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AEC RESEARCH AND DEVELOPMENT REPORT

SAVANNAH RIVER LABORATORY ISOTOPIC POWER AND HEAT SOURCES

QUARTERLY PROGRESS REPORT

OCTOBER – DECEMBER 1968

SRL
RECORD COPY

PART II

CURIUM-244 AND PLUTONIUM-238

DU PONT
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Savannah River Laboratory

Aiken, South Carolina

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SAVANNAH RIVER LABORATORY
ISOTOPIC POWER AND HEAT SOURCES

Quarterly Progress Report

October - December 1968

Part II

Curium-244 and Plutonium-238

H. S. Hilborn, Compiler

March 1969

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PREFACE

This report is one in a series on the applied aspects of isotopes that are under study at the Savannah River Laboratory (SRL), and that are of interest as isotopic heat source materials. Principal emphasis is on isotopes that are produced by neutron addition, since these are the materials for which the production capabilities of the Savannah River Plant (SRP) reactors and other facilities can be used effectively. Data for other materials will be included if pertinent -- such as the isotopic or chemical composition of fission products that can be recovered from Savannah River process wastes.

These reports are intended to present data that are useful to system designers and also to potential or active user agencies. The reports thus deal with the following subject areas of SRL programs:

1. Properties and reactions of isotopes useful or potentially useful as heat sources.
2. Information on the irradiation and postirradiation processing of these materials, when the information is relevant to their use as heat sources and is not in a sensitive area of production technology.
3. Development of design data directed toward manufacturing capability for isotopic heat sources.

The report is issued in two parts: Part I includes only information on cobalt; Part II includes information on the other isotopic heat source materials. Both parts contain principally data from work performed during the report period. Previous reports are listed on the last page.

SUMMARY

CURIUM-244

Page

- The first 36 grams of ^{244}Cm from the Savannah River 5-kg production lot was shipped as curium oxide to Oak Ridge National Laboratory. The product met impurity and gamma radiation specifications. Purity was confirmed by a melting temperature of 2230 ±25°C. 1
- Efficient separations of curium from americium and californium were demonstrated using pressurized cation exchange chromatography. 6
- ^{243}Am is being routinely analyzed in ^{244}Cm by gamma spectrometry using a lithium drifted germanium X-ray detector system. 7
- Spark source mass spectrometry was extended to highly alpha active samples and used to determine simultaneously 70 elements from boron through americium in ^{244}Cm oxide. 8
- Thermal stability studies showed that only minor stabilization of CmO_2 can be obtained by addition of the more stable AmO_2 to form a mixed oxide. 10
- Installation of services and equipping of the extension to the Alpha Materials Facility for small-scale studies of ^{244}Cm fuel materials continued. 11

PLUTONIUM-238

- Conditions were defined for pilot production of ^{238}Pu containing <20 ppb ^{236}Pu from 600 g of Shippingport americium. 13
- Production of >10 kg/yr of ^{238}Pu with low ^{236}Pu content was projected in conjunction with the resonance reactor production of ^{252}Cf using ^{241}Am-^{243}Am from power reactor fuel reprocessing. 14

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CURIUM-244

Approximately 5 kg of ^{244}Cm is being produced at Savannah River; a large fraction will be for the development of ^{244}Cm heat sources. ^{244}Cm, which has a half-life of 18.1 yr and decays principally by emission of alpha particles, generates 2.65 w/g. The isotope can be utilized in applications requiring high power-to-weight ratios, with operating temperatures in the 1500-2000°C range for thermionic conversion systems, as well as in the lower temperature ranges for thermoelectric and thermodynamic conversion systems. Study of refractory forms of the isotope and development of fuel capsules for these applications is under way at Oak Ridge National Laboratory (ORNL) and at Savannah River Laboratory (SRL).

The specific objective of the SRL program is to acquire the technology needed for manufacture of ^{244}Cm fuel capsules. Pending definition of specific product goals, both basic and development studies of candidate systems are being conducted. Close liaison is maintained with ORNL. Current work includes:

- Formation and characterization of refractory ^{244}Cm compounds
- Development of techniques for preparing dense fuel forms
- Provision of facilities for encapsulation process development and testing

PRODUCTION AND SEPARATIONS TECHNOLOGY

Reactor irradiations to produce ~5.5 kg of ^{244}Cm for evaluation as a heat source fuel were completed in May 1967, and the irradiated targets were processed to recover plutonium in the initial stages of the separations operations to produce a high purity curium oxide product. Final separation of the ^{244}Cm, ~2.5 kg of ^{243}Am, and other transplutonium elements from fission product wastes using the Tramex solvent extraction process(1) is in progress. About 1.4 kg of the ^{244}Cm will be blended with ^{243}Am and, as oxides, compacted with aluminum into target slugs for irradiation to produce ^{252}Cf. The remaining ^{244}Cm will be used as oxide for heat source fuel development and other studies.

[redacted]

Curium-244 Oxide Purity

The first 36 grams of curium was shipped as curium oxide to ORNL on November 1, 1968. SRL and ORNL assays are compared in Table I.

TABLE I

Assay of First Curium Oxide Shipment

	SRL	ORNL
Total heat generation	102.07 w	103.6 w
Curium-242 heat	1.49 w(a)	1.3 w(b)
Net curium-244 isotope heat	100.58 w	102.3 w
Net curium-244 isotope weight	36.0 g(c)	36.5 g(c)

(a) Calculated at SRL

(b) Calculated at ORNL from the SRL value allowing for 33-day decay

(c) Calculated at ORNL from the power assuming 2.84 w/g of ^{244}Cm

Isotopic composition of the curium oxide is given in Table II.

TABLE II

Isotopic Composition of SRL Curium-244 Oxide

Isotopic Mass	Isotope % by Weight November 1, 1968
Curium-242	0.0426
Curium-243	0.0238
Curium-244	95.08
Curium-245	0.729
Curium-246	4.02
Curium-247	0.0659
Curium-248	0.0381

Emission spectrographic analyses showed that the impurity content of the curium oxide, summarized in Table III, was within specifications.

TABLE III

Elemental Impurities in SRL Curium Oxide (CmO_2)[a]

Element	ppm	Element	ppm
Li	<50	Sn	350
Na	500	Cd	<50
K	50	B	10
Si	75	As	<10
Zn	<250	Cu	<10
Fe	400	Mg	50
Cr	200	Mo	150
Ni	50	Mn	25
^{243}Am	4300	Co	<50
Zr	7500	Pt	100
Ca	25	Ta	<100
Al	<25	La	570

(a) Specification: Li + Na + K $\leqslant$0.1 wt %; Si + Zr $\leqslant$0.1 wt %; Fe + Cr + Ni $\leqslant$0.2 wt %; any other one element except Am, Zr $\leqslant$0.2 wt %; total of all elements except Am, Zr $\leqslant$1.0 wt %; total, including Am and Zr $\leqslant$3.0 wt %.

The purity of the initial 36 g of ^{244}Cm oxide was confirmed by the melting temperature, which was 2230 ±25°C - - well above the specified 1900°C -- and by satisfactory compatibility with platinum during heating at 1350°C for 2 hr in argon. The melting temperature was determined by optical pyrometry of oxide heated resistively on a tungsten V-ribbon in 4% H_2-He, and calibrated with alumina and thulia. After the 2-hr compatibility heating, X-ray diffraction showed no evidence of reaction of the resultant B-type Cm_2O_3 with platinum; the oxide was removed easily from the platinum with light scraping followed by swabbing with 10% HNO_3 to leave a smooth platinum surface.

Fission products in the purified curium oxide, summarized in Table IV, met the specifications for all nuclides except ^{144}Ce-^{144}Pr, which was slightly high. ^{144}Ce-^{144}Pr in the 36-g batch emitted ~12% of the gamma dose rate of ^{244}Cm; the specified total gamma dose rate from fission products is <10% of the unshielded gamma dose rate from ^{244}Cm.

TABLE IV

Gamma Dose Rate from SRL Curium Oxide

	μCi/g ^{244}Cm
^{144}Ce-^{144}Pr	3000
^{243}Am	720
^{252}Cf	230
^{106}Ru-^{106}Rh	130
^{154}Eu	<30
^{95}Zr-^{95}Nb	<20
^{134}Cs	<10
^{137}Cs	<5
^{103}Ru-^{103}Rh	<5

Cation Exchange Separation of Curium

Curium and americium were separated efficiently from fission products in further tests of the rapid cation exchange chromatographic process. This process is being developed as a potential replacement for Tramex solvent extraction process. The process does not require chloride solutions and the equipment is compact enough to be operated in a small portion of an existing canyon building.

In the first cation exchange separation of curium and americium from a sample of ^{244}Cm solution,(2) the removal of ^{144}Ce-^{144}Pr and ^{95}Zr-^{95}Nb was not adequate -- removal of ^{154}Eu was not measured. In recent tests, improvements were obtained by using chromatographic columns of smaller diameters, by monitoring the curium band with a neutron probe, and by detecting americium and curium in the column effluent with an in-line alpha monitor. The curium effluent met the specifications for removal of fission products.

As in the previous test, two precipitation steps removed interfering cations from the solution and permitted efficient loading of the cation resin. At a lanthanide-actinide concentration of 0.1M, the curium solution contained 1.8M Al^{3+}, 0.22M Fe^{3+}, and 0.16M Hg^{2+}. In the first precipitation, this solution was added to 8M NaOH to precipitate curium, americium, and other insoluble hydroxides for filtration from the aluminate solution. The hydroxide precipitate was then dissolved in nitric acid containing sufficient chloride ion to complex residual mercurous ions added to catalyze the target dissolution.

In the second step, actinides and lanthanides were precipitated as oxalates and filtered from the soluble trioxalato iron and dichloro mercury complexes. The oxalate precipitate degraded radiolytically to carbonate within 24 hr. The carbonate was then dissolved in dilute nitric acid to prepare feed for the cation exchange.

Cation Exchange

The lanthanide-actinide mixture containing 688 mg ^{244}Cm, 238 mg ^{243}Am, and $\sim$2.8 g of lanthanides was sorbed on a column 0.87 inch in diameter and 30-inches long, containing 30- to 60-micron "Dowex"* 50W-X8 resin. This column was subsequently connected in series with two other 30-inch columns of 0.43- and 0.18-inch diameter, respectively; all columns were at 70°C.

The elutriant was 0.05M diethylenetriaminepentaacetic acid (DTPA) buffered to pH 6.0 with NH_4OH and fed at 300 psi and 16 ml/(min)(cm^2). The order of product elution was californium, berkelium, curium, americium, europium, samarium, promethium, neodymium, praseodymium, and cerium. The progress of the curium band down the column was followed with a portable neutron monitor. When the curium neared the bottom of the column, the effluent was valved to the next smaller column. Because the overlap length of adjacent bands was independent of diameter, the eluate fraction of pure curium was increased by decreasing the diameter. Curium and americium were detected in the effluent from the third column with an in-line alpha monitor. Although separation of curium from americium was not expected to be complete with the DTPA elutriant, the cut was made between the curium and americium bands when the alpha monitor registered a sudden decrease.

Curium (in a fraction containing 97% of feed curium) was separated from the major fission products in the ion exchange cycle by the following factors: ^{144}Ce, >3.6 x 10^4; ^{154}Eu, 3.6 x 10^4; ^{106}Ru-^{106}Rh, 1.3 x 10^3. A previous separation factor of 1.3 x 10^3 from ^{106}Ru-^{106}Rh was obtained in the precipitation steps. The curium product purity, summarized in Table V, is within the goal specifications.

TABLE V

^{244}Cm Product from Cation Exchange

Fission Product	Ci/g ^{244}Cm
^{95}Zr-^{95}Nb	$<2.6 \times 10^{-5}$
^{144}Ce-^{144}Pr	$<1.7 \times 10^{-3}$
^{154}Eu	3.9×10^{-5}
^{106}Ru-^{106}Rh	2.0×10^{-4}

In addition to the separation from fission products, the americium and curium were partially resolved: 83% of the curium was eluted with only 4% of the americium; 97% of the curium, with 14% of the americium.

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* Trademark of Dow Chemical Co.

Californium-Curium Separation

Californium was separated more efficiently from curium in further tests using diethylenetriaminepentaacetic acid (DTPA) elutriant in the pressurized ion exchange column. Aluminum ion increased the separation. Californium is usually separated from curium with ammonium α-hydroxyisobutyrate elutriant.

The feed for the californium-curium separation test was the curium-americium-californium product fraction from the previous test, which contained 668 mg of ^{244}Cm, 238 mg of ^{243}Am, and 2.1 μg of ^{252}Cf. Sufficient Al^{3+} (270 mg) was added to form a sorption band ~20 cm long between curium and californium on a 1/2-inch-diameter column. The conditions and elution results are shown in Figure 1; 90% of the eluted californium was associated with only 0.02% of the curium, and 95% of the californium was associated with only 2.5% of the curium. In this test no attempt was made to define the elution of americium, which eluted after curium.

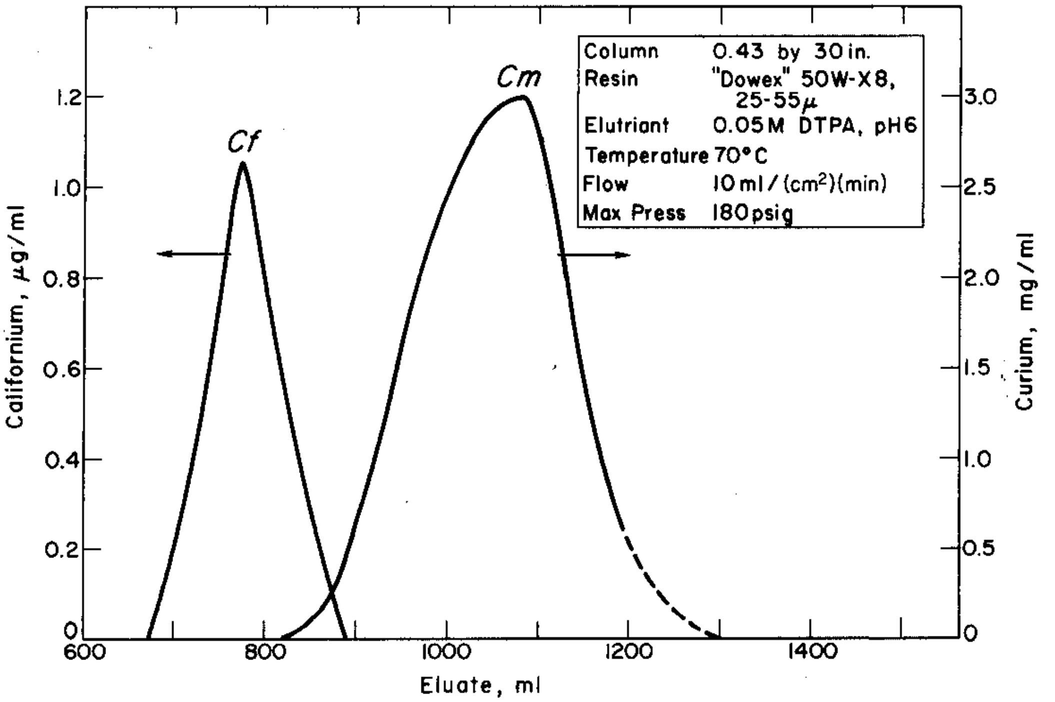


FIG. 1 ELUTION OF CALIFORNIUM AND CURIUM IN CATION EXCHANGE SEPARATION

Pilot equipment is being constructed in a shielded cell to separate ~60 g of curium per test with columns of 4-, 3-, 2-, and 1-inch diameter, each 4-feet long. Laboratory tests are determining the resin service life in the intense alpha radiation of the process.

Spectrographic Determination of Potassium in ^{244}Cm Oxide

An emission spectrographic method was developed to determine potassium in specification analyses of ^{244}Cm oxide. The new method will replace the slower atomic absorption method. The product specifications allow a maximum of 0.1 wt % potassium in ^{244}Cm oxide; the spectrographic method is applicable between 0.0005 and 0.5 wt %, and can be extended. The precision is estimated to be -50 to +100%. The method is combined with previous spectrographic determinations of tantalum, zirconium, americium, and common metal impurities.

In the method, 10 mg of ^{244}Cm oxide and 50 mg of gallium oxide carrier are mixed and mounted on graphite electrodes. The arc spectra of duplicate samples are photographed on an infrared sensitive plate to record the potassium lines at 7664.9 and 7699.0 Å. Second order interference from curium and cyanogen, which have primary lines at 3830 and 3850 Å, is eliminated with a yellow filter. Spectra of a graded series of standards are recorded on the same plate, and the potassium is determined by visual comparison of the spectra. For ease of handling, neptunium oxide is used as a stand-in for curium in the standards.

Gamma Spectrometry of ^{243}Am and ^{244}Cm

A Ge(Li) X-ray detector system has been obtained and is being used routinely for the analysis of ^{243}Am in ^{244}Cm. This detector, a planar lithium-drifted wafer of germanium mounted behind a beryllium window, has much better resolution ($\sim$850 ev peak width at half-maximum at 75 kev) for low-energy gamma and X-rays than Ge(Li) detectors of conventional design. Other applications are being tested that require very high resolution at low photon energies.

Analysis of ^{243}Am in the presence of the much larger quantities of ^{244}Cm using the 74.7-kev ^{243}Am photopeak has been limited by the detector resolution and by the accuracy of the correction for the continuum under the photopeak. In samples containing ^{144}Ce, errors can occur in the integration of the 74.4-kev ^{243}Am photopeak due to the proximity of the 80.1-kev ^{144}Ce peak. With the very high resolution of the X-ray detector, these two peaks are well resolved; thus ^{243}Am may be determined in the presence of large amounts of ^{144}Ce.

Any nuclide with gamma rays above 74.7 kev may also interfere if the continuum counts from these gammas mask the photopeak, or if a gamma-emitting nuclide is present that perturbs the continuum distribution at the photopeak. The latter interference occurs in samples containing ^{144}Ce, ^{243}Cm, and ^{239}Np. Both of the latter nuclides decay into levels of ^{239}Pu and have similar gamma spectra, with a shoulder at the ^{243}Am photopeak. The higher resolution of the X-ray detector resolves the 74.7-kev peak more clearly on this shoulder and makes background correction more accurate.

[redacted]

Spark Source Mass Analysis of ^{244}Cm Oxide

Spark source mass spectrometry was extended for the first time to highly alpha active samples by the addition of a new interchangeable spark source and a small containment unit at the source housing, Figure 2. The spectrometer was used successfully to determine 70 elements from boron through americium simultaneously in ^{244}Cm oxide.

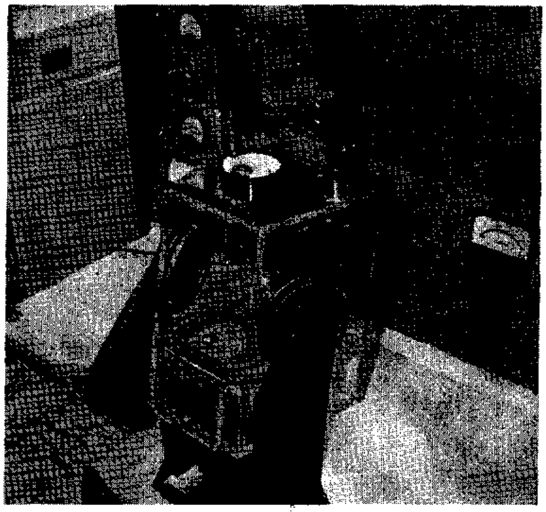


FIG. 2 SPARK SOURCE FOR ^{244}Cm OXIDE ANALYSIS

Sensitivity was 50 ppm or better; precision was estimated to be a factor of 3. The mass spectrometric analyses of ^{244}Cm oxide from the first production campaign are compared with emission spectrographic analyses in Table VI. For most elements, the mass analysis agrees with the more precise emission analysis.

Spark source mass spectrometry is well suited to highly radioactive samples. The method is so sensitive that it requires only microgram samples; thus, simple nonshielded containment of the ion source prevents exposure of personnel. The vaporized sample is deposited almost completely in the housing of the source; only a few picocuries reach the photoplate. No radioactivity was detected on photoplates after analysis of samples containing 10^{10} d/m of ^{244}Cm oxide.

The contained spark source and glove box on the mass spectrometer are shown in Figure 2. For more rapid analysis of nonradioactive samples, the source and glove box are replaced with a standard source in ~8 hr (including decontamination).

[redacted]

[redacted]

In the analyses, 5 to 30 µg of ^{244}Cm oxide is mounted on a pure gold electrode and sparked against a gold counter-electrode to produce a single-exposure spectrum and the spectra are interpreted by comparisons with graded spectra.

The few discrepancies between the mass and emission spectrographic analyses are being resolved, and the mass analysis is being extended to other radioactive compounds.

TABLE VI

Elements in ^{244}Cm Oxide, ppm

	Spark Source Mass Spectrometry	Emission Spectroscopy
C	5,000	-(a)
N	58,000(b)	-
F	<8	-
Na	31	500
Mg	340	50
Al	370	<25
Si	2,500(c)	75
P	130	-
S	440	-
Cl	630	-
K	53	40
Ca	55	25
Cr	140	200
Mn	8	25
Fe	170	400
Co	8	<50
Ni	120	50
Cu	≤26	<10
Zn	20	<250
Zr	7,500	7,500
Mo	<55	150
Pd	54	-
Ag	<30	<10
Cd	<50	<50
Sn	160	350
La	570	-
Ce	20	-
Pr	60	-
Nd	350	-
Ta	≤250	<100
Pb	110	100

(a) No emission measurement

(b) "Nitrox" analyzer shows ~1.0%

(c) Possible contamination from glass

[redacted]

MATERIALS DEVELOPMENT AND TECHNOLOGY

Thermal Stability of Mixed Americium-Curium Oxides

Thermogravimetric analysis and high temperature X-ray diffraction studies of mixed americium-curium oxides have demonstrated that only minor increases in thermal stability of CmO_2 can be achieved by addition of the more stable AmO_2. Stabilization of CmO_2 by other dioxide additives thus does not appear to be a useful means of avoiding effects of reoxidation of B-type Cm_2O_3 at low temperatures.(3)

Americium dioxide is less readily reduced on heating in oxidizing or inert atmospheres than is CmO_2. Temperatures above 850°C are required before significant loss of oxygen occurs from AmO_2, compared to 350°C for CmO_2.(4) Both oxides are readily reduced below 500°C in 4% H_2-He atmospheres.(4) The reduced oxides rapidly reoxidize above 150°C.(3)

The mixed oxide, $Am_{0.48}Cm_{0.52}O_2$, obtained by calcination of a coprecipitated oxalate in air, lost oxygen above about 450°C in oxygen or inert atmospheres to form compositions of variable oxygen content approximately midway between dioxide and sesquioxide compositions, as shown in Figure 3. The face-centered-cubic (fcc) structure of the mixed dioxide was converted to a disordered fcc

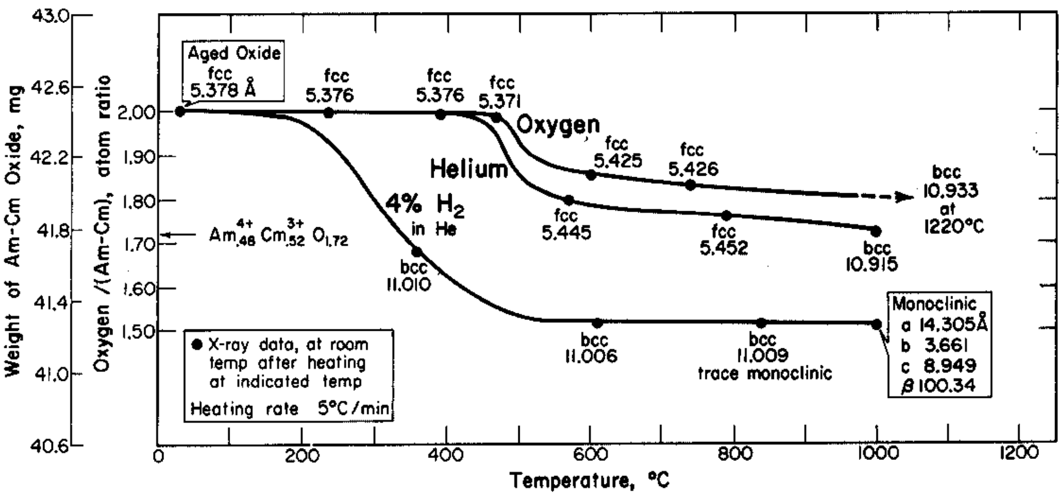


FIG. 3 THERMOGRAMS AND STRUCTURES OF Am-Cm DIOXIDES HEATED IN THREE ATMOSPHERES

phase by this decomposition. At temperatures above 1000°C in inert atmosphere a body-centered-cubic (bcc) (C-type) phase was produced. Formation of phases analogous to Cm_7O_{12} produced by CmO_2 decomposition in oxidizing or inert atmosphere was not observed. Heating in hydrogen-containing atmospheres reduced the mixed dioxides above 200°C and formed the bcc C-type phase with near sesquioxide composition above 500°C. Heating at 800-1000°C in this atmosphere converted the C-type oxide to monoclinic B-type sesquioxide of stoichiometric composition, which was retained on rapid cooling to low temperature. The B-type sesquioxide then exothermally reoxidized rapidly to the dioxide on heating above 150°C in oxygen or slowly (0.5 wt % per day) at room temperature.

FACILITY AND EQUIPMENT DEVELOPMENT

Alpha Materials Facility

The SRL Alpha Materials Facility (AMF) is being extended and modified to provide facilities for small-scale development of ^{244}Cm fuel formation and encapsulation processes. The extended facility will provide space under inert gas cover for hot pressing of small (0.25 g) ^{244}Cm fuel compacts, sealing into capsules by TIG welding, leak testing the capsules, and heating for long times to establish capsule integrity and materials behavior.

Installation of services, particularly piping and controls for the recirculating inert gas system, is continuing. The inert gas purification plant, Figure 4, is undergoing final testing at the vendors, with delivery now scheduled for February 1959. An off-gas exhaust system for the containment boxes has been constructed and installed. Equipment for continuous handling of oxygen and water in the inert gas cover has been constructed in preparation for installation in the next quarter.

Procurement, development, and testing of equipment for the extension proceeded. A 2-watt calorimeter was constructed and calibrated; modification of the design to provide capability down to 0.2 watts is in progress. Construction of a resistance-heated hot press is scheduled to begin in the near future.

A quadrupole mass spectrometer was installed with Knudsen cell apparatus in a presently operating Alpha Materials Facility glove box, to identify and measure curium oxide vapor species.

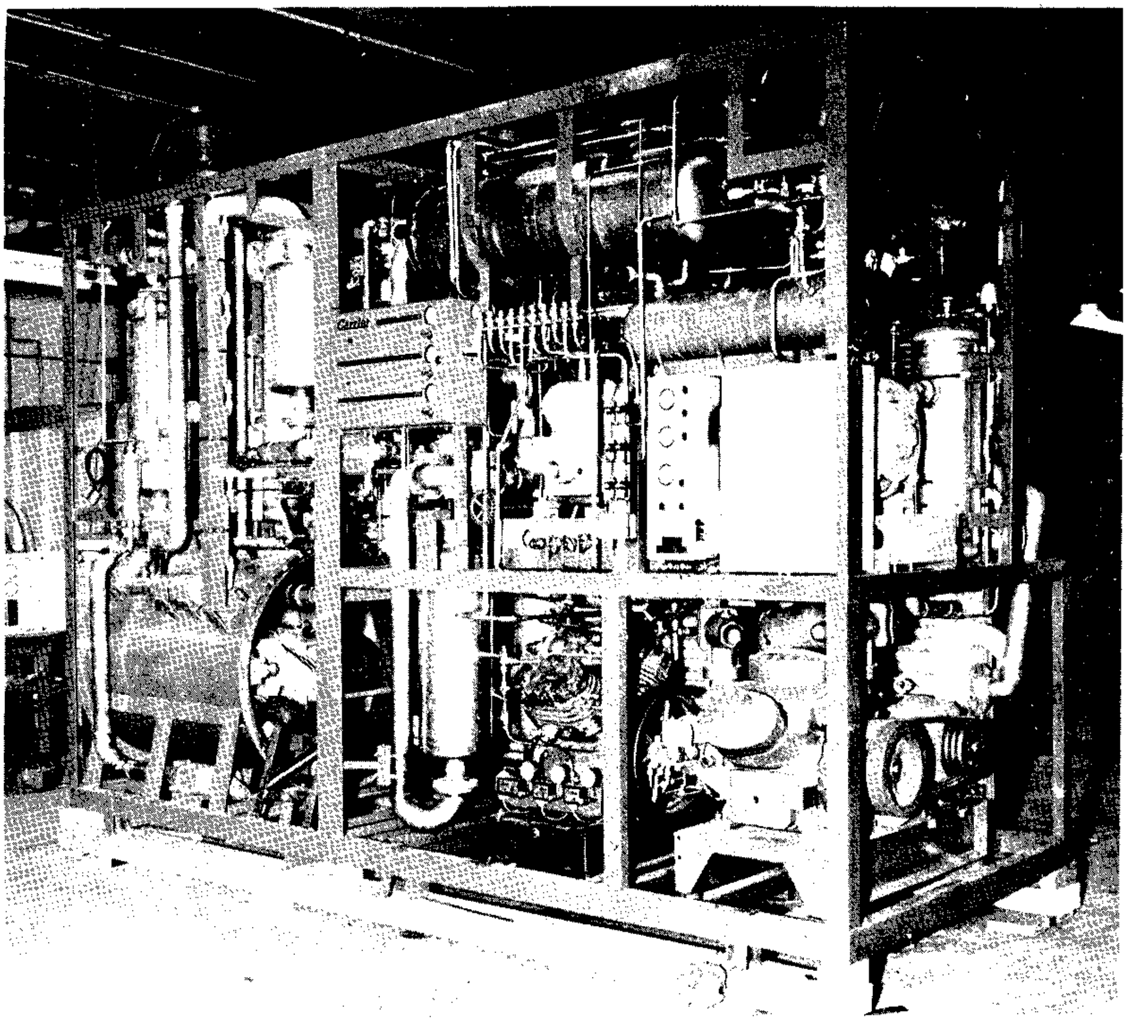


FIG. 4 HELIUM PURIFICATION AND RECIRCULATION PLANT FOR ALPHA MATERIALS FACILITY EXTENSION

[redacted]

PLUTONIUM-238

The Savannah River Plant (SRP) is the principal source of ^{238}Pu, a well established heat source isotope. ^{238}Pu, which decays by alpha emission with a half-life of 87.4 yr, generates 0.56 w/g. The isotope can be utilized as a heat source to operate in thermoelectric and thermodynamic conversion systems for space power and biomedical applications. Use in the biomedical applications requires a high purity ^{238}Pu with a ^{236}Pu content as low as 0.1 ppm.$^{(5)}$

SRP facilities provide the capability for fabrication and irradiation of ^{237}Np targets to produce ^{238}Pu, and to process irradiated neptunium to recover unconverted ^{237}Np for recycle and product ^{238}Pu for further reprocessing elsewhere, principally Mound Laboratory. Production studies and characteristics of the product ^{238}Pu are subjects of major interest in SRL programs.

PRODUCTION TECHNOLOGY

Irradiation of Shippingport Americium to Produce High-Purity ^{238}Pu

Conditions were designed for making ~300 g of ^{238}Pu containing <20 ppb of ^{236}Pu from 600 g of Shippingport americium. The isotopic composition of the americium, separated as oxide at Hanford, is typical of americium that will be produced in power reactors in the early 1970s:

^{241}Am	77.57%
^{242m}Am	0.34%
^{243}Am	22.2%

^{241}Am is a unique target for production of ^{238}Pu with very low ^{236}Pu content: ^{241}Am (n,γ) ^{242}Am (β) ^{242}Cm (α) ^{238}Pu. Low ^{236}Pu is essential in such applications as implantable sources of power for artificial hearts.$^{(5)}$

The program would verify process concepts for target fabrication, irradiation, and separation, and would produce the ~300 g of ^{238}Pu (associated with ~75 g of ^{239}Pu) for study and testing.

The initial americium oxide would be compacted with aluminum powder in ~60 aluminum-clad slugs for irradiation in an SRP reactor. The ^{242}Cm formed would transform to ^{238}Pu during 18 to 24 months of cooling, and the ^{238}Pu would be separated and converted to oxide in the plant.

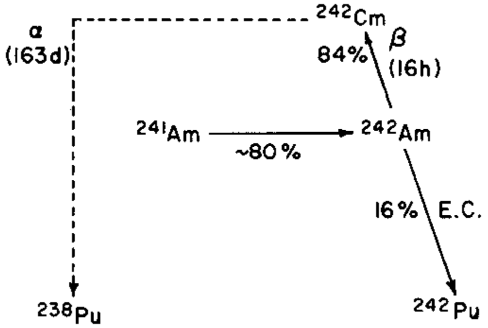
Residual americium and curium (totaling ~200 g) would be purified in the laboratory, converted to oxide, and compacted for further irradiation.

[redacted]

Resonance Production of ^{238}Pu and ^{252}Cf from ^{241}Am

Resonance reactor production of ^{238}Pu and ^{252}Cf from ^{241}Am-^{243}Am mixtures was calculated as a method to accelerate the growth in production of ^{252}Cf. During the early years of a proposed resonance reactor operation, production of ^{252}Cf will be limited by the scarcity of target material in spent fuel from power reactors. The mixtures of americium and curium in such fuel will contain ~90% americium, in which the ^{241}Am/^{243}Am ratio is ~3 during the early years of resonance reactor operation and approaches unity in later years if plutonium is recycled in the power reactors.

^{241}Am causes problems because its capture product ^{242}Am decays to ^{242}Cm, which, because of its high specific alpha activity, limits procedures for separation and fabrication:



Consequently, when the power reactor supply of ^{243}Am reaches the capacity of the resonance reactor, the preferred operation will be to preburn the ^{241}Am-^{243}Am mixture in a thermal reactor, where the ^{241}Am would burn preferentially. However, in the initial resonance operation, such preburning and associated processing would involve a holdup of ~2 yr prior to resonance irradiation of the ^{243}Am.

In the most attractive mode for the early years of resonance operation, the ^{241}Am-^{243}Am mixtures would be irradiated directly in the resonance reactor, in which ^{241}Am and ^{243}Am burn at approximately the same rate (~90% per year). A preliminary schedule for target management was calculated for a californium production facility (resonance reactor, separation, and fabrication). This schedule provides essentially for in-reactor cooling of the ^{242}Cm by extending the initial exposure to 18 months.

In addition to prompt resonance irradiation of ^{243}Am to produce ^{252}Cf, the resonance irradiation of ^{241}Am would provide a valuable second product in ^{238}Pu from decay of ^{242}Cm. This ^{238}Pu would be of high purity -- estimated to contain less than 0.02 ppm ^{236}Pu-- and would be produced at >10 kg/yr in the resonance reactor. These calculations have been completed.

[redacted]

REFERENCES

1. I. D. Eubanks and G. A. Burney. *Curium Process Development I. General Process Description*, USAEC Report DP-1009, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S.C. (1966).

2. H. S. Hilborn, (Comp.) *Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, April-June 1968, Pt II, Curium-244, Plutonium-238, Thulium-170 and 171*, USAEC Report DP-1169-II, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968) (Confidential).

3. H. S. Hilborn, (Comp.) *Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, January-March 1968, Pt II, Curium-244 and Thulium-170*, USAEC Report DP-1155-II, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968) (Confidential).

4. H. S. Hilborn, (Comp.) *Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Report, October-December 1968, Pt II, Curium-244, Plutonium-238, Polonium-210, and Thulium-170*, USAEC Report DP-1143-II, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1968) (Confidential).

5. E. J. Hennelly, "Production of Biomedical-Grade ^{238}Pu." *Trans. Am. Nucl. Soc.*, *11*, 456 (1968).

[redacted]

UNCLASSIFIED

[redacted]

PREVIOUS QUARTERLY PROGRESS REPORTS ISSUED INCLUDE:

DP-1088
DP-1094
DP-1105-I and -1105-II
DP-1120-I and -1120-II
DP-1129-I and -1129-II
DP-1143-I and -1143-II
DP-1155-I and -1155-II
DP-1169-I and -1169-II
DP-1177-I and -1177-II

[redacted]

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