M lithium chloride is necessary before americium begins to elute. Figure III-8 shows that a small column allowed the americium to elute before the fission products had completely eluted. Data also indicate that, if the loading volume of the sample is too large, curium will not be efficiently decontaminated before it breaks through into the eluate. Figures III-9 and -10 illustrate the elution curves obtained for americium and rare-earth fission products after the loading volume was reduced from ten column volumes to three or less. This resulted in a better than 99% separation of americium from fission products. Figure III-11 is a flow sheet received from Lawrence Radiation Laboratories. As can be seen, these results compare favorably with those obtained at Martin. The small differences in elution volumes can be attributed to the fact that the heat produced within the column when large amounts of curium are used can disrupt the elution fractions so that less definition of the phases is obtained. It is believed that a smaller feed volume should be used in conjunction with a longer column for better decontamination. Therefore, when curium becomes available for bench-scale operations, a run will be made following the procedure described by Lawrence.

Fig. III-6. Elution of Rare Earth and Fission Products from Am-FP Mixture

Lactic acid elution for curium purification was carried out on a Dowex 50 x 8 cation resin packed in a 1-centimeter by 10-centimeter glass column. A column temperature of 70°C was maintained by use of a water jacket and heating tape surrounding the column. Americium-241 was used to simulate curium. A solution of 2.5M lactic acid, pH

Fig. III-7. Elution of Americium

Fig. III-8. Separation of Americium from Fission Products

Fig. III-9. Separation of Americium from Fission Products

Fig. III-10. Separation of Americium from Fission Products

Fig. III-11. Fission Product Decontamination of Cm-242 Produced in the Irradiation of 1 gm of Am-241 (Batch C)

4.5, was employed as the eluant to remove americium. Figure III-12 represents a typical elution curve for this system. The americium begins to elute after one column volume, peaks at about three column volumes and continues to elute after eight column volumes. Six column volumes has been chosen as a suitable cutoff point.

The literature reports that the final step in the ion-exchange actinide separation has proved successful with alpha-hydroxyisobutyric acid rather than lactic acid as eluant (Refs. III-1 and -2). A Dowex 50 x 8 resin column, 1 centimeter by 10 centimeters, was used initially to study this system. A 0.4M solution of alpha-hydroxyisobutyric acid was prepared and the pH was adjusted to 4.48 with ammonium hydroxide. The column was heated to 87° C and then pretreated with 0.2M ammonium chloride to give the ammonia form. Americium was subsequently loaded into the column and eluted with the alpha-hydroxyisobutyric acid solution. Samples were periodically obtained and their activity counted in order to establish the elution curve.

A similar experiment was run utilizing a column 0.2 centimeter by 10 centimeters loaded with Dowex 50 x 12 resin. Figure III-13 shows this elution curve. Variations in column dimensions and flow rates had no apparent effect. As reported by other workers (Ref. III-2), americium eluted after at least eight column volumes of eluant in each case.

c. Design, fabrication and equipment installation

Americium processing. Most of the equipment for the processing of americium has been installed; however, the installation of operable solenoid valves is delaying completion of the americium line. Receipt of acceptable valves will be followed by a rapid assembly of the processing apparatus.

Liquid-waste storage. The installation of the liquid-level apparatus in the liquid-waste storage system will enable operating personnel to sample and subsequently determine the discharge rate of the liquid to the county sewer system.

Curium-shipping cask. The curium-shipping cask specifications provide for a well-defined, safe piece of equipment. Performance tests delineated in the specifications will ensure a maximally safe cask capable of dissipating capsule heat with efficiency sufficient to prevent thermal damage to the capsules at any time during containment.

Curium processing. The layout of the curium facility will be initiated as soon as the process specifications have been completed. The final layout will also be affected by the containment philosophy. A number

Fig. III-12. Elution of Americium with Lactic Acid

Fig. III-13. Elution of Americium from Dowex 50 with Alpha-Hydroxyisobutyric Acid

of alternate designs have already been projected; further design awaits establishment of the most desirable criteria.

4. Conclusions

a. Americium processing

Purification. The thiocyanate procedure for the purification of americium has proved successful at other installations. Experimental work already completed indicates this method to be adaptable to americium purification for the irradiation program. However, the literature does not indicate the number of column volumes necessary to achieve satisfactory decontamination of americium. Results obtained in Martin experiments show the necessity for refinement of the procedure, especially in achieving improved decontamination with less eluant.

Encapsulation. The fabrication of aluminum-amerium oxide capsules by the present method has proved most successful, as evidenced by preliminary checkout runs in which dummy capsules were produced. Various modifications implemented to optimize the encapsulation process are as follows:

- (1) The type of aluminum used for the capsule shell was changed to a stronger aluminum alloy.
- (2) A void was incorporated in the capsule, and the core density will be maintained between 85 and 90% of theoretical to prevent excessive helium- and oxygen-pressure buildup.

b. Curium processing

The curium separation procedure appears satisfactory. Either lactic acid or alpha-hydroxyisobutyric acid can be used as eluant for the final ion-exchange separation. However, further work should be performed when curium is made available for tracer studies.

C. SUBTASK 6.3--RADIOISOTOPE FUEL FORM DEVELOPMENT

Investigations were made into the preparation of plutonium carbide (for fuel forms) from plutonium oxide and carbon. Because of the similarity between the compounds of plutonium and cerium, the cerium oxide-carbon system was studied for the purpose of determining optimum reaction conditions and parameters. It was found that almost complete conversion to ceric carbide was achieved if 35% excess carbon was reacted with ceric oxide at a temperature of 1640° C for 16 hours.

Studies were performed to evaluate various matrix materials and to determine a procedure for obtaining a curium fuel core with the proper density. It was found that the lower melting metals had the disadvantage of being susceptible to shrinkage during heat treatment at temperatures in the range required for thermoelectric generators. Such volume shrinkage may constitute a problem in that thermal contact with the cladding would be reduced. Materials such as tantalum and columbium actually undergo slight expansion at thermoelectric temperatures and therefore appear quite attractive for matrix applications.

Compressive-drop impact tests (at temperatures of 1100° and 1700° F) were conducted on composite capsules fabricated from different materials and containing aluminum. (Aluminum is molten at 1700° F.) Type 316 stainless steel exhibited the best performance of all the materials tested; although it suffered the greatest deformation, at no time was it observed to rupture. Compressive-drop tests were also run on solid specimens to evaluate the possibility of a correlation with high-velocity bazooka tests performed at Aberdeen Proving Ground. Recent data obtained from the bazooka tests indicated that high-velocity deformation may be independent of temperature for those materials in which the yield strength is relatively independent of temperature; this lends credence to the hypothesis that high-velocity deformation is related to yield strength. Development work was continued on the fabrication and welding of tantalum liners for fuel capsules.

Air oxidation tests on various container materials at 1400°, 1700° and 2000° F indicated that Hastelloy C was the most oxidation-resistant of all the materials tested. Specimens from the sea water tests were evaluated, and it was found that pitting corrosion was predominant for all specimens tested. Hastelloy C again exhibited the best corrosion resistance; however, the slight attack which did occur was not expected and an investigation is under way to disclose any anomalous environmental conditions.

1. Objectives

The objectives of Subtask 6.3 during the quarter are listed as follows.

a. Curium and plutonium fuel form development

The objectives of this phase of the program were:

- (1) To simulate the production of plutonium and curium carbides from the oxides by employing rare-earth materials and to subsequently establish specifications for these processes.

- (2) To investigate the compatibility of matrix and container materials with the carbide compounds.
- (3) To investigate the effects of compacting pressure on the density of cermets containing selected matrix metals and various percentages of simulated isotope additions.

b. Containment of fuels

The objectives of the fuel-containment studies during the quarter may be listed under the following test efforts:

- (1) Compressive-impact testing--to determine the effectiveness of various alloys in containing simulated isotopes when compressively impacted and to determine the effect of container wall thickness on containment; to determine how the compressive-impact deformation of solid metal cylinders is related to the velocity and weight of the incident ram.
- (2) High-velocity impact testing--to aid in modifying high-velocity impact tests and to evaluate results of the tests.
- (3) Container development--to develop procedures for making tantalum liners for the containment of plutonium metal.
- (4) Corrosion testing--to continue the sea water corrosion testing programs and to study air oxidation of selected alloys at high temperatures.

c. Design, fabrication and equipment installation

The objective of this phase of Subtask 6.3 for the quarter was to design and procure equipment for transforming radioisotopes into appropriate fuel forms.

2. Scope of Work

a. Plutonium fuel form

Effort this quarter was directed toward producing carbides from oxides. Since plutonium carbide is very similar to ceric carbide, the ceric oxide-carbon system was investigated.

Compacting studies were conducted to determine the cermet densities which can be obtained as a function of pressure with the more ductile metal powders. Sintered densities of several refractory metals were obtained.

b. Containment of fuels

Compressive-impact testing. Impact tests were performed on capsules $3/4$ inch in diameter and $1-1/2$ inches in length, as shown in Fig. III-14, which were fabricated from Type 316 stainless steel, Haynes 25, Hastelloy B, Inconel X and Rene 41. These capsules were filled with aluminum and seal welded. Tests were run at 1100° and 1700° F. Also, weights of from 9.5 to 137.5 pounds were dropped from heights of 2, 4, 6 and 8 feet upon solid copper specimens $3/8$ inch in diameter and $3/4$ inch long to determine the relation between deformation of the specimen and velocity and weight of the ram.

High-velocity impact tests. An impact test with the specimens propelled by bazooka rockets was devised and evaluation of the initial tests was made.

Container development. Procedures for making tantalum liners for containment of plutonium metal were developed.

Corrosion testing. Oxidation testing of the four alloys Rene 41, Hastelloy C, Haynes 25 and A-286 has been completed at temperatures of 1400° , 1700° and 2000° F in slow-moving, untreated air. Oxidation rates are being compared, together with physical and metallographic examination of the oxidized specimens. Sea water test specimens were removed from the tank and evaluated as to weight loss and localized pitting.

c. Design, fabrication and equipment installation

Equipment design on this phase of Subtask 6.3 awaits completion of the process specifications for the appropriate fuel forms.

3. Technical Discussion and Evaluation

a. Curium and plutonium fuel forms

Cerium dicarbide has been prepared from ceric oxide and carbon by utilizing the proper conditions of temperature and time and an excess of carbon. A series of pellets were made, some with a stoichiometric amount of carbon and some with 25% excess carbon with ceric oxide. Pellets were sintered at 1640° C for 2, 4 and 8 hours. Pellets containing 25 and 35% excess carbon were also sintered at 1640° C for 16 hours. Samples from the series were analyzed by X-ray diffraction. All of the specimens for examination were prepared under an argon atmosphere with a dew point of -93° F. Table III-1 shows the results of the tests.

All dimensions in inches

Fig. III-14. Design for Simulated Capsule

TABLE III-1
Cerium Dicarbide Conversion Studies

<u>Amount of Carbon</u>	<u>Amount of CeC₂ Conversion (%)</u>			
	<u>2-hour Sintering</u>	<u>4-hour Sintering</u>	<u>8-hour Sintering</u>	<u>16-hour Sintering</u>
Stoichiometric	None	>5	35 ± 15	--
25% excess	>5	35 ± 15	70 ± 20	95 ± 5
35% excess	--	--	--	95 ± 5

From the table it can be seen that at least 8 hours will be required to obtain good carbide. Densities only 59% of theoretical were obtained on the pellets containing 25% excess carbon. Since the X-ray data indicated a very high percentage of cerium dicarbide, the low density is attributed to the carbon dioxide evolved during the formation of the carbide in accordance with the reaction:

Therefore, the pellets must be crushed and resintered to obtain higher densities.

The observed improvement in conversion to the cerium dicarbide is attributed to an increase in reaction temperature from 1550° to 1640° C and the addition of excess carbon. The cerium dicarbide pellets (as a substitute for plutonium carbide) will be tested for compatibility with various container materials at various temperatures in the range expected for thermoelectric generators.

The preparation of gadolinium carbide will be investigated in a manner similar to that of cerium dicarbide. The gadolinium carbide (to simulate curium carbide) will be crushed and checked for compatibility with various matrix materials in the temperature range of thermionic generators. The material will be tested in cermet form.

Density studies were performed on various matrix materials containing uranium dioxide as a substitute for curium oxide. In this case uranium dioxide is employed for the purpose of simulating only the bulk or particle

characteristics of curium oxide and is not to be construed as being chemically similar. Two approaches for obtaining cermets of high density were used:

- (1) Compacting at high pressures.
- (2) Compacting at low pressure, sintering and coining.

Since it may be advisable to have porosity in the cermet to contain the helium released from the isotope during decay, it is believed that bodies which are compacted and not sintered may be satisfactory as fuel forms. Table III-2 gives the effect of compacting pressure on the density of several matrix materials with 20 volume percent uranium dioxide.

TABLE III-2
Effect of Compacting Pressure on Density of Cermets *

Compacting Pressure (tsi)	Density (% of theoretical)			
	<u>Nickel</u>	<u>Tantalum</u>	<u>Columbium</u>	<u>Molybdenum</u>
25	--	--	--	71.4
30	--	--	--	--
40	83.2	--	--	--
50	--	76.3	83.5	76.4
60	85.6	79.4	85.0	80.0
70	86.5	81.9	87.7	--
80	88.6	83.2	88.8	82.0
90	90.8	85.5	89.6	--
100	91.3	86.4	86.6	85.4
110	90.4	--	--	--

* Cermets composed of 20 volume percent UO_2

Table III-3 gives the results of a test performed to determine whether the concept of compacting without sintering was feasible. Ductile metals which were compacted and heat treated at a temperature slightly above the top range of thermoelectric generators were checked for density. It is obvious that nickel, or probably any low-melting metal, would not be usable as compacted since it becomes more dense during treatment. Such density increase and volume decrease would occur during operation and would eliminate thermal contact with the clad. Tantalum or columbium, however, would be satisfactory as compacted. The slight expansion found with tantalum or columbium would, in fact, be advantageous in obtaining better thermal contact. Unfortunately, tantalum and columbium may not be as satisfactory from a burnup standpoint as the lower-melting metals.

In the thermionic temperature range, the use of compacted and unsintered specimens would also be undesirable. Table III-4 gives the results of sintering and coining molybdenum and tantalum at 1700° C (3092° F). This sintering temperature was chosen because it is within the range of the proposed thermionic generators. It can be seen that the density of molybdenum was considerably increased at this temperature and the density of tantalum, only slightly, indicating that sintering and coining will be required for cermets for thermionic application.

b. Containment of fuels

Compressive-impact testing. The results of impact tests on simulated capsules are shown in Table III-5; Figs. III-15 and III-16 show the cross sections of the tested specimens. It can be concluded from Table III-5 that Type 316 stainless steel is the only material which, as a capsule of 1/8-inch wall thickness, contained the molten aluminum at 1700° F even though it suffered the most deformation. Since the Hastelloy B and Rene 41 specimens with the same wall thickness ruptured, these materials may not be suitable as containers. Haynes 25 and Inconel X could be considered useful as container materials when used in thick-walled containers. These conclusions are contingent on the results of high-velocity impact tests.

TABLE III-3
Effect of Heat Treating on Cermet Density

<u>Composition of Cermet Material</u>	<u>Green</u>	<u>First Heat Treatment, 1975°F for 1 hour</u>	<u>Density (% of theoretical)</u>		<u>Second Coin</u>	<u>Third Heat Treatment, 1975°F for 1 hour</u>	<u>Third Coin</u>
			<u>First Coin</u>	<u>Second Heat Treatment, 1975°F for 1 hour</u>			
80 vol % Ni+20 vol % UO ₂ pressed and later coined at 100 tsi	91.3	94.5	97.2	97.2	97.6	97.2	98.3
60 vol % Ni+40 vol % UO ₂ pressed and later coined at 100 tsi	89.8	91.3	95.1	95.1	96.7	96.4	96.8
80 vol % Ta+20 vol % UO ₂ pressed and later coined at 100 tsi	86.4	85.1* 85.8**	87.5	86.0	86.9	86.0	85.9
60 vol % Ta+40 vol % UO ₂ pressed and later coined at 120 tsi	87.4	84.5* 86.8**	88.2	87.0	87.5	***	***
80 vol % Cb+20 vol % UO ₂ pressed and later coined at 90 tsi	89.6	87.8	90.7	90.1	90.5	89.6	90.2
60 vol % Cb+40 vol % UO ₂ pressed and later coined at 90 tsi	88.9	85.8* 89.0**	88.7* 90.9**	88.7	89.3	89.5	89.5

*Charged slowly into furnace
 **Fast charge into furnace
 ***Specimen spalling

TABLE III-4

Effects of Sintering and Coining on Density of Cermets

<u>Cermet Material</u>	<u>Com- pacting Pressure (tsi)</u>	<u>Green Density</u>	<u>Density (% of theoretical)</u>				
			<u>Sintered in Vacuum</u>	<u>Sintered in Argon</u>	<u>Coined at 50 tsi</u>	<u>Coined at 60 tsi</u>	<u>Coined at 70 tsi</u>
Ta	50	74.6	76.6	--	77.5	79.9	81.3
Ta+5% UO ₂	50	75.6	76.0	74.3	78.8	80.1	81.9
Ta+10% UO ₂	50	77.4	79.8	77.4	81.1	83.0	84.1
Ta+20% UO ₂	50	74.9	76.1	--	78.2	79.1	81.5
Mo	50	74.8	84.2	--	91.8	--	--
Mo+5% UO ₂	50	76.4	84.5	86.3	93.2	--	--
Mo+10% UO ₂	50	75.8	85.0	85.2	92.7	--	--
Mo+20% UO ₂	50	76.4	84.6	83.5	91.8	--	--

Sintering temperature--1700° C

Sintering time--1 hour

TABLE III-5
Effect of Impact on Simulated Capsules

<u>Material</u>	<u>1/8-inch Wall Thickness</u>		<u>3/16-inch Wall Thickness</u>	
	<u>Length Deformation (%)</u>	<u>Remarks</u>	<u>Length Deformation (%)</u>	<u>Remarks</u>
Specimen temperature--1700° F*				
316 SS	61.3	No failure	22.9	No failure
Hastelloy B	46.1	Wall ruptured	25.1	No failure
Haynes 25	39.4	Ruptured at weld	23.1	No failure
Rene 41	34.9	Wall ruptured	11.2	No failure
Inconel X	20.0	Ruptured at weld	16.1	No failure
Specimen temperature--1100° F**				
316 SS	39.5	No rupture	16.6	No rupture
Rene 41	20.4	Slight rupture at weld	10.3	No rupture
Haynes 25	20.4	No rupture	13.1	No rupture
Hastelloy B	18.9	No rupture	14.1	No rupture
Inconel X	10.2	No rupture	5.3	No rupture

Specimen weight--137.5 pounds

Height of drop--8 feet

* These specimens were not aged prior to testing as was normally done. Previous data on materials listed showed that aging increased the tendency to fracture on all materials except 316 stainless steel, so the specimens with 3/16-inch wall thickness would probably have failed if aged.

** Specimens tested at 1100° F were aged for 40 hours at 1100° F before testing.

316 stainless steel

Haynes 25

Hastelloy B

1/8-in. wall

3/16-in. wall

1/8-in. wall

3/16-in. wall

1100° F

1700° F

Aluminum plug

Fig. III-15. Simulated Container Specimens

Inconel X

Rene 41

1/8-in. wall

3/16-in. wall

1/8-in. wall

3/16-in. wall

1100° F

1700° F

Aluminum plug

Fig. III-16. Simulated Container Specimens

In determining the relation between impact deformation and the weight and velocity of the incident ram, weights of various masses were dropped from various heights. The percent deformation was plotted against the weight dropped, and a family of curves was drawn for each drop height. It was assumed that the amount of energy imparted to the specimens was constant for a given deformation. Figure III-17 shows the curves plotted from the test data. The energies were calculated at deformations of 25, 35, 45 and 55%. Velocities were calculated from the formula

$$V^2 = 2gs$$

where

V = velocity

g = acceleration due to gravity (32 fps²)

s = the distance the ram fell (2, 4, 6, 8 feet).

Table III-6 gives the energies calculated at the various deformations. It can be seen in Table III-6 that the deformation is essentially a function of the square of the velocity; it may, therefore, be considered a function of the kinetic energy of the ram, at least with the rates of loading shown in the table.

Calculation of the kinetic energy of the specimens in the high-velocity impact tests indicates that considerably less energy is required in these tests to produce deformation similar to that of the compression-impact rate.

TABLE III-6

Dependence of Deformation on Velocity and Weight Dropped *

Length Deformation (%)	Drop Height (ft)	W = Weight (lb) V = Velocity (fps)					
		<u>W</u>	x	<u>V²</u>	= <u>WV²</u>	<u>W</u>	x <u>V</u> = <u>WV</u>
25	8	11	x	512	= 5620	11	x 22.6 = 248
	6	13	x	384	= 4980	13	x 19.1 = 248
	4	19	x	256	= 4860	19	x 16.0 = 304
	2	40	x	128	= 5120	40	x 11.3 = 352

TABLE III-6 (continued)

Length Deformation (%)	Drop Height (ft)	<div>W = Weight (lb)</div> <div>V = Velocity (fps)</div>									
		<u>W</u>	x	<u>V²</u>	=	<u>WV²</u>	<u>W</u>	x	<u>V</u>	=	<u>WV</u>
35	8	18.5	x	512	=	9470	18.5	x	22.6	=	418
	6	23.0	x	384	=	8840	23.0	x	19.1	=	439
	4	32.5	x	256	=	8330	32.5	x	16.0	=	520
	2	100.0	x	128	=	12800	100	x	11.3	=	1130
45	8	26	x	512	=	13300	26	x	22.6	=	588
	6	35	x	384	=	13400	35	x	19.1	=	688
	4	53	x	256	=	13600	53	x	16.0	=	848
55	8	36.5	x	512	=	18700	36.5	x	22.6	=	825
	6	31	x	384	=	19600	51	x	19.1	=	974
	4	95	x	256	=	21860	95	x	16.0	=	1520

* Temperature = 76° F

High-velocity impact tests. High-velocity tests, for which the head was designed to prevent the rocket from crushing the specimen, were found successful in obtaining an angle of impact within the tolerance required.

Tests were performed on nine specimens of Type 316 stainless steel, each 3/4 inch in diameter and 1-1/2 inches in length. The test temperatures were approximately 1100°, 1400° and 1700° F; test velocities were 250, 500 and 650 fps. A speed of 750 fps was desired as the highest test speed but could not be obtained. Type 316 stainless steel was chosen as the initial test material because it exhibited no irregular (cracking or shattering) failure in the compressive-impact tests. Figure III-18 shows the specimens after impact, and Table III-7 gives the percent deformation. Examination of the specimens indicated that the test temperature had no apparent effect on the deformation of the specimens and that deformation changed only with velocity. The deformation of the specimens at 650 fps was unreliable because the specimen holders "mashed" into the specimen, causing additional deformation.

Fig. III-17. Room-Temperature Impact Test on Annealed Copper Rod (3/8-inch diameter, 3/4-inch length)

Fig. III-18. High Velocity Impact Test Specimens

TABLE III-7
High-Velocity Test Deformation of Type 316 Stainless Steel

<u>Temperature (°F)</u>	<u>Velocity (fps)</u>	<u>Deformation (%)</u>
1100	250.1	3.87
	500.0	15.43
	668.9	54.5*
1400	250.1	4.88
	500.03	14.8
	663.2	63.4*
1700	250.1	14.8
	510.2	17.4
	637.0	63.6*

* Results are not accurate because rockets contacted the specimens upon impact and caused additional deformation.

On analyzing the results of the tests, it was evident that either the time duration of impact was too short for the temperature to affect deformation or the deformation was a function of some property which did not change appreciably over the temperature range studied. A review of the properties of Type 316 stainless steel showed that the yield strength was the only property which changed very little over the temperature range from 1100° to 1700° F. Therefore, it is believed that high-speed impact deformation may be related to yield strength.

To determine the validity of the hypothesis that the deformation is a function of yield strength and whether a correlation exists between the bazooka and compressive-impact tests, it will be necessary to conduct further tests. It is recommended that Inconel X, which varies in yield strength by one order of magnitude over the temperature ranges considered, be evaluated and that specimens of different lengths be tested to determine the effect of mass on the amount of deformation.

Container development--cylindrical. Cylindrical tantalum liners (1 inch in diameter and 1-1/2 inches in height with concave, dished ends) were fabricated. The side wall of the container was made from 0.025-inch sheet. The sheet was sheared and rolled into a tube, and the seam was welded by the Tungsten Inert Gas (TIG) process; a copper chill and shaping jig were used. The ends were etched in an acid solution, assembled and welded.

Full penetration was achieved on the seam weld, and approximately 0.020-inch penetration was obtained in the end welds. Figure III-19 shows the components of the tantalum liner and the finished liner.

Liners similar in design to that of Fig. III-19 can be produced in a range of sizes limited only by the welding equipment. Further tests are necessary to determine the temperature rise during welding at various distances from the weld so as to evaluate the possibility of thermal effects on the contained fuel. Therefore, liners will be made and loaded with cerium or aluminum metal as temperature indicators if melting occurs. By this means the void required over the plutonium metal can be determined. These tests will also determine the fit required with the chill block to prevent melting of the metal.

Corrosion testing. The curves obtained in the weight-gain oxidation tests are presented in Figs. III-20 through III-22. All of the alloys tested show a rapid initial gain in weight and then a slower rate of oxidation after about 20 to 40 hours, depending upon the temperature. All of the curves except those for Haynes 25 at 1400° and 2000° F illustrates a parabolic rate law predicted by the Pilling-Bedworth Principle which argues that the initial oxide film formed should be compact and protective. Continued oxidation and thickening of the oxide film is assumed to be diffusion of oxygen inward toward the metal (Ref. III-9). For complex alloys of the type studied in this investigation, the rate-controlling process might be diffusion of some readily oxidized metallic ion to the surface of the alloy. Given sufficient time for the law to apply, the equation for weight gain is:

$$W^2 = A + kt$$

where

W = weight gain (mg/cm^2)

A = constant ($(\text{mg}/\text{cm}^2)^2$)

k = constant ($(\text{mg}/\text{cm}^2)^2/\text{hr}$)

t = time (hr).

Fig. III-19. Fuel Capsule Tantalum Liner Fabrication

Fig. III-20. Weight Gain Versus Time at 1400° F

Fig. III-21. Weight Gain Versus Time at 1700° F

Fig. III-22. Weight Gain Versus Time at 2000° F

The general shape of the oxidation curve of Haynes 25 at 1400° and 2000° F appears to be logarithmic. The direct logarithmic law is

$$W = B + \log (\alpha t + 1)$$

where

$$B = \text{constant (mg/cm}^2\text{)}$$

$$\alpha = \text{constant (hr}^{-1}\text{)}.$$

A discussion of various theories predicting logarithmic oxidation is presented in Ref. III-10. In general, this form of oxidation proceeds by diffusion through pores, slowed by potential gradients with neighboring ions or atoms. The parabolic law is normally obeyed if the temperature is high enough to permit thermal movement in the absence of a potential gradient.

Physical examination of the specimens oxidized at 1400° F indicated that a thin, adherent oxide coating was formed on all the alloys except Hastelloy B, which formed a thick oxide coating that spalled off when cooled to room temperature. At 1700° F, all the specimens formed an adherent oxide coating except alloy A-286, which spalled in localized areas. At 2000° F, there was evidence that some of the oxide coating may have flaked from the specimens while they were still in the hot zone of the furnace. Hastelloy C was speckled with small bits of silvery material, assumed to be a constituent whose oxidation was retarded. Rene 41 formed an oxide coating which spalled completely while cooling, leaving unoxidized metal. A thin scale spalled in localized areas from Haynes 25 when cooled while the oxide coatings formed on Hastelloy C and A-286 did not spall upon cooling.

Metallographic investigations of the oxidized specimens showed uniform penetration of metal for all alloys oxidized at 1400° and 1700° F; however, at 2000° F all the specimens showed deep infiltration of oxide beneath the oxide coating.

Direct comparison of oxidation data is difficult because of the many existing experimental variables. Little data is available for the particular alloys under test; however, oxidation rates have been determined by the National Bureau of Standards for several experimental alloys (Ref. III-11). The oxidation rate of Nichrome V after 100-hour exposure in slowly moving air at 2000° F is presented in Fig. III-23. Nichrome is used in high temperature heating wire because of its good oxidation properties. The rate of oxidation of Hastelloy C compares favorably with that of Nichrome V at 2000° F.

Fig. III-23. Weight Gain Versus Time at 2000° F for Nichrome Alloy

The oxidation rates obtained in this work correspond only to the conditions under which the tests were conducted and should not be used to predict what may happen under other conditions. Oxidation rates generally increase rapidly where specimens are subjected to fluctuating temperatures, moisture and physical stresses.

In selecting the alloy with the best oxidation properties, the type of oxidation curve is most important. The same material may obey different oxidation-rate laws at different temperatures. Evans (Ref. III-10) points out that a material whose oxide thickens according to the parabolic law often is not so acceptable as one which oxidizes by one of the logarithmic laws, generally bringing oxidation almost to a standstill at a small thickness. The risk of flaking and cracking, followed by another rapid gain in weight, increases with increased oxide film. This suggests that oxidation tests of Haynes 25 and Hastelloy C be conducted for a longer period of time--possibly 1000 hours--at the exact temperature that the containment material in a thermoelectric generator would see in case of an abort to establish the validity of these laws.

Sea water corrosion. Test specimens were removed from the sea water tank and thoroughly cleaned to remove surface salt deposits. The specimens were weighed and examined under a microscope for crevice and stress corrosion. Weight loss, crevice corrosion and stress corrosion are summarized in Table III-8. Calculations of surface corrosion rates from the weight losses are meaningless because of the localized pitting characteristic of sea water attack. This pitting was observed on all test specimens, and in one case, with Rene 41, a pit perforated the tubing.

Crevice corrosion occurs within, or adjacent to, a crevice formed by contact between pieces of the same or different materials. In this test, the crevice was formed at the edge of the plastic insulation used to isolate the immersion heaters from sea water. As was evident upon examination of the test specimens, the intensity of attack is more severe in the crevice than on surrounding areas of the same surface. Photographs now being prepared show the pitting which results from the crevices on each test specimen. The mechanism of crevice corrosion involves cell potential differential from either metal-ion or oxygen concentration. The crevice formed by a closed stagnant area between two materials is supplied with water and dissolved salt through capillary action. As a result of the reaction, the crevice surfaces become anodic, with resultant dissolution of metal.

In this testing program, both surface bubbles and surface salt deposits were observed. Surface bubbling was possibly a result of the hot-wall effect often observed on the water side of heater tubes. Often pitting takes place where bubbles of gas repeatedly separate from the water

(usually at scratches or sharp indentations) (Ref. III-12). Hot spots formed on the heated metal surface at those points where the gas bubbles adhere to, and thereby prevent cooling of, the metal surfaces. The formation of salt deposits on the sides of the metal specimens would probably influence the corrosion rates. It is suspected that the initial deposit of salt begins at the crevice edges or at the localized hot spots and spreads over the metal surface.

TABLE III-8
Summary of Results of Corrosion Tests

<u>Material</u>	<u>Length of Time (days)</u>	<u>Total Weight Loss (mg)</u>	<u>Appearance of Crevice Area</u>	<u>Maxi- mum Pit in Crevice Area (in.)</u>	<u>Appearance of Stress Area</u>
Haynes 25	170	4.7	One deeply pitted area	0.021	Deep pitting in weld bead
A-286	134	101.2	Entire crevice edge deeply pitted	0.059	Deep pitting in weld bead and slight attack along edge of weld
Rene 41	71	12.2	Localized deep pitting	0.028	Deep pitting in weld bead and slight attack along edge of weld
Hastel- loy B	134	77.4	Entire crevice area shows corrosion	0.012	Deep pitting in weld bead and along edge of weld
Hastel- loy C	134	2.6	One very small area slightly pitted	0.002	Slight inci- pient attack over a small area

4. Conclusions

a. Curium and plutonium fuel form

Cerium carbide (to simulate plutonium carbide) can be produced by utilizing correct time and temperature and an excess of carbon with cerium oxide. Crushing and resintering is required to produce high-density compacts.

Cermets containing curium compounds cannot be satisfactorily prepared by compacting without sintering as they will become more dense during operation and cause poor thermal contact with the container. Cermets must be sintered and coined prior to encapsulation.

b. Containment of fuels

Compressive-impact deformation appears to be a function of the kinetic energy of the incident ram; however, deformation in the higher-speed bazooka tests occurs with much less energy input.

High-velocity impact tests on 316 stainless steel indicate that the temperature of the test has little effect on the deformation. The only property of 316 stainless steel which shows very little change in the test temperature range is the yield strength. Further tests are required to determine the validity of the hypothesis that high-speed impact deformation is a function of yield strength.

Sea water corrosion of Hastelloy C was unexpected; it is possible that anomalous environmental conditions during the test exerted an adverse effect on this normally corrosion-resistant material. Consequently, effort will continue in which the previous test conditions will be rigorously analyzed, and, subsequently, Hastelloy C specimens will be further tested to verify the above results.

D. SUBTASK 6.4--SAFETY THERMAL AND SHIELDING ANALYSIS

High-velocity impact tests at Aberdeen Proving Ground (APG) were monitored under this effort to assure maintenance of desired test parameters and to provide documentary coverage of the overall program. Several equipment design changes were required before the test specimens impacted within the desired angle of incidence.

The gamma and neutron dose rates associated with the following areas of study were investigated to determine where radiation levels are above tolerance limits:

- (1) Americium processing.
- (2) Plutonium metal production.
- (3) "Getter" materials in irradiation capsules.
- (4) Type 5052 aluminum as capsule material.
- (5) Shipping casks for irradiated capsules.

Shielding requirements for these areas were then calculated.

Thermal analyses were conducted on the irradiation samples employing worst-case conditions during the irradiation, during storage in the reactor canal and during transportation to Martin's Quehanna hot-cell facility.

1. Objectives

The objectives of Subtask 6.4 for the past quarter are listed under their respective work areas.

a. Safety analysis for radioisotopes

The objectives of this phase of the subtask were:

- (1) To monitor the high-velocity impact materials test program performed at Aberdeen Proving Ground (APG).
- (2) To initiate a report on the safety evaluation of Curium-242 to be performed at Quehanna.

b. Shielding analysis

The shielding analysis program had the following objectives for the quarter:

- (1) To investigate dose rates prevailing during purifying, blending and pelletizing of americium into slugs suitable for irradiation.
- (2) To investigate dose rates prevailing during the processing required for production of Plutonium-238 and -239 metal.

- (3) To investigate the radioactivity levels involved when cerium and cesium metals are used as getters for the oxygen liberated when americium dioxide is irradiated with neutrons.
- (4) To investigate dose rates from the americium fuel slugs when 5052 aluminum cladding is substituted for 1100 aluminum.
- (5) To determine dose rates prevailing outside the curium-shipping cask at various slug-cooldown times when the cask is loaded with three irradiated slugs; also, to determine from this information the minimum cooldown time so as to meet the ICC regulations for shipping radioactive materials.
- (6) To compare calculated dose rates with available measured dose rates.

c. Heat transfer analysis

The objective of this phase of Subtask 6.4 was thermal analysis of the americium capsule under the following conditions:

- (1) During irradiation in the Engineering Test Reactor (ETR).
- (2) During underwater storage in the ETR cooling canal.
- (3) During transportation in a shipping cask.

2. Scope of Work

a. Safety analysis for radioisotopes

A test program was devised to measure the effects on several container materials of high-velocity impact with a rigid target. These high-velocity impact tests were performed on a supersonic missile tract at APG. Documentary film coverage was provided for the test setup and runs.

A report entitled "Safety Evaluation of Curium-242 Processing To Be Performed at the Quehanna Site" was initiated during this quarter. It is divided into sections as follows:

- (1) Properties of Curium-242.
- (2) Site and facility description.
- (3) Ventilation system.

- (4) Radioactive-waste system.
- (5) Design criteria.
- (6) Preprocessing operation.

Approximately 30% of the work to be performed on a final report has been completed.

b. Shielding analysis

Americium processing. An investigation of the fuel-slug production line was made to determine areas where operating personnel might experience excessive exposure to radiation. When these areas were found, methods of reducing the exposure were investigated.

Plutonium metal production. The plutonium-processing equipment was analyzed to determine problem areas. In addition to the shielding requirements for Plutonium-238 production, the process equipment was re-examined to determine shielding requirements for production of Plutonium-239 metal. The latter isotope is to be used when the equipment and processing method are checked out.

Cerium and cesium metals as getters. The amount of radioactivity produced when cesium and cerium are irradiated with thermal neutrons was determined. The effect of this additional radioactivity on the shielding of the shipping cask was then ascertained.

Substitution of 5052 for 1100 aluminum. An examination was made of the radioactivity produced by neutron irradiation of the alloying elements in 5052 aluminum; also, the effect of this additional radioactivity on specifications for the shipping container was determined.

Shipping-cask dose rates. Dose rates at intervals up to 30 days after removal of curium slugs from the reactor were determined. This information was then used to calculate the minimum cooldown time required to meet the ICC shipping regulations for radioactive material.

c. Heat transfer analysis

Irradiation. The heat generated in a capsule while under irradiation in a high-flux reactor was calculated. The profile temperature of the capsule was also determined.

Underwater storage in reactor canal. The decay heat of a capsule stored in the reactor canal subsequent to irradiation was calculated; the temperature profile of the capsule was also calculated.

Shipping cask for irradiation capsules. A heat-transfer analysis was made to determine criteria for the shipping cask. The rate of decay heat being generated in the capsules during transit was calculated. The shipping-cask configuration was analyzed to determine the maximum surface temperature of the capsules in the event of complete loss of water from the neutron shield.

3. Technical Discussion and Evaluation

a. Safety analysis for radioisotopes

For the high-velocity impact tests at APG, test specimens--right cylinders, 1.5 inches in length and 0.75 inch in diameter--were placed into cup-type holders and propelled by a bazooka-type rocket toward a rigid metal target. Initial test runs revealed that the specimens impacted on the target medium at an angle greater than 10 degrees from normal because a rotational moment was imparted to the specimens at the time of separation from the cup holder. A modification was made so that the specimen was contained in the holder until the entire assembly impacted on the target medium. Figure III-24 is a diagram of the modified test run used in performing the actual tests.

Nine runs were made in which 316 stainless steel specimens were impacted against a hardened steel plate. The cup holder and specimens were propelled through a 10-foot tube and traveled 30 inches before impacting against the target medium. The information recorded during the runs is tabulated in Table III-9.

TABLE III-9

Firing Record--Materials Impact Test

<u>Specimen Type No.</u>	<u>Length (in.)</u>	<u>Diameter (in.)</u>	<u>Mass (gm)</u>	<u>Preheat Tem- pera- ture (° F)</u>	<u>Time from Furnace to Impact (sec)</u>	<u>Impact Velocity (fps)</u>
4-2	1.490	0.749	86.0292	1740	28	637
4-3	1.497	0.750	86.5647	1770	28	570.2
4-4	1.500	0.749	86.5070	1750	35	250.1
4-5	1.498	0.751	86.5875	1550	28	663.2
4-6	1.498	0.750	86.3860	1500	27	500.03
4-7	1.496	0.750	86.3847	1500	24	250.1
4-8	1.498	0.749	86.3964	1230	20	668.9
4-9	1.499	0.751	86.6295	1220	18	Lost
4-10	1.496	0.750	86.2650	1220	18	250.3

Fig. III-24. Sled Tests at Aberdeen

b. Shielding analysis

Americium processing. An analysis was made on the americium-purification boxes. Dose rates were calculated for 100 grams of Americium-241 in two solution concentrations: 0.1 gram per milliliter and 1.18 grams per milliliter. Since it is assumed that the resin in the ion-exchange column occupies 50% of the volume, the effective americium concentrations must be halved in the columns.

With the exception of the bottles or flasks which feed solutions to the ion-exchange columns, all equipment in these boxes is currently shielded on the sides and in the rear with 1/4-inch sheet lead. The front of the equipment is shielded with plates of leaded glass 1 inch thick. Dose rates from gamma rays through 1/4 inch of lead are very low. At a distance of 1 foot from the center of the vessel, they range between 10^{-6} and 10^{-7} milliroentgen equivalent man per hour (mrem/hr) depending upon the concentration of the solution. Gamma-ray dose rates through the 1-inch lead glass are even lower, ranging between 10^{-8} and 10^{-9} mrem/hr at 1 foot.

Neutron dose rates were calculated for alpha-neutron reactions with oxygen in the ion-exchange columns. Dose rates for other vessels will be approximately the same. The alpha-neutron reaction with oxygen predominates until the dried americium dioxide powder is blended with aluminum.

Neutron dose rates are as follows:

Concentration of americium in column (gm/cm^3)	0.05	0.59
Rate at 1 foot (mrem/hr)	6.8	9.1
Rate at 2 feet (mrem/hr) (approximately at face of dry box)	1.7	2.3

Flask feeding ion-exchange column. Flask dimensions were assumed to be 4 centimeters, diameter, and 22 centimeters, length. It was also assumed that the flask contained 200 milliliters of liquid and two different concentrations of americium--0.5 gram per milliliter and 0.25 gram per milliliter. These concentrations correspond to 100 grams and 50 grams of americium, respectively.

Gamma-ray dose rates at the face of the dry box are as follows:

- (1) No shielding (self-absorption of column only):

<u>Americium Concentration</u> <u>(gm/cm³)</u>	<u>Dose Rate</u> <u>(mrem/hr)</u>
0.5	1.5×10^3
0.25	1.63×10^3

- (2) Shielded with 0.5-inch thickness of 3.3-density glass:

0.5	3.4×10^{-4}
0.25	3.8×10^{-4}

The higher dose rates prevailing with the lower americium concentration are due to the decrease in the self-absorption coefficient of the liquid, caused by the lower americium content. Neutron dose rates at the face of the box are within whole-body allowable dose rates without additional shielding:

<u>Americium Concentration</u> <u>(gm/cm³)</u>	<u>Dose Rate</u> <u>(mrem/hr)</u>
0.5	2.3
0.25	1.1

It is recommended that lead or lead glass (3.3-density) be added to the box in front of the flasks. If sheet lead is used, the minimum thickness should be 1.5 millimeters (0.59 inch). Minimum glass thickness is 0.25 inch. However, if glass is used, it would be advisable to add a 1-inch plate in order to have the same shielding as the other equipment in the box.

Dried americium dioxide powder. To determine the dose rates from the dried americium dioxide powder, it was assumed that 100 grams of the powder with a density of 1 gram per cubic centimeter was contained in a glass vessel 6 centimeters in diameter and 3.54 centimeters high. Gamma-ray dose rates are as follows:

	<u>Dose Rates (mrem/hr)</u>
(1) At surface of container:	
(a) Without shielding (self-absorption only)	2.4×10^5
(b) With lead gloves (0.5-millimeter lead equivalent)	6.3×10^3
(c) With 2 millimeters (0.080 inch) of lead around vessel	8
(2) One foot from center of vessel:	
(a) Without shielding	868
(b) With lead gloves	81
(c) With 2 millimeters (0.080 inch) of lead	0.1

Neutron dose rates are as follows:

(1) At container surface	710
(2) At 1 foot from center	8

It is recommended that 2 millimeters (0.080 inch) of lead sheet be added to the vessel containing the americium dioxide to reduce the gamma-ray dose rate to an insignificant value and that the vessel be handled with tongs to avoid excessive exposure from the neutrons resulting from alpha-neutron reactions with oxygen.

Flask used to blend americium dioxide and aluminum powder. The americium dioxide and aluminum powder are weighed and put into an aluminum flask. The ingredients are then tumbled in the flask to form a homogeneous mixture. When the blending operation is completed, the flask is transferred to the box surrounding the rams of the Denison Press where it is used as a hopper to feed the mixture to a compacting die. The following data were used to determine dose rates from the blending flask:

- (1) Flask dimensions--3.5 inches, diameter; 2-7/8 inches, length; 1/8 inch, aluminum wall thickness; 15.47 cubic inches, capacity.
- (2) Ratio of aluminum to americium dioxide--30:1.

- (3) Total americium content--14.3 grams in the form of the dioxide--enough to make four fuel slugs.
- (4) Composition of blended powder (gram per cubic centimeter)--americium, 0.031; oxygen, 0.0004; aluminum powder, 0.944.
- (5) Neutron production from alpha-neutron reactions in aluminum-- 1.29×10^6 neutrons per second; total, 2.85×10^3 neutrons per cubic centimeter per second (n/cm³-sec).
- (6) Dose rates from gamma rays (mrem/hr):
 - (a) At surface of flask (self-absorption + 1/8-inch aluminum)-- 6.1×10^4 .
 - (b) At surface of flask with 0.5-millimeter lead equivalent gloves-- 2.8×10^3 .
 - (c) At 4 inches from flask center with leaded gloves--720.
 - (d) At 8 inches from flask center with leaded gloves--160.
 - (e) At 12 inches from flask center with leaded gloves--75.
- (7) Dose rates from neutrons (mrem/hr):
 - (a) At container surface--650.
 - (b) At 4 inches from flask center--140.
 - (c) At 8 inches from flask center--36.
 - (d) At 12 inches from flask center--13.
 - (e) At 24 inches from flask center--3.2.
 - (f) At 12 inches from flask center with 6.6 inches of polyethylene shield--2.

Although the dose rates from gamma rays are high, they may be reduced quite readily by the addition of small amounts of lead sheet to the flask. Dose rates from neutrons, however, present problems while the flask is being handled. Distance and short handling times appear to be the best means of reducing the exposure to neutrons. It is

recommended that the flask be covered with a minimum of 2 millimeters (0.080 inch) of lead sheet to reduce the gamma-ray dose rate to negligible amounts when compared with the neutron dosages. Although the neutron dose rate at the surface is high, 650 mrem/hr, the flask can be held in the hands for about 2 hours before the weekly allowable dose to the hands (1500 mrem) is exceeded, provided the gamma dose rate is negligible.

While the flask is fastened to the press dry box, radioactivity in the flask can affect the eyes of the person operating the press. In view of the fact that the eyes are allowed to receive a dose of only 100 mrem in a 40-hour work week, it is necessary to shield the flask with about 6.6 inches of polyethylene to reduce the neutron dose rate to 2 mrem/hr at 1 foot.

Welding and sandblasting. After the fuel slug has been compacted in the press, the remaining end cap is welded in place and the outer surface of the slug is sandblasted (if required). During the time when the slug is removed from the press die and placed in the shipping container, a number of occasions will occur when the slug cannot be shielded. The dose rates from the unshielded slug were calculated, and the results are given in Table III-10. A comparison of these results with those reported in Ref. III-13 shows the present dose rates to be higher in most cases by a factor of about two. It will be assumed that the present results are the more nearly correct. In either case, the gamma-ray dose rates are easily reduced by small thicknesses of lead shielding.

TABLE III-10
Dose Rates from Finished Capsule

<u>Location</u>	<u>Neutrons</u>	<u>Dose Rate (mrem/hr)</u>	
		<u>Gamma Rays</u>	
		<u>Without Gloves</u>	<u>With Gloves</u>
At capsule surface	640.0	8.9×10^4	2×10^3
One foot from capsule center	3.9	1.3×10^3	45
Two feet from capsule center	1.0	320.0	11.0
Three feet from capsule center	0.43	140.0	5.1

During the welding operation the slug is placed in a hollow steel spindle with a wall thickness of about 7/16 inch. Dose rates were calculated for a separation distance of one foot, approximately the distance between the hollow spindle and the surface of the glove box. Dose rates are (mrem/hr):

- (1) From neutrons--3.2.
- (2) From gamma rays (without gloves)--7.2.
- (3) From gamma rays (with gloves)--0.42.

It is recommended that the fuel slug be placed in a lead sheath with a minimum thickness of 0.080 inch whenever possible to reduce the gamma-ray dose rate. When it is not possible or practical to use the lead sheath, it is recommended that the person handling the capsule wear leaded gloves and a leaded rubber apron. The leaded rubber apron is a necessity in performing the sandblast operation. The window of the sandblasting apparatus should be covered with a minimum thickness of 0.25 inch of 3.3-density lead glass.

Plutonium metal production. A shielding analysis was started for the various steps in the processing which leads to the production of plutonium metal. Dose rates were calculated on the basis of 50-gram batches of plutonium as the metal. Previous work has shown that the main problem area for shielding occurs when the plutonium is combined with fluorine. The dose rates listed in Tables III-11 and III-12 were based upon the tetrafluoride of Plutonium-238. The gamma-ray dose rates are applicable to other compounds of Plutonium-238; however, neutron dose rates will differ for each compound. The following dose rates were calculated by assuming that the tetrafluoride powder was contained in a vessel 4 centimeters in diameter and 7.5 centimeters high. The density of the powder was assumed to be 0.7 gram per cubic centimeter (one tenth of crystal density).

TABLE III-11
Gamma-Ray Dose Rates from $\text{Pu}^{238}\text{F}_4$

<u>Location</u>	<u>Dose Rate (mrem/hr)</u>
At surface--no shielding	4.7×10^3
At one foot from center--no shielding	28.3

TABLE III-11 (continued)

<u>Location</u>	<u>Dose Rate (mrem/hr)</u>
At one foot from center with 0.05 centimeter of lead (leaded gloves)	5.0
At one foot from center with 1/8 inch of lead	2.5
At one foot from center with 1/4 inch of lead	1.8
At one foot from center with 1/2 inch of lead	1.0
At one foot from center with 1/4 inch of 3.3-density lead glass	3.8
At one foot from center with 1/2 inch of 3.3-density lead glass	2.6
At one foot from center with 3/4 inch of 3.3-density lead glass	2.1

TABLE III-12Neutron Dose Rates from $\text{Pu}^{238}\text{F}_4$

<u>Location</u>	Dose Rate (mrem/hr)				
	No Shielding	Polyethylene Shield Thickness (in.)			
		<u>4</u>	<u>8</u>	<u>12</u>	<u>16</u>
At surface	7.0×10^5	--	--	--	--
At 1 foot from center	5.8×10^3	1.85×10^3	5.9×10^2	--	--
At 2 feet from center	1.4×10^3	4.5×10^2	1.4×10^2	42	15
At 3 feet from center	6.4×10^2	2.1×10^2	66	21	7

It is recommended that the equipment used to process Plutonium-238 be shielded with at least 1/8 inch of lead on the sides and rear. The front should be shielded with a minimum of 0.5 inch of 3.3-density lead glass. Fluorine compounds of plutonium should be shielded from the operator with 16 inches of polyethylene.

Dose rates prevailing when Plutonium-239 is being processed were investigated. The largest dose rates are, again, encountered when the fluorine compound is formed. However, because of the longer half life of this isotope, dose rates are considerably lower than the dose rates prevailing with Plutonium-238. Dose rates from neutrons generated in a 50-gram batch of Plutonium-239 combined with fluorine are shown in Table III-13.

TABLE III-13

Neutron Dose Rates from 50 Grams of $\text{Pu}^{239}\text{F}_4$

Polyethylene Shield (in.)	Dose Rates (mr/hr)		
	Distance from Center of Container (ft)		
	<u>1</u>	<u>2</u>	<u>3</u>
0	14.3	3.6	1.6
1	10.8	2.7	1.2
2	7.5	1.9	0.83
5	3.4	0.85	0.38

The unshielded gamma-ray dose rate from 50 grams of Plutonium-239 is about 0.5 milliroentgen per hour (mr/hr) at one foot; consequently, no shielding is required.

It is probable that Plutonium-239 can be processed without shielding. If the time required for the operator to be in the close vicinity of the fluoride proves to be too great, an inch or two of polyethylene around the compound will be sufficient.

Cerium and cesium metals as getters. The use of metallic cesium or cerium as a getter for oxygen liberated during the irradiation of americium oxide has been proposed. It was estimated that 0.08 gram of oxygen will be liberated in each capsule. The minimum amounts of these metals required to combine with this oxygen are:

(1) Cesium--0.66 gram.

(2) Cerium--0.70 gram.

To provide an excess of material, about 1.4 grams of either of these metals will be required.

Cesium. Natural cesium is composed of only one isotope, Cesium-133, which has a cross section of 26 barns for capture of thermal neutrons. When Cesium-133 is irradiated with thermal neutrons, Cesium-134 is formed by neutron capture. After 90-day irradiation in a thermal flux of 3×10^{14} n/cm²-sec, 1.4 grams of natural cesium will produce 96 curies of Cesium-134. The unshielded dose rate at one meter from 96 curies of the heavier isotope is about 115 roentgens per hour (r/hr) when the slug containing the cesium is placed in the curium-shipping cask, the dose rate at one meter becomes 7×10^{-4} mr/hr per slug. The half life of Cesium-134 is 2.3 years.

Cerium. Natural cerium contains four isotopes: Cerium-136, -138, -140 and -142. Pertinent data for these isotopes and the products resulting from neutron irradiation are given in Table III-14. Also included in the table are the activities and dose rates resulting from the irradiation of 1.4 grams of natural cerium in a flux of 3×10^{14} n/cm²-sec for 90 days.

When the capsules are placed in the curium-shipping cask, the dose rate drops to 5.5×10^{-3} mr/hr per capsule at one meter.

Substitution of 5052 aluminum for 1100 aluminum. In order to gain an increased strength advantage, it was decided to fabricate the irradiation capsule from 5052 aluminum in preference to 1100 aluminum. The principal difference between 1100 and 5052 aluminum is that the latter alloy contains the following constituents: 2.5 nominal weight percent magnesium and 0.25 nominal weight percent chromium.

Examination of activation data shows Chromium-51 to be the only isotope which might affect the shipping-cask shielding. All other radioactive isotopes will have decayed to negligible amounts several hours after removal from the neutron flux. After 90-day irradiation in a flux of 3×10^{14} n/cm²-sec, the amount of Chromium-51 present in 60 grams of 5052 aluminum will produce the following gamma rays:

TABLE III-14
Activation of Cerium During Irradiation

Target	Abundance in Natural Element (%)	Product	Thermal Neutron Cross Section (barns)	Activity (curies)	Half Life	Gammas Emitted		
						Energy (mev)	Disin- tegra- tion (%)	Dose Rate at 1 Meter (mr/hr)
Ce-136	0.19	Ce-137	25	2.4	9 hr	0.445	3	19.5
Ce-138	0.26	Ce-139	0.4	0.018	140 d	0.166	100	1.6
Ce-140	11.0	Ce-141	0.39	1.4	32 d	0.142	30	22
Ce-142	89.0	Ce-143	1.0	43.0	34 hr	1.10	6	2570
						0.86	12	
						0.72	5	

- (1) 3.1×10^{10} gamma rays per second at 0.32 million electron volts.
- (2) 1.75×10^6 gamma rays per second at 0.65 million electron volts.

After attenuation by the present shielding of the shipping cask, the additional radiation from the chromium will increase the dose rate at one meter by only 10^{-15} to 10^{-16} mr/hr per irradiated slug.

Photon production due to the other constituent elements in 1100 and 5052 aluminum was calculated with the aid of tables given in Ref. III-14. The calculated activities in 60 grams of each type of material irradiated for 90 days in a flux of 3×10^{14} n/cm²-sec indicated that no problem existed with respect to excessive radiation levels.

Shipping-cask dose rates. The present shipping cask is designed for seven irradiated capsules; however, immediate plans require the shipment of only three capsules. Consequently, to determine radiation levels involved with three capsules, gamma-ray dose rates were determined by rescaling data for seven capsules given in Ref. III-15.

Neutron production was calculated to be 2.23×10^8 neutrons per second per slug from alpha-neutron reactions with aluminum and 3.1×10^7 neutrons per second from spontaneous fissioning. The shipping cask is shielded with 8-1/4 inches of lead, 11 inches of water and 3/4 inch of steel. Dose rates for different times after removal of slugs from reactor are as shown in Table III-15.

TABLE III-15

Shipping-Cask Gamma Ray Dose Rates

Time after removal (days)	Dose rates in air at 0.4 feet from slug center (mrem/hr x 10^5)		Dose rates for cask with 3 slugs (mrem/hr)	
	1 slug	3 slugs	At cask surface	One meter from slug surface envelope
5	3.7	11.0	285	79
10	3.2	9.6	113	31

TABLE III-15 (continued)

Time after removal (days)	Dose rates in air at 0.4 feet from slug center (mrem/hr x 10 ⁵)		Dose rates for cask with 3 slugs (mrem/hr)	
	1 slug	3 slugs	At cask surface	One meter from slug surface envelope
15	2.7	8.1	49	14
20	2.4	7.2	25	7
30	1.75	5.2	6.3	2

When the above data are plotted, it is found that:

- (1) The minimum cooling time to meet the ICC requirement of 200 mrem/hr maximum at cask surface is seven days.
- (2) The minimum cooling time to meet the ICC requirement of 10 mrem/hr maximum at one meter from slug envelope is about 17-1/2 days.

Comparison of calculated and measured dose rates. Comparison of calculated dose rates for heavy elements with measured dose rates supplied by Lawrence Radiation Laboratories was started. Preliminary results indicate excellent agreement between the two.

c. Heat transfer analysis

Irradiation in the reactor. Curium-242 is produced by the irradiation of Americium-241 in a high-flux reactor. A conversion of 43% of the Americium-241 to curium is desired, but the actual conversion will probably be less. It has been found that irradiation of americium capsules (1-inch diameter, 5-inch length) in an unperturbed thermal neutron flux of 4×10^{14} n/cm²-sec for five cycles in the Materials Testing Reactor will provide an optimum balance between the cost of irradiation and the amount of Curium-242 produced. At this gross flux intensity, the effective or perturbed flux will be 2.8×10^{14} n/cm²-sec (i.e., the capsules incur a flux depression factor of 0.7). Figure 5 of Ref. III-15 shows the approximate variation of the products of americium while in the reactor.

The total heat production in the capsule during irradiation is made up of the following:

- (1) Fission of americium and its products.
- (2) Decay of Curium-242.
- (3) Decay of fission products.
- (4) Gamma heating.

The largest heat production rate will probably peak upon 30 days of irradiation since most of the heat produced in the capsule during irradiation is due to fission. Americium-242, the major contributor, will be at maximum concentration at this time. Table III-16 presents the heat production rate due to fission of the products of Americium-241 after 30 days in the reactor. A summary of the heat production rate, including gamma heating, fission, Curium-242 decay and fission-product decay follows:

Fission of americium products	9080 watts
Decay of Curium-242	108 watts
Decay of produced fission products	120 watts
Gamma heating	<u>1645 watts</u>
Total heat rate	10,953 watts

TABLE III-16

Heat Production Rate Due to Fission of Am-241 Products
After 30 Days in the Reactor

<u>Material</u>	<u>Fission cross section (barns)</u>	<u>Weight (mg)</u>	<u>Heat production rate (watts)</u>
Cm-242	< 5	895	128
Cm-243	700	5.35	107
Cm-244	---	0.86	---
Am-241	3.2	2320	212

TABLE III-16 (continued)

<u>Material</u>	<u>Fission cross section (barns)</u>	<u>Weight (mg)</u>	<u>Heat production rate (watts)</u>
Am-242	6390	44.5	8144
Am-242m	2500	4.1	277
Am-243	<25	---	---
Pu-243	<0.2	179	103
Pu-238	17	60.6	31.6
Pu-240	<0.1	25	0.072
Pu-239	746	3.57	77.2
Pu-241	1100	70	--
		Total	9080

With this heat rate (10,953 watts), the following maximum temperatures were found during reactor operation:

- (1) At outer surface of aluminum clad--330° F.
- (2) At inner surface of aluminum clad--349° F.
- (3) At centerline of capsule--422° F.

No boiling in the reactor will take place since the saturation temperature of the water is about 390° F.

Underwater storage in the reactor canal. After the capsules of Americium-241 have been irradiated for 84 days, they are removed from the reactor and allowed to cool in the reactor canal for 30 days before shipment. Maximum heat production rate, and, therefore, maximum temperatures, will occur immediately after insertion in the canal. The heat is produced by the decay of Curium-242 and fission products. The following is a summary of the heat production rates:

Decay of Curium-242	182 watts
---------------------	-----------

Decay of fission products	<u>120 watts</u>
Total heat rate	302 watts

In the cooling canal the mode of heat transfer is natural convection. The resulting temperatures are as follows:

- (1) At outer surface of aluminum clad--134° F.
- (2) At inner surface of aluminum clad--134.5° F.
- (3) At centerline of capsule--136.5° F.

Since the boiling point of water in the canal is 212° F, no problem exists.

Transporting in shipping cask. After the required 30-day cooloff period, the capsules will be ready for shipment. During this time the fission products will have decayed to the point where they are contributing only 1.63% of the total heat produced. At the end of 30 days the following heat-production rates will be expected:

(1) Decay of Curium-242	160 watts
(2) Decay of fission products	<u>2.6 watts</u>
Total	162.6 watts

Preliminary specifications given to the shipping cask manufacturers stated that the heat-production rate per capsule would be 186 watts. Therefore, analysis of the proposed shipping casks was made on the basis of this requirement. Two companies quoted that the surface temperature of the capsule while in the shipping cask would never exceed 300° F. However, based on the specifications sent to these companies, an analysis of the cask shows that, in the event of complete loss of neutron shield water in the outer container, a curium capsule surface temperature of 325° F can be expected. The following specifications on the cask must also be met to assure that this temperature will not be exceeded:

- (1) The outside surface must be painted with white enamel.
- (2) All inside surfaces must be nearly black.
- (3) The maximum number of fins must be used both on outer and inner surfaces.

It is also advised that the outside surface be protected by one of the following methods (in order of increasing preference):

- (1) Cadmium plate.
- (2) Nickel plate.
- (3) Zinc plate.
- (4) Tin plate.
- (5) Tin-zinc hot dip..

These surfaces assure protection against rusting and excessive temperatures in the event of loss of the white enamel paint due to peeling. Since a lower heat rate than was specified actually will exist, it is felt that the temperature of the capsule at the surface will not exceed 300° F. (Of course, this temperature represents a maximum value after loss of the neutron shield; the normal temperature is considerably less.)

4. Conclusions

a. Safety analysis for radioisotopes

The incident angle of specimen impact obtained on the bazooka tests was within desired tolerances. The technique developed for this program is readily adaptable to future test programs.

Hot cells for the processing of curium are available to The Martin Company at the Quehanna Laboratory. The greatest potential hazard of the radioisotope is release to the environment. The concept of multiple containment being followed by the Task 6 program provides the best possible means for preventing such release. To prevent the spread of contamination to the surrounding area, the existing ventilation system is designed for containment of any contamination accidentally released inside the facility during processing. Also, an elaborate system is available for the disposal of low radioactive waste which accumulates during processing of the radioactive fuel. The high-level wastes are to be absorbed and prepared for solid packaging.

b. Shielding analysis

Americium processing. An analysis of the fuel-slug production line revealed only one area where radiation levels required special attention. This area exists where the blending flask is mounted to the press dry box. Methods of mounting the flask must be devised which will require a minimum of handling by the press operator to reduce dose rates.

Plutonium metal production. The analysis of the Plutonium-238 metal producing process revealed no area in which shielding could not be provided to protect personnel. Because of the lower dose rates prevailing when Plutonium-239 is being processed, it is probable that no shielding will be required during processing of this isotope.

Cerium and cesium metals as "getters". The use of either of these metals as an oxygen "getter" would be satisfactory from a shielding viewpoint. It would seem that Cesium-134 may prove more desirable because of its lower dose rate in the shipping cask. However, if the half lives are considered, cerium would be preferable. The amount of activity from either of these metals should not present any additional problem when the irradiated slugs are processed in the Quehanna hot cells.

Substitution of 5052 aluminum for 1100 aluminum. The substitution of 5052 aluminum for 1100 aluminum will not affect the shielding of the shipping cask.

Shipping cask. The minimum cooling time required before shipment of three irradiated slugs is controlled by the maximum permissible dose rate (10 mrem/hr) at one meter from the shipping cask. About 17-1/2 days are required for the activity to decay sufficiently to meet this requirement. The minimum cooling time to meet the required 200 mrem/hr at the surface of the container is 7 days.

Comparison of calculated and measured dose rates. Preliminary results indicate good agreement between calculated and measured dose rates.

c. Heat transfer analysis

Due to the high radiation level necessary for converting Americium-241 to Curium-242, the temperature of the capsule will range from a maximum of 330° F at the surface to 422° F at the centerline. During cooling in the canal the temperature will be a maximum 134° F at the surface and 136° F at the centerline. Investigation of shipping casks proposed by two commercial suppliers revealed that neither met the requirements of maintaining the capsule surface temperature at less than 300° F in the event of complete loss of the cooling water. However, it is felt that their designs will assure a maximum not higher than 325° F at the capsule surface.

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APPENDIX A

ACTIVATION UNIT 2

1. Introduction

The method of heating the emitter of a thermionic generator, in the past, has been by conduction from the heat source. While the approach is satisfactory for most applications, the isotope-heated prototype being developed must withstand an environment which makes the assembly of a conduction-heated device extremely difficult. The proposed solution is to heat the emitter by thermal radiation from the isotope casing. An analysis of this method of heating and methods for increasing the emissivities of the emitter and the isotope container were reported in the preceding quarterly report (Ref. A-1).

This appendix describes a test program undertaken to confirm the prediction of the temperature required for heating the emitter. The test program also had two secondary objectives: to confirm the existence of a minimum in the relation between the collector work function and the collector temperature, and to achieve a low collector work function by using a metal vacuum system and bell jar, rather than the vacuum tube approach used in previous collector work function tests.

2. Description of the Apparatus

The apparatus is shown in Fig. A-1. The isotope casing is simulated by a radiator (5) which is heated by electron bombardment from the emitter filament (attached to 38). The emitter (2) is heated by the radiator (5). The opposing surfaces of the emitter and radiator are grooved to increase their emissivities. The collector (1) is equipped with a collector filament (35). During operation the collector can be radiation-heated by the filament. During activation the collector is heated by electron bombardment from the same filament. An inter-electrode spacing of 0.0011 ± 0.0001 inch is achieved by the use of 0.040-inch diameter sapphire rods brazed into the collector. The emitter is held against the spacers by a 50-gram weight (40) attached to the emitter by the emitter lead (27) and the emitter lead plug (25).

The emitter and radiator temperatures are read on an optical pyrometer sighted through a hole in the shields and the radiator holder (4). Collector temperatures are measured with a Pt-Pt + 10% Rh thermocouple (36). During activation the collector temperature is also measured with an optical pyrometer when temperatures permit.

Fig. A-1. Activation Unit 2

The entire unit is mounted on a vacuum system and enclosed in a Pyrex bell jar, and all electrical leads are attached to a leadthrough plate to permit changes in the circuit without the necessity of opening the unit to air.

3. Experimental Procedure

a. Activation

After the pressure in the bell jar had reached approximately 5×10^{-7} millimeter of mercury, heat was applied gradually to both emitter and collector. The temperature was increased slowly so as to keep the pressure in the bell jar below 3×10^{-6} millimeter of mercury. The two temperatures were increased together to 1150°C , as measured with a pyrometer on both emitter and collector. This temperature was held for one hour, and then the collector heat was removed and the collector allowed to cool.

During the first attempt at activation the collector thermocouple shorted against a high-voltage lead and burned out. The unit was cooled and the thermocouple was replaced. The unit was then activated. During the second activation it became apparent that the collector thermocouple was giving an erroneous reading. However, operation continued.

b. Operation

The electron-bombardment power to the radiator was controlled to give an emitter temperature of 1150°C , as read with an optical pyrometer. The temperatures of the emitter and the radiator were monitored continuously with the optical pyrometer.

The converter's output characteristics were measured in the following manner:

- (1) The output current was held at 100 milliamperes while the variation in output voltage was recorded as a function of time.
- (2) The output current was varied, and the output current and voltage were recorded to obtain the I-V relationship.
- (3) A retarding voltage was applied across the diode, and the current across the diode was recorded to obtain the back emission of the diode.

Each of the three procedures was undertaken a number of times during the test period to give the diode characteristics as a function of time. The total operating time of the converter was 14.7 hours. At the end of this period, it was decided to open the tube and replace the collector thermocouple; then, the diode was to have been reactivated to establish a second set of operating characteristics. Unfortunately, as the tube was cooling, a loss of power to the forepump of the vacuum system permitted oil vapor to rise into the bell jar and deposit on the diode. Since the repair of the damage would require considerable time, the test was terminated.

4. Analysis of Data

Representative curves from the data obtained are presented in this appendix (Figs. A-2, A-3, A-4, A-5, A-6 and A-7).

a. Temperatures

Collector temperature. Due to the failure of the collector thermocouple, the temperature of the collector could not be read directly from the data. However, during the activation of the unit, pyrometer measurements on the collector were taken, both at the emitter end and at the location of the thermocouple. These readings were used to correct the thermocouple reading to a reasonable degree.

Emitter temperature. The emitter temperature was corrected from the pyrometer reading by the addition of 47°C . This correction takes into account both the emitter's emissivity and the transmission losses through the glass bell jar. The correction factor was based on previous emissivity measurements on type-B emitters.

Radiator temperature. The correction of the pyrometer reading of the radiator temperature is somewhat involved. The theoretical analysis is presented in Section 7 of this appendix. This analysis gives a radiator temperature of 1700°K for this unit during operation.

b. Diode characteristics

Sample calculations are shown in Section 8 of this appendix.

Collector work function versus collector temperature. The relation ϕ_C versus T_C is shown in Fig. A-2. This curve shows a minimum ϕ_C at a collector temperature of $900^{\circ} \pm 25^{\circ}\text{K}$. The value of the work function at this collector temperature is 2.02 ± 0.01 volts.

Fig. A-2. Output Voltage Versus Collector Temperature

Current-voltage characteristics. A family of I-V curves is presented in Fig. A-3 for an emitter temperature of 1470° K. Figure A-4 shows the output and collector work function of the unit as a function of time. The collector temperature is also recorded at various times. The shape of this curve is similar to data obtained from other tubes. Curves A-2, A-3 and A-4 are similar to the results achieved in previous experiments. Unfortunately, however, the work function is higher than that measured on previous diodes activated with the collector at a high temperature.

5. Discussion of Results

a. Radiation heat transfer

The test data confirm, within experimental accuracy, the analysis reported in the preceding quarterly progress report (Ref. A-1). The radiator temperature measured was 1700° K for an emitter temperature of 1463° K. This temperature agrees, to within 10° C, with that predicted by the method of analysis used in Ref. A-1.

b. Collector work function versus collector temperature

The test data confirmed the existence of a minimum in the curve of collector work function versus collector temperature. Due to the erroneous readings of the collector thermocouple, the temperature at which this minimum work function occurs could not be accurately determined. However, with the error observed in the thermocouple readings during activation as a base, a correction can be made in the collector temperature readings. Use of the correction factor places the minimum collector work function at approximately 900° K.

c. Collector work function

The lowest collector work function achieved during this test was 2.02 volts. No explanation of this unexpectedly high work function is readily available.

6. Recommendations

- (1) The emitters for Unit 2B should be heated by thermal radiation from the isotope casing.
- (2) The analysis presented in Ref. A-1 should be used to predict the required isotope casing temperature.
- (3) Use of coatings on emitter and radiator to increase their respective emissivities should be investigated. The purpose of

Fig. A-3. Output Voltage Versus Output Current

Fig. A-4. Output Voltage and Collector Work Function Versus Time and Collector Temperature

these increased emissivities is to reduce further the required isotope casing temperature.

7. Consolidation of Analysis of Correction Factor for Radiator Temperature

W_T = total heat radiated from radiator (1)

T_{1A} = actual radiator temperature ($^{\circ}\text{K}$)

T_{2A} = actual emitter temperature ($^{\circ}\text{K}$)

T_1 = observed radiator temperature = $1350^{\circ}\text{C} + 20^{\circ}\text{C}$
(glass correction factor) = 1370°C (1643°K)

T_2 = observed emitter temperature = $1150^{\circ}\text{C} + 40^{\circ}\text{C} =$
 1463°K (corrected for ϵ_2 and glass losses)

$J_{\lambda T}$ = brightness at $\lambda = 0.65$ micron (A-1)

= $C_1 (\lambda^{-5}) \exp (-C_2 / \lambda T)$, (Ref. A-2) (A-2)

W_T (observed) = W_{TR} (radiated directly) + W_{T1} (radiated
by radiator and reflected out) + W_{T2} (radiated
by emitter and reflected out) (A-3)

$W_{TR} = \epsilon_1 \sigma A T_{1A}^4$ (A-4)

$W_{T1} = \sigma A T_{1A}^4 \epsilon_1 \left[\sum_{n=0}^{n=\infty} (1-\epsilon_2)^n (1-\epsilon_1)^n - 1 \right]$

$$\begin{aligned}
&= \sigma A T_{1A}^4 \epsilon_1 \left\{ \left[1 - (1 - \epsilon_2)(1 - \epsilon_1) \right]^{-1} - 1 \right\} \\
&= \sigma A T_{1A}^4 \epsilon_1 \frac{1 - \epsilon_1 - \epsilon_2 + \epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2 - \epsilon_1 \epsilon_2}
\end{aligned} \tag{A-5}$$

$$\begin{aligned}
W_{T2} &= \sigma A T_2^4 \epsilon_2 (1 - \epsilon_1) \sum_{n=0}^{\infty} \left[(1 - \epsilon_1)(1 - \epsilon_2) \right]^n \\
&= \sigma A T_2^4 \epsilon_2 (1 - \epsilon_1) \left[1 - (1 - \epsilon_2 - \epsilon_1 + \epsilon_1 \epsilon_2) \right]^{-1} \\
&= \sigma A T_2^4 \frac{\epsilon_2 (1 - \epsilon_1)}{\epsilon_2 + \epsilon_1 - \epsilon_1 \epsilon_2}
\end{aligned} \tag{A-6}$$

Equations (A-3) through (A-6) may be combined to give

$$\begin{aligned}
W_T &= \epsilon_1 \sigma A T_{1A}^4 + \sigma A T_{1A}^4 \epsilon_1 \frac{1 - \epsilon_1 - \epsilon_2 + \epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2 - \epsilon_1 \epsilon_2} \\
&\quad + \sigma A T_2^4 \frac{\epsilon_2 (1 - \epsilon_1)}{\epsilon_2 + \epsilon_1 - \epsilon_1 \epsilon_2} = \epsilon_1 \sigma A T_{1A}^4 \left(1 + \frac{1 - \epsilon_1 - \epsilon_2 + \epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2 - \epsilon_1 \epsilon_2} \right) \\
&\quad + \sigma A T_2^4 \frac{\epsilon_2 (1 - \epsilon_1)}{\epsilon_2 + \epsilon_1 - \epsilon_1 \epsilon_2} = \sigma A T_{1A}^4 \frac{\epsilon_1}{\epsilon_1 + \epsilon_2 - \epsilon_1 \epsilon_2} \\
&\quad + \sigma A T_2^4 \frac{\epsilon_2 (1 - \epsilon_1)}{\epsilon_2 + \epsilon_1 - \epsilon_1 \epsilon_2}
\end{aligned} \tag{A-7}$$

Check for $\epsilon_2 = 0$, $\epsilon_1 \neq 0$:

$$W_T = \sigma A T_{1A}^4 \text{ (black body conditions).}$$

Check for $\epsilon_2 = 1$, $\epsilon_1 = 1$:

$$W_T = \sigma A T_{1A}^4 \text{ (black body conditions).}$$

Assume $\epsilon_1 = \epsilon_2$. Then Equation (A-7) becomes

$$W_T = \sigma A T_{1A}^4 \frac{1}{2 - \epsilon} + \sigma A T_2^4 \frac{1 - \epsilon}{2 - \epsilon} \quad (A-8)$$

From Equation (A-2),

$$\sigma A T_{1A}^4 = J_{\lambda T_{1A}} = C_1 (\lambda^{-5}) \exp \left(- \frac{C_2}{\lambda T_{1A}} \right)$$

$$\sigma A T_2^4 = J_{\lambda T_2} = C_1 (\lambda^{-5}) \exp \left(- \frac{C_2}{\lambda T_2} \right)$$

$$W_T = J_{\lambda T_1} = C_1 (\lambda^{-5}) \exp \left(- \frac{C_2}{\lambda T_1} \right)$$

and Equation (A-8) becomes

$$\exp \left(- \frac{C_2}{\lambda T_1} \right) = \exp \left(- \frac{C_2}{\lambda T_{1A}} \right) (2 - \epsilon)^{-1} + \exp \left(- \frac{C_2}{\lambda T_2} \right) \frac{1 - \epsilon}{2 - \epsilon} \quad (A-9a)$$

or

$$\begin{aligned} (2 - \epsilon)^{-1} &= \exp \left[- \frac{C_2}{\lambda} \left(T_1^{-1} - T_{1A}^{-1} \right) \right] \\ &+ \frac{1 - \epsilon}{2 - \epsilon} \exp \left[- \frac{C_2}{\lambda} \left(T_2^{-1} - T_{1A}^{-1} \right) \right] \end{aligned} \quad (A-9b)$$

Both sides of Equation (A-9b) are solved independently and the difference is reduced to zero to determine T_{1A} . The initial assumption is that $\epsilon = 0.6$. (Note: The analysis in the preceding quarterly report predicts $\epsilon = 0.63$ for the given surface conditions.)

$$\frac{C_2}{\lambda} = 2.21 \times 10^4, \text{ for } \lambda = 0.65 \text{ micron.}$$

$$\frac{1 - \epsilon}{2 - \epsilon} = 0.286 \text{ (see Table A-1).}$$

$$T_{1A} = 1700 \pm 5^\circ \text{ K}$$

Therefore,

$$\Delta T \text{ (correction factor)} = 1700^\circ \text{ K} - 1463^\circ \text{ K} = 237^\circ \text{ K.}$$

TABLE A-1

$(2 - \epsilon)^{-1}$	$T_{1A} (^{\circ}\text{K})$	$\exp \left[-\frac{C_2}{\lambda} (T_1^{-1} - T_{1A}^{-1}) \right]$	$\left(\frac{1 - \epsilon}{2 - \epsilon} \right) \exp \left[-\frac{C_2}{\lambda} (T_2^{-1} - T_{1A}^{-1}) \right]$	ϵ'	$\epsilon - \epsilon'$	High-Low + -	
0.715	1650	0.957	0.053	0.904	0.304	+	
0.715	1700	0.631	0.0348	0.596	0.004		-
0.715	1800	0.312	0.071	0.241	0.359		-
0.715	1725	0.529	0.029	0.500	0.100		-
0.715	1710	0.604	0.033	0.571	0.029		-
0.715	1690	0.688	0.037	0.651	0.051	+	
0.715	1695	0.664	0.036	0.628	0.028		-

For an emitter temperature of 1463° K and a collector temperature of 800° K, the heat transferred across the diode is approximately 38 watts. For $Q = 38$ watts, T_{1A} for $T_2 = 1463^\circ \text{K}$ is given by

$$Q = \sigma A (T_{1A}^4 - T_2^4) \left(\frac{1}{\epsilon} - 1 \right)^{-1} = A (T_{1A}^4 - T_2^4) \frac{\epsilon}{2 - \epsilon}$$

$$38 = 5.71 \times 4.3 \times 0.428 \left[\left(\frac{T_{1A}}{1000} \right)^4 - 1.463^4 \right]$$

$$\left(\frac{T_{1A}}{1000} \right)^4 = \frac{38}{5.71 \times 4.3 \times 0.428} + 1.463^4 = 3.62 + 4.57 = 8.19$$

$$\frac{T_{1A}}{1000} = 8.19^{1/4} = 1.692$$

$$T_{1A} = 1692^\circ \text{K}.$$

8. Calculation of Diode Characteristics

Equations used below are standard in the field of thermionics.

$$\phi_C = KT_1 \ln \left(\frac{A_1 T_1^2}{J_0} \right) - V_0 \quad (\text{A-10})$$

where

ϕ_C = collector work function (volts)

K = Boltzmann's constant = 8.61×10^{-5} volt / °K

T_E = emitter temperature (°K)

A_1 = Richardson's constant = $120 \text{ amp/cm}^2 - ^\circ\text{K}^2$

J_0 = diode current density (amp/cm^2)

V_0 = diode output voltage (volts)

and where J_o and V_o are determined at any point on the straight-line portion of the I-V curve.

To study ϕ_C over a given length of time, it is appropriate to choose an arbitrary value for J_o on the straight-line portion of the I-V curve and to hold this value constant by varying the load resistance across the diode. Equation (A-10) becomes

$$\phi_C = C_C - V_o \quad (A-11a)$$

where C_C is a constant that can be determined easily for constant values of T_E and J_o . Then ϕ_C is a function of only V_o . For $J_o = 0.1/4.3 = 0.0233 \text{ amp/cm}^2$

$$\begin{aligned} C_C &= KT_E \ln \left(\frac{A_1 T_E^2}{J_o} \right) = 8.61 \times 10^{-5} \times 1463 \ln \left(\frac{120 \times 1463^2}{0.0233} \right) \\ &= 0.126 \ln \left(\frac{2.57 \times 10^8}{0.0233} \right) = 0.126 \ln (1.1 \times 10^{10}) \\ &= 0.126 \ln (1.1 \times 10^{10}) = 0.126 \ln (1.1) + 10 \times 2.3026 \\ &= 0.126 (0.0953 + 23.026) = 0.126 \times 23.12 = 2.91. \end{aligned}$$

Therefore,

$$\phi_C = 2.91 - V_o \quad (A-11b)$$

At a current of 100 milliamperes, the minimum work function occurred when V_o was 890 millivolts. Then

$$\phi_C = 2.91 - 0.890 = 2.02 \text{ volts.}$$

The maximum work function occurred when V_o was 475 millivolts. Then

$$\phi_C = 2.91 - 0.475 = 2.44 \text{ volts.}$$

These values are individual points taken when the emitter temperature was approximately 1463° K and after the unit had warmed up. The higher work function occurred immediately after activation of the tube.

9. Experimental Procedure and Results Pertaining to Diode Performance

Collector temperature was increased to 900° K in 5 hours. Testing was then discontinued because of thermocouple burnout. The thermocouple was repaired.

Emitter and collector temperatures were increased to 1150° C in 4 hours and maintained at equal brightness. After one hour at this temperature the collector was cooled down and an I-V curve obtained (Fig. A-6). At this time the collector work function was 2.42 volts at a temperature of 730° K. With the emitter at operating temperature, the output voltage was recorded at an output current of 100 milliamperes. The behavior of the diode is shown in Fig. A-4, where output voltage is plotted against time. Collector work function and temperatures are also indicated.

After 30 minutes of operation, another I-V curve (Fig. A-7) was obtained. The collector work function was 0.15 volt lower than after the initial activation, at approximately equal collector temperatures.

After 55 minutes of operation, the collector was heated, the work function decreased by 0.29 volt and the I-V curve of Fig. A-7 obtained. The work function was now 2.02 volts at 920° K.

The tube was shut down for one hour and the emitter and collector then brought up to operating temperatures. The output of the tube was exactly as before the shutdown.

The collector temperature was now varied and work function variations determined by output voltage at 100 milliamperes (Fig. A-5) and I-V curve (Fig. A-6). The sequence used can be seen from the table of Fig. A-6. Figure A-5 shows the collector work functions versus collector temperature, with the I-V curve points indicating steady-state and transient temperature conditions.

The emitter was again brought up to operating temperature. The diode was allowed to reach steady state, and the collector was then heated to 915° K. Points marked by squares in Fig. A-2 show the change in work function during this heating. Collector temperature was further varied as shown by the table of Fig. A-2 and output at 100 milliamperes was recorded and I-V curves taken (Fig. A-3).

Fig. A-5. Output Voltage Versus Collector Temperature and Collector Work Function

Fig. A-6. Current Versus Voltage

Fig. A-7. Current Versus Voltage

This terminated testing of the unit.

Data obtained on collector work function versus collector temperature in the two sets of test runs were similar but not identical. In particular, there is a considerable difference between the two sets of data at the higher collector temperatures. This may have been due to malfunction of the thermocouple, as explained earlier.

Data of Fig. A-5 show the collector minimum to be located at 920° K, with a 0.03-volt increase at 55° C above and below this temperature.

The data of Figs. A-2 and A-5 locate the minimum at 890° K with a 0.03-volt change for 45° C above and below 890° K.

Data from a previous diode in a bell jar show the collector work function minimum to be at 920° K, with a 0.03-volt rise 55° C above and below this temperature.

The following observations were notable:

- (1) Collector work function was high following the initial activation (2.42 volts at 730° K).
- (2) The tube was left overnight under high vacuum and emitter raised to operating temperature. Collector work function decreased to 2.27 volts at 715° K.
- (3) Heating of the collector caused a further decrease (2.02 volts at 920° K). This initial decrease was larger (290 millivolts) during heating from 760° K to 920° K than the collector work function variations later observed for the same temperature interval.
- (4) Collector emission data were obtained. At a collector temperature of 780° K an emission current of 0.33 milliamperes per cm² was observed. A 1.76-volt collector would account for this current, but this was not supported by the I-V data. A 2-volt collector at this temperature emits less than 10 microamperes. If the reading of 0.33 milliamperes per cm² is considered spurious electron and ion current and is subtracted from the observed currents at higher collector temperatures, the following collector work functions can be calculated.

Collector Temperature (°K)	Total Current (ma/cm ²)	Emission Current (ma/cm ²)	Collector Work Function (volts)
780	0.33	0.00	--
920	0.63	0.30	2.11
990	1.42	1.09	2.17

The collector work functions thus obtained were in excellent agreement with the I-V curve in Fig. A-6.

- (5) Operating temperature of the emitter was maintained constant at 1150° C brightness, as measured with an optical pyrometer on the edge of the emitter. This temperature was controlled by maintaining the input power at an electron bombardment voltage of 430 volts and a current of 300 milliamperes. The emitter temperature was also checked with the pyrometer and showed no variation during the operation of the tube. Darkening of the bell jar glass was very slight because a bell jar of large diameter was used.

All curves presented are based on a true emitter temperature of 1470° K. This temperature is the pyrometer temperature, to which 47° C has been added to include the correction for emissivity and glass losses due to clean glass.

Reflections from the shields and supports of the device will tend to reduce this correction because of the additional radiation seen by the pyrometer. This correction, however, is not large on account of the small areas of the emitter and emitter-collector gap, as compared with the cold support structures.

The uncertainty of the true emitter temperature may be ±40° C; the best estimate is the 47° C correction used. This is made up of a 10° C correction for glass losses from clean glass and a 37° C emissivity correction. This corresponds to a spectral emissivity of the emitter of 0.67. If the actual emitter emissivity is 0.45, the correction must be increased by 40° C. This is estimated to be the upper limit of the true emitter temperature.

The other correction extreme will occur for a glass loss of 10° C, emissivity loss of 37° C (emissivity 0.67), and a gain due to reflected radiations of 40° C.

This true emitter temperature uncertainty of 40° C causes a ±0.09-volt uncertainty in the collector work function.

10. References

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APPENDIX B

HEAT TRANSFER ANALYSIS

1. Introduction

The heat transfer analysis proceeds with the calculation of the necessary heat input to each diode. This establishes the required heater temperature which, in turn, controls the losses through the heater supporting structure. The sum of the heater supporting losses and diode inputs gives the total heat input into the unit. Nomenclature used in this appendix is listed in Table B-1.

2. Heat Requirements of the Diodes

a. Radiation from emitter to collector

$$q_r = \bar{\epsilon} A \sigma (T_E^4 - T_C^4)$$

where

$$\bar{\epsilon} = 0.15^*$$

$$A = 10 \text{ cm}^2$$

$$\sigma = 5.67 \times 10^{-12} \text{ watt/cm}^2 - \text{K}^4$$

$$T_E = 1500^\circ \text{K}$$

$$T_C = 900^\circ \text{K}$$

$$q_r = 38 \text{ watts.}$$

b. Electron cooling

$$q_e = A J V_T \left(\frac{\phi_R}{V_T} + 2 - 2.3 \log \frac{J}{J_R} \right)$$

where (with $\phi_C = 2.00$ and $w = 10$ microns)

* See Fig. A-17 of Ref. A-1.

TABLE B-1
Definition of Terms in Appendix B

<u>Symbol</u>	<u>Description</u>	<u>Unit</u>
A	Area	cm ²
C	Area factor	cm ²
F ₁₂	View factor	--
J	Current	amp/cm ²
J _R	Current at critical point	amp/cm ²
k	Thermal conductivity	watt/cm - °C
n	Number of heat shields	--
q _{r, e, c}	Heat flow	watts
r	Radius	cm
T _{E, C, R, H}	Temperatures	°K
t	Wall thickness	inches
w	Spacing	microns
ε	Thermal emissivity	--
σ	Stefan-Boltzmann constant	watt/cm ² - °K ⁴
φ	Work function	electron volts
R	Thermal resistance	°C/watt

$$J = 1.67 \text{ amp/cm}^2$$

$$\phi_R = 2.60 \text{ volts}$$

$$V_T = 0.129 \text{ volt}$$

$$J_R = 0.48 \text{ amp/cm}^2$$

$$A = 10 \text{ cm}^2.$$

Then

$$q_e = 41 \text{ watts.}$$

c. Conduction loss through sapphire

$$q_c = \frac{T_E - T_C}{R}$$

where R is the total thermal resistance of sapphire. R is computed to be

$$R = \frac{L}{kA}$$

where

L = conduction path length

k = thermal conductivity of sapphire

A = cross-sectional area.

For the three sections of the sapphire, shown in Fig. B-1,

$$R_1 = 150^\circ \text{ C per watt}$$

$$R_2 = 120^\circ \text{ C per watt}$$

$$R_3 = 25^\circ \text{ C per watt}$$

$$R = \frac{1}{6} \Sigma R_i = 49.2^\circ \text{ C per watt}$$

$$q_c = \frac{1500 - 900}{49.2} = 12.2 \text{ watts.}$$

Fig. B-1.

d. Conduction loss through lead

$$R = \frac{1}{2} \frac{L}{kA}$$

where

$$L = 0.170 \text{ inch}$$

$$A = 0.00134 \text{ square inch}$$

$$k = 0.5 \text{ watt/cm} - ^\circ\text{C}$$

$$R = 50^\circ \text{ C per watt}$$

$$q_c = \frac{T_E - T_C}{R} = \frac{1500 - 900}{50} = 12 \text{ watts.}$$

e. Total heat requirements of the diodes

$$q = q_r + q_e + q_c = 38 + 41 + (12 + 12) = 103 \text{ watts.}$$

3. Calculation of Heater Temperature

For the heat transfer by radiation,

$$q = C\sigma(T_R^4 - T_E^4)$$

or

$$\sigma T_R^4 = \frac{q}{C} + T_E^4 \sigma,$$

where

$$C = \frac{A_1}{F_{12}^{-1} + (\epsilon_1^{-1} - 1) + A_1 A_2^{-1} (\epsilon_2^{-1} - 1)}$$

$$q = \text{heat to be transferred, 50 watts}$$

$$A_1 = \text{heating area, } 10 \text{ cm}^2$$

$$A_2 = \text{heated area, } 9.14 \text{ cm}^2$$

$$F_{12} = \text{view factor of } A_1 \text{ and } A_2, 0.85$$

$$\epsilon_1 = \epsilon_2 = \text{emissivity of } A_1 \text{ and } A_2, 0.60$$

$$\sigma = 5.67 \times 10^{-12} \text{ watt/cm}^2 \cdot \text{°K}^4$$

$$T_E = 1500^\circ \text{K.}$$

Then

$$\sigma T_R^4 = 41.7 \text{ watts/cm}^2$$

and

$$T_R = 1650^\circ \text{K.}$$

4. Losses Through Heater Support Structure

a. Conduction loss

The total thermal resistance of the structure is equal to the sum of the thermal resistances of each cylinder:

$$R = \Sigma R_i$$

where

$$R_i = \frac{L_i}{k_i 2\pi r_i t}$$

and

$$t = 0.005 \text{ inch.}$$

TABLE B-2

Characteristics of Support-Structure Cylinders

Cylinder	Radius, r (cm)	Length, L_i (cm)	Thermal Conductivity, k_i watt/cm - °C	Thermal Resistance, R (°C/watt)	Temperature, T_i (°K)
1 (inner)	1.59	1.16	0.67	13.5	1520
2	1.72	2.64	0.63	30.0	1255
3	1.85	2.75	0.59	31.0	1040
4	1.97	2.84	0.54	34.0	840
5	2.10	2.96	0.52	32.0	670
6 (outer)	2.23	1.36	0.52	14.5	600

$$\Sigma R = 155.0$$

The heat loss is

$$q_c = \frac{\Delta T}{R} = \frac{1650 - 600}{155} = 6.8 \text{ watts.}$$

b. Radiation loss

The radiation loss of the heater to the casing without heat shields is

$$q_r = A\sigma\epsilon (T_R^4 - T_H^4)$$

where

$$A = 45 \text{ cm}^2$$

$$\epsilon = 0.1$$

$$\sigma = 5.78 \times 10^{-12} \text{ watt/cm}^2 - ^\circ\text{K}^4$$

$$T_R = 1650 ^\circ\text{K}$$

$$T_H = 600 ^\circ\text{K.}$$

Then

$$q_r = 180 \text{ watts.}$$

The effectiveness of radiation shields in reducing this heat loss is given by

$$q_r' = \frac{q_r}{n+1}$$

where

$$q_r' = \text{heat loss with shields}$$

$$q_r = \text{heat loss without shields}$$

$$n = \text{number of shields}$$

if it is assumed that the shields are of equal thermal emissivity and that no conduction loss exists between the shields. To reduce the radiation heat loss to four watts, a value small in comparison with the conduction loss, the number of shields needed is

$$n + 1 = \frac{q_r}{q_r} = \frac{180}{4} = 45$$

$$n = 44.$$

If the six structural cylinders are counted as shields, 38 more shields must be introduced into the structure. An additional two shields are added as a safety factor and to simplify calculations.

The temperatures of the shields, as determined by radiation, will drop from 1650° K, the heater temperature, to 600° K, the housing temperature, by a fourth-power law. The temperatures of the structural cylinders, as determined by conduction, will drop off linearly. For minimum heat loss, the 40 additional cylinders are, therefore, distributed among the six structural cylinders in such a fashion as to match the fourth-power distribution to the linear distribution (see Figs. B-2 and B-3).

c. Radiation end losses

The gap between the diode and the heater support structure constitutes a black-body radiator. The effective temperature of this black-body hole is the mean temperature of its walls. When the gap is shielded by a ridge machined in the collector, the heat loss can be estimated to be

$$q_r = A\sigma\epsilon (T_B^4 - T_C^4)$$

where

$$A = \text{radiating area of the gap} = 0.4 \text{ cm}^2 \text{ per diode}$$

$$\sigma = 5.78 \times 10^{-12} \text{ watt/cm}^2 - \text{K}^4$$

$$\epsilon = 0.3$$

$$T_B = 1500^\circ \text{ K}$$

$$T_C = 900^\circ \text{ K.}$$

Then

$$q_r = 3 \text{ watts per diode or 6 watts.}$$

Fig. B-2. Temperature Distribution over Upper Stage

Fig. B-3. Temperature Distribution over Lower Stage

APPENDIX C

ACTINIDES COMPENDIUM FOR SUBTASK 6.1

The compendium covering available literature (classified and unclassified) on radioisotopes for space power will have the following format, with a bibliography, to include nuclides of interest to the program. The outline will be as follows:

- (1) Introduction--a short preface containing program and background information, not highly technical in character.
- (2) A review of properties of substances of interest--a complete unit, organized according to materials covered, to be prepared in tabular form similar to that of a handbook, with necessary references.
- (3) A review of process information--the separation chemistry of materials of interest, to be organized according to method of separation. This will form another complete unit with references.
- (4) An annotated bibliography of selected documents--informative abstracts will be provided for documents of major importance; indicative abstracts, for documents of lesser importance; titles and bibliographic information, for marginal items such as progress reports. The bibliography will be organized according to subject.

Classified documents will be treated in separate sections in each division of the outline to facilitate publication of a two-part report.

Each review section will be prepared completely so as to facilitate eventual publication in journals. The bibliography will be complete so as to constitute a suitable report publication in itself.

The following isotopes will be covered: Radium-228, Actinium-227, Thorium-228, Uranium-232, Neptunium-237, Plutonium-238, Plutonium-239, Plutonium-241, Americium-241, Curium-242, Curium-243, Curium-244. In addition, Polonium-210, Cesium-137, Strontium-90, Cerium-144 and Promethium-147 will be included insofar as these materials are applicable to isotopic power uses.

APPENDIX D

ISOTOPES OF INTEREST FOR THE COMPENDIUM

Radium-228	}	Research completed in Bibliography as of June 30; selection and review needed.
Actinium-227		
Thorium-228		
Uranium-232		
Neptunium-237		
Plutonium-238		
Plutonium-239		
Plutonium-241		
Americium-241		
Curium-242		
Curium-243	}	Nonactinides
Curium-244		
Polonium-210		
Cesium-137		
Strontium-90		
Cerium-144		
Promethium-147		

APPENDIX E

TOPICS EXCLUDED FROM THE SEARCH

- (1) All biological aspects of the actinides.
- (2) All Physics Progress Reports (all laboratories).
- (3) Nearly all Reactor Engineering Reports (progress and hardware).
- (4) All trip reports.
- (5) Most references on specific separation processes (e.g., Purex or Redox) which did not indicate summary, development, evaluation or comparison information.
- (6) Most general information on cerium, cesium, polonium, promethium and strontium.
- (7) All general information except bibliographies on thorium and uranium.
- (8) All nuclides or the elements listed which were not specifically cited (Pu-240, U-233, U-235 and Th-232, for example).
- (9) Foreign publications which are secondary discussions of other nations' work and results.
- (10) Abstracts for all progress reports cited. Lists of report numbers and abstract references have been compiled for the most important series, but series having sporadic information or interest have been excluded.
- (11) Most analytical chemistry reports (other than compilations and reviews).
- (12) All hazards, safety and critical reports (except compendia and bibliographies).
- (13) Reports and journal articles dealing entirely with nuclear properties have been included but marked to indicate limited interest. It is intended to cut most of these references from the final bibliography.