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SNAP PROGRAMS

MASTER

Quarterly Progress Report No. 6

Task 6

Fuel Technology Development Program

January 1 through March 31, 1961

MND-P-3014-III

Approved by

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Advanced Development Section

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FOREWORD

This is the sixth unclassified SNAP Quarterly Progress Report prepared by The Martin Company. It covers the work performed during the period January 1, 1961 through March 31, 1961 under Contract AT(30-3)-217, Task 6, Fuel Technology Development Program, for the U. S. Atomic Energy Commission.

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SUMMARY

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

The Fuel Technology Development Program, Task 6, is to develop suitable radioisotope fuels and heat source containment capsules for SNAP-type generators with land, sea and space applications. Curium-242 and Plutonium-238 are currently being investigated, and methods for purification, compound conversion and fuel form are being established. A Heavy Elements Processing Facility capable of handling and preparing gram quantities of Am-241 in an aluminum matrix for irradiation to Cm is being activated. In the Facility, high integrity aluminum capsules have been produced through special encapsulation and welding techniques. Curium purification by ion exchange techniques is under study to determine the operating conditions and design parameters for Martin's Quehanna Hot Cell Facility. Following a screening process, these container materials were selected for further investigation: Hastelloy C and Inconel X for thermoelectric applications and a Mo-W combination for the curium thermionic generator. Safety, thermal and shielding analyses have been conducted on the processes being developed as well as during handling and shipping.

I. INTRODUCTION

The third Quarterly Progress Report submitted under the requirements of Contract AT(30-3)-217 for Fiscal Year 1961 covers an appraisal of the work done under Task 6--Fuel Technology Development Program. This report covers the effort for the period January 1, 1961, through March 31, 1961. The report is prepared by W. M. Bowes, based on principal contributions from J. B. Zorn, A. Abriss, R. Corcoran, J. Kane, P. Hess, S. Zwickler, E. Smith, A. Spamer and G. Samos of The Martin Nuclear Division. The Subtasks of the Fuel Technology Development Program are:

- (1) Subtask 6.1--General Development and Material Requirements.
- (2) Subtask 6.2--Americium-Curium Radioisotopes Preparation Processes.
- (3) Subtask 6.3--Radioisotope Fuel Form Development.
- (4) Subtask 6.4--Safety, Thermal and Shielding Analysis.
- (5) Subtask 6.5--Americium Irradiation.

II. TASK 6--FUEL TECHNOLOGY DEVELOPMENT

Techniques for purifying multikilocurie quantities of transuranic radioisotopes have been under development for a little over a year. The search for methods of purifying these materials--as well as methods of obtaining and isolating them--grew out of the need for alpha-emitting radioisotopes suitable for use as energy sources in auxiliary power systems. As an essential phase of the development of the SNAP (thermoelectric) and SNAP-TIP (thermionic) generator systems, fuel technology has advanced in two additional areas: transformation of radioisotopes into suitable fuel forms, and selection of fuel containment materials appropriate to both the fuel and the energy conversion method.

Such radioisotopes as Americium-241 (target material), Curium-242, Plutonium-238 and Uranium-232 are currently being evaluated to determine the feasibility of producing suitable quantities, encapsulating in high-integrity containers and assuring the integrity of the capsules in the anticipated environments. Methods of handling the high levels of activity in hot-cell operations are being developed to provide the necessary process equipment for production operations. Safety evaluations of each phase of the effort are also being conducted to ensure safe radiological monitoring during all handling, purification, encapsulation, shipping, irradiation and mission applications.

A major portion of this effort has been devoted to the study of Curium-242 and Plutonium-238, short- and long-lived isotopes, respectively.

A. SUBTASK 6.1--GENERAL DEVELOPMENT AND MATERIALS REQUIREMENTS

A progress report describing work on the compilation of a compendium of technical information pertinent to isotopic power sources appeared in the preceding quarterly report (Ref. 1).

During the period of this report certain modifications in the plan for the compendium were made. A new plan for production of the compendium was devised, and research was continued in accordance with the plan. The original target date for completion of the compendium proved impossible to meet because of the heavy clerical and editing loads required. At the close of the quarter, a new set of target dates was selected. The list includes:

- (1) Completion of Phase 4, Subject Classification--March 31, 1961. This will permit transmittal of the bibliography for presentations processing.

- (2) Completion of Phase 3, Study and Annotation of Selected References--about April 30, 1961.
- (3) Completion of Properties and Processes Reviews--about May 31, 1961.
- (4) Completion of Compendium, all parts--June 1961.

1. Objectives

The objectives of this subtask are:

- (1) To perform literature reviews on all data--including physical, chemical and nuclear properties--pertaining to Americium-241, Curium-242, Plutonium-238 and other heat producing radioisotopes which can be used in auxiliary power sources and to compile this data in the form of a compendium.
- (2) To review and evaluate literature on purification and energy conversion processes tried with isotopes suitable for use in kilocurie quantities; to eliminate those radioisotopes that are unsuitable from a handling, shielding, economic, or availability viewpoint.
- (3) To continue the necessary liaison between The Martin Company and the various Government agencies and national laboratories so as to keep them informed of the Task 6 effort.
- (4) To direct the effort of Task 6 and to coordinate the various support, department, procurement and subcontracted phases of this program.

2. Scope of Work

a. Literature reviews

During this quarter the bulk of the compendium literature search was performed and the necessary arrangement, editing and indexing of the report was accomplished. The bibliography of unclassified references (without abstracts) was assembled in its final form and sent to the project for review. The compendium will be published in three volumes, one of which will be classified.

Phase 1 of the compendium program, selection of references, was completed during the preceding quarter.

Phase 2, typing of material not available for thermofax copying, was completed early in the quarter of this report.

Phase 3, study of annotation of selected references, proceeded rather sporadically. A list of about 850 references was selected for examination. At the close of the period, 143 had been examined and 91 annotations prepared. About half of these have been typed. The goal of this study is a collection of 300 to 350 items, on which the reviews are to be based.

Phase 4, subject classification, commenced late in February and was completed by the end of March. A 2400-item bibliography is being sent to Presentations for processing while the study and reviews are already completed. Since classified abstracts cannot be sorted in the existing office arrangement, the one volume of ACR abstracts is to have special handling; this means 378 items arranged according to abstract number. The lists of process report references only are complete and typed. This is the last component of the unclassified bibliography.

Phases 5 and 6, properties and processes reviews, were not active during this period. The only significant change is a decision to omit all classified information in preparing the reviews.

Phase 7, author and report number indices for the bibliography, has been cancelled.

In Phase 8, completion of the compendium, the planning guide has been extended from 21 to 41 items. All work will be delivered to the publishers during the next quarter.

In Phase 9, postmortems, the plan to use the assistance of Office of Technical Information Services, Oak Ridge National Laboratory (OTIS), assistance was dropped early in this period, due to a long delay in their publication date. A new plan to use the Martin Presentations Section for production was evolved in mid-February. The Martin Company has selected "Isotopic Power Sources" as a provisional title for the compendium.

b. Project direction

Under a system for assuring adequate project review of the Fuel Technology Development Program, all processes being developed are first presented to the Design group in the form of a process specification. After project review and approval, the process specification is translated into a process design specification, which is similarly reviewed by Health Physics and the Quehanna Hot Cell committee.

Prior to actual activation all facilities that are to be licensed receive a review by the Martin Radioisotopes Committee to ensure that compliance with all regulations set forth under the licensing agreement have been met.

During this quarter several process specifications were developed and reviewed. One process design specification (americium encapsulation) was also completed.

The project direction also included the preparation of the monthly, quarterly and topical reports, required under the contract, to keep the AEC informed of the progress of the program and of the technical advances accomplished.

3. Technical Discussion and Evaluation

The nature of the compendium requires a highly generalized discussion in order to adequately convey the information on radioisotope application to auxiliary power heat sources.

The special objective of Phase 3 is the critical evaluation of experimental work and data reported in the literature. The properties of radioisotopes do not have the same constancy expected for stable isotopes. Autoradiolysis is a pervasive limiting factor in experiments on a large scale. The isotopic composition of a particular sample of a radioisotope can vary with treatment history, with a significant influence on properties. The reliability of much of the thermodynamic and kinetic data for the radioisotopes must be considered very skeptically. The hazards involved in handling some of the isotopes, notably Plutonium-239, make experimentation difficult. As a result, process information is even less well developed and less reliable than properties information.

Phase 4--Subject Classification. This work deals with indexing. The chemical elements form a basic index, and the alloys and compounds, properties and processes are the major reference categories. Attention to the individual isotopes is given wherever possible, with miscellaneous subdivisions to collect references covering more than one isotope. Without a detailed subject index, the arrangement of abstracts determines ease of reference.

Phases 5 and 6--Properties and Process Reviews. No evaluation can be made on the reviews until the work on them has progressed farther. Completions and postmortems (Phase 8 and 9) are routine and do not require technical discussion.

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

During the last quarter the processing equipment in the Heavy Elements Processing Facility (HEPF) was installed and testing of the operation was started. For the americium purification system the valves originally procured would not operate properly. Development of a new valve which would withstand 10^8 roentgens (r) total gamma radiation without failure was initiated. For the encapsulation of target material (americium) the process equipment, including the Denison Press, was activated. Dry runs using gadolinium oxide to simulate americium in the encapsulation and welding steps were conducted. Some design modifications were necessary to reduce the shrinkage of the aluminum shell during compaction. Die lubricants, although they assisted, did not wholly resolve the difficulties. Further testing of the system included the determination of the number of compacts of aluminum powder, ram pressures and differential pressure between rams.

The problem of pressure buildup due to helium and oxygen formation during the conversion of americium to curium led to a reanalysis of the stresses in the aluminum container for the americium target material. The use of a void space or a getter to absorb the oxygen was evaluated, and it was decided that, although cesium would be satisfactory, the additional contamination was undesirable. A new capsule design using Type 5052 aluminum instead of the softer Type 1100 F18 aluminum is being considered.

After investigations of the use of gaseous chlorine for the sublimation of aluminum away from the curium oxide, it was revealed that a volatile chloride of curium may also be produced. This phase of the investigation must be deferred until volatility data on curium chloride are available. As an alternate method of removing curium, an alkaline dissolution method appears to be feasible if the evolution of hydrogen can be eliminated. Sodium nitrate added to the alkali will reduce the hydrogen content through an oxidation-reduction method. Studies will be conducted during the coming quarter to establish the effectiveness of this process.

1. Objectives

a. Nuclear chemistry

For the Nuclear Chemistry Section the objectives for this period were:

- (1) Americium purification process--to continue experiments on the purification of americium by ion exchange methods,

with ammonium thiocyanate, in order to determine the operating conditions of the process to be used in the HEPF; to revise and update the americium processing manual (Ref. 2); to further test the Valcor valve for operational stability during liquid transfer; to make necessary installations and alterations in the americium processing boxes and install the Valcor valves.

- (2) Americium encapsulation--to check out and modify equipment for americium encapsulation and determine procedures for handling. Additional dry runs are to be performed using gadolinium oxide to demonstrate the full scale encapsulation and welding of the americium target capsules with the new end-cap design, Type 5052 aluminum shell and ultrapure aluminum powder.
- (3) Alkaline dissolution process--to continue experimental work on the caustic dissolution of aluminum target capsules so as to establish the temperature conditions, gas evolution rate, reaction time and purity of the precipitate and to determine the method for controlling the rate of reaction.
- (4) Curium purification process--to run curium purification experiments in order to establish the process parameters. Americium has been used to simulate curium in those experiments testing the efficiency of the hydrochloric acid and alpha-hydroxyisobutyric acid eluants. To prepare a preliminary curium process specification for design study; to construct an alpha sample volatilizer and counting apparatus for curium analysis.

b. Design--equipment and facilities

For the period January through March 1961 the design objectives were:

- (1) To review the radioactive waste disposal procedures for the relatively high activity waste from the processing facility.
- (2) To provide the following drawings for the design of HEPF equipment:
 - (a) Installation drawing of Facility.
 - (b) Hood installation in waste disposal cell.
 - (c) Equipment in hood (with specifications).

- (d) Alarm systems in glove boxes (with specifications).
- (3) To specify the materials for installation of the system for liquid waste disposal from the americium purification boxes.
- (4) To initiate layout and design of the dissolver system for dissolution of the irradiated curium capsule.
- (5) To initiate layout and design of the purification system to separate americium and curium from the aluminum, plutonium and fission products after dissolution of the target capsules (after irradiation).

2. Scope of Work

a. Americium purification

Experiments have been conducted in the laboratory following the proposed procedure for the purification of americium (Fig. 1). The process does, however, present a waste problem in that large volumes of waste are generated. Efforts are directed toward modifying the procedures to reduce this waste volume.

The americium processing manual (Ref. 2) was revised and brought up-to-date.

Modifications and alterations were made in the americium processing boxes to accommodate the new Valcor valves.

Testing of the Valcor valves for both operational reliability and radiation damage has been completed.

The processing procedures for the preparation of the americium capsule were reviewed and modified where necessary. The disposal system for the liquid wastes was designed. Appropriate drawings were prepared and equipment was ordered and installed to prepare the facility for "hot" operations.

b. Americium encapsulation

Each of the operations in the encapsulation process was checked and, where necessary, modified until all were considered satisfactory. The operations are mixing, compacting (new capsule design), removing the oxide layer from the top of the capsule, welding, ultrasonic cleaning, and helium leak detecting. Welding procedures were established for joining the bottom and top caps to the newly designed capsules. Thermal conductivity data were obtained for a powdered aluminum product with a density 83% of theoretical.

Column I

Column II

Column III

10

Fig. 1. Flow Sheet for Americium Purification

c. Alkaline dissolution

In the dissolution of aluminum pellets, the most serious problems encountered are controlling the rates of reaction and hydrogen gas evolution. The reaction rate can be controlled either by temperature regulation or by maintaining a minimum concentration of sodium hydroxide in solution; hydrogen evolution can be eliminated by the use of NO_3^- to react with the hydrogen, forming ammonia. The literature (Refs. 3 and 4) indicates that addition of sodium hydroxide to a nitrate solution will control the reaction rate; however, the evolution of gases will be at a minimum if the following equation is adhered to:

d. Curium purification process

For adequate decontamination, the curium should be processed through three separate ion exchange columns in the following order:

- (1) Dowex 1 column with lithium chloride and hydrochloric acid as eluants.
- (2) Dowex 50 column with 0.5M hydrochloric acid and concentrated hydrochloric acid as eluants.
- (3) Dowex 50 column with alpha-hydroxyisobutyric acid as the eluant.

Experiments were therefore run to determine the operating conditions of these columns using americium tracer to replace curium. Although experiments using curium are planned, appropriate facilities to handle the curium must first be secured. Two glove boxes have been made available and are being modified to allow for a complete run of the process as described above.

From the data obtained thus far, a preliminary purification process specification has been drawn up to include all steps from dissolution of a pellet to final product.

Construction of the sample volatilizer is being held up, pending the arrival of those parts which had to be ordered. The construction of the counting apparatus has been completed and testing of the assembly is under way.

The dissolution and purification of the irradiated americium capsule is recorded in a flow diagram. The design of some of the vessels to be used in the processes was initiated, as was the design of the containment box in which the process vessels are to be located.

3. Technical Discussion and Evaluation

a. Americium purification

The ammonium thiocyanate elution of fission products from a Dowex 1 column as a means of purifying americium yields large volumes of wastes and must be evaluated further. To reduce this waste volume, several modifications in the procedure were tried, in which resin, particle size and column areas were varied. Figure 2 shows the results of varying the resin size. Although the number of column volumes necessary to elute the fission products remains about the same, the overall volume is reduced by a factor of approximately 2. The data also indicate that the large particle size (50-100 mesh) gives more definitive separation of the fission products than the 200-400 mesh resin (Fig. 2).

Modifications of the valve brackets and inlet-outlet tubing in the americium processing boxes have been made to accommodate the Valcor valves, which differ in size from the valves formerly used. Installation of the valves should be completed in April.

The Valcor valves have been tested by exposure to a total gamma flux of 1.6×10^8 r. The irradiated valve was then cycled 200 times under vacuum. There was no evidence of leakage while the valve was closed. Current and voltage requirements for opening the valve did not show any change after irradiation. The Vitron A diaphragm used in the valve showed no loss of flexibility, although there was some physical change in the material (Fig. 3). Double barriers on solution lines carrying radioactive solutions between boxes have also been installed.

b. Americium encapsulation

Initial mixing tests, of one-hour duration, of aluminum powder and uranium dioxide powder (as a substitute for americium oxide) revealed many large agglomerates of the oxide present after mixing. Drying and sieving the uranium oxide powder addition prior to mixing it with aluminum produced a uniform distribution. With the drying and sieving method the discrete oxide particles were not apparent in a radiograph of a compacted capsule. Since sieving of the americium oxide powder was not considered desirable from a contamination standpoint, a screen was incorporated in the mixing container to eliminate the operation. Although this technique did not produce so good a mixture as sieving, it is acceptable. Figure 4 shows the mixture obtained when the oxide powder was dried and blended, utilizing a screen in the mixer. This method is considered satisfactory for blending the americium oxide and aluminum powder.

Fig. 2. Fission Product and Americium Removals in Americium Purification System--Variations with Mesh Size

Fig. 3. Americium Purification Box Valve

Fig. 4. Aluminum-UO₂ Blend in Capsule

Welding of the newly designed capsules, as shown in Fig. 5, was accomplished with a filler metal of Type 5052 aluminum, rather than the conventional Type 43 alloy. A more desirable weld was obtained with this filler material. It was also found that an Air Reduction-Wilson welding machine produced much cleaner and sounder welds than the P & H welding machine for this application.

Completed capsule shells of the new design were helium-leak checked and found leakfree. Pressurization of a capsule with hydraulic oil revealed that at 750 psi the outside diameter had expanded 0.0005 inch and that 1200 psi was required for an 0.001 inch expansion. The capsule failed because of a leak through the top weld at 2700 psi. The outside diameter of the capsule, at room temperature, had expanded 0.012 inch at the weakest point at this pressure.

The newly designed capsules were checked on the Denison press and found to work satisfactorily. The particle size of the ultrapure aluminum powder was suitable for obtaining the required density of 83 to 84% of theoretical. The dispensing unit on the press was altered so that 15 cycles would give the desired height of the core in the capsule. This height allows the top lid of the capsule to be positioned within $\pm 1/16$ inch from the top of the shell, a distance which is satisfactory for welding. The compacting operation is now considered satisfactory for making capsules containing americium oxide-aluminum mixtures.

The initial method for removing the oxide layer from the seal area of the capsule prior to welding was to etch the capsule in a sodium hydroxide solution. Since the sodium hydroxide could come in contact with americium core by this method, it was abandoned and a standard reamer was used to remove the oxide layer. The reamer removes 0.001 inch of stock, $1/8$ inch in depth, from the inside diameter of the capsule shell, and removes 0.001 inch from the top face of the shell. Tests have proved this method satisfactory.

Ultrasonic cleaning of capsules was successfully accomplished with a mixture of green soap and water (5 cc to 1000 cc, respectively). Thirty seconds of cleaning time was sufficient to decontaminate the test capsules.

The top lids of the capsules have a center vent hole to permit seal welding of the capsule without blowout of the edge seal due to the heated-gas pressure buildup. Blowout during welding of this center hole at the time of final closure did occur. Therefore, an aluminum wedge was driven into this hole prior to welding to ensure a sound weld and prevent blowout.

The helium leak check procedure was perfected with the equipment to be used in the americium line. A filter has been incorporated between

Fig. 5. Revised Design of Aluminum Shell for Aluminum Targets

the chamber which contains the capsule and vacuum pump and the mass spectrograph so that no radioactive material will be discharged outside of the box. Thirty minutes is required to test each capsule.

Lead shields and containers were made to reduce gamma activity exposure. Special handling tools were fabricated to permit handling of the isotope at a distance to reduce the neutron activity exposure.

Dry runs were made in the system to determine whether the handling equipment worked satisfactorily and whether the required distances from the isotope source can be maintained.

Thermal conductivity tests were run comparing the conductivity of wrought aluminum and a specimen of compacted aluminum powder (with rare earth oxide) cut from a capsule. Figure 6 shows the difference in conductivity. Deviation in the results is $\pm 5\%$. As a result of the lower thermal conductivity, the centerline temperature of the capsules will be higher than originally calculated.

4. Design

a. Heavy elements processing facility

The disposal procedure for the liquid waste from the americium purification box was modified. An earlier procedure required mixing of the liquid waste with a concrete aggregate. The attendant dusting problem that arises during mixing was eliminated by substituting vermiculite as the retention medium. The radioactive waste solutions will be treated in the purification box with sodium hydroxide. The resultant alkaline solution will be transferred by vacuum to a head tank and will flow by gravity to a 55-gallon tank containing vermiculite. The ratio of 3 volumes of vermiculite to 1 volume of active waste solution will be used, with the solution being absorbed in the vermiculite. The final mixture yields the waste in a form acceptable both for transportation and final burial at ORNL.

A number of drawings were prepared in connection with installations at the HEPF:

- (1) An assembly drawing locates the individual glove boxes in the facility, including the purification boxes for americium, the encapsulating and welding boxes for the americium capsule, as well as the laboratory bench area.
- (2) A detailed drawing was released for the installation of a hood. This hood is to be used for analysis of samples from the purification area and to check the surface contamination of the americium capsule prior to shipment for reactor irradiation.

Fig. 6. Comparison of Thermal Conductivities of Wrought Aluminum Versus Compacted Aluminum Powder

Equipment was specified and purchase requisitions were issued for the equipment to be installed, as follows:

- (1) Appropriate fittings were specified to retain fire alarm probes in the glove boxes. The probes are connected to the ADT alarm system installed in the facility.
- (2) Flexible metal hose was specified for the air-exhaust system from the glove boxes. The metal hose will cover the rubber hose connectors between the box exhaust and the HEPA air-exhaust manifold and will serve as a secondary barrier in case of fire.
- (3) Regulators and associated tubing were specified for an argon purge system. The argon system will be installed to serve as an inert atmosphere purge in the americium encapsulating boxes in the event of a fire.

b. Curium dissolution process

The dissolution of aluminum pellets in sodium nitrate-sodium hydroxide solution involves two major difficulties:

- (1) The control of the rate of reaction to prevent solution from vigorous boiling and entrainment in the condenser. If excessive boiling occurred, the accompanying high rate of gas evolution might drive the solution through the condenser with the subsequent loss of product.
- (2) The hydrogen gas evolved in dissolving aluminum should be kept minimized to eliminate the explosion hazards.

The first problem may be overcome by the addition of sodium hydroxide at a constant rate or by cooling the reaction vessel, when large concentrations of sodium hydroxide are present. Experiments to control the concentration of sodium hydroxide have resulted in precipitates heavily loaded with aluminum. Table 1 shows a typical analysis of a precipitate from one of the experiments. It can be seen that substantial quantities of silicon are also present, indicating some reaction with the pyrex glass vessel. Vycor may be substituted for pyrex glass to reduce the alkaline attack on the glass vessel.

TABLE I

Analysis of Precipitate after Dissolution of Aluminum Pellets

Conditions:140 gm Al + 4.2 gm Gd_2O_3 dissolved in 1 liter of 3.5M $NaNO_3$

+ 450 ml 30% NaOH

<u>Element</u>	<u>Run 1 (gm)</u>	<u>Run 2 (gm)</u>	<u>Average (gm)</u>	<u>% of Precipitate</u>
Si	3.045	3.105	3.075	17.8
Gd_2O_3	2.828	2.796	2.812	16.2
Al	10.990	11.431	11.210	64.5
Ni	Not detectable			
Fe	0.326	0.3245	0.325	1.9

Three full-sized pellets were dissolved in the following manner:

- (1) The first, in 1.5 liters of 3.5M sodium nitrate with 450 ml of 50% sodium hydroxide added dropwise.
- (2) The second, in 1.0 liter of 3.5M sodium nitrate with 450 ml of 50% sodium hydroxide added dropwise.
- (3) The third, in 1200 ml of 5.9M sodium nitrate with 600 ml of 50% sodium hydroxide added dropwise.

In all cases the volume of precipitate was excessively large. The last experiment yielded a precipitate containing less aluminum. Current analyses still show evidence of silicon. The literature (Refs. 5, 6) states that mole ratios of 1.65 NaOH/1.65 $NaNO_3$ /1.00 Al at the start of the reaction will yield suitably stable solutions. Therefore, dissolution experiments using these ratios with control of reaction rate by cooling will be made to test the efficiency of separation of the aluminum from the rare earths (fission products).

c. Curium purification

For the decontamination of Curium-242 a process involving three ion exchange columns has been proposed. The column conditions being studied are, respectively:

- (1) Column 1--lithium chloride solution elution of fission products from a Dowex 1 column, followed by an 8.5M hydrochloric acid elution of the curium.
- (2) Column 2--An 0.5M hydrochloric acid elution of light fission products from a Dowex 50 column, followed by a concentrated hydrochloric acid elution of curium.
- (3) Column 3--A 0.4M alpha-hydroxyisobutyric acid elution from a Dowex 50 column for final decontamination.

Accordingly, a number of experiments have been run to demonstrate the operation of each of the steps in the process. Figures 7, 8 and 9 show the results of Step 1. Satisfactory separations and reproducibility are evident from the curves with either 50-100 mesh or 200-400 mesh Dowex 1 resin. Decontamination factors (300 to 700) in accordance with that quoted in Ref. 16 were obtained.

Figures 10 and 11 show the results of Step 2 using Dowex 50 resins. Although two different resins (Dowex 50 x 4 and Dowex 50 x 8) were used, elution volumes are very nearly the same while considerably greater decontamination is evidenced with the Dowex 50 x 8 resin. It should be noted at this point that the decontamination factors of 10 and 100, obtained for the 50 x 4 and 50 x 8 columns, respectively, may not be wholly realistic because the feed solutions had not been previously processed with lithium chloride.

In the next series of experiments different feed solutions and different resins were used, but in all cases 0.4M alpha hydroxyisobutyric acid was the eluant. Figures 12 and 13 show the elution of fission products and americium (Cm)*, respectively, from Dowex 50 x 12 resin. However, considerable improvement in separation and elution fractions is seen when the feed solution is made to 0.4M alpha-hydroxyisobutyric acid, rather than 0.1M hydrochloric acid. The same observations are true with Dowex 50 x 8 (Figs. 14, 15) resins with even sharper separations evidenced.

Figure 16 shows the results of a duplicate run of the experiment shown in Fig. 10, except that in this case sampling was done by two different methods. The first method of sampling (Ref. 7) was to withdraw an aliquot of solution and prepare a planchet. In the second method (Ref. 8) the sample, after being withdrawn, was precipitated first as the oxalate and then as the hydroxide before the planchet was prepared. This was done in an attempt to determine whether the fission product peak under the americium (Cm)* peak might be due to such elements as cesium or strontium. A finer cut of the elution solutions from that portion of separation where the fission products and curium are eluted together shows a short-lived beta activity with an E (max)

*Curium tracer.

Fig. 7. Ion Exchange Separation of Fission Products from Americium-241

Fig. 8. Ion Exchange Separation of Fission Products from Americium-241

Fig. 9. Ion Exchange Separation of Fission Products from Americium-241

Fig. 10. Ion Exchange Separation of Fission Products from Americium-241

C. SUBTASK 6.3--RADIOISOTOPE FUEL FORM DEVELOPMENT

Investigations to evaluate fuel compounds of curium and plutonium for compatibility with the matrix and container materials at elevated temperatures and under the various conditions of operation have been conducted. Gadolinium, cerium, uranium and rare earths were used as substitutes for curium and plutonium. Pellets of the carbides and oxides were prepared for determining the methods of manufacture, and for establishing the purities and yields of the compounds. Distribution of fuel in the matrix materials was determined by X-ray, with uranium dioxide particles simulating particles of curium oxide. Satisfactory compactings, up to 98.3%, could be achieved by the techniques developed. Further investigations of the fuel form compatibility with the container must be pursued.

The container material evaluations during the last quarter were devoted to establishing a correlation between the high velocity impact conditions of a re-entry capsule and the physical properties associated with the deformation of the material under test conditions of velocity and temperature. For the material selected, Type 316 stainless steel, the temperature of the capsule had little effect on deformation at high impact velocities, but the yield strength of the material appears to be one of the controlling factors. However, further evaluation of the test results will be required. The testing of Inconel X, a more temperature-dependent material, may also be required.

For the plutonium fuel form experiments, Plutonium-239 is to replace the more expensive 238 isotope in all evaluations. Processes for conversion of plutonium to an insoluble, stable compound are being sought. Although the oxide is suitable, it has two major drawbacks:

- (1) It is slightly soluble in water.
- (2) It is a high contributor to alpha-neutron reactions for producing neutrons.

The carbide is similar to the oxide, but withstands a higher temperature than the oxide. Depending on the operating temperature of the generator, either or both may be ruled out on the basis of stability within the container. Therefore, other compounds will also be sought. The present plans are to develop equipment for installation in the HEPF which will permit investigation of the properties of the plutonium compounds at the higher temperatures. The containment boxes to be developed must have an inert atmosphere to prevent oxidation of compound during fabrication. Process specification for this system is currently being formulated.

1. Objectives

The objectives of this subtask are:

- (1) Curium fuel form development--to investigate the preparation of curium compounds, with gadolinium compounds as substitutes, and determine the problems involved in, and the procedures for, making the curium compounds; to prepare gadolinium compounds to simulate curium compounds for compatibility studies with matrix and container materials.
- (2) Plutonium fuel form development--to investigate the preparation of plutonium carbides, with cerium carbides as substitutes, and determine the problems involved in, and the procedures for, making plutonium carbides; to prepare cerium carbides to simulate plutonium carbides for compatibility studies with matrix and container materials.
- (3) Containment of radioisotope--to present some of the problems involved in producing containers and fuel forms in light of the present philosophies of containment; to determine the compatibility of the compounds prepared under Items (1) and (2) with container and matrix materials for thermoelectric and thermionic generators.
- (4) Equipment and facilities (design, fabrication and installation)--to provide installation drawings for the location of glove boxes in the HEPF.

2. Scope of Work

a. Fuel form development

An investigation of the preparation of gadolinium carbide to decrease the time required to produce the carbide was undertaken during the quarter of this report. Time and temperature were varied in locating an optimum point at which the best carbide could be produced in the shortest time. X-ray diffraction patterns were studied to determine the purity of the carbide.

The parameters for producing dense cerium carbide pellets which would simulate plutonium carbide were established.

b. Container material development

The problems involved in meeting the requirements of the various philosophies of containment were evaluated. Compatibility tests of simulated compounds and metals at the temperatures of thermionic and

Fig. 13. Elution Characteristics of Americium-241 in the Presence of Fission Products

Fig. 14. Elution Characteristics of Americium-241 in the Presence of Fission Products

Fig. 15. Elution Characteristics of Americium-241 in the Presence of Fission Products

Fig. 16. Elution Characteristics of Americium-241 in the Presence of Fission Products

of approximately 0.95. The results, however, are not conclusive because of the low level of activity. No evidence of any associated gamma activity was found on a sample analyzed with a multichannel analyzer.

The two glove boxes and additional ventilation to permit the use of curium have been set up (Fig. 17) and are being tested for the operation. A complete curium purification in accordance with the preliminary specifications will be run. The feed will consist of a sodium aluminate solution containing 3 millicuries of Curium-242 and 1 millicurie of fission products, and a rare earth carrier. After separation of aluminum by centrifugation, the solution will be adjusted to 12.5M in lithium chloride and 0.5M in hydrochloric acid and the mixture passed through a Dowex 1 column. An elution with lithium chloride and hydrochloric acid solutions follows the loading of the Curium-242 onto the column. The recovered curium fraction will be evaporated to dryness, taken up in 0.5M hydrochloric acid and passed through another Dowex 50 column, to be followed by elutions with 0.5 hydrochloric acid and concentrated hydrochloric acid. The curium fraction will again be evaporated to dryness and taken up in 0.4M alpha-hydroxyisobutyric acid at pH 4.48, then passed through a second Dowex 50 column, followed by elution with 0.4M alpha-hydroxyisobutyric acid, pH 4.48. Analytical samples will be taken at intervals throughout the entire run, and decontamination factors and recoveries will be calculated on the basis of analytical results.

A preliminary process specification has been submitted to the project. It covers the entire process from pellet dissolution to final product, curium oxide, and is based upon the results of the experiments conducted in the laboratory, in accordance with available literature. From this process specification a preliminary design will be drawn up.

The alpha radioisotope volatilizer for the preparation of analytical samples is currently being fabricated in the Nuclear Components Department. The alpha counting apparatus has also been constructed, including the solid state detector and housing, preamplifier and power supply. It is now being tested. There are some electronic difficulties still to be resolved, such as radio-frequency noise and shielding. With a single channel analyzer, pulses have been recorded, but they do not give satisfactory amplitude or resolution. The electrical circuitry and tubes for the unit are being tested and checked.

d. Alkaline dissolution of aluminum capsules and curium purification

A flow diagram showing the steps and equipment to be used in the dissolution and purification of an irradiated americium capsule was prepared. The flow sheet is based on the specifications supplied by the Materials Section. The flow diagram includes provisions for:

Fig. 17. Laboratory Experimental Curium Containment Boxes

- (1) One dissolver vessel, to dissolve the capsule.
- (2) Three resin columns, to effect a separation of the americium-curium fraction from fission products and plutonium.
- (3) Three feed adjustment vessels, to prepare solutions for loading on the resin columns and for their elution.
- (4) Two filters, to separate precipitates from supernatant solutions.
- (5) Adequate receiver vessels, to store all solutions, in recognition of the need for sampling all solutions to prevent inadvertent discharge of americium and curium bearing solutions.

Design effort was initiated to size the dissolution vessel to handle the caustic dissolution of one capsule. Features of the design will include the following provisions:

- (1) To insert a metal capsule into the system.
- (2) To heat the dissolved solution to 90° C before dissolution.
- (3) To cool the solution during dissolution.
- (4) To measure the solution temperatures.
- (5) To air-agitate the combined solutions.

Design effort was initiated to prepare a containment box which will house the equipment in Cell 3 at the Quehanna Facility (QHCF). Initial design features will provide for:

- (1) Transfer of samples from box.
- (2) Transfer of capsule into box.

5. Conclusions

a. Americium purification

The coarser resin size (50 to 100) in the Dowex ion exchange column used for thiocyanate elution serves to reduce the overall volume of solution necessary for the separation of fission products from americium.

The Valcor solenoid valves have satisfactorily passed the irradiation and cycling tests to which they were subjected.

b. Americium encapsulation

The mixing, compacting, preweld cleaning (oxide layer), welding and helium leak detection procedures are all ready for hot operation. Other phases of the system which have been explained in the previous quarterly report (Ref. 1) are also ready for hot operation. No difficulties are foreseen in the preparation of capsules for irradiation.

c. Alkaline dissolution of aluminum targets

Difficulties encountered in the dissolution of aluminum pellets in sodium nitrate-sodium hydroxide indicate that further investigation is needed. Although reaction rates and gas evolution have been controlled satisfactorily, excessive amounts of precipitate result. A survey of the literature has shown that various procedures were used to accomplish dissolution. Additional experiments will therefore be conducted to find the method that will minimize the amount of impurities in the curium-amerium precipitate.

d. Curium purification processes

The individual steps in the curium purification process have proceeded satisfactorily, with the objectives being accomplished. No further work is anticipated for bench-scale operations. A complete three-column run will be made in the glove box setup using curium levels of approximately 7×10^9 disintegrations per minute.

In the next quarter enough information should be available for a final draft of the curium process specifications.

More work on the pulse-height counting assembly will be required before satisfactory results will be obtained. The Health Physics and Electronics Sections will collaborate with the project to eliminate all the difficulties.

e. Equipment and facilities

Design, fabrication and installation. A satisfactory method for handling liquid wastes from americium purification is achieved by absorption in vermiculite. A satisfactory solenoid valve is produced by the Valcor Company. It has withstood radiation doses of 10^8 r without functional deterioration.

C. SUBTASK 6.3--RADIOISOTOPE FUEL FORM DEVELOPMENT

Investigations to evaluate fuel compounds of curium and plutonium for compatibility with the matrix and container materials at elevated temperatures and under the various conditions of operation have been conducted. Gadolinium, cerium, uranium and rare earths were used as substitutes for curium and plutonium. Pellets of the carbides and oxides were prepared for determining the methods of manufacture, and for establishing the purities and yields of the compounds. Distribution of fuel in the matrix materials was determined by X-ray, with uranium dioxide particles simulating particles of curium oxide. Satisfactory compactings, up to 98.3%, could be achieved by the techniques developed. Further investigations of the fuel form compatibility with the container must be pursued.

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- (3) Containment of radioisotope--to present some of the problems involved in producing containers and fuel forms in light of the present philosophies of containment; to determine the compatibility of the compounds prepared under Items (1) and (2) with container and matrix materials for thermoelectric and thermionic generators.
- (4) Equipment and facilities (design, fabrication and installation)--to provide installation drawings for the location of glove boxes in the HEPF.

2. Scope of Work

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The parameters for producing dense cerium carbide pellets which would simulate plutonium carbide were established.

b. Container material development

The problems involved in meeting the requirements of the various philosophies of containment were evaluated. Compatibility tests of simulated compounds and metals at the temperatures of thermionic and

thermoelectric generators were undertaken. Tests were performed with both matrix and container materials.

Oxidation tests were run on metals considered for thermionic heat sources at the equilibrium temperature in air which would exist for a bare heat source. This condition could exist in case of an abort if the heat source was freed from the generator.

c. Design, fabrication and installation

The location of the glove boxes for plutonium processing was fixed in the HEPF.

3. Work Accomplished and Evaluation of Effort

a. Curium fuel form

Gadolinium carbide, as a substitute for curium carbide, was first prepared by reacting a mixture of gadolinium oxide and 25% excess carbon in an argon atmosphere at 1800° C for 4 hours. The carbide thus formed produced a good X-ray pattern. Carbide formed from a mixture of gadolinium oxide and a 15% deficiency of carbon, a stoichiometric amount of carbon, and a 15, 35 and 50% excess of carbon reacted at 2000° C for 16 hours to also produce good X-ray patterns. These carbides, however, had melted and some carbon and other compounds were found. A series of tests was then performed to determine the minimum time required to produce good carbide. The results are given in Table 2. All tests to date have been performed in a carbon tube furnace.

The tests indicate that at least two hours of sintering time is required to produce satisfactory carbide and that a stoichiometric mixture and carbon contents up to 35% excess are satisfactory. Since melting was observed in pellets sintered at 1950° C, and a sintering at 1800° C produces good carbide, a temperature of 1875° C for two hours will be used for further carbide preparation. Further testing will be conducted in an eddy-current concentrator induction furnace, of the type to be used in the hot cells, which has been fabricated for this purpose.

Since the high specific power of curium necessitates material dilution, compacts of high density curium carbide are not required. Therefore, no attempt has been made to produce high density pellets.

b. Plutonium fuel form

Cerium dicarbide, as a substitute for plutonium dicarbide, was successfully prepared during the last quarter; however, the density of the compacts prepared was low; they would therefore be unsatisfactory for

TABLE 2
Results of Gadolinium Carbide Studies

Composition	Sintering		X-ray Pattern		Number of Other Phases Present	
	Time (hr)	Temperature ($^{\circ}$ C)	Run 1	Run 2	Run 1	Run 2
Stoichiometric	1	1800	P	P	2	4
	2	1800	F	F	1	0
	4	1800	F	F	2	2
	1/4	1950	F-G	P	0	5
	1/2	1950	F-G	F	0	1
	1	1950	F	F-G	1	0
	2	1950	F-G	G	0	0
35% excess carbon	1	1800	P	P	1	4
	2	1800	F-G	G	0	0
	4	1800	F	G-E	1	0
	1/4	1950	P	F	1	5
	1/2	1980	P	G	2	1
	1	1950	F	F-G	2	4
	2	1980	G	G	0	3

P = poor

F = fair

G = good

E = excellent

use in heat sources. During this quarter, tests were performed to find the conditions required to produce highly dense compacts. The parameters studied were pressure and sintering time. The raw materials (cerium oxide and carbide) were blended, pressed at 20 tons per square inch (tsi) and sintered; they were then crushed, repressed and resintered to produce the final product. All sintering was at 1625° C (2957° F) in a carbon tube furnace, under an argon atmosphere. Table 2 gives the parameters used and the results of these tests. All compacts, except those noted, were prepared from small pellets which were crushed and repressed individually. Two density values are given in the final sinter column because the pellet was removed from the furnace and checked for density and then returned to the furnace for further sintering.

Analyzing the data in Table 3 indicates that 40 tsi was the best pressure for the second compacting. An initial sintering time of eight hours to prepare the carbide and a final sinter of eight hours to increase the density of the compacts produced pellets with densities equal to, or better than, 90% of theoretical. Discussions with personnel at Los Alamos Scientific Laboratory revealed that they required approximately the same sintering times to produce good plutonium carbide. Further tests will be run in an induction furnace, utilizing the optimum parameters found. Since there are conflicting data on the melting point of plutonium carbide, several experiments with Pu-239 carbide will be run to verify the optimum sintering temperature.

c. Containment of radioisotopes

There are two design approaches to the containment of radioisotopes for space missions. At the end of space duty, the missile will re-enter the atmosphere at high velocity and will be subjected to extremely high temperatures. One approach to container design favors burnup of the container and the isotope to particles at least as small as 10 microns before the unit drops to a 100,000-foot altitude. Particles of this size would remain suspended in air above 100,000 feet. The other design approach favors retaining the isotope, unexposed, in the container throughout re-entry, and landing the unit intact. The state of missile art for the thermoelectric generator units has not progressed to a point where the return of the radioactive fuel and container from one orbital flight to any designated location on the earth can be definitely assured. The assurance of placing the isotope and container in a position where they can be burned up after orbital flight has been mathematically shown to be 100%; but the unit must be ejected from its vehicle. Neither total burnup nor safe impact at terminal velocity, with total containment, can be assured above a certain altitude on a nonorbital flight for a thermionic system at the present time.

The burnup of isotopes requires:

TABLE 3

Effect of Sintering Time and Compacting Pressure on Carbide Density

Carbon in Mixture	Time		Repress Pressure (tsi)	Pellet Height (in.)	Density (% of theoretical)	
	Initial Sinter (hr)	Final Sinter (hr)			4-Hour Final Sinter	8-Hour Final Sinter
15% deficient	8	8	40	1/5	--	75.5
Stoichiometric	4	8	40	1/5	--	87.2
	8	8	40	1/5	--	89.2
	4	4-4	40	1/2	75.2	85.4
	4	4-4	40	1/2	75.0	81.6
	8	4-4	40	1/2	78.4	93.3
	8	4-4	40	1/2	87.2	97.1
	4	4-4	60	1/2	80.6	91.0
	8	4-4	60	1/2	88.7	78.4
25% excess	4	8	40	1/5	--	86.3
	8	8	40	1/5	--	89.4
	4	4-4	40	1/2	77.2	76.8
	4	4-4	40	1/2	73.0	77.1
	8	4-4	40	1/2	83.5	95.3
	8	4-4	40	1/2	--	96.7
	4	4-4	60	1/2	82.0	80.8
	8	4-4	60	1/2	89.2	88.7
50% excess	4	8	40	1/5	--	74.3
	8	8	40	1/5	--	82.9
*25% excess	8	4-4	40	1/3	75.2	88.3
	8	4-4	40	1/3	75.2	96.0
	8	4-4	60	1/3	77.5	84.0
	8	4-4	60	1/3	77.7	83.8
	8	4-4	80	1/3	77.9	80.7
	8	4-4	20	1/3	70.7	75.9

*Pellets prepared from carbide originally made in large pellets.
Sintering temperature 1625° to 1650° C.

- (1) The use of readily vaporized or low melting metals in the container or matrix.
- (2) A material which will aid in rapidly dispersing the isotope.

Very unstable isotope compounds or matrix compounds, for example, would be beneficial in aiding this disintegration. The use of these unstable materials, however, would present considerable difficulties in the fabrication of heat sources in hot cells where techniques are severely limited. The use of external aids to disintegrate the heat source is another possibility. Here again, the reliability of mechanical or electrical systems may be somewhat questionable. In high temperature thermionic applications, low melting metals are not applicable; therefore, burnup becomes a problem unless the container can be externally removed.

Similarly, the retention of the isotope presents as many problems as, if not more than, that of burnup route. The limitations of weight and volume, the need for designs based on electrical efficiency rather than containment, and the extremes of temperature and pressure (both external and internal) acting on the isotope container make absolute containment of the isotope very difficult. For absolute containment the container material must be able to withstand the gas pressure buildup, must be strong enough to withstand impact, must be oxidation-resistant enough to prevent exposure of the isotope, and must be sufficiently corrosion-resistant to prevent exposure of the isotope. A multilayer radioisotope container with a protective layer to prevent air oxidation may provide a plausible system.

For the thermoelectric or low temperature heat sources, many, if not all, of these requirements can be met; however, the high temperature heat sources for thermionic generators cannot readily meet these requirements at present. The only metals applicable in these high temperature sources are the refractory metals, whose high velocity impact resistance at elevated temperature is unknown and must be evaluated. The oxidation resistance of these materials is very poor, and attempts at alloying to improve oxidation resistance have not been successful to date. Also, coatings on these materials have not proved successful at the thermionic heat source operating temperatures.

These are only some of the problems confronting the design and materials engineer. The problems are outlined to establish the magnitude of the work required to achieve and ensure the desired containment of isotope heat sources for generators for space application.

d. Compatibility tests

The compatibility tests for the low temperature or thermoelectric heat sources are divided into a study of the compatibility of container materials with cerium compounds as a substitute for plutonium compounds, and a study of the compatibility of matrix materials with gadolinium compounds as a substitute for curium compounds. Tests of the compatibility of container materials with cerium and gadolinium compounds have been run. The testing temperature was 1093° C (2000° F) and the duration of the tests was 124 hours. The gadolinium and cerium oxides and carbides were tested in an argon atmosphere. Any material which did not react with the simulated isotope compounds at this temperature within 124 hours was considered to be acceptable for long time usage at lower temperatures. The compatibility was determined by observing the metal-isotope interface, metallographically. Table 4 gives the results obtained.

The compatibility of cerium compounds with Hastelloy C, Inconel X, Type 316 SS and Haynes 25 appears to be satisfactory. If cerium is a valid substitute for plutonium, these alloys should contain the plutonium compounds. A search of the literature (Ref. 15) has revealed that iron, stainless steel and columbium reduced the oxide PuO_2 to the oxide Pu_2O_3 at 1400° C (2552° F). The maximum amount of reduction was approximately 20%. The reference stated that iron, stainless steel and columbium should be suitable with PuO_2 under reactor conditions. This temperature (1400° C) is far in excess of anything expected in thermoelectric generators at the present time. No data on tantalum and molybdenum compatibility nor data on plutonium carbide compatibility were found in the unclassified literature.

The isotope compounds to be used in thermionic generators were subjected to compatibility tests with various metals. Only substitutes for curium compounds were tested, since plutonium does not produce enough power to be used in thermionic generators. Gadolinium oxide and carbide were used as substitutes for the curium compounds. The test temperature in this case was 1625° C (2957° F), since this temperature approaches the centerline temperature to be obtained in proposed thermionic generators. Testing time was 64 hours. Testing was performed with the simulated fuels in the following forms:

- (1) Cermets of 6 volume percent gadolinium compound and 94 volume percent metal.
- (2) Pellets of gadolinium compounds placed on pieces of metal.

All testing was performed in a carbon tube furnace under an argon atmosphere. Compatibility was confirmed by metallography. The results of the test are given in Table 5.

TABLE 4
Compatibility of Container and Matrix
Materials at 1093° C (2000° F)

Plutonium Compounds

<u>Container Material</u>	<u>Simulated Isotope Compound</u>	<u>Results</u>
Hastelloy C	CeO ₂	No attack
	CeC ₂	No attack
Inconel X	CeO ₂	No attack
	CeC ₂	No attack
		} excessive grain growth of alloy
Type 316 stainless steel	CeO ₂	No attack
	CeC ₂	No attack
Haynes 25	CeO ₂	No attack
	CeC ₂	No attack
Tantalum	CeO ₂	Attack (intergranular)
	CeC ₂	Attack
Molybdenum	CeO ₂	Attack (intergranular)
	CeC ₂	Attack

Curium Compounds

<u>Matrix Material</u>	<u>Simulated Isotope Compound</u>	<u>Results</u>
Nickel	GdO ₂	No attack
	GdC ₂	No attack
Iron	GdO ₂	Dislocation of oxide
	GdC ₂	Dislocation of carbide

TABLE 5
High Temperature Compatibility Test with Simulated
Curium Compounds

<u>Material Simulated</u>	<u>Isotope Compound</u>	<u>Results</u>
Tantalum	GdO ₂	Questionable
	GdC ₂	
Molybdenum	GdO ₂	No attack
	GdC ₂	
Platinum--5% Rhodium*	GdO ₂	Questionable (crystal phase forms)
	GdC ₂	

Pellet Tests

Zirconium	GdO ₂	Badly attacked
	GdC ₂	
Tungsten	GdO ₂	Attacked
	GdC ₂	
Molybdenum	GdO ₂	No attack
	GdC ₂	
Tantalum	GdO ₂	Questionable
	GdC ₂	
Columbium	GdO ₂	Questionable
	GdC ₂	

*Test at 1575° C (2850° F) for 64 hours.

Molybdenum appears to be the only material which is satisfactory beyond question. Zirconium and tungsten are definitely unsatisfactory. Further testing will be required on the questionable metals before their use as matrix materials is confirmed.

Oxidation tests of refractory metals were performed at 705° C (1300° F) in air. This temperature is the minimum to be expected in the event a heat source is ejected from its generator. The tests give an idea of the length of time unalloyed refractories would contain isotopes at this temperature. Table 6 gives the results of the test.

TABLE 6
Oxidation Tests of Refractory Metals

<u>Material</u>	<u>Weight Loss</u> <u>(mg/cm²-hr)</u>	<u>Mils/side-hr</u> <u>Lost</u>
Molybdenum	38.21	1.5
Tantalum	38.74	0.9
Tungsten	4.83	0.1
Columbium	17.94	0.8

From these data, it is apparent that tungsten is the only refractory metal which would contain the isotope any appreciable time at 705° C (1300° F).

e. Equipment and facilities--design, fabrication and installation

Installation drawings were released for the connection of glove boxes into the air exhaust system of the HEPF. The boxes will be used for the processing and subsequent encapsulation of plutonium fuel. The plutonium boxes were installed at this time to obviate the necessity of having to tie them in later into a "hot" exhaust system.

5. Conclusions

a. Curium fuel form development

Two hours of sintering time at 1875° C produces good gadolinium carbide. It is expected that these same conditions will be directly applicable to curium carbide, except for a slight variation in temperature. Carbon contents from stoichiometric to 35% excess can be utilized for the conversion of the oxide to carbide.

b. Plutonium fuel form development

Dense cerium carbide pellets (density at least 90% of theoretical) can be produced by sintering a compacted mixture of cerium oxide and 35% excess carbon for eight hours at 1625° C, crushing the formed carbide, and repressing and sintering for eight hours at 1625° C. All parameters, with the possible exception of temperature, should be directly applicable to the production of plutonium carbide. Several tests with Plutonium-239 should determine the temperature required.

c. Container materials

Cerium oxide and cerium carbide are compatible with Hastelloy C, Inconel X, Haynes 25 and Type 316 stainless steel at thermoelectric operating temperatures. References reveal that they should also be compatible with plutonium compounds.

Nickel will be satisfactory as a matrix material for curium oxide or carbide at thermoelectric operating temperature.

At 1300° F, the temperature of the thermionic generator heat source at atmospheric conditions, the only refractory metal which would contain the heat source for any reasonable length of time is tungsten. However, it should be noted that since both gadolinium oxide and carbide attack tungsten, it will be necessary to provide an inner liner of molybdenum to eliminate attack at the operating temperature of 1575° C (2867° F).

D. SUBTASK 6.4--SAFETY, THERMAL AND SHIELDING ANALYSIS

The safety analysis of the curium process progressed to a point where specific handling data were required before the work could proceed; therefore, this phase of the study has been temporarily deactivated. The physical properties of the radioisotopes presented in Ref. 9 were reviewed and updated to reflect recent data on specific activity (curies/gm) for compounds of the radioisotopes (see Table in Appendix A).

During the last quarter, sled and bazooka tests were conducted at Aberdeen Proving Ground (APG) to determine the effect of velocity and temperature on the alloys being considered for container materials. The evaluation and correlation of these tests with various physical properties are currently being studied. If successful, a correlation will also be attempted between the compressive drop tests conducted in the laboratories on similarly heated specimens and the physical properties.

The shielding analyses calculated to date include the americium processing system in the HEPF, the plutonium fuel form and chemical conversion systems in the same facility, and the necessary shielding for the americium and curium casks. Further studies are required to determine the effective neutrons and gammas from the irradiated americium target in the hot cells at Quehanna and to establish the amount of additional shielding required for the protection of operating personnel.

1. Objectives

For the shielding analysis effort, the objectives are:

- (1) To determine the shielding requirements for storing and shipping wastes from the americium purification process.
- (2) To compare calculated dose rates with experimental dose rates for encapsulated americium slugs.
- (3) To compare neutron attenuation by fast removal theory for neutrons from Curium-242 spontaneous fission with actual measurements.
- (4) To determine shielding requirements for storing and shipping wastes generated during curium processing.
- (5) To complete the shielding analysis for the plutonium processing line.

2. Scope of Work

For a comparison of calculated and experimental radiation dose rates, Dr. R. W. Hoff, consultant to The Martin Company, provided data of some shielding experiments performed at Lawrence Radiation Laboratory. One set of results gave the measured dose rates from an unirradiated slug containing a mixture of americium oxide and aluminum powder. The other set of results concerned the attenuation by polyethylene of neutrons from Curium-242.

Shielding requirements were reviewed for wastes from the americium purification absorbed on a vermiculite clay. Shielding and storage requirements were also reviewed for the wastes from the curium separations process in order to prepare them for ultimate disposal.

3. Technical Discussion and Evaluation

a. Americium processes

The present plan is to store and ship the waste solutions from the heavy element processes in 55-gallon drums filled with vermiculite. To perform the calculations, it was assumed that the isotopes were distributed uniformly throughout the drum.

The weights of pure Am-241 which could be contained in the vermiculite-filled drums for various surface dose rates are:

<u>Dose Rate (mr/hr)</u>	<u>Am-241 in Solution (gm)</u>
10	0.064
100	0.64
200	1.28

It is estimated that the amount of Americium-241 lost in the process wastes will be less than 1%. For 50 grams of americium, the maximum amount lost will be less than 0.5 grams; the surface dose rate would be 78 mrem/hr from gammas and 0.6 mrem/hr from neutrons.

The waste from the plutonium process line was conservatively assumed to be Plutonium-238, since it requires more shielding than the 239 isotope. For a dose rate of 10 mr/hr at the drum surface, about 28 grams of Plutonium-238 would have to be distributed throughout the drum. Since the amount estimated to be in the waste is only 0.5 gram after processing 50 grams, there will be no problem in storing the waste as proposed. For the estimated maximum Plutonium-238 content (0.5 gram), the dose rate at the surface will be 0.2 mrem/hr from gammas and 5.6 mrem/hr from neutrons.

b. Americium-241 dose rates

At Lawrence Radiation Laboratory, dose rates were measured at 10 centimeters from a slug 5/8 inch in diameter and 4 inches long, containing approximately one gram of relatively pure Americium-241 mixed with aluminum powder. Dose rates were also calculated with the aid of tables and graphs given in Ref. 10. The slug was assumed to have an aluminum casing with a 5/8-inch outside diameter and 0.050-inch walls. The overall length of 4 inches was assumed to include two end caps 1/4-inch thick. The fuel form in the capsule was assumed to be a mixture of 30 grams of aluminum powder and 1.133 grams of

americium dioxide (1.0 gram of pure americium). Dose rates were calculated for a point located 10 centimeters radially from the center of the capsule.

Measured and calculated results are given in Table 7.

Table 7 contains calculated values without and with buildup because of the uncertainty in the buildup factors for low energy gamma rays. Buildup factors were estimated by extrapolating and cross-plotting data given in Ref. 11.

c. Curium processes

A series of measurements (Ref. 12) of the spectral distribution of neutrons being emitted from a source of Cm-242 were made. The neutrons originate from spontaneous fissions and from alpha-neutron reactions with oxygen. Neutron attenuation was measured by using a Nemo boron trifluoride counter and a series of polyethylene absorbers of varying thicknesses. The experimental curves were then compared with similar curves of known spectra. By finding the "e" folding (relaxation) length, the energy was resolved with a calibration curve (E versus relaxation length). Two components were found:

- (1) A high energy component with $E = 5.8$ mev and an "e" folding length of $5\frac{1}{3}$ inches of polyethylene.
- (2) A low energy component with $E = 0.5$ mev and an "e" folding length of $1\frac{5}{8}$ inches of polyethylene.

It is probable that the high energy component is due to the alpha-neutron reaction of the 6.1-mev alphas of Curium-242 with oxygen. Compare the 5.8-mev component with the calculated maximum neutron energy of 6.3 mev (see Fig. 18). The low energy component is probably due to the neutrons from spontaneous fission of Curium-242.

Prior to the time the information was received from Dr. Hoff, it was assumed that all the neutrons were "fast" and that fast removal theory was applicable (Refs. 13 and 14). For conservation, the relation $1 \text{ mrem/hr} = 7 \text{ neutrons/cm}^2\text{-sec}$ was used to convert flux to dose rate for neutrons. The fast removal cross section Σ_n of polyethylene was calculated to be $0.112/\text{cm}^{-1}$. From this, the "e" folding length was calculated to be 3.51 inches. Fast neutron dose rates at one meter, calculated by using these same assumptions, are plotted in Fig. 19 for one gram of Curium-242 combined with oxygen for various thicknesses of polyethylene shielding. Also plotted in Fig. 19 are the dose rates found by using the high and low energy components measured in the experiments.

TABLE 7
 Calculated and Measured Dose Rates from Capsule
 Containing Americium and Aluminum Powder

<u>Shield</u>	<u>Measured</u>	Calculated	
		<u>Without Buildup</u>	<u>With Buildup</u>
None	2.40	3.75	--
3 in. graphite	1.20	0.33	2.17
0.125 in. steel	0.11	0.13	0.14
0.032 in. copper	0.52	0.74	0.85
0.007 in. lead	--	0.45	0.54
0.008 in. lead	0.96	0.34	0.42
0.066 in. lead	No measurable amount	0.00084	0.00095

Fig. 18. Maximum Energy of Neutrons from (α, n) Reactions Versus Alpha Particle Energy

Fig. 19. Fast Neutron Dose Rates at One Meter from Point Source of One Gram of Curium-242 Combined with Oxygen

E. THERMAL ANALYSIS FOR CURIUM DISSOLUTION AND PURIFICATION

During the last quarter, thermal analysis of the americium capsule in the reactor was further verified by the Safety Section to show a total heating rate of 10,953 watts. The thermal analysis also showed a centerline temperature of 422° F during reactor operation at a flux of 2.8×10^4 neutrons/cm²-sec. Thermal effects of the irradiated target capsule in the cooldown canal and in shipping cask were also evaluated.

1. Objectives

The objectives of this study were:

- (1) To determine heating and cooling requirements for dissolution and separation of the aluminum capsule from curium.
- (2) To determine pressure drop in the ion exchange columns during curium separation.
- (3) To evaluate a circulation system for the fuel form, inert atmosphere dry box.
- (4) To evaluate the calorimetry requirements for the final fuel capsule.

2. Scope of Work

Calculations were performed to determine heating requirements for the dissolver vessel in the aluminum capsule dissolution. Pressure drop calculations were considered for fluid flow through the purification resin columns. Systems were considered for purifying and maintaining an inert gas system in the planned fuel conversion containment box. The inert atmosphere may contain a maximum of 50 ppm of oxygen and must have a -50° C dew point. Several calorimetry systems were analyzed for use in the curium fuel capsule processing system.

3. Technical Discussion and Evaluation

a. Curium dissolution

The reaction vessel for the fuel capsule dissolution in the hot cells at Quehanna was estimated to be a 20-inch glass vessel, 4 inches in diameter, enclosed in a 5-inch diameter water jacket. Bare nichrome wire is coiled around the inner tube inside the water jacket to heat the dissolver solution to 90° C. The wire is approximately 50% efficient, and a 1-kw service will produce the desired reaction temperature in 15 minutes. An automatic on-off switch system can be used to maintain the reaction temperature after addition of the reactants. Data on cooling requirements during the reaction were not available.

b. Curium purification and fuel form processing

Calculations are in progress to determine the pressure drops to be anticipated when a variety of eluant solutions are passed through the resin columns in the curium purification system for the hot cells.

Cursory studies of various methods of maintaining the chemically inert, moisture-free atmosphere required for curium, uranium, or plutonium processing were made. Three methods were suggested:

- (1) Complete recirculation of box atmosphere with occasional purges.
- (2) A "bleed and feed" system where a certain percentage of the box atmosphere is replaced per unit time.
- (3) A "once-through" system when the dry, inert gas has a comparatively short residence time in the box.

The desired circulation rate was six complete changes per hour or 840 cfh. Tolerance levels given were 50 ppm for oxygen and a dew point of -50° F for water. For reliability and a minimum of maintenance, an electrical moisture removal system had previously been recommended.

For oxygen removal, a quick survey indicated that a catalytic reduction with hydrogen was the most efficient system. Purities of less than 1 ppm oxygen were guaranteed by the vendor, assuming no appreciable leakage of oxygen into the box. With a tolerance of 50 ppm and the recirculation rate prescribed, a leak of 0.21 cfh of air can be tolerated.

The oxygen removal system will cost from \$3500 to \$7000, depending upon the refinements desired. The choice of the three circulation systems suggested will depend on the amount of time in which the dry box is in circulation. Argon of less than 15 ppm impurities costs 7 cents/ft³. If the box is in operation only 100 hr/yr, the cost of operation of a "once-through" system would be less than \$6000/yr and all gas purification and drying could be eliminated. This type of system is highly insensitive to leaks in the box wall or glove punctures.

The gas recirculation system is recommended if the box is to be used more than 100 hr/yr, while the "bleed and feed" or "once-through" systems are suggested for more sporadic utilization.

Three general types of calorimeters were examined. In order of decreasing accuracy and probable cost, they were:

- (1) A flow-type calorimeter for extremely precise measurements with accuracy of the order of $\pm 0.1\%$.

Fig. 20. Neutron Dose Rate and Gamma Shielding Requirements for Curium Processing

loading the dissolver and removal from the evaporator vessel. Dose rates were calculated for distances of one and two feet from the centers of the vessels for this part of the process. For procedures where physical handling is required, dose rates at the surface of the container were also calculated. All calculations were based upon a plutonium content of 50 grams. The results are given in Tables 8 through 12.

Dose rates at the surface and at one foot from the center of the containing vessel were calculated to estimate the dose to the operator's hands and forearms. The dose rates at two feet were calculated to estimate the whole-body dose. Figure 21 shows the position of the operator in relation to the glove box.

The permissible dose to the whole body from ionizing radiation is 100 mrem for any seven-day period. With exposure for 40 hours during the seven-day period, a field intensity of 2.5 mrem per hour will produce 100 mrem. The permissible dose to the hands, forearms, feet and legs is 1500 mrem in a seven-day period. This is equivalent to exposure for 40 hours in a field intensity of 35 mrem per hour. Radiation fields higher than this can be tolerated if the exposure time is reduced.

The results do not vary greatly for vessels of different diameters and heights. If less than 50 grams of plutonium is present, dose rates would be lower by approximately the ratio of the weight present to 50.

4. Conclusions

The comparison of calculations with experimental data indicates good agreement. Examination of Table 12 shows that for dose rates from an americium capsule the calculated results are all higher than the measured values, except for the case of the 0.008-inch lead shield. No explanation can yet be offered for this difference. This point requires further investigation. The agreement between measured and calculated values is considered to be good, since the difference is less than a factor of two.

The use of fast-removal theory to calculate neutron dose rates gives results that are conservative for thicknesses of polyethylene up to ten inches. For greater thicknesses, this method of calculation gives low dose rates.

There are no apparent shielding problems associated with the proposed method of absorbing the waste from the heavy element processes in a 55-gallon drum filled with vermiculite. The drums containing the americium and plutonium wastes will meet shipping regulations without additional shielding on the basis of assumed maximum product losses of 1%.

TABLE 8

Gamma Ray Dose Rates Associated with the Processing of
50 Grams of Plutonium

	Dose Rates (mrem/hr)	
	<u>Pu-238</u>	<u>Pu-239</u>
50 gm plutonium nitrate (density assumed = 1) in vessel, 4 cm in diameter and 8.1 cm in height		
At surface } self-	4.7×10^3	200
At 1 ft } absorption	28.3	0.22
At 2 ft } only	7	0.055
Lead gloves (0.05-cm lead) at 1 ft	4.98	--
1/8-in. lead at 1 ft	2.54	--
1/4-in. lead at 1 ft	1.83	--
1/2-in. lead at 1 ft	1.0	--
1/4-in. lead glass ($\rho = 3.3$) at 1 ft	3.8	--
1/2-in. lead glass ($\rho = 3.3$) at 1 ft	2.6	--
3/4-in. lead glass ($\rho = 3.3$) at 1 ft	2.1	--
50 gm plutonium nitrate (1000-ml solution)		
Dose rate at 1 ft } self-absorption	34.8	0.11
Dose rate at 2 ft } only	10	0.027
Dose rate at 1 ft, 1/8-in. lead shield	1.2	--
50 gm plutonium in ion exchange column		
Dose rate at 1 ft	55	0.53
Dose rate at 2 ft	14	0.13
Dose rate at 1 ft, 1/8-in. lead shield	9.2	--

TABLE 9

**Dose Rates at Two Feet from Powdered Salts
Containing 50 grams of Combined Plutonium-238**

<u>Salt</u>	<u>Unshielded Dose Rates at 2 ft (mrem/hr)</u>			<u>Shield* Thickness (in.)</u>	<u>Shielded Dose Rates at 2 ft (mrem/hr)</u>		
	<u>Neutrons</u>	<u>Gamma</u>	<u>Total</u>		<u>Neutrons</u>	<u>Gamma</u>	<u>Total</u>
Carbide	12	7	19	5.0	3.0	2	5
Nitrate	2	7	9	None required	2.0	7	9
Oxide	8	7	15	3.4	3.0	3	6
Fluoride	1350	7	1360	21.5	3.0	-	3
Chloride	12	7	19	5.0	3.0	2	5

*Either water or polyethylene--thickness
required to give 3 mrem/hr at 2 ft

TABLE 10

**Dose Rates at Four Feet from 50 grams
of Plutonium-238 as Powdered Fluoride Salt**

<u>Shield* Thickness (in.)</u>	<u>Neutron (mrem/hr)</u>	<u>Gamma (mrem/hr)</u>	<u>Total</u>
16.6	3	0.014	3
12.3	10	0.034	10
9.9	20	0.061	20

* Either water or polyethylene--thickness required to give
3 mrem/hr at 2 ft.

TABLE 11
Neutron Dose Rates Associated with 50 grams of Plutonium-238
in Powdered Salt Form

<u>Salt</u>	<u>α, n</u>	<u>Neutron Production</u>			<u>Dose Rate (mrem/hr)</u>	
		<u>Spontaneous Fission</u>	<u>Total</u>	<u>Surface</u>	<u>1 ft</u>	<u>2 ft</u>
Carbide	4.06×10^6	2.2×10^5	4.28×10^6	$1.62 \times 10^{4*}$	49.7	12.4
Nitrate	3.67×10^5	2.2×10^5	5.86×10^5	$2.2 \times 10^{4*}$	7.2	1.8
Oxide	2.57×10^6	2.2×10^5	2.79×10^6	$1.05 \times 10^{4*}$	31.4	7.9
Fluoride	4.40×10^8	2.2×10^5	4.40×10^8	$1.7 \times 10^{6*}$	5380	1350
Chloride	4.06×10^6	2.2×10^5	4.28×10^6	$1.62 \times 10^{4*}$	49.7	12.4
Metal	--	2.2×10^5	2.2×10^5	8.54×10^3	2.57	0.67

* 4-cm diameter vessel; all materials in powder form

TABLE 12
Neutron Dose Rates Associated with 50 Grams of Plutonium-239
in Powdered Salt Form

<u>Salt</u>	<u>Neutron Production</u>			<u>Dose Rate (mrem/hr)</u>		
	<u>α, n</u>	<u>Spontaneous Fission</u>	<u>Total</u>	<u>Surface</u>	<u>1 ft</u>	<u>2 ft</u>
Carbide	1.09×10^4	24.6	1.09×10^4	41.0*	0.13	0.033
Nitrate	9.84×10^2	24.6	1.008×10^3	3.8*	0.012	0.003
Oxide	6.90×10^3	24.6	6.90×10^3	26.1*	0.084	0.021
Fluoride	1.18×10^6	24.6	1.18×10^6	4460*	14.4	3.6
Chloride	1.09×10^4	24.6	1.09×10^4	41*	0.13	0.033
Metal	--	24.6	--	--	--	--

*Vessel 4-cm in diameter; all materials in powder form.

Fig. 21. Position of Operator in Relation to Plutonium Glove Box

The radioactive wastes from the curium purification will require concrete shielding combined with the waste disposal container. Three inches of concrete will permit transportation of the container after approximately six months of storage. Storage is required to allow for the decay of the short-lived radioisotopes and to minimize the shielding requirement during transit.

There are no shielding requirements for the plutonium line when Pu-239 is processed. Vessels containing the fluoride salt, however, should not be held with the hands; one-foot tongs are recommended.

When the 238 isotope is being processed, dose rates are higher than for corresponding salts or operations with Plutonium-239. With the two exceptions of the neutron dose rates at container surfaces and the neutron dose rates from the fluorine salt, dose rates are not excessive. However, it is advisable to shield the sides and backs of the processing vessels with 1/8 inch of lead, and the fronts with 1/2 inch of 3.3-density lead glass to reduce the gamma dose rates to negligible amounts compared to neutron dose rates. Table 11 gives the thicknesses of water or polyethylene required to reduce neutron dose rates from powdered salts to 3 mrem/hr at two feet.

The fluorine salt is actually the only salt for which shielding is an absolute necessity. The 21.5 inches of material required to reduce the dose rate from the fluoride to 3 mrem/hr at two feet will probably involve considerable design problems. The thickness of shielding can be reduced by increasing source to operator separation distance and by increasing the field intensity. Table 12 gives the shield thickness at a 4-foot separation distance for several field intensities.

Higher radiation fields are permissible since the hydrofluorination process takes more than four hours to complete, and the operator is not required to remain in the vicinity of the process during most of this time. When the fluoride is converted to plutonium metal, the neutron dose rate will decrease by a factor between 10^2 and 10^3 .

If the fluorine process is used, special consideration will have to be given to the design of the apparatus to determine the best way to prevent overexposure of the operator during the time the Plutonium-238 is combined with fluorine.

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During the last quarter, thermal analysis of the americium capsule in the reactor was further verified by the Safety Section to show a total heating rate of 10.953 watts. The thermal analysis also showed a centerline temperature of 422° F during reactor operation at a flux of 2.8×10^4 neutrons/cm²-sec. Thermal effects of the irradiated target capsule in the cooldown canal and in shipping cask were also evaluated.

1. Objectives

The objectives of this study were:

- (1) To determine heating and cooling requirements for dissolution and separation of the aluminum capsule from curium.
- (2) To determine pressure drop in the ion exchange columns during curium separation.
- (3) To evaluate a circulation system for the fuel form, inert atmosphere dry box.
- (4) To evaluate the calorimetry requirements for the final fuel capsule.

2. Scope of Work

Calculations were performed to determine heating requirements for the dissolver vessel in the aluminum capsule dissolution. Pressure drop calculations were considered for fluid flow through the purification resin columns. Systems were considered for purifying and maintaining an inert gas system in the planned fuel conversion containment box. The inert atmosphere may contain a maximum of 50 ppm of oxygen and must have a -50° C dew point. Several calorimetry systems were analyzed for use in the curium fuel capsule processing system.

3. Technical Discussion and Evaluation

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The reaction vessel for the fuel capsule dissolution in the hot cells at Quehanna was estimated to be a 20-inch glass vessel, 4 inches in diameter, enclosed in a 5-inch diameter water jacket. Bare nichrome wire is coiled around the inner tube inside the water jacket to heat the dissolver solution to 90° C. The wire is approximately 50% efficient, and a 1-kw service will produce the desired reaction temperature in 15 minutes. An automatic on-off switch system can be used to maintain the reaction temperature after addition of the reactants. Data on cooling requirements during the reaction were not available.

b. Curium purification and fuel form processing

Calculations are in progress to determine the pressure drops to be anticipated when a variety of eluant solutions are passed through the resin columns in the curium purification system for the hot cells.

Cursory studies of various methods of maintaining the chemically inert, moisture-free atmosphere required for curium, uranium, or plutonium processing were made. Three methods were suggested:

- (1) Complete recirculation of box atmosphere with occasional purges.
- (2) A "bleed and feed" system where a certain percentage of the box atmosphere is replaced per unit time.
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The desired circulation rate was six complete changes per hour or 840 cfh. Tolerance levels given were 50 ppm for oxygen and a dew point of -50° F for water. For reliability and a minimum of maintenance, an electrical moisture removal system had previously been recommended.

For oxygen removal, a quick survey indicated that a catalytic reduction with hydrogen was the most efficient system. Purities of less than 1 ppm oxygen were guaranteed by the vendor, assuming no appreciable leakage of oxygen into the box. With a tolerance of 50 ppm and the recirculation rate prescribed, a leak of 0.21 cfh of air can be tolerated.

The oxygen removal system will cost from \$3500 to \$7000, depending upon the refinements desired. The choice of the three circulation systems suggested will depend on the amount of time in which the dry box is in circulation. Argon of less than 15 ppm impurities costs 7 cents/ft³. If the box is in operation only 100 hr/yr, the cost of operation of a "once-through" system would be less than \$6000/yr and all gas purification and drying could be eliminated. This type of system is highly insensitive to leaks in the box wall or glove punctures.

The gas recirculation system is recommended if the box is to be used more than 100 hr/yr, while the "bleed and feed" or "once-through" systems are suggested for more sporadic utilization.

Three general types of calorimeters were examined. In order of decreasing accuracy and probable cost, they were:

- (1) A flow-type calorimeter for extremely precise measurements with accuracy of the order of $\pm 0.1\%$.

- (2) A vacuum flask calorimeter with a maximum error of about 5%.
- (3) A very crude measuring device in which the significant causes of error were not available.

For curium processing where accurate calorimetric measurements of the heat source at the time of loading are required, it was recommended for use with the alpha isotopes in Cell 5 that the most accurate calorimeter be fabricated. The cost of this calorimeter, based on an estimate supplied by the UCRL personnel who designed and built lower capacity models, is about \$5000.

4. Conclusions

A 1-kw electrical service will be adequate to produce a 90° C reaction temperature in the dissolution bath within one hour.

For extensive use of an inert atmosphere box, a recirculation system is most economical. The system requires a catalyzer to remove oxygen to the desired level of 50 ppm followed by a drier to remove the water formed in the catalyzer.

The choice of a calorimeter is governed by the accuracy and heat capacity required in the unit. An accuracy of $\pm 0.1\%$ can be obtained with the UCRL Twin Unit.

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