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SNAP PROGRAMS

Quarterly Progress Report No. 7

Task 6

Fuel Technology Development Program

April 1 through June 30, 1961

MND-P-3015-III

Approved by

William M. Bowes

Assistant Project Engineer

Advanced Development Section

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FOREWORD

This is the seventh unclassified SNAP quarterly progress report prepared by The Martin Company. It covers the work performed during the period April 1, 1961 through June 30, 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

During this report period, the Task 6 Program made several noteworthy accomplishments towards the ultimate development of isotope power sources for thermoelectric and thermionic conversion to electrical energy.

A comprehensive literature search on suitable isotopes to supply power for land, sea and space applications resulted in the completion of a compendium entitled "Isotopic Power Sources." The manuscript for this compendium has been delivered for publication.

The Martin Company's Heavy Elements Processing Facility for processing americium and fabricating americium capsules for irradiation was officially opened, and hot operation has commenced. Five fully loaded americium capsules were packaged and shipped to the National Reactor Testing Station, Idaho Falls, Idaho, to await insertion in the Materials Testing Reactor. These capsules will be irradiated in a high thermal neutron flux to convert the Americium-241 to the power isotope Curium-242.

Most of the modifications to the americium purification system were completed, and checkout operations were begun during this report period. Design of the waste disposal system for americium purification was also completed.

Process specifications have been established for dissolution of the irradiated capsules, and the major equipment has been designed for this operation at Martin's Hot Cell Facility at Quehanna, Pennsylvania. Experimental work on the purification of curium was continued with tracer level quantities of radioactive materials. Equipment design for this process was initiated. Blending and crushing studies were conducted to establish techniques and procedures for preparing curium carbide and for producing homogeneous cermet fuel forms. Investigations were also made into the compatibility of various matrix and container materials with curium carbide and curium oxide at temperature levels characteristic of both thermoelectric and thermionic devices.

Flow diagrams for the fuel form and isotope encapsulation processes at Quehanna were completed. Specification control drawings were also prepared on the containment boxes for these processes. Design and procurement of equipment for the fuel form effort proceeded on schedule with the bulk of the design work nearing completion.

A safety analysis was conducted which verified integrity of the americium capsules during all stages of the irradiation program. This analysis will be submitted to the Safeguards Committee at the National Reactor Testing Station, Idaho Falls, Idaho, for their approval of the Task 6 irradiation program.

I. INTRODUCTION

This is the progress report for the last quarter of Fiscal Year 1961, as required under Contract AT (30-3)-217. It includes an appraisal of the work accomplished under Task 6--Fuel Technology Development--of the SNAP programs. The work covered in this report is divided into separate subtasks:

- 6.1--General Development and Materials Requirements.
- 6.2--Americium-Curium Radioisotope Preparation Processes.
- 6.3--Radioisotope Fuel Form Development.
- 6.4--Safety, Shielding and Thermal Analyses.
- 6.6--Irradiation of Americium.

The report was edited by W. M. Bowes and J. B. Zorn, with principal contributions from G. Samos, A. Abriss, J. Kane, P. Hess, S. Zwickler, E. Smith and J. Zorn.

The first batch of Americium-241 capsules have been processed in the Heavy Elements Processing Facility this quarter, after only three months of hot operation, to provide the target material for the curium work to be performed at the Quehanna Hot Cell Facility.

II. SUBTASK 6.1--GENERAL DEVELOPMENT AND MATERIAL REQUIREMENTS

The AEC has requested a literature search of radioisotopes useful in providing thermal power for conversion to electrical power and subsequent compilation of available information in an isotopes compendium. For those isotopes that are of practical use, and where availability is sufficient to warrant further investigation, the methods of processing were also reviewed; included in the search were such elements as curium, plutonium, americium, actinium, thorium, radium and uranium.

A. OBJECTIVES

During this quarter, the goals of Subtask 6.1 were as follows:

- (1) Completion of final annotation of the abstracts.
- (2) Assembling of the classified section and tabulation in suitable form for publication.
- (3) Completion of annotation on the selected abstracts with the text of selected reference.
- (4) Preparation of the final manuscript of the compendium for publication.
- (5) Direction and coordination of the subtasks under Task 6 and liaison between the project and the AEC.

B. SCOPE OF WORK--EVALUATION OF THE LITERATURE

The scope of the literature search is based on a statement of work prepared October 6, 1960. The 18 nuclides selected as most suitable for review have been studied, and the physical properties and methods of processing evaluated, to facilitate final selection of a radioisotope heat source for any specific mission. The final compendium has been prepared in accordance with the outline shown:

Title--Isotopic Power Sources--a Compendium

1. General introduction

- a. A statement of the purpose of the compendium
- b. Annotation on the application of isotopes as power sources, based on selected references

- c. A list of topics included
- d. The organizational plan of the compendium

2. Properties of the selected isotopes (one volume)

Isotopes arranged in order of increasing atomic number (Z); twelve sections (one per element); each containing:

- a. General discussion, including history, applications, etc.
- b. Nuclear properties
- c. Physical properties
- d. Metallurgy
- e. Chemistry (preparation, properties, analysis, etc.)
- f. Miscellaneous properties of technical interest (i.e., handling, storage, hazards)

3. Processes for separation of the isotope (one volume)

- a. General discussion
- b. Ion exchange purification processes
 - (1) General
 - (a) By groups
 - (b) By elements
- c. Solvent extraction (same arrangement as Item b)
- d. Precipitation processes (same arrangement as Item b)
- e. Other processes (same arrangement as Item b)
- f. Industrial aspects--plant information, handling and safety
- g. Concluding remarks on fabrication techniques, etc.

4. Abstracts (in two volumes unclassified and classified) arranged in the order of the bibliography and numbered serially for reference purposes, with an annotated list of general references, including the abstract.

C. TECHNICAL DISCUSSION AND EVALUATION

The compendium, when published, will be valuable to the chemist, engineer and designer concerned with producing a heat source to be incorporated in a direct conversion energy unit (such as the thermoelectric and thermionic auxiliary power units of the SNAP type). The annotation will be extremely useful in the evaluation of the properties and processes as applied to the specific isotope or system. The compendium comprises the major contributions in the field of transuranic and transplutonic chemistry, even where only milligram quantities of the specific isotope have been prepared to date. Curium-242 is still favored as the most suitable short-lived isotope for thermal power output, while Plutonium-238 is suitable for the longer missions.

D. CONCLUSIONS

In its final form, the compendium is composed of four volumes: Volume I, containing the unclassified bibliography of over 2000 titles; Volume II, consisting of 378 abstracts from classified sources; Volumes III and IV, comprising short review texts with tabulated properties data and 466 abstracts, some expanded with notes and comments, and selected illustrations.

The compendium was completed and the manuscript was delivered for publication on June 23, 1961.

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

The Heavy Elements Processing Facility (HEPF), which was officially opened April 3, 1961, by the AEC at The Martin Company's Middle River Plant, has been used to process Americium-241 dioxide into capsules to be irradiated to Americium-242. By beta decay, Americium-242 is transmuted to Curium-242. Whereas dummy capsules had been tested in this facility successfully, the radioactive material required considerable additional protection to insure its safe handling. Experiments to be performed during the next quarter will be conducted first at the tracer level, and then at full scale.

The curium purification procedure is still under development; an apparatus is being prepared to handle greater than tracer quantities of curium in the HEPF. In the interim, the use of americium as a stand-in for curium has permitted the checkout of the procedures for both dissolution of aluminum capsules in sodium hydroxide-sodium nitrate solutions, and ion exchange separation of gross fission products from americium-curium. In the latter procedure, various Dowex resins were used, and separation was tested with several recommended eluants such as lithium chloride, alpha-hydroxyisobutyric acid and hydrochloric acid.

The specifications for the containment boxes in the hot cells at the Quehanna site, for handling kilocurie quantities of the alpha isotope of curium, have been prepared and submitted to the design section; but the definition of the process steps and the analytical procedures are still to be prepared. The biggest problem to be resolved in the design of all containment boxes is the selection of the materials for the box windows, the seals, and the coatings to be applied to the inner walls of the boxes. Another design area under study, paramount to the success of the process manipulations in the box, is the booting of the Argonne Model 8 manipulators. Several consultants and manufacturers have been contacted to assist in selection of the best system. Of equal importance is the mechanism for transferring chemical samples, equipment and waste in and/or out of the containment box, cell and isolation areas. The work of the current quarter is to be devoted primarily to resolving these problems, designing and specifying the equipment, and establishing the procedures for operation within the cell.

A. OBJECTIVES

1. Americium Purification and Encapsulation (6.2.1)

For the final quarter of Fiscal Year 1961, the objectives of this sub-subtask were:

- (1) To continue assembling the americium process equipment in preparation for purifying gram quantities of Americium-241.
- (2) To check the equipment used for converting americium oxalate to the oxide in the HEPF.
- (3) To produce two capsules containing trace amounts of americium for checking the process, and to produce five fully loaded capsules for irradiation in the reactor.
- (4) To provide an installation drawing and procure the equipment for a liquid waste disposal system.

2. Curium purification (6.2.2)

The program objectives for this phase were:

- (1) To test the effectiveness of Dowex resins of 100 to 200 mesh by performing the curium purification with tracer quantities of Americium-241 and fission products.
- (2) To determine the effectiveness of deacidifying a lithium chloride-hydrochloric acid solution by boiling off the excess acid in connection with the ion exchange purification.
- (3) To develop analytical procedures for the curium process with respect to sampling points, volume of sample, dilutions necessary for counting the samples and activities to be analyzed.
- (4) To compare the final Martin Company curium purification process step-by-step with other reported processes.
- (5) To continue with the design and procurement of equipment required in the processing of the initial irradiated Americium-241 capsule, including the process sampling system, in the hot cells at Quehanna.
- (6) To confirm the design of the chill block for the Curium-242 shipping cask and initiate the design of equipment required to unload and transfer capsules from the storage pool to the hot cells at Quehanna.
- (7) To prepare specification control drawings for the containment boxes required to process Curium-242 at the Quehanna site.

B. SCOPE OF WORK

1. Americium purification (6.2.1)

Work has proceeded on the americium purification containment boxes, and the following steps have been accomplished:

- (1) Installation of the solenoid valves was completed, with all connecting polyethylene lines adjusted to fit the smaller valves, and the system was leak checked.
- (2) Stainless steel double barrier lines were installed on all lines between boxes.
- (3) New doors were placed inside the boxes at openings between the boxes.
- (4) A sampling line was installed from the assay valve to the dilution box.
- (5) Millipore filters were placed in the vacuum lines within the boxes.
- (6) Two sets of ion exchange columns, one inch and two inches in ID, respectively, were designed and fabricated. The one-inch ID columns were installed; the two-inch ID columns will be installed later to handle larger quantities as the need arises.
- (7) A glass condenser for use during the drying of the oxalate precipitate was designed, fabricated and installed.
- (8) Tests were conducted to determine the ease of removing spent ion exchange resin from the columns. The resins can be readily removed through the 1/4-inch lines if they are slurried and drawn out by vacuum.
- (9) The liquid volume of the assay valve was calibrated.

In order to investigate the effectiveness of the equipment designed to convert americium oxalate to the oxide, experiments were conducted with rare earths substituted for americium to prove feasibility.

2. Americium Encapsulation

Two test capsules containing americium dioxide were made. No modifications of the process were found to be necessary in fabricating these aluminum capsules with tracer amounts of isotope present. Following the successful

processing of the tracer capsules, five fully loaded capsules were also prepared, helium leak checked and decontaminated to remove most of the loose and fixed contamination. These fully loaded capsules were finally enclosed in aluminum sleeves to increase the outer diameter to 1-1/8 inches to accommodate them to the X-basket in the Materials Testing Reactor. X-rays were made to show the americium oxide distribution. Methods of removing the fixed contamination, adapting the shell to the capsule and x-raying the loaded capsules were developed; and tests were performed to ensure the integrity of the capsule design.

Engineering drawings for the liquid waste disposal system to handle effluents from the americium purification boxes were completed and released to manufacturing.

3. Alkaline Dissolution of the Irradiated Capsules (6.2.2)

Work continued, during the quarter, on the study of the alkaline dissolution of aluminum-curium capsules in the presence of sodium nitrate. Aluminum-gadolinium sesquioxide capsules (used to simulate curium) were placed in 1500 ml of a solution 6.7M in sodium hydroxide and 4.1M in the nitrate. The solution was first heated to 90°C to start the reaction and then maintained at this temperature by cooling to control the highly exothermic reaction. After one hour of cooling, the reaction rate subsided sufficiently to permit reheating for completing the dissolution. After an additional hour of heating, one liter of water containing one gm of hydrated ferric chloride was added and the solution boiled for another hour, at which time the dissolution was complete. This solution was allowed to sit overnight, then filtered and washed. Four aluminum-gadolinium sesquioxide capsules were dissolved in this manner. In a fifth experiment a mixture of one half of a solution from a dissolved capsule and an equivalent half quantity of solution spiked with Americium-241 was processed to determine the losses of americium in the filtering and washing.

4. Curium Purification

For the ion exchange separation of curium from the fission products produced during irradiation of the capsules, the use of larger-mesh resins (100 to 200) has been recommended (Ref. 1) as more practical for large-scale column operation than the 200 to 400-mesh resins used in the laboratory experiments at Martin. An experiment with 100 to 200-mesh resin was performed, simulating the three-column operation of the curium process, to determine what effects, if any, this size resin would have on the process.

A recommended method for preparing for the lithium chloride solution was used to load the curium on the Dowex 1 resin without eliminating the excess hydrochloric acid in the curium solution by concentrating nearly to dryness. The loading was accomplished by adding the appropriate volume of 10.5M lithium chloride to the acid solution containing the curium and boiling until the volume of the solution was again equal to the 100-ml volume of the chloride

solution. Therefore, to a 100-ml solution of 6N hydrochloric acid, 100 ml of 10.5M lithium chloride was added, and the combined solutions were boiled until the volume was reduced to 100 ml. Titration of the evaporated solution showed it to be 0.1M in hydrochloric acid--satisfactory for operation of the column.

An analytical procedure for the curium process has been written, defining:

- (1) The volume of sample to be taken from the containment box into the dilution box.
- (2) The dilutions necessary to prepare each sample for counting.
- (3) The particular activities to be analyzed in each sample, taking into consideration the locations from which the samples were taken.

The objectives of the procedure are to determine the curium recovery and decontamination and to determine plutonium recovery.

The curium purification process, as has been proposed, is the result of an experimental program using some of the techniques and methods of other workers in the field. A survey of the literature of the various process phases has been made to include the latest information.

Design for curium purification system. Nonstandard authorization forms were prepared and issued for obtaining cost estimates on all the chemical purification equipment. These tentative specifications were reviewed during the period with outside vendors and suitable procurement sources were selected for the majority of the items.

Shipping cask for irradiated capsules. Negotiations were completed with Knapp Mills, Inc., on the fabrication of the chill block in the curium shipping cask. Design studies were initiated with respect to the equipment and techniques to be employed to transfer capsules from the storage pool to the hot cell at the Quehanna site.

Vent and vacuum system. A process vent and vacuum system schematic was prepared. The vent and vacuum system will be used to effect the normal transfer of process fluids and for sampling. Since process solutions in Cell 3 contain hydrochloric acid, both the vent and vacuum systems have been provided with alkaline scrubbers, condensers and moisture separators to protect the absolute filters. Nonstandard authorization forms have been prepared for the equipment and negotiations are being conducted with outside vendors.

C. TECHNICAL DISCUSSION AND EVALUATION

1. Americium purification system (6.2.1)

With the installation of the new equipment in the americium purification containment boxes, most modifications were complete. The ion exchange columns previously supplied were defective and inadequate, and, as a result, new columns were designed, fabricated and installed in the boxes. Other defects, such as in the seals for the heating wires leading into the water jacket and in the transfer of resin in and out of the columns through the 1/8-inch lines, have been remedied on the new columns. The new valve system was leak tested and found satisfactory; however, a large number of the polyethylene Swagelok fittings developed cracks while being tightened, and, as a result, all the polyethylene nuts will be replaced by brass nuts.* Aside from changing of fittings, the only remaining work to be done before activating the system is installing the waste disposal system and caustic scrubber, and conducting the cold run.

In experiments to check out the equipment for oxide conversion yields and process conditions, the following procedure was carried out. Fifty grams of rare earth oxides were dissolved in hydrochloric acid and water. The solution was neutralized with concentrated ammonium hydroxide to approximately 0.1N free acid, and a saturated oxalic acid solution was slowly added. Upon completion of the precipitation, a 100% excess of oxalic acid was added and the slurry was agitated for one hour to promote crystal growth and to complete the reaction. The oxalate precipitate was then filtered through the inverted sintered glass filter stick and washed with ten volumes of water. The oxalate cake was partially dried by drawing air through it and was further dried in the same vessel by heating at 150°C for one hour. The temperature was then increased to 800°C and held for one hour. The resultant oxide, after cooling, was a dense brown powder much the same in appearance as the original powder. The americium dioxide was then transferred to a weighing bottle using the specially designed weighing bottle cover. In an experiment with neodymium, the recovery was 98.8%. A subsequent experiment using mixed rare earth oxides gave 92% recovery.

A vapor condenser was found necessary to handle the steam produced during the drying and decomposition steps and a detachable condenser was designed, fabricated and installed for this purpose.

2. Encapsulation of americium--system modification

The powder compacting operation was modified by decreasing the amount of each powder per charge and increasing the number of compactions. This was

*Nicholson and Coogler of the Savannah River Laboratory have also adopted this change.

accomplished by decreasing the volume of the dispenser charging chamber. This modification improved the accuracy of the weight of the final compacted powder, thus ensuring the proper positioning of the lid for welding. A dial indicator was mounted on the bottom ram of the press to measure the amount of capsule shortening during compacting; a powder fill level indicator was mounted on the top ram to determine when the capsule was filled to the correct height. To ensure their release from the die, the capsules were coated with Molycoat, a die lubricant.

In the ultrasonic cleaning tanks a wetting agent was substituted for green soap and was found to be more satisfactory.

3. Capsules Containing Trace Amounts of Americium Oxide

A trace amount of americium oxide (10 millicuries) was added to 10 grams of gadolinium oxide. The oxides were prepared by coprecipitation of the oxalate and firing to the oxide. Ten grams of gadolinium oxide has a volume equivalent to 16 grams of americium dioxide, enough for a four-capsule charge. All operations--mixing, compacting, ultrasonic cleaning, reaming and welding--proceeded without difficulty. A hot water leak test, rather than a helium leak test, was performed on the tracer capsules. The ultrasonic cleaner completely removed the loose contamination from both capsules. The first capsule was devoid of fixed contamination, but the second had a fixed count of 2000 alpha disintegrations per minute (dpm). Etching the capsule surface with approximately 6N hydrochloric acid solution, followed by a water rinse, was found to be the best method for removing the fixed contamination. The fixed contamination on the second capsule was due to radioactivity picked up from the face of the compacting die which, purposely, had not been cleaned between capsule pressings.

To reduce fixed contamination, the die faces were decontaminated, prior to insertion of each new capsule, with Kleenex tissues moistened with perchloroethylene. Smears were taken to determine the amount of activity on the die, and the die was cleaned to a predetermined low level of activity.

4. Capsules Containing Americium Dioxide for Curium Production

A container with 72 curies (22.7 gm) of relatively pure americium dioxide was introduced into the weighing glove box. Considerable difficulty was encountered in opening the weighing bottle containing the oxide because of a frozen glass stopper. To avoid the possibility of spillage or excessive contamination during weighing, the entire contents of the bottle (sufficient to make 5 capsules) was directly charged into the mixing container with an appropriate amount of dried aluminum powder. Previously, a test had been made to determine whether this size charge could be effectively mixed. It was noted that some americium dioxide stuck to the lid and sides of the weighing bottle above the bulk powder and could not be removed by scraping. No further attempt was made to remove it at the time. No problems were encountered in any of

the operations within the glove box system (i.e., mixing, compacting, ultrasonic cleaning, reaming, helium welding, loose contamination smearing and leak checking).

The length of time required for ultrasonic cleaning was increased from 30 seconds to as much as 15 minutes to remove loose contamination. The cleaning bath was changed once during the operation. No capsules introduced into the helium leak detection chamber carried loose contamination of more than 600 alpha dpm. To meet this requirement, several capsules had to be re-cleaned.

A smear of the floor of the helium leak detection glove box revealed that the activity was less than 1000 alpha dpm, indicating that the air flow toward the contaminated boxes and the precautionary procedures were adequate for keeping activity low.

Activity measurements of the completed capsules revealed a level of less than 600 alpha dpm for the loose contamination and a fixed contamination of about 20,000 alpha dpm. Several procedures were tried to remove fixed contamination without etching the capsules, but none was successful. Therefore, etching tests were conducted on one capsule to establish the correct etching time. The final decontamination procedure consisted of a 70-second immersion in an approximately 6N hydrochloric acid solution followed by a three-minute water rinse and drying. This brought the fixed contamination count down to less than 5000 alpha dpm.

The capsules were then placed in a lathe chuck and sanded with Nos. 180 and 240 emery cloth, followed by abrasion with medium and fine aluminum wool. This phase of the cleaning operation was accomplished in six minutes. The capsules were then etched for 10 seconds in the 6N hydrochloric acid bath, thoroughly rinsed in water and dried. The final fixed count on the capsules was less than 500 alpha dpm.

The capsules were originally designed with an outside diameter of one inch to fit the water flow channel in the Engineering Test Reactor. Since space in this reactor was not available, the first group of capsules was rescheduled to be irradiated in the MTR. To provide sufficient capsule cooling in this reactor, a 1-1/8-inch diameter is required. This requirement was not established until after the boxes had become radioactively hot; therefore, altering the capsule size by changing the die was impractical in the time available. Therefore, an aluminum shell (1/16-inch wall) was shrunk over each capsule to increase the diameter from 1 inch to 1-1/8 inch. The shrinking was accomplished by making eight weld passes along the length of the shell. Tests of this method of shrinking had shown that the welds penetrated from one third to one half of the shell thickness and decreased the inside diameter 0.006 inch. Several other methods of adapting the shell to the tube were tried, but this method was found to be best.

To prepare the capsules for the shell, the weld overhang on the outside diameter was machined off on a lathe. A Type 6061-T6 aluminum tube was honed to a bore 0.004 inch greater than the capsule diameter. The shell was made 6 inches long to allow a 1/2-inch extension over each end of the capsule. Machined aluminum end plugs, one inch long, were fitted into the end of the shell and butted against the ends of the capsule. A jig was built to hold the capsule and shell during welding. The first four welds were made from one end of the capsule to the other; the direction was reversed in the last four welds. The second weld bead was laid 180 degrees from the first; the third, 90 degrees from the second; and the fourth, 180 degrees from the third. The last four weld beads were placed, alternately with the first four, in a similar manner. The capsule was then held in a lathe chuck where the outside diameter was machined to 1.125 ± 0.002 inches. After machining, the capsules were lightly sanded with 180-grit emery paper to eliminate the machining marks. The excess shell material and end plugs were then cut off flush with the ends of the capsule. Figure 1 is a micro-photograph of a capsule and shell; and Fig. 2, a photo-micrograph of the interface between the capsule and outer shell.

Fig. 1. Sleeve of Simulated Americium Capsule

Fig. 2. Interface of Sleeve of Simulated Americium Capsule

The ends of the shell were not welded to the capsule because there would then be no way of determining whether a pinhole-free weld was produced. It was then postulated that, if there were a pinhole in the weld, water could enter the hole on cooldown, and that on startup the rapid temperature rise might cause rupture of the shell due to rapid expansion of the water.

The capsules without shells produced a gamma activity of 80 r/hr; and with shells, 60 r/hr at the surface. Smears indicated no loose alpha contamination on the surface.

Table 1 summarizes all pertinent data concerning the capsules. Figure 3 shows the distribution of americium oxide in the capsule as determined by X-ray. To produce an X-ray negative of these capsules, which emitted 60 r/hr of soft gammas, an 0.042-inch piece of lead was laid between the X-ray film and the capsules. An exposure time of 7-1/2 minutes at 200 kilovolts and a distance of two feet produced a satisfactory X-ray negative.

Integrity testing. Besides the tests mentioned above, a number of tests were made to determine the depth of penetration and integrity of the welds at each end of the capsule. A hydraulic pressure test of a capsule revealed that 2700 psi was required before leaking occurred in the weld at the top sealed end of the capsule. No appreciable deformation was noted at pressures below 500 psi. At 1000 psi there was a 0.00075-inch expansion of the OD; and at 2000 psi, a 0.007-inch deformation. Permanent distortion after release of the 2700-psi pressure amounted to 0.0093 inch. Since the internal pressure should not exceed 250 psi and shells have been placed on the capsules, there should be no problems from internal pressure buildup within the prescribed periods of irradiation, cooldown and shipment.

5. Equipment Design

The design of the waste disposal system to handle effluents from the americium purification boxes is complete. Liquids will be neutralized in the glove box, transferred to a head tank and gravity fed into a drum where they will be absorbed in vermiculite. This system was used in lieu of the previously proposed concrete disposal to eliminate the problems of mixing and dusting, and it adheres to the double containment philosophy. The all-plastic primary system is contained in an all-metal secondary system. Provisions have been made to permit the operator to bag-off all connections that must be broken during maintenance or normal replacement of components.

6. Alkaline Dissolution of Curium-Aluminum Capsules (6.2.2)

In the caustic dissolution of aluminum capsules, the dropwise addition of sodium hydroxide to a sodium nitrate solution containing the capsule was found to yield a precipitate with a high aluminum content. However, when a capsule was placed in a nitrate solution containing a large excess of sodium hydroxide and the reaction rate controlled by cooling, a precipitate very low in aluminum resulted. Furthermore, the addition of a small amount (1 gm/liter) of ferric chloride to the solution after dissolution of the capsule aided considerably in yielding a stable filtrate (i.e., no further precipitation occurred in the filtrate after a period of several days). Four aluminum-gadolinium sesquioxide capsules were dissolved, and no difficulties were encountered in either the

TABLE 1
Summary Sheet--Americium Capsules

<u>Capsule No.</u>	<u>MND-1-1</u>	<u>MND-1-2</u>	<u>MND-1-3</u>	<u>MND-1-4</u>	<u>MND-1-5</u>
<u>Identification No.</u>	79	83	80	84	82
Material					
Capsule	5052 A1	5052 A1	5052 A1	5052 A1	5052 A1
Shell	6061T6 A1	6061T6 A1	6061T6 A1	6061T6 A1	606TT6A1
Matrix	Ultra- pure A1	Ultra- pure A1	Ultra- pure A1	Ultra- pure A1	Ultra- pure A1
Weight of AmO ₂ (gm)	3.845	4.124	4.115	3.998	3.931
Capsule weight (gm)	191.598	199.760	197.880	195.598	194.130
Powder height (in.)	4.139	4.201	4.233	4.139	4.139
Capsule OD less shell (in.)	1.002	1.000	1.001	1.000	1.005
Capsule length (in.)	4.957	4.958	4.956	4.958	4.958
Final OD, shell (in.) *	1.124	1.124	1.124	1.124	1.124
Density					
(gm/cc)	2.24	2.39	2.34	2.33	2.29
(% of theoretical) (100% = 2.788 gm/cc)	80.34	85.72	83.93	83.57	82.14

* Tolerance $\pm$ 0.001 in.

Fig. 3. Distribution of Americium Dioxide in Aluminum Capsule

dissolution or the filtration steps. A typical analysis of a capsule dissolved in this manner is the following:

<u>Precipitate Constituent</u>	<u>Weight (gm)</u>
R E ₂ O ₃ (rare earth oxides)	3.50
Silicon	0.05
Aluminum	0.00
Iron	0.33
Chromium	0.01
Magnesium	0.64

The residue, after filtration and drying at 110°C, weighed approximately nine grams.

To determine the losses of americium, one half of a capsule was added to 750 ml of a solution 6.7M in sodium hydroxide and 4.1M in sodium nitrate, spiked with Americium-241. The loss of americium to the filtrate and washings was found to be $2.6 \times 10^{-2}\%$ and $4.6 \times 10^{-3}\%$, respectively.

7. Curium Purification System (6.2.2)

For large-scale operation, the use of larger particle size resins has been recommended to provide for flow rates through the column which would allow practical operation. In the laboratory, resins of 200 to 400 mesh had been used, but it has been calculated that about 15 psi would be needed to maintain a flow rate of 25 ml/cm^2 through the production column 5 cm in diameter and 60 cm long. With resin 100 to 200 mesh, flow rates can be maintained with much less pressure (3 to 5 psi). An experiment was therefore run using the 100 to 200-mesh resin, which simulated the curium purification process. The results appear in Figs. 4, 5 and 6. Figure 4 shows the elution of fission product and Americium-241 through an anion column, with solutions of 10.5M lithium chloride-0.1M hydrochloric acid and 8.5M hydrochloric acid as eluants, in which excellent recovery and decontamination are achieved. Figures 5 and 6 show the undesirable tail effects which require further investigation.

The effectiveness of deacidifying 10.5M lithium chloride in a hydrochloric acid solution has been amply demonstrated in the laboratory. Use of this method of obtaining the proper acidity in the feed solution to the lithium chloride column will eliminate the problem of evaporating the curium solution to a very small volume where heat and hydrolysis are of concern.

Fig. 4. Elution of Americium and Fission Products from Dowex 1 Resin with 10.5M LiCl-0.1M HCl and 8.5M HCl

Fig. 5 Elution of Americium with 0.5M HCl and 13M HCl from Dowex 50 Resin

Fig. 6. Elution of Americium with 0.4M Alpha-hydroxyisobutyric Acid, pH 4.48, from Dowex 50 Resin

An analytical procedure details all the necessary information for determining the activity levels in each volume of eluate. The analysis calls for both control and quantitative determinations. For control, a gross count for alpha and gamma radiation determines what is happening in each column. Quantitative measurements are determined to analyze more precisely the yields and the decontamination factor achieved. This quantitative analysis requires the use of a multichannel analyzer for both alpha and gamma energies.

A literature survey of processes for the separation of actinides from lanthanides was made to correlate the Martin process with the work done by other workers in the field. The Martin process, as it has been proposed, consists of three separate ion exchange columns, each to purify by a different mechanism:

- (1) An anion exchange column based upon the chromatographic elution of lanthanides and actinides from a chloride complex in the presence of a high concentration of Li^+ . Figures 7 and 8 summarize the elution positions of the elements in such a system and indicate that good separation may be achieved.
- (2) A cation exchange column based upon the exchange mechanism of the individual elements and their elution as a function of acidity. Figures 9, 10 and 11 summarize elution positions as a function of hydrochloric acid concentration and again indicate good separation.
- (3) A cation exchange column based upon the chromatographic elution of an organic complex. Table 2 shows the separation factors of the lanthanides and actinides with alpha-hydroxyisobutyric acid. Although this process has been used primarily for actinide separations, the peak positions between most of the lanthanides and actinides is sufficient to give some separation. Based on data in the literature, the three exchange systems have a sufficient spread in their separation operations to effect a good overall decontamination.

TABLE 2*

Separation Factors with Ammonium α -hydroxyisobutyrate
(Basis: curium factor = 1.0)

<u>Element</u>	<u>Relative Peak Position</u>
E	0.13
Cf	0.19
Bk	0.37

* Ref. 2.

<u>Element</u>	<u>Relative Peak Position</u>
Cm	1.00
Am	1.39
Lu	0.011
Yb	0.016
Tm	0.022
Er	- -
Ho	0.039
Y	0.069
Dy	0.076
Tb	0.14
Gd	0.22
Eu	0.34
Sm	0.7
Pm	1.1
Nd	2.3
Ce	3.4

D. CONCLUSIONS

1. Americium Purification System (6.2.1)

Replacement of the polyethylene Swagelok fittings and installation of the waste system and caustic scrubber will conclude the preparation of the americium purification boxes.

The equipment for converting oxalate to oxide is now in satisfactory operating condition.

Fig. 7. Elution of Lanthanides and Tripositive Actinides from Dowex 1

Fig. 8. Summary of the Elution Order of the Actinides and Lanthanides from Dowex 1 Resin as a Function of Atomic Number

Fig. 9. Elution of Americium, Curium, Lutecium, Promethium and Cerium Tracers with 6M Hydrochloric Acid from Dowex 50 Resin

Fig. 10. Elution of Americium, Curium, Promethium and Cerium with 9M Hydrochloric Acid from Dowex 50 Resin

Fig. 11. Elution of Americium, Curium, Lutecium and Promethium with 13.3M Hydrochloric Acid from Dowex 50 Resin

2. Americium Encapsulation (6.2.1)

Americium capsules may be made by the established process. Only minor modifications will be made in any future work so as to minimize fixed contamination on the capsules. The diameter of the capsule can be increased by shrinking an aluminum shell over the capsule; however, if future capsules of a larger diameter are to be made, a larger die should be provided to increase production and to reduce the exposure time during processing. If possible, the weight of americium dioxide per container should be specified so that the weighing operations can be eliminated from the process.

3. Curium Dissolution System (6.2.2)

Experimental work on the alkaline dissolution of aluminum capsules has been satisfactorily concluded.

4. Curium Purification System (6.2.2)

Additional work will have to be done on the cation exchange columns to find the cause of the tail effects noted in the last experiments with the 100- to 200-mesh resin.

5. Equipment Design (6.2.2)

Chemical process equipment for separating the curium in the irradiated americium capsule from the fission products and converting the curium oxalate to curium oxide will be procured as soon as possible. It is to be installed in Cell No. 3 at Quehanna. With the exception of the following equipment, the design details have been discussed in previous reports:

- (1) Dissolver--A dissolver was designed to dissolve the aluminum capsule in a sodium hydroxide-sodium nitrate solution. The vessel will be made of Vycor for chemical and thermal resistance. Electrical heaters and a cooling jacket will be provided to control the solution temperature during operation.
- (2) Resin column--A resin column was designed. The resin is contained between two filter frits, one free-floating to allow for the expansion or contraction of the resin. The pressure gradient across the frit maintains sufficient preload on the resin bed during operation. The column is provided with heating coils and a cooling jacket to control column operating temperatures and is designed as an expendable unit--the column, less the end caps, with the resin contained between the two frits, is to be periodically replaced.
- (3) Storage vessels--Storage vessels were designed to contain process solutions. Each storage vessel is provided with an air sparger to agitate solutions for sampling purposes. Where necessary, the

vessels have been provided with cooling jackets to dissipate decay heat.

- (4) Filters--Two filters were designed to separate precipitates. Since both filters may be subjected to solutions of hydrochloric acid, platinum filter frits are used.
- (5) Curium shipping cask--Negotiations were completed with Knapp Mills, Inc., on the Curium-242 shipping cask chill block. It was mutually agreed to use a segmented block that could be disassembled in the event capsule seizure occurred. In conjunction with this operation a study was made of the procedure to be employed at Quehanna to transfer the capsule into the cell:

The capsules will be removed from the shipping cask and stored under water at Quehanna. Upon demand they will be transferred to the cell in the transfer can installed within a lead-shielded coffin. Within the transfer can the capsule will be surrounded by a slug of aluminum and water. It has been calculated that the mass of the system will permit a lag time of one hour before the capsule attains a temperature of 300° F. Within the pool the mechanism for removing and replacing the can cover on the containment box will be used. The coffin and mechanism mounted on a bed will be hoisted from the pool, transferred to the appropriate isolation room and adjusted to the transfer port elevation for loading into the containment box.

IV SUBTASK 6.3 -- RADIOISOTOPE FUEL FORM DEVELOPMENT

Blending studies were conducted on gadolinium sesquioxide and carbon and various ceramic-metal powder mixtures to establish the basic parameters involved in the preparation of the curium fuel form. In the initial steps of preparing curium carbide, curium oxide must first be thoroughly mixed with carbon to obtain maximum yield; also the ceramic and metal matrix system planned for the curium fuel form requires maximum homogeneity to achieve an even powder distribution.

The conversion of curium oxide to curium carbide results in rigid, sintered pellets which must be crushed to the powder form for dispersion in the proper fuel matrix. The optimum crushing procedure has been established and the resulting particle size distribution determined.

A preliminary flow diagram for the fuel form process was completed. On such items as the press and dies, blender, loading rack for the blender and dies, inert gas system, welder for final encapsulation, and fuel capsule storage vessel, design was initiated. A detailed flow diagram for encapsulation of the curium fuel form was also completed. Design and procurement of equipment for the fuel form effort is proceeding on schedule, with the bulk of the design work near completion.

Compatibility studies in which gadolinium oxide and carbide were used to simulate the respective curium compounds were continued. Metals for thermoelectric applications were tested at 2000°F for 150 hours, during which oxide and carbide pellets were in intimate contact with the metal surfaces. Evidence of incompatibility was observed by means of metallographic examination. Materials for thermionic applications were tested at a temperature of 2950°F.

A. OBJECTIVES

The objectives of the fuel form development program for this quarter were:

- (1) To initiate the design and procurement of the equipment required to produce fuel forms from the oxide of Curium-242.
- (2) To investigate the parameters of the blending operation to be used for preparing the carbides and cermets of both curium and plutonium.
- (3) To investigate methods of crushing curium carbide applicable to "hot cell" operation.
- (4) To design and fabricate pass-ports for the plutonium glove box system in the HEPPF.

- (5) To design and build the experimental apparatus for the plutonium system, including the process sampling system.
- (6) To determine the compatibility of gadolinium compounds used to simulate curium compounds with various matrix and container materials for thermionic and thermoelectric applications.
- (7) To investigate the heating of thermionic containment materials for compressive impact tests.
- (8) To initiate the design and procurement of the equipment required to encapsulate the Curium-242 fuel form.

B. SCOPE OF WORK

1. Curium Fuel Form Development (6.3.1)

Blending studies were conducted with various size containers at various speeds to determine the optimum blending parameters. Crushing of sintered carbide pellets in a die was considered to be the cleanest and simplest method for hot cell operation. The methods for, and type of product from, this operation were established.

Specification control drawings were prepared for containment boxes to be installed in Cells 3 and 4 at Quehanna. These drawings were employed to develop engineering and manufacturing cost estimates. A preliminary flow diagram was prepared showing the equipment required in the containment box to produce fuel forms from the oxide of Curium-242. Design effort was initiated on the following items:

- (1) Press and Dies
- (2) Blender
- (3) Loading rack
- (4) Inert gas system.

A preliminary flow diagram was prepared, showing the equipment required in the containment box to encapsulate the Curium-242 fuel form. In this case, design effort was initiated on these items:

- (1) Welder
- (2) Shielded storage vessel for capsule
- (3) Inert gas system (same size as that in the fuel form containment box).

A preliminary flow diagram was prepared for the facility process sampling system, and design effort was initiated to procure the required equipment. In conjunction with the sampling system, engineering drawings were forwarded to Quehanna, specifying the cell penetrations required to route all tubing. All sampling will be performed in the isolation room behind Hot Cell 4.

2. Plutonium Fuel Compound Development (6.3.2)

Designs for the pass-ports for the plutonium glove box system have been completed, and initial fabrication of the ports has begun. An eddy-current-concentrator induction furnace has been constructed and tested.

3. Containment and Compatibility Investigation (6.3.3)

The compatibility of gadolinium carbide and oxide at thermoelectric operating temperature (2000°F), (as substitutes for curium compounds) with Hastelloy C, Inconel X, Type 316 stainless steel, Haynes 25, zirconium, columbium, molybdenum, and tungsten was determined. The compatibility of gadolinium oxide and carbide with tungsten, tantalum, columbium and platinum at thermionic operating temperatures (2950°F) was also determined. Studies on a 2.5-kilowatt portable induction unit were made to determine whether thermionic operating temperatures could be obtained for compressive drop testing work.

C. WORK ACCOMPLISHED AND EVALUATION OF EFFORT

1. Fuel Form Development (6.3.1)

Blending studies were conducted on mixtures of gadolinium oxide and carbon, and a metal-ceramic mixture. These powders were mixed in a rotating cylindrical chamber, tilted at a 20-degree angle to improve the mixing. Tests were initially performed with bottles of various diameters to determine the optimum diameter. The amount of material mixed was enough to produce three grams of gadolinium carbide. It was found, in all cases, that incorporation of a screen in the mixing bottles improved the blending. The screen had 20 openings to the lineal inch. It was found that a container having a 1-1/2-inch inside diameter produced the most uniform mixture. The rotational speed for this container was 36 rpm.

Blending times from 10 minutes to one hour were employed, and it was found that one hour produced the best blend. A stainless steel weighing bottle, as shown in Fig 12, was fabricated. The cap of the mixer was made to facilitate pouring the mixture into the compacting die. Tests performed on this mixer indicated that cermet mixtures could be satisfactorily blended in a container of this size. Figure 13 shows the distribution of UO_2 and carbon as obtained by X-ray. A cooling jacket capable of cooling over 360 watts of heat must be provided for this blender.

Fig. 12. Mixing Container for Curium Compounds and Cermets

Fig. 13. As-Blended Distribution of
 UO_2 and Carbon

Crushing of sintered carbide pellets with mortar and pestle was considered too dirty an operation for hot cell use. Crushing in a ball mill would cause excessive loss of material; therefore, a die and punch was chosen for crushing as it was the cleanest and least complicated method. Initial crushing tests were made with dies of 3/8-inch and 1-inch diameters. The 3/8-inch die did not allow enough spreading of the material for crushing while the larger 1-inch die produced satisfactory results. The crushing procedure is as follows:

- (1) The pellet is placed on its side in the die.
- (2) The top punch is inserted and pressure is applied (approx. 250 psi).
- (3) Pressure is removed and punch is rotated several times.
- (4) Pressure is reapplied.
- (5) Steps 3 and 4 are repeated.
- (6) Top punch is removed and powder is transferred into either storage container or blender.

Crushing over three times did not appreciably reduce the carbide particle size. Rotating the top punch helped to spread the crushed particles across the face of the punch and aided in producing a higher percentage of fine size particles.

Table 3 gives the particle size distribution obtained when various numbers of gadolinium carbide pellets, each pellet equivalent to one gram of curium carbide, were crushed in a one-inch die.

TABLE 3
Particle Size Distribution of Crushed Gadolinium Carbide

Weight of Screened Gd C ₂						
<u>Mesh Size</u>	Batch 1		Batch 2		Batch 3	
	<u>(gm)</u>	<u>(% of charge)</u>	<u>(gm)</u>	<u>(% of charge)</u>	<u>(gm)</u>	<u>(% of charge)</u>
+35	--	--	--	--	0.22	3.1
-35 +45	--	--	--	--	0.51	7.0
-45 +80	--	--	--	--	1.87	25.6
-80 +100	--	--	1.77	34.8	0.72	1.0
-100 +170	4.5	54.6	0.96	18.9	1.41	19.3
-170 +230	0.78	9.5	0.59	11.6	0.51	7.0
-230 +270	0.49	5.9	0.32	6.3	--	--
-270 +325	0.75	9.1	0.44	8.7	1.00	13.7
-325	1.71	20.8	1.00	19.7	1.06	14.5
Total charge	8.23		5.08		7.3	

A study was made to determine whether the large particles obtained would further break up and disperse, in the blending operation, with the matrix material. Iron powder was used as the simulated matrix so that separation could be achieved magnetically. The mixture of gadolinium carbide and iron was blended for one hour in an argon atmosphere, and then an attempt was made to separate the iron. Unfortunately, the carbide adhered to the iron, interfering with the analysis and negating the results.

Experiments are being conducted on gadolinium carbide pellets having a volume equivalent of three grams of curium carbide to establish the die size needed, and the particle size distribution which can be obtained, with this amount of material.

2. Design of Fuel Form Containment Boxes at Quehanna Site (6.3.1)

A preliminary flow diagram for the fuel form operations was completed, and design efforts were initiated on a number of pieces of equipment.

a. Press and dies

The layout of the press and dies neared completion. Present plans call for procurement of an off-the-shelf press from Parker-Hannifin Corp. It will have a capacity of 125 tons and will be able to compact one-inch diameter pellets at pressures in excess of 100,000 psi. Additional effort, however, is required to ensure that the dies are integrated with the loading rack to be discussed later in the report.

b. Blender

Several blender designs were investigated. The design of the blender is greatly influenced by the sequence and manner in which operations will be carried out. The blender finally selected will be mounted at an angle of 20° to the horizontal plane. The mixing chamber will be a tubular can approximately 1-1/2 inches in diameter 4 inches in length. The chamber will rotate within a water jacket designed to remove the decay heat of Curium-242. The mixing chamber and cooling jacket can be easily disengaged from the motor and moved to the loading rack discussed below. Plans include leaving the water hoses attached to simplify the operations and increase reliability.

c. Loading rack

A concept for loading the powder in the blender and die was developed. The most significant problem during the transfer operations is that of dusting, particularly in the case of the oxide of curium. To overcome this problem, the powder will be transferred through the use of sliding covers and other devices; e.g., dispensers, from one unit to another, will be arranged in such a manner that the vessel containing powder always remains covered. Special covers will be designed, and operations will be performed in a special loading rack. The system

will require interchangeability of components among the oxalate vessel, blender and dies. Consideration was given to using dust enclosures similar to those used at the University of California. It was felt, however, that operation within an enclosure in the containment box would be unnecessarily complex.

d. Inert gas system

Design effort was initiated on the inert gas system to be installed in the containment boxes of Cells 4 and 5. No firm decisions on the selection of equipment will be made until the project has contacted the following laboratories for a review of the efficiency of their systems:

- (1) Savannah River National Laboratory
- (2) Argonne National Laboratories
- (3) Los Alamos Scientific Laboratory.

A preliminary flow diagram for the encapsulation of the curium fuel form was completed. Detail design of the inert gas containment boxes has been initiated.

e. Welder

The layout of the welder for sealing the final fuel form neared completion; the unit will have the capability of encapsulating either the Subtask 5.3 or Surveyor capsule. Water cooling will be provided to absorb the decay heat of Curium-242 or any excessive energy produced by the welding operation. The welder itself is similar to that used for strontium work; modifications are required, however, to adapt it to handling the Subtask 5.3 capsule configuration.

f. Capsule shielded storage vessel

A preliminary layout was completed for a vessel in which the final capsule could be stored. The vessel is water cooled and shielded to permit access of personnel to Cell No. 4. The vessel will also be equipped to serve as a leak detector to evaluate weld integrity by the helium check method. This will eliminate the requirement for an additional piece of equipment. At present it appears that, with minor modification, the vessel can also be used for pellet storage in the fuel form containment box.

g. Inert gas system

The system will be the same as that used for the fuel form work.

3. Containment and Compatibility Studies (6.3.3)

The development of curium compounds for use in thermoelectric generators prompted the investigation of container material behavior at thermoelectric operating temperatures. Compatibility studies on the carbide and sesquioxide of gadolinium (GdC_2 and Gd_2O_3) with various materials were conducted at 2000°F for 150 hours. Since thermoelectric operating temperatures presently are lower than 2000°F, 150 hours at this temperature should be comparable to a long-time operation at the lower temperatures (1300° to 1500°F). The following materials were compatible with gadolinium oxide and carbide:

- (1) Haynes 25
- (2) Hastelloy C
- (3) Inconel X
- (4) Type 316 stainless steel
- (5) Columbium
- (6) Molybdenum
- (7) Tungsten
- (8) Zirconium.

Tantalum was not tested, but it should be satisfactory as it has been proved compatible with other similar materials at the high temperatures. Compatibility was determined by metallographic observation.

In the preceding quarterly report (Ref. 3) it was stated that the compatibility tests of tantalum, columbium and platinum with gadolinium oxide and carbide (as substitutes for curium compounds) at 2952°F were not conclusive because of superimposed atmosphere effects. A test was devised, therefore, to eliminate these effects. Pellets were placed in tubes of the container materials; the tubes were then evacuated and crimped shut. Figure 14 shows the design of the test specimens.

Fig. 14. Design of Compatibility Test Specimens

The results of the test at 2952°F are given in Table 4.

TABLE 4

Compatibility of Container and Matrix Materials
with Gadolinium Oxide and Carbide at 2952°F

<u>Material</u>	<u>Time (hr)</u>	<u>Simulated Isotope Compound</u>	<u>Results</u>
Columbium	112	Gd ₂ O ₃	No reaction
		GdC ₂	Reaction observed
Tantalum	64	Gd ₂ O ₃	No reaction
		GdC ₂	Test capsule leaked and inner surface oxidized
Platinum	112	Gd ₂ O ₃	Reaction-- melted
		GdC ₂	Reaction-- melted

Several additional tests were performed on platinum. In all cases there was reaction, indicating that platinum cannot be used as a matrix material. Tantalum and columbium can be used as matrix materials for curium oxide if gadolinium oxide is a valid substitute. It is doubtful whether these materials could be used with the carbide because of excessive reactions observed.

It was previously reported that tungsten was compatible with neither gadolinium oxide nor carbide. However, since molybdenum was compatible and

tungsten usually reacts similarly to molybdenum, further testing was conducted. Arc-melted tungsten was tested since the sheet tungsten previously used was thought to contain impurities which reacted with the simulated fuel. An eight-hour test in vacuum at 2952°F indicated that no reaction occurred. A 64-hour test in argon has been run, but metallographic test results are not yet available.

Tests run with a 2.5-kilowatt induction unit indicated that the unit would be capable of heating refractory metals to thermionic operating temperatures. A unit of this type could be used in conjunction with the compressive drop test for testing refractory metals.

D. CONCLUSIONS

1. Curium Fuel Form (6.3.1)

Blending of curium compound and a metal-ceramic mixture can be accomplished in a container with a 1-1/2-inch inside diameter and a 2-1/4-inch length, containing a screen. Cooling facilities will have to be provided.

Fuel pellets maybe successfully crushed in a die, although several applications of pressure will be necessary.

2. Design of Fuel Form Containment Boxes for Quehanna Site (6.3.1)

Selection, design and procurement of equipment are proceeding according to schedule. The bulk of the design work is under way and nearing completion. Detailed design requirements for the equipment are being established concurrently with this design. Much more detailed information will be given in the next quarterly report, at which time final engineering diagrams and drawings will be available.

3. Fuel Form Containment (6.3.3)

If gadolinium oxide and carbide are valid substitutes for the corresponding curium compounds, then Hastelloy C, Inconel X, Type 316 stainless steel, Haynes 25, zirconium, columbium, molybdenum, and tungsten should be satisfactory as container materials at thermoelectric operating temperature. Tantalum and columbium should make satisfactory matrices at thermionic operating temperatures with curium oxide. With curium carbide their use is unlikely. Platinum can be used as a matrix material with neither the oxide nor the carbide. Testing of arc-melted tungsten indicates that it might be compatible with gadolinium oxide and carbide.

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

The safety aspects of irradiating Americium-241 capsules in the MTR at a flux of 4×10^{14} neutrons/cm²-sec have been thoroughly investigated. The worst possible conditions were assumed throughout the analysis; a hot-spot safety factor of approximately 2 was ultimately applied to the thermal values. Because this approach was more stringent than necessary, the capsules were found to be safe with a satisfactory margin. A safety analysis has been completed and transmitted to the National Reactor Testing Station, Idaho Falls, Idaho, for their appraisal of the Task 6 irradiation program.

Shielding analyses were conducted to determine the various requirements for transfer of the irradiated capsules at Quehanna and for waste disposal of fission products, also at Quehanna.

The Task 6 calorimetry requirements for determining curium content at various stages of the processing were investigated, with the consequent recommendation of two types of calorimeters. Also, several items of process equipment were thermally analyzed to aid in the design effort. Special techniques were outlined where overheating could become a problem or where additional safety features appeared desirable.

A. OBJECTIVES

The objectives for the Safety, Thermal and Shielding Analysis Sections were:

- (1) To determine whether the americium capsule can be safely irradiated in the MTR at a thermal flux of 4×10^{14} neutrons/cm²-sec.
- (2) To determine and specify shielding requirements and materials during the processing of irradiated Americium-241.
- (3) To review the Task 6 calorimetry requirements and recommend suitable equipment to meet thermal requirements.
- (4) To conduct thermal analyses applicable to the design of Curium-242 process equipment.

B. SCOPE OF WORK

1. Safety Analysis

An analysis was made to determine the maximum heat flux in the americium capsule during irradiation in the MTR for the purpose of substantiating that the

capsule would withstand the internal temperatures and pressures generated.

2. Shielding Analysis

An analysis was made to determine the shielding required during the transfer of irradiated capsules from the storage pool to the hot cell. Shielding requirements for the disposal of wastes containing fission products were determined.

3. Thermal Analysis

A study was made of three general types of radioisotope calorimeters, an extremely accurate thermopile type, a vacuum flash thermometer type and an ambient equilibrium temperature type.

C. WORK ACCOMPLISHED AND EVALUATION OF EFFORT

1. Americium Capsule Preparation

A safety analysis on the americium capsules at several points in the irradiation process--during irradiation at an unperturbed flux of 4×10^{14} nv* (thermal neutrons), during postirradiation cooling in the reactor canal, and during shipment to Quehanna--was made during this report period. Worst-case conditions were employed in all phases of the analysis, and the equations used reflected the greatest possible conservatism to assure a maximum factor of safety. It was found that the capsules would safely contain all radioactive constituents throughout the entire program and would at no time have an adverse effect on reactor operation.

2. Curium Process Shielding

The analysis of the shield required for transferring a single irradiated slug from the shipping cask to the hot cell showed that a minimum of 7-1/4 inches of lead would be required to reduce the combined neutron and gamma dose rate to 10 mr/hr at one meter. This shielding was determined for slugs cooled 30 days after removal from the reactor.

Dose rates from the process wastes will be predominantly from fission product gamma rays. The neutron activity will not be significant.

*nv = number of neutrons per unit volume x mean neutron velocity:

$$\frac{n}{\text{cm}^3} \times \frac{\text{cm}}{\text{sec}} = \frac{n}{\text{cm}^2 - \text{sec}}$$

Previous shielding analyses were based upon information given (Ref. 4, Fig. 6). This figure was constructed from rate readings taken from a slug containing 250 milligrams of Americium-241 after 84 days irradiation in the MTR. The readings were started 16 days, and stopped 42 days, after the slug was removed from the reactor. This information is excellent for the range of decay times it covers. An extrapolation of this curve to longer decay times would produce very inaccurate results. Therefore, it became necessary to calculate activities from the fission rate and yields of fission products. Examination of the fission cross sections of the heavy elements formed during the irradiation of Americium-241 (Ref. 4, Fig. 5) shows that the largest number of fission products result from fission of Americium-242. Its fission cross section for thermal neutrons is 6400 barns. Of all the fission products formed, only about a dozen have half lives greater than 30 days. Comparison of the dose rates from these fission products with the dose rates given in Ref. 4, Fig. 6, shows that this group produces about 60 percent of the total dose rate 42 days after shutdown.

At the end of 360 days of decay, the fission products resulting from irradiation of 3.57 grams of americium produce a dose rate of about 860 mr/hr at four feet from the center of a 55-gallon drum lined with 3 inches of ordinary concrete and filled with vermiculite to absorb the liquid waste.

3. Thermal Analysis-- Curium Purification, Fuel Form Processing and Containment

A study was made of three different general types of radioisotope calorimeters: an extremely accurate thermopile designed and built originally by UCRL, a vacuum flask thermometer type measuring the temperature rise of a known fluid volume with time, and an ambient equilibrium type where the temperature is measured in a holder while heat is given up by conduction, convection and radiation to the surroundings.

The vacuum flask type was recommended for normal requirements such as the evaluation of the thermal output of a radioisotope sample. For precise measurements such as required for radiological analysis, the thermopile type was suggested. The ambient equilibrium type was not recommended because of inherent unreliability.

The process equipment analysis included sizing the containers, condensers and heaters for evaporation steps during the process (no distribution), thermal analysis of the curium-loaded ion exchange column, thermal analysis of the dry curium-loaded platinum filter (initial filtrations), and a discussion of the choice of an air filtration system for the curium containment box.

Recommendations were made as to heater capacities required for various process times. Reflux condenser columns in standard sizes were chosen and special equipment suggested where required.

The temperature difference between center and surface of the curium-loaded ion exchange column was found to be 24°F at a flow rate of 40 ml/min through the two-inch column. Based on this analysis, it was recommended that the column wall be held at 80°C to avoid boiling at the center of the column. The equilibrium temperature of the dry curium-loaded filter was found to be approximately 900°F. This temperature exceeds the thermal limit of the proposed irradiated polyethylene cover of the filter. It was recommended that an emergency water supply be provided to prevent the filter cake from drying out and overheating the polyethylene during operation.

A dual filtration system was recommended for the curium containment box. It included a single internal prefilter-filter combination replaceable through the use of the seven-pound (weight limit) manipulator. Snap-clamps rather than threaded joints are used to facilitate replacement since tight seals are not required. For air flow in and out of the box, large-capacity, highly damped (normal operation) filters are suggested. Should a leak occur, the dampers would open immediately for maximum air flow. It is not anticipated that these filters will require changing during the operational life of the containment box. However, provision should be made for a dual system which will permit changing one set of high capacity filters while the other is in use.

D. CONCLUSIONS

1. Safety Analysis

The irradiated americium capsule will safely sustain the maximum temperatures and pressures encountered during irradiation in the MTR and subsequent shipment from the reactor to the Martin Nuclear Facility at Quehanna, Pennsylvania.

2. Shielding Analysis

The shielding determined for the slug transfer cask was based upon 30 days' decay. Since it is probable that a longer decay time will be involved due to shipping and processing of other slugs, the dose rate through the 7-1/2 inches of lead will usually be less than the 10 mr/hr used as the design criterion. To maintain a dose rate of 10 mr/hr after a year's decay, about 100 drums would be required for the waste from each slug. Consequently, it has been recommended that the fission products be concentrated by some method and that the concentrate be shipped to a burial ground in a lead shipping cask.

VI. SUBTASK 6.6--IRRADIATION OF AMERICIUM-241

The AEC-21 and AEC-22 Irradiation Request Forms were submitted for irradiation of five Task 6 capsules in the MTR. A safety analysis covering all aspects of the irradiation (i.e., shipment to the National Reactor Testing Station, during irradiation at 4×10^{14} nv, during post-irradiation cooldown in the MTR canal, and during shipment to Quehanna). This analysis was transmitted to personnel at the MTR for their review and subsequent presentation to the NRTS Safeguards Committee.

Five fully loaded americium capsules were packaged and shipped to the MTR; arrangements for the irradiation will be made in July.

A. OBJECTIVES

The objectives for the americium irradiation program were:

- (1) To complete arrangements for irradiating five capsules of Americium-241 in a high thermal neutron flux to form Curium-242.
- (2) To prepare a comprehensive safety analysis on the irradiation program to prove capsule integrity and to maintain liaison with the MTR on the overall effort.

B. SCOPE OF WORK

1. Irradiation of Americium-241

AEC-21 and AEC-22 Irradiation Request Forms for irradiation of five americium capsules in the MTR at a thermal neutron flux of 4×10^{14} nv were originated by The Martin Company and transmitted to the New York Operations Office for approval and appropriate action.

2. Safety Analysis of Capsule Irradiation

A safety analysis report was prepared and submitted to the National Reactor Testing Station for review by the MTR personnel and the NRTS Safeguards Committee. Included in this analysis are such things as:

- (1) Capsule description, including composition, dimensions and methods of fabrications.
- (2) Establishment of irradiation parameters; i.e., neutron flux levels

required for maximum conversion of Americium-241 to Curium-242, the irradiation periods involved, and the corresponding power output at different intervals throughout and after irradiation.

- (3) Capsule modifications and test results.
- (4) Thermal analyses for the periods during shipment to NRTS, during irradiation, during cooldown in the MTR Canal and during transportation to Quehanna.
- (5) Pressure buildup in the capsules due to decay helium, and possible formation of oxygen from the americium dioxide when curium oxide is produced. This analysis covered periods of time identical to those listed in Item (4).
- (6) Stress analysis on all aspects of the capsule design for the worst condition encountered in Item (5).

C. WORK ACCOMPLISHED AND EVALUATION OF EFFORT

1. Selection of Reactor

Although the Engineering Test Reactor (ETR) is capable of furnishing the high thermal neutron flux (4×10^{14} nv) required for the Task 6 Program, the danger of unscheduled shutdowns dictated that the Irradiation Request Forms, AEC-21 and AEC-22, specify that the irradiation be performed in the MTR. The net result of the various nuclear reactions such as fission, capture, decay, etc., occurring during the irradiation make it necessary to irradiate the americium capsules at a high flux intensity for a minimum period of time so maximum conversion of Americium-241 to Curium-242 can be achieved. The higher flux facilities in the MTR can satisfy this requirement; effort is therefore under way to schedule all five of the Task 6 capsules in the MTR. The reasons for this choice are as follows:

- (1) Unscheduled shutdowns during the irradiation can result in excessive decay of the relatively short-lived Curium-242 isotope. Such an occurrence could seriously hamper the Task 6 Program.
- (2) Since Americium-241 is quite expensive, maximum conversion in a high flux field is most desirable. Irradiation at low flux levels will burn out the americium without giving a proportional amount of curium.

2. Irradiation Safety Analysis

A safety analysis was prepared on the Task 6 irradiation program to establish integrity of the capsules under the worst conditions possible at any time during or after the irradiation. This analysis was initiated with a rather comprehensive nuclear investigation of the capsules during irradiation. Due to the relatively large amount of Americium-241 present in each capsule, the flux depression within the capsules was considered quite significant. Consequently, a cell correction factor, or flux depression factor, was calculated by means of a standard method in current use in reactor analysis. This method is programmed for the IBM 7090 computer and is known as Program I-2. The mathematics and method of programming are fully developed in Ref. 5. The Boltzmann equation is solved for cylindrical geometry by the P_3 spherical harmonic approximation. The thermal flux distribution across the fuel capsule is calculated and averaged. Results of this analysis indicated that the average flux depression factor for the americium capsules is 0.7; that is, in an unperturbed flux of 4×10^{14} nv, the effective flux will be 2.8×10^{14} nv.

During the irradiation, numerous isotopes are formed; the more significant of these and their relationships are shown in Fig. 15. However, interest for the Task 6 program is centered on the reaction which produces Curium-242:

The differential equations that define this process are:

$$\frac{dA}{dt} = -A_0 \sigma_{A_T} \phi \quad (1)$$

$$\frac{dB}{dt} = F_{A_B} A_0 \sigma_{A_T} \phi - B \left(\sigma_{B_R} + \frac{\lambda_B}{\phi} \right) \phi \quad (2)$$

$$\frac{dC}{dt} = F_{B_C} B \lambda_B - C \left(\sigma_{C_R} + \frac{\lambda_C}{\phi} \right) \phi \quad (3)$$

where

$$A = \text{Am-241}$$

$$B = \text{Am-242m}$$

$$C = \text{Cm-242}$$

$$\sigma_{A_T} = \text{total absorption cross section for A}$$

σ_f = thermal neutron fission cross section (cm^2)

σ_c = thermal neutron capture cross section (cm^2)

λ = decay constant (sec^{-1})

SF = spontaneous fission half life

b = cross section (barns)

τ = half life

Fig. 15. Nuclear Properties and Relations of the Isotopes Produced by the Neutron Irradiation of Americium-241

σ_{A_B} = absorption cross section for A to B

σ_{B_R} = removal cross section for B

σ_{C_R} = removal cross section for C

λ_B, λ_C = decay constants for B and C, respectively

F_{A_B} = branching fraction of A to B

F_{B_C} = branching fraction of B to C

ϕ = effective thermal neutron flux.

Equation (4) is the general solution which is programmed on the 7090.

$$N_L(t) = N_0 \prod_{J=1}^{L-1} \sigma_J F_J \sum_{J=1}^L \frac{\exp \left[- \left(\sigma_J \phi + \lambda_J \right) t \right]}{\left[\prod_{K=1}^J \left(\sigma_K + \frac{\lambda_K}{\phi} - \sigma_J - \frac{\lambda_J}{\phi} \right) \right] \left[\prod_{m=J+1}^L \left(\sigma_m + \frac{\lambda_m}{\phi} - \sigma_J - \frac{\lambda_J}{\phi} \right) \right]} \quad (4)$$

$$\text{When } K \geq J, \quad \left(\sigma_K + \frac{\lambda_K}{\phi} - \sigma_J - \frac{\lambda_J}{\phi} \right) = 1.0$$

$$\text{When } m > L, \quad \left(\sigma_m + \frac{\lambda_m}{\phi} - \sigma_J - \frac{\lambda_J}{\phi} \right) = 1.0$$

Equations (1), (2) and (3) can be solved by hand; however, because of the intermittent irradiation method (the reactor is expected to be shut down for a significant portion of the total irradiation period) and to permit numerous calculations if necessary, the solution was generalized and programmed for the IBM 7090. Equation (4) is the general solution which was programmed. This program can calculate the amount of each isotope in any burnup and decay chain whether the irradiation is continuous or intermittent.

Subsequent calculations were made to determine the rate of burnup of Americium-241 and percent conversion to Curium-242 employing a basis of constant effective neutron flux. It should be noted that the MTR operates on a 21-day cycle; i.e., 15 days of actual operation and 6 days of shutdown. This fact was taken into account in the calculations.

The percent of Americium-241 consumed and the percent of the original Americium-241 available as Curium-242 at the end of various irradiation cycles were calculated and are summarized in Table 5. The actual weight per capsule of Americium-241 and Curium-242 versus time is also given in Table 5.

The actual percentage of the original Americium-241 converted to the different isotopes by continuous irradiation at 2.8×10^{14} nv (perturbed flux) is given in Fig. 16. Of these, only Americium-242, Americium-242m, and Curium-242 are important for this study from the viewpoint of heat production. Since Americium-242 and Americium-242m have very large thermal fission cross sections, they are the most important sources of fission heat; the primary source of heat from Curium-242 is the decay process. Table 6 summarizes the analysis made to determine the total heat produced by a single capsule at different periods during the irradiation. It is seen that this revised analysis results in a peak heat flux 10 days after start of the irradiation.

Based on the maximum power output listed, a heat transfer analysis was conducted on an americium capsule in the L-47 position of the MTR. A hot-spot factor was developed along the same lines as recommended for irradiations in the MTR; the value of the hot-spot factor was 1.94, with an additive factor of 14°F. These factors were then applied to the capsule surface temperature obtained by using the Sieder-Tate Correlation for turbulent fluid flow.

Results indicated that neither core-melting nor boiling on the capsule surface would occur at any time during irradiation. The thermal analysis was then extended to include periods of time after irradiation (i.e., during cooldown in the MTR Canal and during shipment of the irradiated capsules to the Quehanna Hot Cell Facility). In this case, fission is not the dominant factor, but considerable heat is generated from decay of contained fission products and the Curium-242. It was found that maximum temperatures would occur (360°F on the capsule surface and an average core temperature of 418°F) during shipment of the capsules to Quehanna. These figures represent the worst possible case--all the cask shield water (11 inches thick) is lost and a maximum barrier exists to heat transfer through the walls of the cask.

TABLE 5

Percent of Americium-241 Burned and Curium-242 Available
from MTR Irradiation at Thermal Neutron Flux of
 2.8×10^{14} nv

cycle--15 days operation, 6 days shutdown

<u>Cycle</u>	<u>Days</u>	Percent Conversion		Production (gm)	
		<u>Am-241</u>	<u>Cm-242</u>	<u>Am-241</u>	<u>Cm-242</u>
	0	100	0.0	3.57	0.0
1	15	77.0	13.7	2.75	0.49
2	36	59.3	22.1	2.12	0.79
3	57	45.6	27.5	1.63	0.98
4	78	35.1	30.7	1.25	1.10
5	99	27.0	32.2	0.96	1.15
6	120	20.8	32.6	0.74	1.16
7	141	16.0	32.2	0.57	1.15

TABLE 6

Heat Produced in the Capsule by Fission of Americium Products
During Irradiation in the MTR at Thermal Neutron Flux of

$$2.8 \times 10^{14} \text{ nv}$$

<u>Isotope</u>	σ_f (barns)	Heat Produced			
		in 10 Days (watts)	in 15 Days (watts)	in 20 Days (watts)	in 30 Days (watts)
Cm-242	5	40	51	60	86
Cm-243	700	-	-	-	-
Cm-244	-	-	-	-	-
Am-241	3.1	208	191	178	147
Am-242	6390	7598	7783	7700	7388
Am-242m	3000	2661	2271	2000	1749
Am-243	0.075	-	-	-	-
Pu-238	18.4	4	8	12	27
Pu-239	746	-	8	18	60
Pu-240	0.1	-	-	-	-
Pu-241	950	-	-	-	-
Pu-242	0.3	-	-	-	-
Np-237	-	-	-	-	-
Total power from fission in the capsule (watts)		10,511	10,312	9968	9457
Power from reactor * (Watts)		1120	1120	1120	1120
Total power per capsule (watts)		11,631	11,432	11,088	10,577

* 7 watts/ gram x 160 gram/capsule

On the basis of this worst-case condition, the maximum pressure developed in the capsule from decay helium and oxygen formation was then calculated. It is doubtful that any free oxygen will result from the reaction

but it was included in the pressure analysis to provide a margin of safety. Since the core density is 85% of theoretical and an additional void was incorporated in the capsule for the sole purpose of accommodating evolved gases, the total pressure buildup was minimal:

Pressure from helium at 418°F = 228 psi

Pressure from oxygen at 418°F = 113 psi

Maximum 341 psi

Actually, the capsule pressure is less than the estimated 341 psi for the three following reasons:

- (1) There will be less free oxygen than indicated by the chemical equation since some of the americium exists as Am_2O_3 and some of the curium may exist as CmO_2 .
- (2) Most of the free oxygen will probably combine with the fission products and with the aluminum matrix.
- (3) The curium buildup is gradual over a 99-day period during irradiation and then decays over a 60-day period during which the capsules are shipped to Quehanna. However, the assumption made for these calculations was that all the curium was present the first day of the irradiation, decaying over a subsequent 160-day period. Obviously, the helium pressure from the curium decay is significantly overestimated.

Based on 341 psi and the maximum temperatures involved, stress analyses were conducted on all aspects of the capsule design. It was found that the seal weld was the most critical area of the entire capsule and that, even with the worst-case conditions employed, there exists a margin of safety of 38%. Furthermore, the stress analysis was based on a weld depth of 0.025 inch while the actual weld is closer to 0.05 inch. No consideration was given to the extra strength provided by the sleeve which was ultimately shrunk over the capsules. Both these factors afford an additional margin of safety.

The Task 6 capsules were initially designed for irradiation in a one-inch diameter facility in the ETR; however, the decision to irradiate in the MTR

made it necessary to change the capsule diameters from 1 inch to 1-1/8 inches so they could be readily accommodated in a standard X-basket facility in the MTR. As a result, sleeves with wall thicknesses of 1/16 inch were shrink-fitted over the capsules to achieve the proper diameter. Development work was first conducted on shrink fitting sleeves over dummy capsules to optimize the procedures; these dummy samples were then tested to determine whether any water leakage into the capsule-sleeve interface would cause blistering due to thermal cycling. All tests proved the capsule to be safe in every respect.

The thermal barrier presented by the capsule-sleeve interface was rigorously analyzed by the methods described in Ref. 6. Results indicated that, with the alloys and surfaces actually used and with the good mechanical contact achieved, the temperature drop across the capsule-sleeve interface is probably around 10° to 15°F. The very worst case gave a maximum temperature drop of 95°F; this figure was employed in the calculations without adverse effects to either the thermal or structural aspects of the capsules.

Details were also provided in the safety analysis on the americium shipping cask, the cask for transfer of the irradiated capsules, the Bureau of Explosives Approval, the properties of Americium-241 and Curium-242, etc.; but these have been covered in earlier quarterly reports and will not be discussed at this time.

D. CONCLUSIONS

It is concluded that the most desirable facility for irradiating the Task 6 capsules is the MTR. The reasons are that a continuous high neutron flux over a rather long period of time is necessary to achieve maximum conversion of Americium-241 to Curium-242. Also, a thorough safety analysis has established that no radioactive materials will escape at any time during the entire irradiation program.

At present it appears desirable to irradiate capsules at flux levels up to 5.5×10^{14} nv. Consequently, new calculations will be performed along the same lines as the procedure followed for the 4×10^{14} nv flux.

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