TOP DRAFT**MASTER**

DO NOT TYPE ON THIS LINE

CONF - 800814 - - 19 (Draft)

Start typing — 50
on this line.Do Not Type
Past this
LinePREPARATION OF CURIUM-AMERICIUM OXIDE MICROSPHERES
BY RESIN-BEAD LOADING*F. R. Chattin, D. E. Benker, M. H. Lloyd, P. B. Orr,
R. G. Ross and J. T. WigginsStart First — 35
Page Here.Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37830

DISCLAIMER

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

To be presented at the Symposium on "Industrial-Scale Production - Separation - Recovery of Transplutonium Elements,"
2nd Chemical Congress, San Francisco, August 24-29, 1980

By acceptance of this article, the
publisher or recipient acknowledges
the U.S. Government's right to
retain a nonexclusive, royalty-free
license in and to any copyright
covering the article.

REPRODUCTION OF THIS DOCUMENT IS UNLIMITED

*Research sponsored by the Office of Basic Energy Sciences,
U. S. Department of Energy, under contract W-7405-eng-26
with the Union Carbide Corporation.

Do Not Type — 1
Below This
Line.

DO NOT TYPE ON THIS LINE

Start typing _____
on this line.

50

PREPARATION OF CURIUM-AMERICIUM OXIDE MICROSPHERES
BY RESIN-BEAD LOADINGF. R. Chattin, D. E. Benker, M. H. Lloyd, P. B. Orr,
R. G. Ross and J. T. WigginsDo Not Type
Past this
LineStart First _____
Page Here.

35

A resin-bead loading and calcination technique developed at the Transuranium Processing Plant (TRU) of the Oak Ridge National Laboratory (ORNL) is used routinely for producing uniform particles of curium-ameridium oxide in the size range desired for the fabrication of targets for irradiation in the High Flux Isotope Reactor (HFIR). TRU is the storage, production, and distribution center for the heavy-element research program of the U. S. Department of Energy. Target rods are remotely fabricated at TRU, irradiated in the HFIR, and then processed chemically at TRU for the separation and purification of the heavy actinide elements. Berkelium, californium, einsteinium, and fermium are distributed to researchers. Unburned curium and americium that is recovered during the chemical processing is refabricated into targets for additional irradiation. About 200 g of curium-ameridium oxide is produced annually for use in fabricating HFIR targets. Detailed descriptions of TRU and the overall production program

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 have been published previously (1,2,3,4,5) and a summary of the program is presented in another paper at this symposium (6).

A HFIR target is a 9.4-mm-diam by 0.89-m-long aluminum rod encased in a cylindrical aluminum tube which serves to channel the flow of cooling water in the reactor. The central target rod contains some end hardware plus thirty-five 6.3-mm-diam by 14-mm-long pellets that contain a blend of curium-ameridium oxide and aluminum powder encased in an aluminum jacket. A pellet is made by placing a weighed amount of blended actinide oxide-aluminum powder in a liner of aluminum tubing that has had one end closed with aluminum powder pressed to theoretical density. Then clean aluminum powder is added to form a top cap and the pellet is cold pressed in a pellet die.

The size of the curium-ameridium oxide particles is an important criterion in the production of HFIR targets. The oxide particles must be uniformly dispersed in the aluminum pellets and the pressed pellets must have a continuous aluminum phase to assure adequate heat transfer during irradiation. When oxide particles at some size smaller than 10 μm in diameter are mixed with -325 mesh (<44 μm) aluminum powder and pressed into pellets, the oxide phase may be continuous and the thermal conductivity low. With oxide particles ranging from 20 μm to 210 μm in diameter, the aluminum phase is continuous and the thermal conductivity is satisfactory for irradiation in high neutron fluxes. The resin-bead loading and calcination

Do Not Type
Below This
Line.

1

<R1
Do Not Type
Past this
Line <R2

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50 technique allows this criterion to be met easily even though remote operation is necessary for curium oxide production.

Do Not Type
Past this
Line

In the following sections, a brief chronology of the development of this process is presented; the materials, equipment, and basic operations relating to the resin-bead loading and calcination method of producing sized curium-ameridium oxide microspheres at TRU are described; and typical production data are presented.

Start First
Page Here.

35 Process Development

During the mid-1960's, the feed material for the HFIR targets was ^{242}Pu . Plutonium oxide was prepared in glove box operations using hydroxide precipitation, calcination, and grinding (7). When curium became available for use in HFIR targets, a sol-gel method for preparing curium oxide remotely was developed and was used from 1968 through 1970 (8).

<R1

<R8

However, the sol-gel process was not well-suited for the small batch sizes that are involved in the TRU program. Equipment startup and shutdown were a major part of the operating time. The processing was plagued with erratic operation, poor yields, and production of oversize oxide particles that required undesirable grinding and screening operations. A simpler, more reliable oxide preparation method that was adaptable to remote operation was needed.

Do Not Type
Below This
Line.

1 The resin-bead loading and calcination process was developed and evaluated during 1971 in a series of test runs in

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

Do Not Type
Past this
Line

Start First
Page Here.

50 which 150 g of curium oxide was successfully prepared and subsequently used in HFIR targets. Two resins, which required different process steps, were tested. The process using Dowex 50W resin, which contains sulfonic acid exchange groups, was selected for continued curium oxide production over a similar process using Amberlite IRC-50 resin, which contains carboxylic acid exchange groups. The Dowex 50W process (Fig. 1) requires fewer steps and is better suited for remote operation. In the
35 Dowex 50W process, pre-sized Dowex 50W-X8 resin beads are loaded to saturation from a dilute nitric acid solution of curium-ameridium. The resin is rinsed with water and then the resin matrix is destroyed by calcination to form dense actinide oxide microspheres. An early problem of inadequate sulfur removal at reasonably low calcination temperatures was resolved by reacting the loaded resin with hydrogen at 800°C. Curium-ameridium oxide for HFIR targets has been produced exclusively by this process since 1971. Until 1975, the production was accomplished in special production runs in the development equipment. In 1975, the present production-scale equipment was installed in the hot cells.

Materials

Feed Solutions. The process is applicable to both ameridium and curium, which are not usually separated from each other at TRU. The curium and ameridium recovered from the processing of irradiated HFIR targets is supplemented with

Do Not Type
Below This
Line.

<F>

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50 additional americium or curium obtained from other programs. The supplemental materials may be blended with the recycle material or processed separately. The curium and/or americium may be processed through a variety of purification steps, depending on the source of the material, to remove other actinides, rare earths, or ^{240}Pu , the decay daughter of ^{244}Cm . However, the final step before oxide production is always a double oxalate precipitation for removal of ionic contaminants and the product from that step is collected in strong HNO_3 in a tantalum-lined evaporator. All curium-americium solutions for a series of oxide production runs (typically containing 50-100 g of actinides) are collected, mixed, and sampled to determine the americium and curium isotopic compositions of the feed and ultimately the isotopic composition of the actinides in HFIR targets prepared from the oxide products.

Do Not Type
Past this
LineStart First
Page Here.

35

Resin Beads. The resin beads are prepared from commercially available Dowex 50W-X8, a sulfonic acid cation exchange resin. The 8% cross-linked resin was chosen because it does not shrink or expand too much in acid-water systems and because it is readily available. Resin cross-linkage had no detectable effect on the final oxide products in the development test runs. The resin is hydraulically classified to produce a size fraction that is predominately 60-80 μm in diameter. The classified resin is washed with 6 M HCl to

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50

remove contaminants such as iron and rinsed thoroughly with water.

Do Not Type
Past this
LineEquipment

The equipment for curium-ameridium oxide production (illustrated schematically in Fig. 2) is located on a rack within a master-slave manipulator-equipped hot cell in the TRU cell bank. The feed adjustment and raffinate collection vessels are located in a tank pit in another part of the cell bank. General design considerations and operating philosophies for chemical process operations at TRU have been described previously (4). Only equipment items that are unique to the curium-ameridium oxide production are discussed below.

LF2

Start First
Page Here.

35

Resin Loading Equipment. The feed is prepared in a 25-litre tantalum-lined evaporator. Small, run-size batches (about 22.5 g of actinides) of adjusted feed solution are transferred by vacuum to a 4-litre glass-pipe head tank on the equipment rack. The feed solution is then drawn through a quartz column containing the resin beads. A TRU pump is used to control the rate for loading the actinides on the resin. The effluent solution from the column passes through the pump and is discharged through an in-line alpha detector which is used to determine when curium breakthrough occurs. The effluent solution is collected in a 25-litre Zircaloy-2 raffinate tank until breakthrough occurs and is diverted back into the

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 tantalum-lined feed adjustment tank following breakthrough. In this way actinides in the column effluent can be conveniently recycled to the feed. This equipment arrangement facilitates the complete loading of resin and overcomes the problem experienced in the development equipment in which about 5% of the resin usually was not loaded because curium containing raffinates could not be recycled and curium in the raffinates would have been lost to rework.

Do Not Type
Past this
LineStart First —
Page Here.

35 The overall length of the quartz column is 0.4 m. The upper portion of the column has an inside diameter of 30 mm and is 0.25 m long. About 145 mL of resin can be supported on the quartz frit at the bottom of the upper section. The 0.15-m-long by 12-mm-diam lower portion is provided for access to the bottom of the quartz column when it is placed in the cylindrical furnace for the calcination of the resin. Both ends of the column have ground ball joints for connections to other equipment. For loading operations the column is clamped to the equipment rack and is connected with flexible polyethylene connections. The top of the column can either be connected to the feed head tank or be left open for adding resin and solutions directly to the column.

Calcination and Product Characterization Equipment. The resin drying, burning, sulfur removal, and oxidation steps are carried out in a cylindrical furnace built specifically for heating the quartz column. A Kanthal heating element is used

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50

to provide adequate furnace lifetime. The 63-mm-diam by 0.3-m-high firing cavity has a removable top and a 25-mm-diam access hole in the bottom. In the firing position the lower section of the column extends through the bottom of the furnace and a flexible polyethylene gas purge line is attached well away from the heat-affected region through a tempered glass tube extension. A quartz frit top is placed on the column to provide an exit path for the purge and combustion gases while preventing the resin from popping out like popcorn during the drying step.

Do Not Type
Past this
LineStart First
Page Here.

35

The sintering step (to 1050°C) is carried out in a laboratory muffle furnace with the curium-ameridium oxide contained in an open 50 mL platinum-lined Inconel crucible.

The oxide characterization steps are carried out with laboratory equipment that has been adapted as necessary to facilitate handling in cell with the manipulators. The items include a 210- μ m-opening stainless steel screen assembly, a 300-g-capacity triple-beam balance, a 10 mL glass graduated cylinder and assorted weigh pans, spatulas, and oxide containers. A remotely-operated calorimeter is used to assay the content of ^{244}Cm .

Processing Steps

Feed Adjustment. The tantalum-lined evaporator used to collect the actinide product solutions from a series of double

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50 oxalate precipitation runs also serves to adjust the composite product into a feed solution for a series of oxide production runs. Excess acid is removed by boiling the solution slowly to near dryness. The temperature is held at greater than 119°C for 5 hours or more during boiling to assure the destruction of all oxalates. After approximate dryness is reached, the evaporator is cooled, 0.01 M HNO₃ is added to dilute the actinides to less than 10 g/L (usually to 10-15 litres total volume) and a sample is taken to determine the acidity (typically 0.05-0.10 M) and verify that the actinides are still in solution. The adjustment is completed by an acid addition and evaporation to give an actinide concentration of about 10 g/L and a free acid concentration of 0.20-0.35 M at the final volume.

Start First
Page Here.

35 The acidity and actinide concentrations of the feed solution has to be readjusted between loading runs because the breakthrough raffinates and rinse solutions are recycled to the feed adjustment tank during the resin loading step and various solutions are added during the preparations for the next run of the series. When the quartz column is returned for the subsequent loading run, it contains a small amount (up to 200 mg) of curium-ameridium oxide on the frit. To prevent this material from being dissolved in the raffinate and lost, the column is given a preliminary leach with 250 mL of 0.25 M HNO₃ to the feed adjustment tank. Then, a new batch of resin is poured directly into the column as a slurry. The excess water is

Do Not Type
Past this
LineDo Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50

pumped to the feed adjustment tank. Finally, the top connection to the column is made and 200 mL of water is transferred through the system as the final test of the flow path in preparation for the next loading run. An accounting is kept of the acid and actinides recycled and the readjustment is made by evaporation between runs and dilution at the start of each loading run without repeating the original feed adjustment procedure and without resampling. The acid concentration is held in the required range but the curium concentration is allowed to decrease if necessary in the readjustments. For the final run of a series, the feed solution is diluted to about 10 litres with water and sampled to determine the exact acidity and the total curium-amerium remaining in the feed. The actinide content is used to size the final resin batch (7.7 mL of resin per gram of actinide plus some excess to prevent breakthrough) and the acidity is used to determine the final volume of feed. The feed solution is then evaporated to approximately 1 litre and transferred to the head tank. The feed is diluted in the feed head tank by a series of small water flushes that are made in the feed adjustment tank to flush out all of the feed solution. Six readjustments for a seven-run series have been handled in this manner without problems.

Do Not Type
Past this
LineStart First
Page Here.

35

Do Not Type
Below This
Line.

1

Resin Loading. The resin has a capacity for curium and americium of about 0.13 g/mL and 145 mL will therefore sorb

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50

about 19 g of actinides. An excess of adjusted feed (typically 2.25 litres) is transferred to the feed head tank, then drawn through the column at about 1.5 L/h (3.5 mL/cm²/min). The loading progress can be followed easily by darkening the hot cell and observing the orange-red glow from ²⁴⁴Cm if ²⁴⁴Cm

Do Not Type
Past this
Line

Start First
Page Here.

35

is present in sufficient quantity or less distinctly with the lights on by observing a dark ring of radiation damage on the resin bed which is located just above the loading front. The in-line alpha detector will detect curium breakthrough but is usually not relied on for the diversion of the raffinate stream when the curium glow can be seen distinctly. As the loading front nears the bottom of the resin bed, the effluent is diverted from the raffinate catch tank to the feed adjustment tank to collect the curium-amerium-containing raffinate and the resin washes, thus, eliminating loss of actinides from the feed system. The excess feed is transferred through the column and is followed by two small water flushes of the feed head tank and a final 200-mL water flush that is added directly to the top of the column. The resin is then allowed to dry for 10 minutes while the pump continues to pull air through the resin before the column is removed to the calcining furnace.

Raffinate batches from the individual loading runs are analyzed for ²⁴⁴Cm and usually can be discarded to waste.

Do Not Type
Below This
Line.

1

A final run of the series is made with as little as 3 g of curium-amerium remaining in the feed. For this partial size

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 run, the resin volume is sized with as much as 20 mL of excess resin to prevent curium-ameridium loading breakthrough. This change in the loading philosophy for the final run allows all the actinides in the feed solution to be converted to oxides and eliminates rework solutions from the feed system.

Do Not Type
Past this
Line

Start First —
Page Here.

35 Calcination. The quartz column containing the loaded resin is removed from the equipment rack and placed upright in the cylindrical furnace with the top frit in place. The resin is dried for one and one-half hours at 150°C without a purge through the bed to remove most of the remaining water which tends to bridge against an air flow and lift out the resin bed. After the initial drying period, the purge line is attached to the bottom of the column, air flow is established at 0.3 L/min (superficial velocity of 7 mm/sec) from outside the hot cell, and the resin is thoroughly dried in the air stream for one hour at 150°C. After the drying period, the temperature is increased to 800°C and held during a four hour period (including the heat up time) in the air stream to remove carbon. Then, sulfur is removed by changing (outside the hot cell) to a 0.3 L/min flow of 4% H₂-Ar purge gas while the temperature is maintained at 800°C for four hours. The purge is then changed back to air at 0.3 L/min and the heating is continued at 800°C for four additional hours to promote further oxidation.

Do Not Type
Below This
Line.

At the end of the firing schedule, the cylindrical furnace is cooled, the quartz column is removed, and the actinide oxide

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 microspheres are transferred by pouring into the platinum-lined crucible. The open-top crucible is placed in a muffle furnace and the oxide is sintered at about 1050°C for 16 hours.

Although there is no forced air flow during the sintering step, the approximately 10-mm-deep bed of oxide is exposed to air inside the furnace. After sintering is completed the oxide is cooled prior to the oxide characterization steps.

Do Not Type
Past this
LineStart First — 35
Page Here.

Curium-Americium Oxide Characterization. Large clusters of microspheres which have sintered together are broken apart and passed through a 210- μ m-opening, stainless steel screen using vibration and a sliding weight on the screen. The free-flowing, dust-free product, consisting of microspheres and small clusters of microspheres, is characterized by noting the color, weighing the oxide on a triple-beam laboratory balance and measuring the oxide volume ("tapped" for compaction) in a 10-mL glass graduate. A typical product which is predominately curium oxide, is usually a dark grey color indicating a mixture of CmO_2 (black) and Cm_2O_3 (white). Products which are predominately americium oxide have the same physical appearance and cannot be easily distinguished from curium oxide. The accepted stoichiometry of a typical curium oxide product is approximately $\text{CmO}_{1.71}$. The tap density obtained from the weight and volume measurements, is compared against a minimum standard of 2.0 g/mL as the key indicator of the acceptability of the oxide for target fabrication. The actinide oxide is placed in a

1

Do Not Type
Below This
Line.

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50 stainless steel storage container and the ^{244}Cm content (2.832 W/g) is measured using an in-cell calorimeter. The ^{244}Cm content, the curium and americium mass analyses from the feed, and the weight of oxide are used to calculate the product purity, usually in the range of 85-93% actinide oxide. The major impurity is believed to be unburned carbon since analyses for other possible impurities give minimal indications. A composite product sample made up of a small sample from each batch in a series of runs is taken for spark source mass spectrometric analysis and a weight loss on ignition test. The impurity analyses are used to verify that the calculated limits for heat production and neutron absorption in the HFIR are not exceeded.

Do Not Type
Past this
LineStart First
Page Here.

Process Variables

The free acid content of the feed solution is an important processing parameter. Free acidities in the range 0.01-0.70 M were investigated in the development tests. In each test the curium sorbed on the resin was sufficient to produce acceptable oxide products. However, the free acidity of the feed is maintained in the range 0.20 to 0.35 M in the production runs. In one of the earlier production runs at lower acidity, a precipitate formed in the feed solution. This was thought to be caused by an unknown contaminant, possibly phosphorus. In the production runs the reduced actinide capacity of the resin is noticeable at the higher acidities. Convenient batch sizes and short loading times for the current scale of production are

Do Not Type
Below This
Line.

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 achieved with actinide concentrations of about 10 g/L but it is not considered an important variable. When a choice has to be made as sometimes occurs when the breakthrough loading raffinates are recycled to the feed tank, the free acid is held in range and the actinide concentration is allowed to decrease. Process operation and product quality are unaffected.

Do Not Type
Past this
Line

Start First —
Page Here.

35 The sequence of drying and calcination steps is important to reliable operation and to the production of acceptable product. The initial air drying period without purge removes most of the free water and avoids operational difficulties. The drying at 150°C with air purge thoroughly dries the resin and avoids shattering the beads with internally-generated steam during subsequent heating. The calcination with an air purge at 800°C removes the carbon from the resin matrix. The sulfur content is then reduced to less than 1% during the 4% H₂-Ar purging at 800°C. This is a crucial step in producing oxide having adequate density and particle strength for use in fabricating HFIR targets. In test runs, the product tap densities were increased from 1.0 g/mL to values as high as 2.5 g/mL when 4% H₂-Ar was used. The calcination in air after the sulfur removal helps to oxidize Cm₂O₃ to CmO₂, a much more desirable product because of its higher density. (Americium is believed to have similar behavior.) Tap densities of greater than 2.9 g/mL are generally an indication of acceptable product for HFIR targets while lower tap densities, especially lower

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50

than 1.5 g/mL, indicate that the product will have unacceptably low crushing strength and will contain fines created from excessive particle breakage. The low density also makes it difficult or impossible to fit the desired amount of actinides into a target. Curium oxide with tap densities of 2.0-3.0 g/mL are estimated to be 40-60% of the theoretical crystal density by comparison with tap densities and mercury densities obtained for rare earth oxides which were made by comparable methods.

Start First
Page Here.

35

Curium oxide with a density of only 30-40% of the theoretical crystal density is barely adequate to obtain the desired curium loading in the target.

Curium oxide particles formed from large resin beads, 200-300 μm , show acceptable tap densities but have large internal voids. Such particles are more fragile than solid particles and are therefore less desirable. The voids decrease with decreasing resin size and do not exist in oxides made from smaller size resins of 50-150 μm diameter. Resin beads averaging 60-80 μm in diameter are now used to eliminate the internal voids and produce sintered oxide spheres (although not completely spherical) of about 20 μm diameter. Although this is on the low side of the desired range (20-210- μm -diam) spheres of this size sinter together at 1050°C and produce small clusters of spheres that are sized to less than 210 μm in diameter by screening.

Do Not Type
Below This
Line.

1

Typical tap densities from routine oxide production runs are 2.9-3.1 g/mL within a 2.7-3.6 g/mL range. This is an

Do Not Type
Past this
Line

DO NOT TYPE ON THIS LINE

Start typing —
on this line.

50 increase from the typical 2.3-2.4 g/mL within a 2.0-2.7 g/mL range experienced in the development test runs. This increase is not attributed to any one fundamental change in a process parameter but is believed to be a result of using a slightly smaller, more uniform size resin, loading all resin to saturation (to column breakthrough) and the ability to control and reproduce all run parameters in the present equipment.

Do Not Type
Past this
LineStart First — 35
Page Here.

Results

The production and oxide characterization data for a series of curium-ameridium oxide production runs are presented in Table I. The table includes the composite feed analyses, the product data for each run and a summary of the product data. Totals and averages are presented to indicate performance even though the products are not usually combined. Note that 93.7% of the feed material was converted into product. The normal losses of actinides from the product are the result of oxide particles that stick to the product handling equipment. These are not actual process losses because they are eventually returned to rework.

<TI

The analysis of the composite sample from the same series of curium-ameridium oxide production runs is presented in Table II. The content of carbon, the major impurity, is inferred rather than directly analyzed. The analyses of curium-ameridium oxide products generally reflect the purity of the

<TII

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50

Table I. Typical Curium-Americium Oxide Production Data

[Data from a series of runs made March 2-8, 1978]

Do Not Type
Past this
Line

Feed Composition

Curium: 66.45 g [37.4% ^{244}Cm , 0.4% ^{245}Cm ,
53.2% ^{246}Cm , 1.4% ^{247}Cm ,
and 7.6% ^{248}Cm]

Americium: 17.39 g [100% ^{243}Am]

Total Actinides: 83.84 g

Individual Run Data

Start First
Page Here.

35

Run No.	Actinide Losses to Loading Raffinate (mg)	Oxide Product Measurements and Observations						
		Color, Shade of Gray	Weight (g)	Vol. (mL)	Tap Density (g/mL)	^{244}Cm (g)	Actinide Oxide (%)	Sample Weight (g) ^a
54-CO-1	2.3	Med.	24.46	8.1	3.02	5.84	89.6	0.11
54-CO-2	5.7	Med.	23.54	7.6	3.10	5.58	88.9	0.11
54-CO-3	26.8	Dark	22.70	7.8	2.91	5.39	89.1	0.14
54-CO-4	18.2	Med.	22.93	8.6	2.67	5.30	86.7	0.12
54-CO-5 ^b	12.8	Lt.	5.09	1.9	2.68	1.18	87.0	0.09
TOTALS ^c (or weighted averages)	65.8		98.72 ^d	34.0	2.90	23.29 ^d	88.5	0.57 ^e

a

Samples are removed before product measurements are taken.

b

Partial batch as final run of the series.

c

Individual batches are usually kept separate.

d

Product yield is 93.7%.

e

Composite sample analyzed for impurities - see Table II.

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50

Table II. Typical Curium-Americium Oxide Impurity Levels
[Analysis of the composite sample from a series of runs
made March 2-8, 1978]

Do Not Type
Past this
Line

General Impurities ^a			Rare Earth Impurities ^a	
Element	ppm ^b	% ^b	Element	ppm ^b
C	c	10 ^d	Gd	450
S	4500	0.45	La	450
Zr	200		Ce	300
Fe	150		Eu	150
Ca	75		Nd	45
P	75		Sm	30
C	30		Pr	20
Pb	30		Tb	15
Al	15		Dy	15
Si	15		Other(total)	150
Na	10			
Ti	10			
Zn	10			
Other(total) 25				

a

Analysis by spark source mass spectrometer.

b

Based on total product weight.

c

Analysis not applicable for carbon.

d

Carbon content is inferred from sample weight loss during
ignition, typically 5-12%, and from the difference between
the gross impurity level and the total of other impurities.

Start First
Page Here.

35

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing
on this line.

50

feed, except for carbon and sulfur from the resin and a few potential corrosion products.

Do Not Type
Past this
LineSummaryStart First
Page Here.

35

Resin-bead loading and calcination techniques have been used to produce all curium and americium oxide feed material (about 2.2 kg) for HFIR targets since 1971. The process based on Dowex 50W resin has progressed from a series of test runs, through special production runs, into routine production in permanent equipment beginning in 1975. Key attributes of this process are its reliability, high yields, and ease of operation. The process is well suited for remote operation in hot cells although some delicate handling of the oxide product is still necessary. Yields approaching 95% are routinely obtained and only one unacceptable product has been generated during routine production operations. There have been no problems in fabricating targets from this oxide or in the subsequent irradiation of these targets.

The present scale of production of 150-250 g/yr supplies the present need and is comparable with the level of other chemical process operations at TRU. Since the annual production is accomplished in two 8-12 day periods, there has been no reason to consider further scale-up of the process. However, the rate of production could easily be doubled by simply adding a second set of calcination equipment.

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing _____
on this line.50 Acknowledgement

This research was sponsored by the Office of Basic Energy Sciences, U. S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.

Literature Cited

1. Burch, W. D.; Arnold, E. D. Chetham-Strode, A.
Production of the Transuranium Elements," Nucl. Sci. Eng.,
17, 1963, 438-442. See also, the following ten articles
that describe details of the program.
2. King, L. J.; Matherne, J. L. "Containment of Radioactive
Material in the Transuranium Processing Plant," Proc.
14th Conf. Remote Syst. Technol., 1966, 21-27.
3. Bottenfield, B. F.; Hahs, C. A.; Hannon, F. L.; McCarter,
R.; Peishel, F. L. "Remote Maintenance Systems in the
Transuranium Processing Plant," Proc. 14th Conf. Remote
Syst. Technol., 1966, 172-184.
4. Chattin, F. R.; King, L. J.; Peishel, F. L. "Remote
Processing Practices and Equipment in the Transuranium
Processing Plant After Ten Years of Operation," Proc.
24th Conf. Remote Syst. Technol., 1976, 118-129.
5. Collins, E. D.; Bigelow, J. E. "Chemical Process
Engineering in the Transuranium Processing Plant," 24th
Conf. Remote Syst. Technol. 1976, 130-139.

Do Not Type
Past this
LineStart First _____
Page Here.

35

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing _____
on this line.

50

6. King, L. J.; Bigelow, J. E.; Collins, E. D. "Experience in the Separation and Purification of Transplutonium Elements in the Transuranium Processing Plant at the Oak Ridge National Laboratory," Industrial-Scale Production-Separation-Recovery of Transplutonium Elements," ACS Symposium 2nd Chem. Congress North American Continent, 1980.

Do Not Type
Past this
Line

Start First _____
Page Here.

35

7. Sease, J. D. "The Fabrication of Target Elements for the High-Flux Isotope Reactor," ORNL-TM-1712, 1967.

8. Burch, W. D.; Bigelow, J. E.; King, L. J. "Transuranium Processing Plant Semiannual Report of Production, Status, and Plans for Period Ending June 30, 1968," ORNL-4376, 1970.

Do Not Type
Below This
Line.

1

DO NOT TYPE ON THIS LINE

Start typing — 50
on this line.

FIGURE TITLES

Do Not Type
Past this
Line

Figure 1. Curium-ameridium oxide microsphere production by
Dowex 50W-X8 resin-bead loading and calcination

Figure 2. Schematic arrangement of equipment for the
production of curium-ameridium oxide microspheres

Start First — 35
Page Here.

Do Not Type — 1
Below This
Line.

Figure 1. Curium-ameridium oxide microsphere production by Dowex 50W-X8 resin-bead loading and calcination

Figure 2. Schematic arrangement of equipment for the production of curium-ameracium oxide microspheres