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**THE SEPARATION  
AND PURIFICATION OF AMERICIUM  
AND CURIUM ON THE MULTIGRAMME SCALE**

by

K. BUIJS, W. MULLER, J. REUL and J.Cl. TOUSSAINT

1973



Joint Nuclear Research Centre  
Karlsruhe Establishment - Germany

European Institute for Transuranium Elements

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During a five year period, different techniques, including precipitation, ion-exchange and extraction chromatography, were applied to the purification of various gramme size samples containing  $^{241}\text{Am}$  and also  $^{242}\text{Cm}$ . All of these techniques together were put to use for the separation of 6 g of  $^{244}\text{Cm}$  from 5 g

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## ABSTRACT

Experience obtained in separation and purification of transplutonium elements is discussed.

During a five year period, different techniques, including precipitation, ion-exchange and extraction chromatography, were applied to the purification of various gramme size samples containing  $^{241}\text{Am}$  and also  $^{242}\text{Cm}$ . All of these techniques together were put to use for the separation of 6 g of  $^{244}\text{Cm}$  from 5 g of  $^{243}\text{Am}$ , followed by the purification of each nuclide. The separation was carried out by precipitation of  $\text{K}_3\text{AmO}_2(\text{CO}_3)_2$  from  $\text{K}_2\text{CO}_3$  medium with ozone. Final purifications were achieved by extraction chromatography (Aliquat-336 and HDEHP), cation-exchange and oxalate precipitation.

## KEY WORDS

PRECIPITATION  
PURIFICATION  
AMERICIUM 241  
CURIUM 242  
EXTRACTION CHROMATOGRAPHY  
ION EXCHANGE  
OZONE

## 1. Introduction

At the European Institute for Transuranium Elements, the Transplutonium Programme is aimed mainly at the preparation of selected compounds (including metals) and the study of their physical and physico-chemical properties. Many of these studies (e.g. those concerning electrical and magnetic properties) require samples consisting of several hundred milligrammes of high purity material. At the present time, the elements americium and curium are available in sufficient amounts, so that our attention is centered on them. Of prime importance for such a programme are the procedures used for purification of the materials and for their recovery and recycling. These separations and purifications are different in character from mere production operations, because their smaller scale and the variety of their starting materials preclude the application of semi-automated procedures, such as liquid-liquid extraction with mixer-settlers. On the other hand, our separations and purifications differ from microscale preparations as regards the effects caused by intense radiation. High levels of alpha-radiation pose problems of radiolysis and alpha erosion and require stringent measures for the prevention of contamination. The shielding for protection against neutrons and gamma-rays puts an additional burden on the operator. Thus, the gramme-scale purifications present additional problems in comparison to both bigger and smaller scale operations. In this paper a review will be presented of the techniques used for these various gramme-scale operations and the experience gained with them. Finally, a description is given of the same techniques as applied to the purification of 6 g of  $^{244}\text{Cm}$ , starting from a mixture of this nuclide with an equal quantity of  $^{243}\text{Am}$  and with some fission products.

## 2. Precipitation of Potassium Americyl Carbonate

### 2.1. General features

The pronounced differences in oxidation-reduction behaviour between americium and curium, which are observed in aqueous solutions, suggest immediate applications for the separation of

these two elements. The double carbonate of potassium and americium(V), which is insoluble in concentrated carbonate solutions [1] is especially suited for this purpose and, therefore, was used already in the early era of americium research [2]. Precipitation of this double carbonate, which was analysed to be  $K_3AmO_2(CO_3)_2$  [3], leaves curium in solution. The effect of radiolysis on the separation will be less than in an opposite case when curium would be precipitated (as e.g.  $CmF_3$ ). Moreover, the pentavalent state in alkaline medium is by far the most stable higher oxidation state of americium.

A detailed study of this precipitation of gramme quantities of americium was made by Burney [4], who examined oxidations with persulphate, hydrochlorite or ozone in 3.5 - 5 M  $K_2CO_3$  solutions at temperatures between 65 and 80°C. The solubility of  $K_3AmO_2(CO_3)_2$  in 3.5 M  $K_2CO_3$  was found to be 10 to 40 mg Am/l.

As we intended to develop a separation and purification scheme without recourse to hydrochloric acid as explained later, we discarded hypochlorite as a potential oxidant. Preliminary studies [5] indicated that the oxidation of americium in  $K_2CO_3$  solution by persulphate leads to coprecipitation of potassium salts other than  $K_3AmO_2(CO_3)_2$ , which may carry some curium and reduce the separation factor. Thus, in agreement with Burney, it was concluded that the most suitable oxidant is ozone, which does not introduce any new substance into the solution.

## 2.2 Handling of ozone in glove-boxes

Before taking up work with ozone in glove-boxes and hot cells we had to develop a safe method of disposing of it, in order to avoid attack of manipulator bootings, gloves and other objects. As it was not considered safe to pass a gas stream ( $O_2+O_3$ ) possibly carrying fine particles of high specific activity out of the alpha-containment, the excess ozone had to be destroyed before release of the oxygen into the atmosphere. The selective

reduction of ozone in an  $O_2-O_3$  mixture by reaction with aqueous reducing solutions is feasible only in not too strongly reduced solutions which do not react with oxygen. After unsuccessful attempts with  $H_2O_2$  with and without addition of heavy metals, a satisfactory method was found, which involved unbuffered KI-solutions. The KI concentration had to be kept under  $0.2 \text{ M}$  to avoid crystallisation of reaction products and blocking of the sintered glass filters of the wash bottles. Serious drawbacks of the ozone destruction in dilute aqueous solutions, however, are the big volumes of liquid waste that are produced and the frequent renewings of solution in the wash bottles. The possibility of catalytic ozone destruction, therefore, was looked into. A suitable catalyst was found to be "on carrier activated granular  $MnO_2$  for elementary analysis" produced by Merck, Darmstadt. Two drying towers filled with a total of 350 g of granulated  $MnO_2$ -catalyst were amply sufficient to deal with a 100 l/h stream of  $O_2-1.7 \% O_3$ . After 20 hours of operation no sign of decreased efficiency was visible. Adsorption of moisture reduces the efficiency of the catalyst; the gas stream was, therefore, passed through a reflux condenser directly after passage through the  $K_2CO_3$  solution.

### 2.3. Purification of Americium via $K_3AmO_2(CO_3)_2$

Before the application of the ozone precipitation to the separation of gramme quantities of  $^{243}Am$  and  $^{244}Cm$ , experience had been gathered at a couple of less complicated operations. A purification run for 7 grammes of  $AmO_2$  (Fig.1) gave the opportunity for studies of kinetics and of solubility. At  $80^\circ C$ ,  $O_2 - 1.7 \% O_3$  was passed at a rate of 100 l/h through 620 ml of  $3.5 \text{ M } K_2CO_3$  solution containing 9.2 g Am/l. The concentrations of Am(III) and total Am were measured at regular intervals using a slightly modified version of a standard spectrophotometric method [6]. The concentration of Am(III) decreases exponentially with a half value time of 41 minutes, the concentration

of Am(V) is about 200 mg/l during the whole ozonisation. The reaction may, thus, be considered complete after 7 hours; in practice a safety margin of another 2-3 hours was added. After cooling, the filtrate contained 40 mg Am/l, a value which agrees with those given by Burney [4].

#### 2.4. Separation of Curium-242 from Americium-241 via $K_3AmO_2(CO_3)_2$

The recovery of actinides from a spent isotopic power source [7] offered an occasion for testing the separation with macro amounts of nuclides, viz. 3.30 g of  $^{241}Am$ , 18 mg of  $^{242}Cm$  and 380 mg of  $^{238}Pu$ . The precipitate consisted of 3.15 g of Am, 1.5 mg of Cm and 323 mg of Pu. Prior to this precipitation run it was established that the preparation of a solution of actinides in 3.5 M  $K_2CO_3$  is best carried out by dissolving the hydroxides in a minimum of  $HNO_3$  and adding an appropriate amount of 5 M  $K_2CO_3$ . Direct reaction of the hydroxides with a carbonate solution leads to colloidal solutions which are not suitable for ozonisation. The actinides were recovered from the 3.5 M  $K_2CO_3$  filtrate by neutralisation with 11 M  $HNO_3$  followed by precipitation with 10 M KOH. The results obtained during separation and purification experiments were good enough to warrant the successful application of the precipitation method to the separation of multigramme amounts of americium and curium.

### 3. Extraction Chromatography

#### 3.1. General features

The advantage of extraction as a technique for carrying out separations lies in its high degree of adaptability to specific problems, since the composition of the two phases may be varied at will. Obviously then a technique which combines the versatility of extraction with the many successive equilibria occurring in a chromatography column is a very powerful tool for small-scale separations. In fact, the separation of transplutonium elements was one of the first applications of this technique, when it was developed some 10 years ago [8].

Kooi, at the first campaign in Western Europe for isolation of weighable quantities of transuranium elements from irradiated americium, used extraction chromatography for the final separations. He separated Cf and Bk from each other and from Am+Cm by sorption of the nuclides at 85 °C on columns of di-(2-ethylhexyl)phosphoric acid (HDEHP) from 0.1 M HCl followed by elution with HCl of concentrations increasing from 0.1 M to 2 M [9,10].

Important achievements in actinide separations research were the developments of an Am-Cm separation by extraction chromatography on a column containing a quaternary amine (Aliquat-336) [11,12] and the actinide-lanthanide separation by the TALSPEAK extraction method using HDEHP [13]. Extraction chromatography versions of the latter procedure were developed at the Institute for Transuranium Elements [14] and at the Institute for Radiochemistry [15], both at Karlsruhe.

### 3.2. Tricaprylmethylammonium (Aliquat-336) nitrate

#### 3.2.1. Preliminary measurements

Am-Cm separations at the milligramme scale [7,16] carried out by extraction chromatography proved promising for later gramme-scale applications. We compared Am-Cm separations by two methods: extraction chromatography [16] on a column of Aliquat-336 (20%) supported on kieselgur and anion exchange [17] on Dowex 1 x 8. The product solutions contained 5 mg of  $^{241}\text{Am}$  and 5 mg of  $^{244}\text{Cm}$ . The various parameters and results are listed in Table I.

#### 3.2.2. Partial purification of Americium-241 on an Aliquat-column

The application of extraction chromatography to gramme scale separations of Am and Cm requires operation of big columns. The operating characteristics of a big Aliquat-336 column could be studied at the occasion of the above mentioned campaign for the purification of 7 g of  $\text{AmO}_2$  (Fig.1). Preliminary experiments had shown that transition metals (Fe, Cr, Ni) are not retained on an Aliquat-336 column in 8 M  $\text{LiNO}_3$  in contrast to Pu(IV) and Ce(IV), which in the presence of  $\text{KBrO}_3$  stay fixed

also when Am is being eluted with 1 M HNO<sub>3</sub> [18]. The various data pertaining to this column run which are shown in Table II, indicate that the operation presented no major difficulties. A detail, however, which merits some consideration, is the preparation of the feed solution. Partial neutralisation of an 8 M HNO<sub>3</sub> solution of Am with solid Li<sub>2</sub>CO<sub>3</sub> proved very cumbersome in practice: dissolution of Am(OH)<sub>3</sub> in a minute amount of HNO<sub>3</sub> followed by addition of 8 M LiNO<sub>3</sub> - 0.01 M HNO<sub>3</sub> was far more efficient.

### 3.3. Di-(2-ethylhexyl)phosphoric acid

#### 3.3.1. Preliminary measurements

The actinide-lanthanide separations with HDEHP columns are based on the combined effects of differences in extraction coefficients and in degrees of complex formation between trivalent actinides and lanthanides. As a complex former, generally, diethylenetriaminepentaacetic acid (DTPA) is used. The solubility of the trivalent ions under the conditions of the separations is assured by the addition of lactic acid. Thus, a typical TALSPEAK-like separation involves feed and washing solutions which are 1 M in lactic acid at pH = 2.8, followed by a solution of 1 M lactic acid - 0.05 M DTPA at pH = 2.8 which elutes the actinides first.

In view of the fact that HDEHP acts as a strong cation exchanger and generally extracts very efficiently at pH values between 2 and 3, it seemed interesting to test the feasibility of various types of feed solution. The Cm-containing eluant solution of an Aliquat-column is 4 M LiNO<sub>3</sub> - 0.01 M HNO<sub>3</sub>. Experiments with <sup>241</sup>Am and <sup>152</sup>Eu have shown that such a solution, after a slight adjustment of the pH, can directly be used as feed solution for an HDEHP column. Similarly, a feed solution of 0.3 M Al(NO<sub>3</sub>)<sub>3</sub>, 0.6 M citric acid and 4 M NH<sub>4</sub>NO<sub>3</sub> (pH = 2.8), which might be obtained by acid dissolution of Al-actinide cermet, does not interfere with the separation. In this case, citric acid is added to keep Al in solution.

### 3.2.2. Purification of 25 g of $\text{AmO}_2$ on one HDEHP column

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After the successful purification of 6 g of Cm on a HDEHP column, described later in this paper, it was decided to use the same type of column for a separation of  $^{241}\text{Am}$  at a larger scale. In preliminary experiments it had been established, that the elution speed of trivalent actinides from the HDEHP column with lactic acid-DTPA solutions depends very strongly on the pH. At pH = 2.8 elution proceeds fairly rapidly, whereas at pH = 2.0 the actinides stay practically fixed in their position on the column. In contrast, Pb, Mn and Zn are eluted both at pH = 2.0 and at pH = 2.8, and Fe is not eluted at either pH. Consequently, an efficient purification of Am may be achieved by an extra washing at pH = 2 in presence of DTPA. On this basis a campaign for the purification of about 30 g of impure  $\text{AmO}_2$  was undertaken with a 3.7 l extraction chromatography column. Data relevant to this purification are shown in Table III. The big quantity of  $^{241}\text{Am}$  involved in this purification made it possible to discern its position on the column by observing its radioluminescence. The complete column run took 60 hours. The purity of the final product after precipitation and calcination was better than 99.7 %.

## 4. Ion Exchange

### 4.1 General remarks

The separation technique based on ion exchange, both cation and anion exchange, has been introduced into transplutonium chemistry and processing at an early stage. The most prominent applications of cation exchange include columns eluted with HCl [19,20] or ethanol-HCl [21] for general clean-up and for lanthanide-actinide separations, and those eluted with alpha-hydroxyisobutyric acid [22,23] for interactinide separations. Anion exchange columns eluted with 10 M LiCl [24] serve for lanthanide-actinide separations, whereas in HCl or  $\text{HNO}_3$  medium Pu may be isolated.

#### 4.2. Processing of irradiated Americium by ion-exchange

At the time of the transuranium processing campaign by Kooi and co-workers [9], the best studied small-scale separation techniques involved ion exchange. Therefore, they applied such techniques for their separations involving about 250 mg of heavy actinides (including microgramme quantities of Cf) and about 2 g of fission products. More recently, gramme quantities of irradiated  $^{241}\text{Am}$  have been treated [25], following a flow-sheet (Fig.2), which is essentially the same as that used by Kooi et al. At the start of this separation, the irradiated  $^{241}\text{Am}$ , which originally was incorporated in cermet with Al, consisted of about 6 g of Pu-isotopes, 4 g of  $^{241}\text{Am}$ , 800 mg of  $^{242}\text{Cm}$ , and fission products with a gamma dose rate at contact of 35,000 Rem/h. After dissolution and removal of Al by precipitation methods, the Am-Cm fraction was isolated by anion exchange.

A first anion exchange column served to separate Pu and some fission products (e.g. Zr, Ru). Its operating characteristics are given in Table IV. A good separation was obtained, although the column seemed literally burned by the high activity which passed through it. Part of the darkening, however, may be attributed to the presence of Ru-nitrato-complexes which were irreversibly retained on the column. A second anion exchange column was used to separate the remaining actinides from the lanthanide fission products in LiCl-medium. At each run, only half of the product from the HCl-column was processed on the LiCl-column. Data on its operation are presented in Table V. During the elution of lanthanides, the separation was clearly visible by the occurrence of two differently coloured zones of radioluminescence. The high degree of decontamination from fission products is seen in Fig. 3.

#### 4.3. Purification of $\text{AmO}_2$ by ion exchange

Before the introduction of extraction chromatography for the purification of  $^{241}\text{Am}$ , up to 5 g quantities of this nuclide

were treated by ion exchange. The flowsheet of this procedure is shown in Fig.4 [26]. Characteristic parameters and results for the thiocyanate anion-exchange column, which is the cornerstone of the purification, are shown in Table VI. Especially these thiocyanate ion exchange columns gave rise to problems as the combined actions on the thiocyanate ions by alpha-radiation and HCl resulted in considerable precipitates of sulphur, which were very difficult to eliminate. After oxalate precipitation and calcination 75 % of the Am were obtained with a purity of 99.8 %. The main loss of Am is due to retardation in the HCl anion exchange column.

## 5. General Conclusions Based on Practical Experience

### 5.1. Advantage of nitrate medium.

Because of the favourable dissolving properties of HCl and the relatively high complexing power of the chloride ion, the greater part of early ion exchange separations were carried out in chloride media. The practical consequences of this frequent use of hydrochloric acid or chlorides are, however:

- heavy corrosion of metal equipment
- strong radiolytic decomposition of solutions containing high levels of alpha activity; the resulting evolution of Cl<sub>2</sub>-gas then enhances the corrosion,
- risk of inclusion of chloride ions in the calcined purified product.

These drawbacks are sufficient reason to try to abandon chloride media in favour of nitrate media.

### 5.2. Advantage of extraction chromatography

As indicated in the foregoing section, the preference for nitrate media in high-activity separations is sometimes hard to reconcile with application of ion exchange techniques. The greater flexibility of extraction chromatography, however, makes it much easier to adapt separation procedures to nitrate media.

A second advantage of extraction chromatography columns over ion exchange columns is their constant free column volume, which is independent of high salt or acid concentrations (no swelling).

Finally, the lower capacity per unit of volume of extraction chromatography columns as compared to ion exchange columns diminishes the risk of burning the columns with high alpha-activities. Explosion risks are eliminated by using an inorganic support material.

## 6. Characterization of the Americium-243/Curium-244 Mixture

The experience described in the foregoing sections was applied to the selection and execution of a procedure for the separation and purification of a 6 gramme  $^{244}\text{Cm}$  sample. This  $^{244}\text{Cm}$  was contained in a mixture of  $^{243}\text{Am}$  and  $^{244}\text{Cm}$  which had been obtained from the U.S. Atomic Energy Commission. It was intended to separate  $^{244}\text{Cm}$  from  $^{243}\text{Am}$  and then purify it, in order to obtain the starting material for carrying out the research programme indicated in the introduction.

### 6.1. Composition of the Americium-243/Curium-244 mixture

The main components of the mixture were:

- 5.93 g of Cm (94,16 %  $^{244}\text{Cm}$ , 484 Ci)
- 5.11 g of Am (99,81 %  $^{243}\text{Am}$ )

The principal "inactive" impurities were Pu (1.33 %), S (1.05 %), Fe (0.55 %), Nd (0.46 %), Al (0,27 %) and La (0.18 %).

As an active impurity 0.13 Ci of  $^{154}\text{Eu}$  should be mentioned.

### 6.2 Radiation intensity of the Americium-243/Curium-244 mixture

Radiation dose rates at a distance of 50 cm, which form the basis for radiation protection measures, were calculated from the data given in section 6.1. The results agreed with total dose rate measurements on part of the material. The following values were found:

$^{244}\text{Cm}$ , Neutrons:	200 mRem/h
$^{243}\text{Am}$ ( $^{239}\text{Np}$ ), 0.20 - 0.80 MeV $\gamma$	80 mRem/h
$^{154}\text{Eu}$ , 0.8 - 1.3 MeV $\gamma$	320 mRem/h

The half-value layers were found to be 5.2 cm of water for the neutrons and 7 cm of water or 1.5 mm of lead for the gamma-rays. It was concluded that working with telemanipulators behind a 25 cm watershielding and application of lead-shielded glassware would keep the radiation dose rates, to which the operators would be exposed, to a tolerable level. In fact, these dose rates were found to be 1 mRem/h and 3-6 mRem/h for neutrons and gamma rays, respectively.

#### 7. Equipment used for Purification of 6 g of Curium-244

As indicated in the foregoing section, the chemical operations were carried out behind 25 cm watershielding (Fig.5). At the backside, however, the 1.4 x 1.3 m<sup>2</sup> alpha-tight boxes were equipped with gloves, so that short time manual interventions were possible. These interventions, alleviated by the presence of paraffin blocks stacked in the rear part of the boxes, were very useful for carrying out quick repairs. Active materials could be stored temporarily in wells, welded to the bottoms of the boxes and surrounded by 2 cm of Pb and 15 cm of H<sub>2</sub>O.

All chemical operations were carried out in glassware except the final precipitation of Cm-oxalate, which was made in a polyethylene beaker, and its filtration which was done in a sintered platinum filter crucible. Tightness of glass joints between glass parts was assured with Teflon-covered O-rings. Filtrations were carried out with filter-candles featuring G 4 sintered glass filters. Liquids were transported through polyethylene tubing with detachable polyethylene joints. Over- and under-pressures were provided by a membrane pump placed inside the alpha-containment. Neutron or gamma activities of samples were monitored with rem-counters placed outside the boxes against the rear walls.

## 8. Flowsheet for the Purification of Curium-244

Fig. 6 shows the flowsheet which has been applied to obtain the Cm in a purified state. The general principles, as outlined in section 5 have been followed; maximum use of extraction chromatography and chemistry in nitrate medium. As in the case of the  $^{241}\text{Am}$  purification (2.2), the addition of  $\text{KBrO}_3$  to solutions passing the Aliquat-column serves to assure the retention of traces of Pu (and Ce).

The Dowex-50 cation exchange column which constitutes the last purification step could, in principle, be replaced by a second HDEHP column. However, Si-containing erosion products from the support material of such a HDEHP column might then be carried along into the final product. Therefore, an organic cation exchange column is better suited for this final purification step.

All changes of medium were made by hydroxide precipitation followed by redissolution in the desired medium.

## 9. Americium-Curium Separation

### 9.1. Dissolution of the oxide mixture

Without heating, the Am-Cm oxide dissolved in 11 M  $\text{HNO}_3$  within 2 hours. A change of medium to dilute  $\text{HNO}_3$  and addition of 5 M  $\text{K}_2\text{CO}_3$  gave a solution of the actinides in 3.5 M  $\text{K}_2\text{CO}_3$ , suited for precipitation of  $\text{K}_3\text{AmO}_2(\text{CO}_3)_2$ .

### 9.2. Precipitation of $\text{K}_3^{243}\text{AmO}_2(\text{CO}_3)_2$

According to the experience obtained before (section 2), ozone was passed through the 3.5 M  $\text{K}_2\text{CO}_3$  solution of  $^{243}\text{Am} + ^{244}\text{Cm}$  at 80 °C for 10 hours. During this operation, the solution grew more and more cloudy and foamed increasingly. After filtration, the crystalline precipitate was washed once with 3.5 M  $\text{K}_2\text{CO}_3$ . In 11 M  $\text{HNO}_3$  the precipitate dissolved giving a

clear green solution, which subsequently was reduced with hydroxylamine and converted to  $3.5 \text{ M K}_2\text{CO}_3$ . After a second ozonisation, the  $^{243}\text{Am}$  was put aside for later treatment. The  $^{244}\text{Cm}$  was precipitated from its  $\text{K}_2\text{CO}_3$  solution and stored as  $\text{Cm}(\text{OH})_3$  awaiting further purification by extraction chromatography. The first ozonisation precipitated 98.5 % of the total Am and 2.1 % of the Cm. After the second ozonisation and filtration a jelly-like precipitate in the filtrate formed, which contained some Cm. After addition of the jelly-like product to the total Am-fraction, the latter contained 0.4 % Cm. The Cm fraction contained 2.7 % of Am. Fission products  $^{144}\text{Ce}$  and  $^{154}\text{Eu}$  were detected only in the Cm-fraction.

## 10. Final Purification of Curium-244

### 10.1. Aliquat-336 column

The  $^{244}\text{Cm}$ -product of the Am-Cm separation stored as hydroxide, was washed and dissolved in the feed-solution for the Aliquat-336 column. Data relevant to the operation of this column are given in Table VII. Fig. 7 shows the elution diagram. An eluate fraction of 3.5 l containing 4.5 g of  $^{244}\text{Cm}$  with 0.28 % of  $^{243}\text{Am}$  was collected for further purification.

After sorption of Cm on the (white) column, its position was clearly visible as a dark zone, which coincided exactly with the region of intense radioluminescence. During the elution of the curium the grayish coloration advanced together with the activity moving down the column. This colour effect and the occurrence of yellowish bands originating from the presence of  $\text{KBrO}_3$ , were the only visible signs of the Cm-containing solutions passing the column.

### 10.2. HDEHP-column

The 3.5 l yellow  $3.5 \text{ M LiNO}_3$  eluate from the Aliquat-336 column present in different fractions with pH varying between 3.5 and 4.3, were brought to pH - 2.8 by addition of lactic acid and ammonia. The curium then was sorbed on the HDEHP-column. Data

regarding this column are presented in Table VIII. The elution diagram is shown in Fig. 8. At this sharp separation no loss of  $^{244}\text{Cm}$  occurred. The influence of the medium on the radio-luminescence was clearly shown by a considerable increase in intensity upon addition of lactic acid at  $\text{pH} = 2.8$ . Like the Aliquat-column the HDEHP-column showed only some change of colour as a result of the passage of the curium. The purification on the HDEHP-column took somewhat less time (32 h) than that on the Aliquat-336 column (42 h).

### 10.3 Cation exchange column

The 1750 ml of colourless eluate from the HDEHP-column were made 1 M in  $\text{HNO}_3$  in order to have a feed solution for the cation exchange final clean-up column. Table IX shows data characteristic of the operation of this column. The elution diagram is shown in Fig. 9. During sorption of  $^{244}\text{Cm}$  on the column, the latter considerably darkened. The fact that the dark zone extended further than the Cm-containing zone indicates that radio-lysis products carried along with feed and washing solutions, are partly responsible for the darkening. The structure of the column was severely disturbed by the development of big gas bubbles, so that considerable mixing of loaded and unloaded resin occurred. As the column only served for hold-up of Cm and not for chromatographic separation, the only consequence was a certain broadening of the elution peak.

### 11. Oxalate-precipitation and Calcination

The curium was precipitated as the hydroxide from the 1750 ml eluate of the cation exchange column and taken up in a small volume of dilute  $\text{HNO}_3$ . After addition of a solution of oxalic acid, Cm oxalate precipitated from 0.5 M  $\text{HNO}_3$  - 0.5 M oxalic acid. After filtration and drying in a Pt-filter crucible the oxalate was calcined under air at  $600^\circ\text{C}$  for 16 hours to  $\text{CmO}_2$ . Preliminary experiments with  $\text{AmO}_2$  had indicated these calcination conditions as the most favourable ones as regards complete destruction of the oxalate and reactivity of the resulting oxide.

## 12. Results and Conclusion

As a result of the purification, 4.8 g of purified  $^{244}\text{CmO}_2$  were obtained. The final product contained 0.28 %  $^{243}\text{Am}$ . An analysis by spark source mass spectrometry carried out at Oak Ridge National Laboratory indicates the possibility of 4000 ppm of Fe as the only inactive impurity.

A number of practical conclusions may be drawn from this Am-Cm separation:

- Considerable quantities of  $^{244}\text{Cm}$  can be handled in boxes with simple neutron shielding.
- Extraction chromatography with liter-size columns permits good separations and eliminates radiolysis problems.
- Routine work with ozone within alpha-enclosures may be done without risk, provided a leak-free gas system and efficient catalytic destruction such as with  $\text{MnO}_2$  are used.

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List of Figures

- Fig. 1 Purification of 7 g of  $\text{AmO}_2$  by precipitation and chromatography techniques
- Fig. 2 Processing of irradiated  $^{241}\text{Am}$
- Fig. 3 Gamma-spectra of product solutions before and after passage through 2 anion exchange columns during processing of irradiated  $^{241}\text{Am}$
- Fig. 4 Purification of 5 g of  $\text{AmO}_2$  by ion exchange
- Fig. 5 Watershielded cells used for  $^{244}\text{Cm}$  purification
- Fig. 6 Isolation and purification of 5.9 g of  $^{244}\text{Cm}$
- Fig. 7 Purification of 5.9 g of  $^{244}\text{Cm}$  on Aliquat-336 column;  $^{244}\text{Cm}$  concentration in eluate as function of volume eluted
- Fig. 8 Purification of 4.5 g of  $^{244}\text{Cm}$  on HDEHP-column. Neutron and gamma dose rates (at 12 cm) per 250 ml fraction as function of volume eluted
- Fig. 9 Purification of 4.5 g of  $^{244}\text{Cm}$  on cation exchange column. Neutron dose rate (at 12 cm) per 250 ml fraction as function of volume eluted

	Aliquat-336 (20 % on Kieselgur)	Dowex 1 x 8
Column diameter	7 mm	7 mm
Column height	7 cm	10 cm
Column capacity	10 fold excess	150 fold excess
Feed solution	8 <u>M</u> LiNO <sub>3</sub> , 0.01 <u>M</u> HNO <sub>3</sub>	
Feed volume	unlimited (Am and Cm fixed)	1 free column volume (fcv)
Working temperature	10 °C	Room temperature
Eluent	3.5 <u>M</u> LiNO <sub>3</sub> 0.01 <u>M</u> HNO <sub>3</sub>	4 <u>M</u> LiNO <sub>3</sub> 0.01 <u>M</u> HNO <sub>3</sub>
Cm-Peak Position	6 fcv	7.5 fcv
Cm-Peak Halfwidth	5 fcv	12 fcv
Am content of 80 % of Cm	< 1 %	23 %
Am content of 92 % of Cm	11 %	35 %

Table 1: Comparison of Am-Cm separations by extraction chromatography with Aliquat-336 and anion exchange with Dowex 1 x 8.

Column diameter	6 cm
Column height	40 cm
Bed volume	1.13 l
Free column volume	0.6 l
Weight of Kieselgur	500 g
Weight of Aliquat	100 g
Feed and washing solution	3.2 l 8 M $\text{LiNO}_3$ - 0.5 M $\text{KBrO}_3$ - 0.01 M $\text{HNO}_3$
Eluent	1.4 l 0.5 M $\text{KBrO}_3$ - 1 M $\text{HNO}_3$
Am loss: feed and washing	100 mg (1.5%)
Am loss: recovered from column after elution	5 mg (0.07 %)
Purity of final product	> 99.7 %

Table 2: Partial purification of 7 g of  $\text{AmO}_2$  on Aliquat-336 column.

Diameter of Plexiglass column	9 cm
Column height	60 cm
Bed volume	3.7 l
Free column volume (fcv)	1.6 l
Weight of Kieselgur	1600 g
Weight of HDEHP	250 g
Feed solution	1 l 1M lactic acid (pH= <u>2.8</u> )
Wash solution I	1 l 1M lactic acid (pH= <u>2.8</u> )
Wash solution II	9 l 1M lactic acid 0.05 M DTPA (pH= <u>2.0</u> )
Eluent	11 l 1 M lactic acid 0.05 M DTPA (pH= <u>2.8</u> )
Strip solution	3 l 5 M HNO <sub>3</sub>
Pure AmO <sub>2</sub> after precipitation and calcination	25.4 g
AmO <sub>2</sub> recovered from strip solution	3.1 g

Table 3: Purification of 30 g of AmO<sub>2</sub> on single HDEHP-column.

Diameter of glass column	4 cm
Column height	40 cm
Bed volume	400 ml
Anion-exchange resin	Dowex 1 x 8, 100 - 200 mesh
Feed solution	8 <u>M</u> HCl (+ KNO <sub>2</sub> + sulphamic acid)
Eluent for Am + Cm	700 ml 8 <u>M</u> HCl (+ KNO <sub>2</sub> + sulphamic acid)
Eluent for Pu	400 ml 0.5 <u>M</u> HCl

Table 4: Separation of 6 g of Pu from gramme quantities of irradiated <sup>241</sup>Am containing <sup>242</sup>Cm and fission products.

Diameter of glass column	4 cm
Column height	40 cm
Bed volume	400 ml
Anion exchange resin	Dowex 1 x 8, 100 - 200 mesh
Working temperature	85 °C
Feed solution	10 <u>M</u> LiCl, 0.1 <u>M</u> HCl 0.1 <u>M</u> NH <sub>2</sub> OH·HCl, 5% CH <sub>3</sub> OH
Wash solution	1000 ml 10 <u>M</u> LiCl, 0.1 <u>M</u> HCl 0.1 <u>M</u> NH <sub>2</sub> OH·HCl, 5 % CH <sub>3</sub> OH
Eluent for Am + Cm	600 ml 8 <u>M</u> HCl
Eluent for Pu-traces	0.5 <u>M</u> HCl

Table 5: Separation of lanthanide fission products from  
2 g of <sup>241</sup>Am + 200 mg of <sup>242</sup>Cm.

Diameter of glass column	4 cm
Column height	40 cm
Bed volume	300 ml
Anion exchange resin	Dowex 1 x 8, 100 - 200 mesh purified
Feed solution	300 ml 8 <u>M</u> NH <sub>4</sub> SCN
Wash solution	500 ml 8 <u>M</u> NH <sub>4</sub> SCN
Eluent I	200 ml H <sub>2</sub> O
Eluent II	1000 ml 1 <u>M</u> HCl

Table 6: Thiocyanate - anionexchange column for purification of 5 g of AmO<sub>2</sub>.

Column diameter	5.8 cm
Column height	45 cm
Bed volume	1030 ml
Free column volume	600 ml
Weight of Kieselgur	430 g
Weight of Aliquat-336	108 g
Working temperature	10 °C
Feed solution	4 ml 11 <u>M</u> HNO <sub>3</sub> + 250 ml 9 <u>M</u> LiNO <sub>3</sub> , 0.005 <u>M</u> HNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Feed washings (residue in feed container)	250 ml 9 <u>M</u> LiNO <sub>3</sub> 0.005 <u>M</u> HNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Wash solution	1500 ml 8 <u>M</u> LiNO <sub>3</sub> 0.01 <u>M</u> HNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Eluent for Cm	5500 ml 3.5 <u>M</u> LiNO <sub>3</sub> 0.01 <u>M</u> HNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Eluent for Am	2000 ml 1 <u>M</u> HNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Pu-Ce strip solution	2000 ml 0.1 <u>M</u> HNO <sub>3</sub> , 0.1 <u>M</u> NH <sub>2</sub> OH·HCl
Fraction of column height occupied by Cm	~ 25 %

Table 7: Purification of 6 g of <sup>244</sup>Cm on Aliquat-336 column.

Column diameter	5.8 cm
Column height	45 cm
Bed volume	910 ml
Free column volume	500 ml
Weight of Kieselgur	425 g
Weight of HDEHP	45 g
Working temperature	Room temperature
Feed solution	3700 ml, pH = 2.8 3.5 <u>M</u> LiNO <sub>3</sub> , 0.5 <u>M</u> KBrO <sub>3</sub>
Wash solution	2500 ml 1 <u>M</u> lactic acid, pH = 2.8
Eluent for Cm	2500 ml, pH = 2.8 1 <u>M</u> lactic acid, 0.05 <u>M</u> DTPA
Lanthanides strip solution	2000 ml 1 <u>M</u> HNO <sub>3</sub>
Fraction of column height occupied by Cm	~ 30 %

Table 8: Purification of 4.5 g of <sup>244</sup>Cm on HDEHP column.

Column diameter	5.8 cm
Column height	45 cm
Volume of resin ( 1 <u>M</u> HNO <sub>3</sub> )	800 ml
Cation exchange resin	A.G. 50 W x 8, Biorad 100 - 200 mesh
Working temperature	Room temperature
Feed solution	1900 ml 0.85 <u>M</u> HNO <sub>3</sub> 1 <u>M</u> lactic acid, 0.04 <u>M</u> DTPA
Wash solution	2000 ml 0.5 <u>M</u> HNO <sub>3</sub>
Eluent for Cm	3500 ml 6 <u>M</u> HNO <sub>3</sub>
Fraction of column height occupied by Cm	~ 10 %

Table 9: Purification of 4.5 g <sup>244</sup>Cm on cation exchange column.

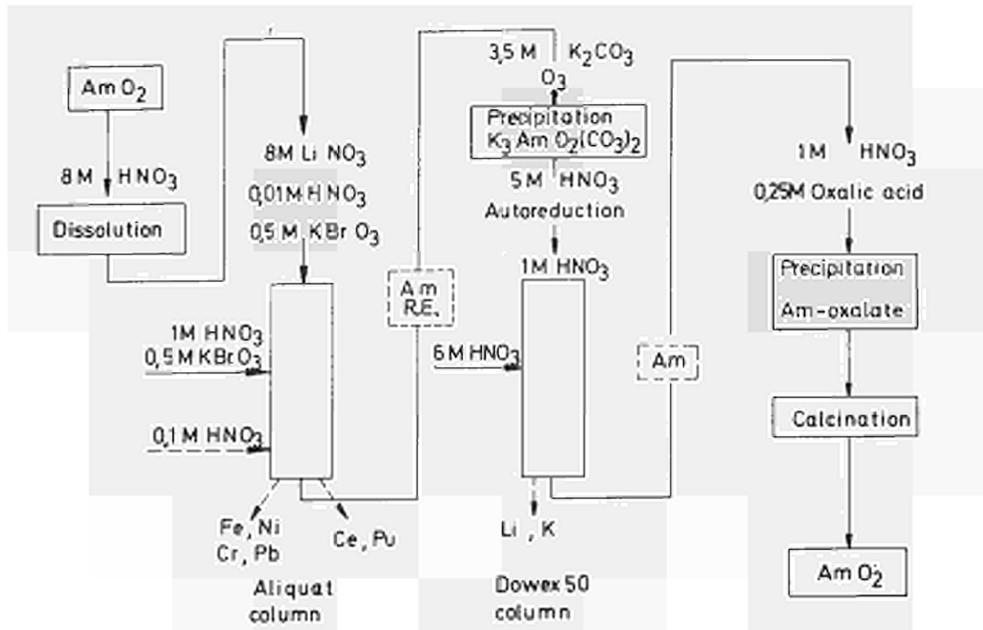


Fig. 1 Purification of 7 g of AmO<sub>2</sub> by precipitation and chromatography techniques

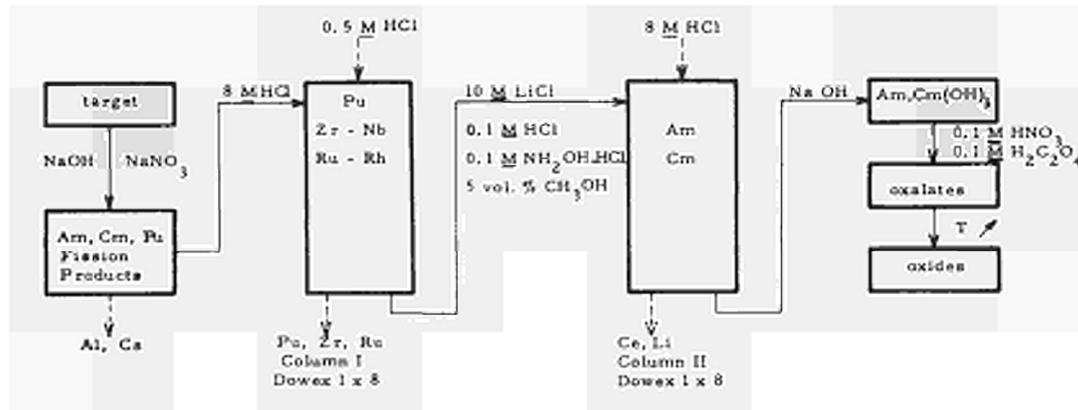


Fig. 2 Processing of irradiated <sup>241</sup>Am

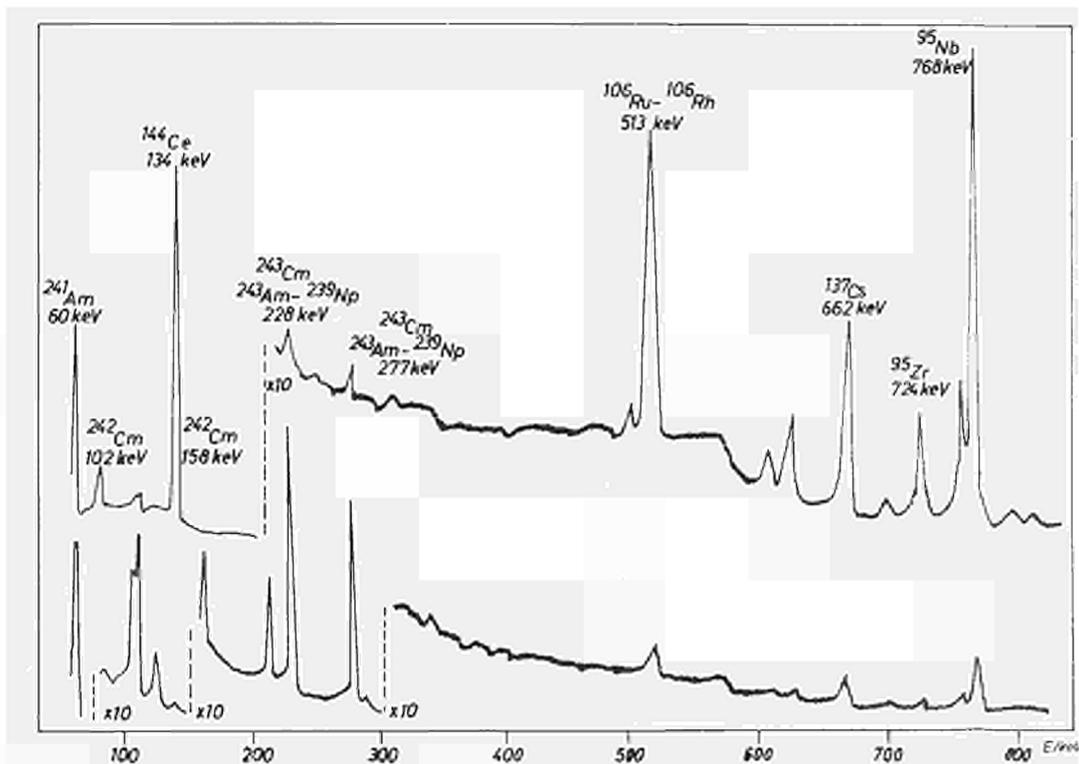


Fig. 3 Gamma-spectra of product solutions before and after passage through 2 anion exchange columns during processing of irradiated  $^{241}\text{Am}$

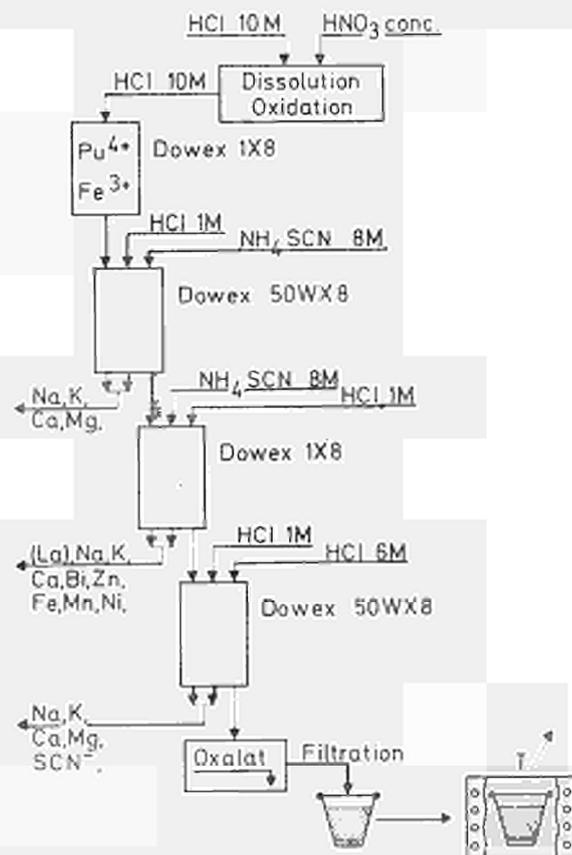


Fig. 4 Purification of 5 g of  $\text{AmO}_2$  by ion exchange

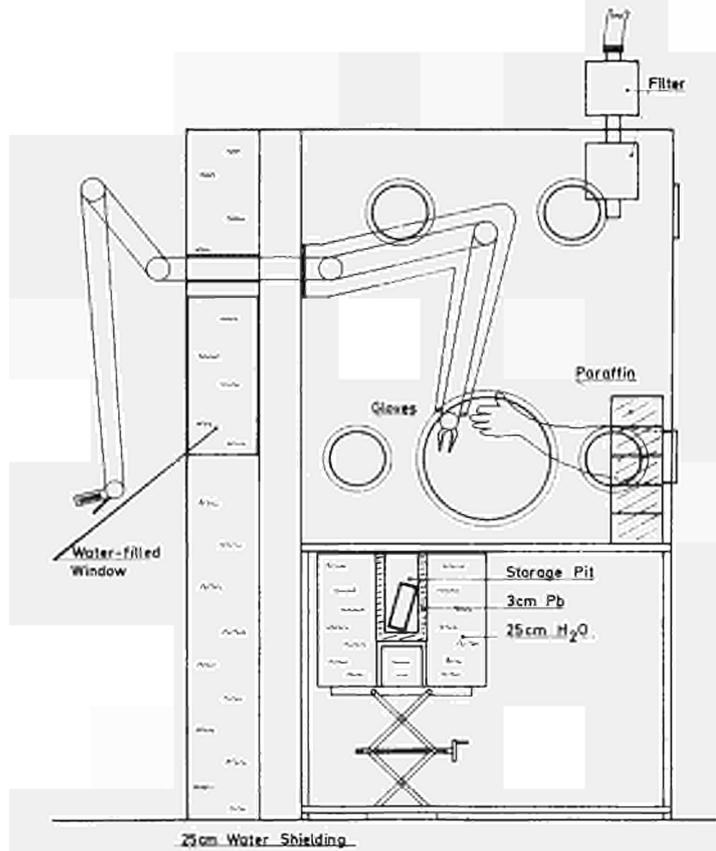


Fig. 5 Watershielded cells used for  $^{244}\text{Cm}$  purification

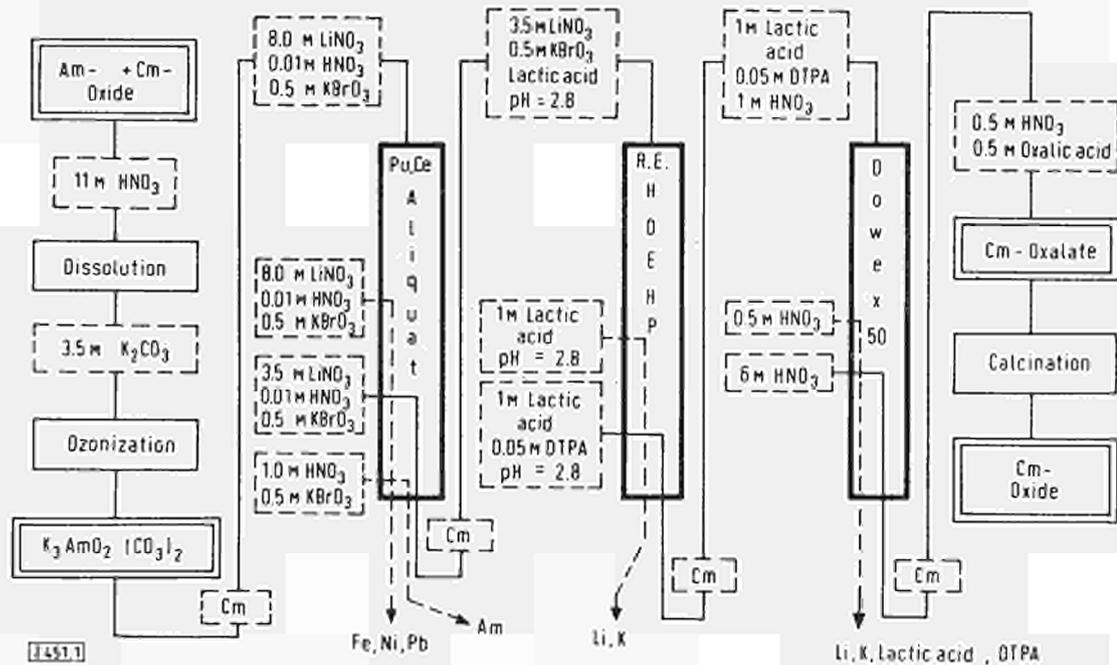


Fig. 6 Isolation and purification of 5.9 g of  $^{244}\text{Cm}$

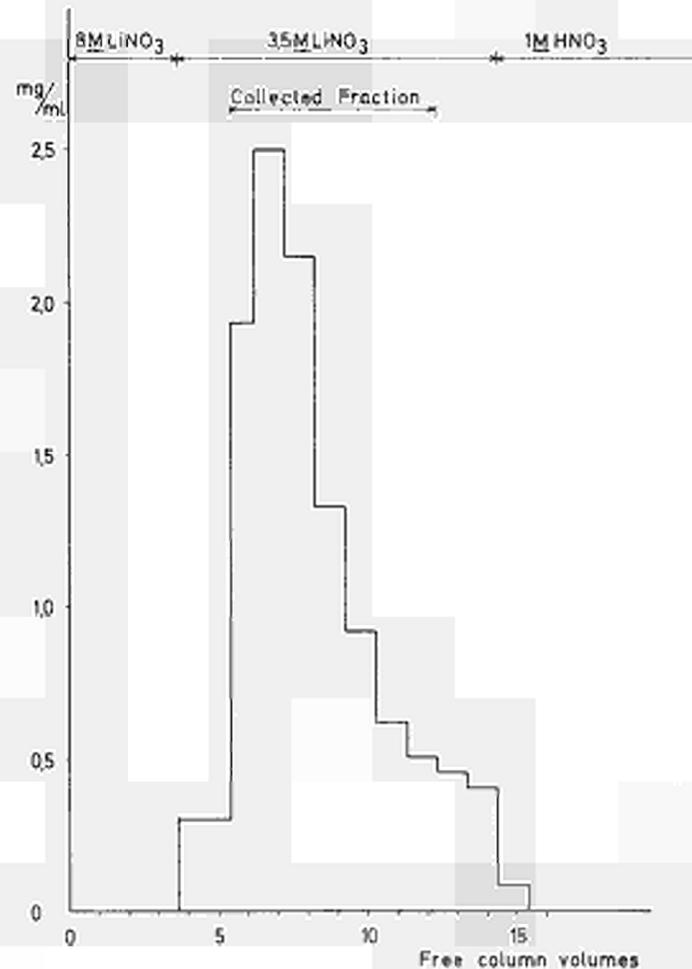


Fig. 7  
Purification of 5.9 g of  $^{244}\text{Cm}$  on Aliquat-336 column;  $^{244}\text{Cm}$  concentration in eluate as function of volume eluted

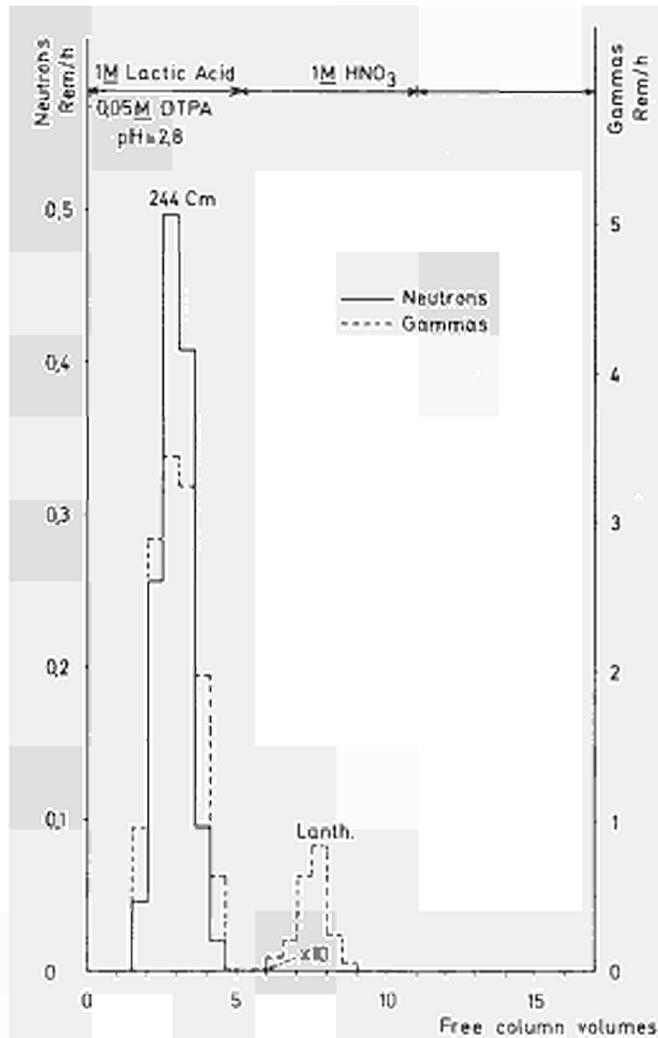


Fig. 8  
Purification of 4.5 g of  $^{244}\text{Cm}$  on HDEHP-column. Neutron and gamma dose rates (at 12 cm) per 250 ml fraction as function of volume eluted

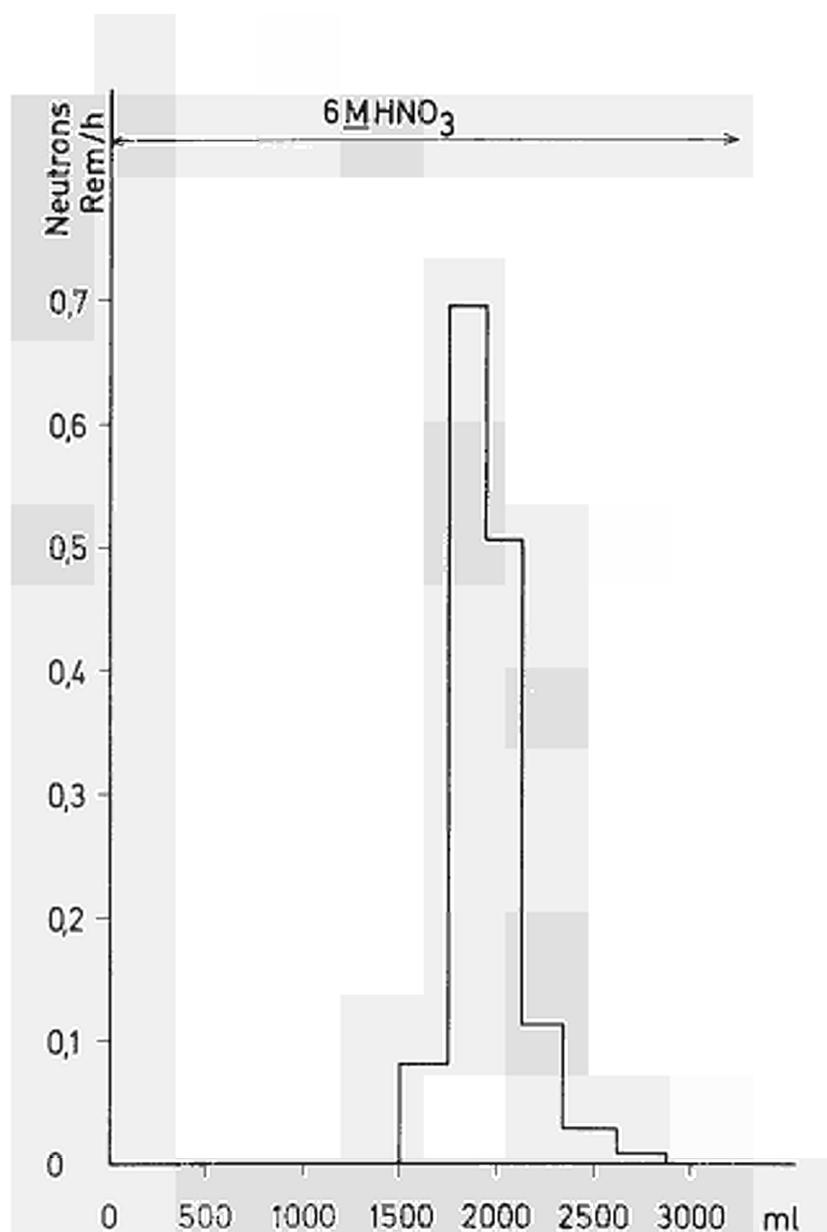
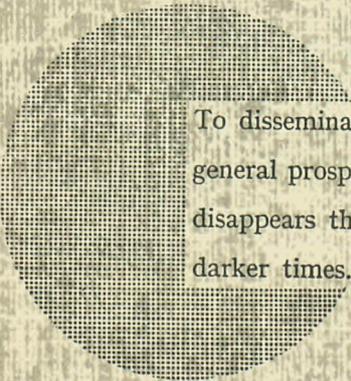


Fig. 9 Purification of 4.5 g of  $^{244}\text{Cm}$  on cation exchange column. Neutron dose rate (at 12 cm) per 250 ml fraction as function of volume eluted

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Alfred Nobel

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