

Fast Radiochemical Separation of Am, Pu, Np, U, Pa, Th, Ac and Ra in Heavy Ion Reactions with Actinide Targets

By A. TÜRLE, F. WEGMÜLLER, H. R. VON GUNTEN*, Radiochemisches Laboratorium, Universität Bern, CH-3000 Bern 9, Switzerland

K. E. GREGORICH, D. LEE, D. C. HOFFMAN, Nuclear Science Division, Lawrence Berkeley Laboratory, 70A-3307, University of California, Berkeley, California 94720, USA

and M. M. FOWLER, Isotope and Nuclear Chemistry Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

(Received July 31, 1987)

Heavy ion reactions / Actinide targets / Fast radiochemical separation / Ra to Am

Abstract

A procedure is presented for a relatively fast (20 to 30 min) separation of trace amounts of americium, plutonium, neptunium, uranium, protactinium, thorium, actinium and radium. Isotopes of these elements, produced in heavy ion reactions with actinide targets and collected by recoil in copper catcher foils, can be separated from each other and from contaminating activities. The element fractions were sufficiently clean for determinations by gamma-ray and/or alpha spectroscopy.

1. Introduction

In recent years we performed several reaction mechanism studies of heavy ion induced reactions with heavy actinide targets [1–4]. Radiochemical methods have been used throughout these experiments which were performed either at the Lawrence Berkeley Laboratory (LBL), Berkeley, California, USA or at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, FRG. Some of the chemical procedures used in these experiments were published recently in this journal [5]. In an attempt to determine relatively short-lived products (half-lives > 10 min) below the actinide target ($Z_{\text{Product}} < Z_{\text{Target}}$) faster procedures were developed and successfully used. These procedures are described in detail in the experimental section.

2. Experimental

The recoil products from the heavy ion reactions were collected in copper catcher foils. The chemical processing of the copper foils for the separation and recovery of the products produced during the bombardments is described in the following sections and in Figs. 1–8.

2.1. Dissolution of the recoil catcher foils and pre-separation of the elements

The copper catcher foil (up to 50 mg, usually 15–20 mg) was dissolved in 1 ml of conc. HNO_3 containing 50 μl of 70% HClO_4 and appropriate yield tracers (Fig. 1). The

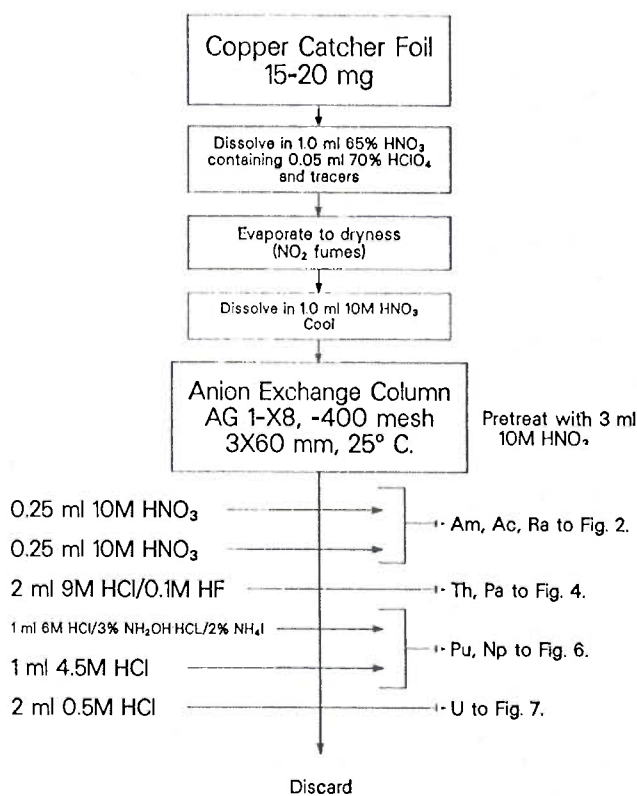


Fig. 1. Dissolution of copper recoil catcher foil and initial separation of elements Am through Ra.

solution was evaporated to dryness until the evolution of NO_2 fumes. The residue was dissolved in 1 ml of 10M HNO_3 , the resulting solution was cooled to room temperature, transferred to an anion exchange column (AG 1-X8, -400 mesh, 3 mm \times 60 mm) pretreated shortly before the experiment with 3 ml 10M HNO_3 . The column was washed twice with 0.25 ml of 10M HNO_3 at a flow rate of 10–12 drops/min. The first effluent from the column, containing the trivalent actinides, Ra and Cu was used for the determination of Am, Ac and Ra. Thorium and Pa were eluted together with 2.0 ml of 9M $\text{HCl}/0.1$ M HF. Plutonium and Np were stripped simultaneously using 1.0 ml of a solution containing 6M $\text{HCl}/3\%$ $\text{NH}_2\text{OH}\cdot\text{HCl}/2\%$ NH_4I and 1.0 ml of 4.5M HCl . Finally U was eluted with 2.0 ml of 0.5M HCl [6].

* Also at Eidgenössisches Institut für Reaktorforschung Würenlingen, Switzerland.

2.2. Separation of americium, actinium and radium

The HNO_3 effluent from the anion column from section 2.1 (Fig. 1) containing the Cu, Am, Ac and Ra was evaporated to near dryness (Fig. 2). The residue was picked up in 0.5–1.0 ml 9 M HCl and loaded on an anion exchange column (AG 1-X8, 200–400 mesh, 10 mm \times 50 mm), pretreated with 9 M HCl shortly before the experiment. Radium, Ac, Am and other +3 actinides were eluted ahead of the Cu with 9 M HCl. The fraction containing the Ra, Ac and Am was dried, dissolved in 0.1 ml H_2O and dried a second time to remove any residual acid. The residue was picked up in 0.2 ml of 0.02 M HNO_3 and loaded onto a 2 mm \times 50 mm HDEHP chromatographic column (HDEHP on Celite 535 prepared as in Ref. 7). The Ra along with other ions in +1 or +2 charge states comes off the HDEHP column in the first two free column volumes of 0.02 M HNO_3 . The Ra fraction was evaporated on a Pt disc for alpha counting. Actinium comes off the HDEHP column in the third free column volume of 0.1 M HNO_3 and was collected directly on the top of a cation exchange column. Americium comes off the HDEHP column in the second free column volume of 0.4 M HNO_3 and was also collected directly on the top of a cation exchange column. The other +3 actinides can then be eluted with 0.04 M HCl from this column, if desired.

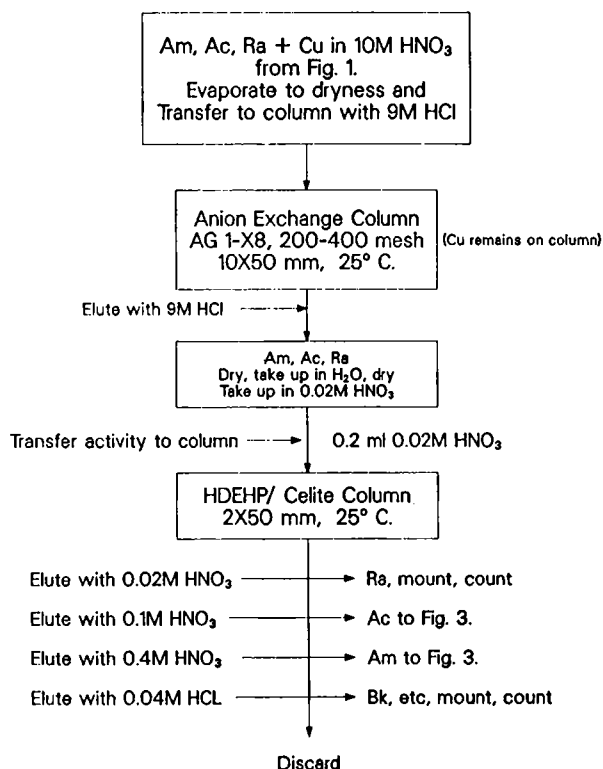


Fig. 2. Separation of Ra, Ac, Am and heavier actinides.

The Ac and Am fractions were passed through their respective cation columns (MP 50, 200–400 mesh, 2 mm \times 50 mm) and were eluted in the second through

fourth free column volumes ahead of the lanthanide activities with freshly saturated 13 M HCl (Fig. 3). The Ac fraction from the cation column was prepared for alpha and gamma-ray counting by evaporating on a Pt disc. The Am fraction from the cation column was coprecipitated with LaF_3 . Lanthanum carrier and Zr/Ba hold-back carriers were added and LaF_3 precipitated with 1 ml of 1 M HF. After five minutes the precipitate was filtered and mounted for gamma counting.

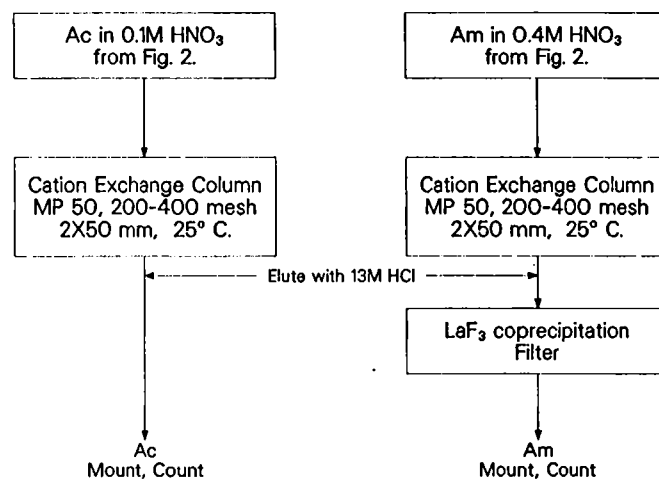


Fig. 3. Purification of the Ac and Am fractions.

2.3. Thorium and protactinium separation

0.1 ml of 40% HF was added to the column effluent from section 2.1 (Fig. 1) containing Th and Pa (Fig. 4). This solution was extracted twice with 2.0 ml of methyl isobutyl ketone [8] which had been pre-equilibrated with 9 M HCl/0.1 M HF. 10 ml centrifuge tubes with screw caps were used for all extractions. The phases were separated by centrifuging and the organic phase was discarded.

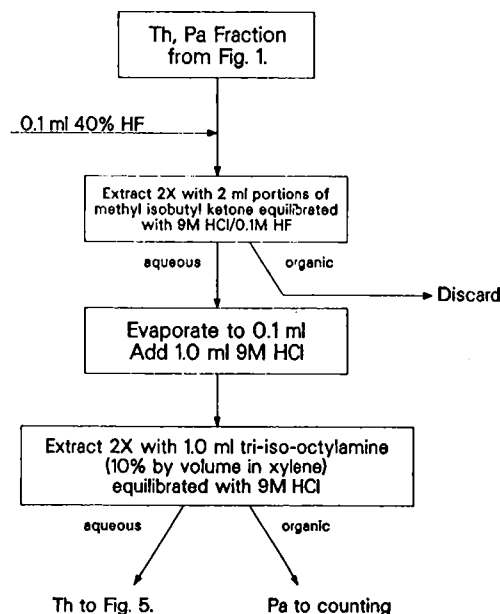


Fig. 4. Separation of Th and Pa.

The aqueous phase was evaporated to less than 0.1 ml, was taken up in 1.0 ml of 9 M HCl and Pa was extracted with two times 1.0 ml of tri-iso-octylamine (10% by volume in xylene) [6] which had been pre-equilibrated with 9 M HCl. The organic phases containing the Pa were combined and gamma counted.

Thorium was extracted from the aqueous phase (Fig. 5) with 1.0 ml of 0.1 M tri-n-octyl-phosphine oxide in cyclohexane [6] which had been pre-equilibrated with 9 M HCl and was back-extracted twice with 1.0 ml of 0.1 M HCl/1 M HF. The aqueous solutions were combined and gamma counted or electroplated for alpha counting (see section 2.6).

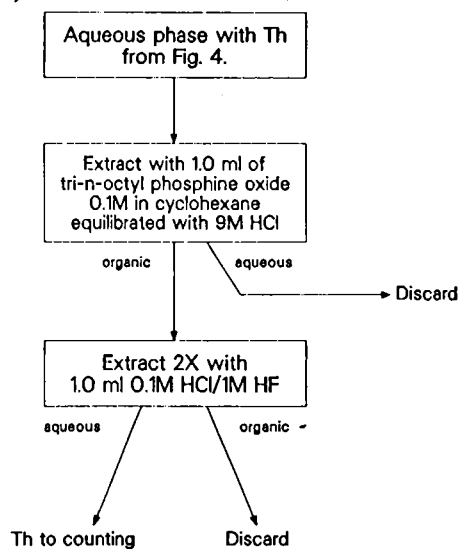


Fig. 5. Purification of the Th fraction.

2.4. Neptunium and plutonium determination

Neptunium and Pu from section 2.1 (Fig. 1) were either coprecipitated with LaF_3 (Fig. 6) by the method de-

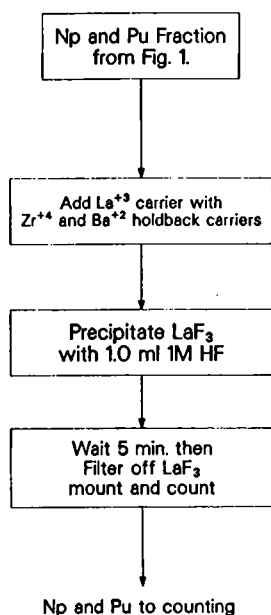


Fig. 6. Purification of the Np and Pu fraction.

scribed previously in the Am procedure and mounted for gamma-ray counting or electroplated for alpha counting (see section 2.6).

2.5. Uranium determination

1.0 ml of conc. HNO_3 and 0.1 ml NaNO_3 (11 wt%) were added to the column effluent from section 2.1 (Fig. 1) containing the uranium (Fig. 7). This solution was evaporated twice to dryness to minimize the Cl^- concentration. The residue was dissolved in 2.0 ml of a solution containing NaNO_3 (11 wt%) and EDTA (0.03 wt%), having a pH of 2.3. The uranium was extracted with 3.5 ml of triphenyl arsine oxide (1 wt% in chloroform, pre-equilibrated with 6 M HCl) and back extracted with 3.0 ml of 6 M HCl [9]. The solution was gamma counted or electroplated for alpha counting (see section 2.6).

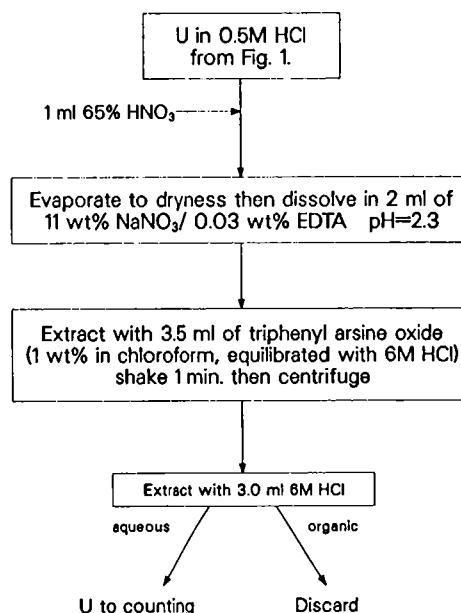


Fig. 7. Purification of the U fraction.

2.6. Electroplating of neptunium/plutonium, uranium and thorium samples

The electroplating cell used for the preparation of samples suitable for alpha counting is shown in Fig. 8. It was machined from teflon. The two parts were held together by a ball-joint clamp. A "O"-ring, pressed onto the platinum disk, ensured against leaking. Convection within the solution was accomplished with a stirrer.

2.6.1. Electroplating of neptunium/plutonium

The column effluent containing Np and Pu from section 2.1 (Fig. 1) was evaporated to dryness several times with 2 ml of conc. HCl. The residue was picked up with 0.1 ml of conc. HCl, and 4 ml of 5 M NH_4Cl were added. Two

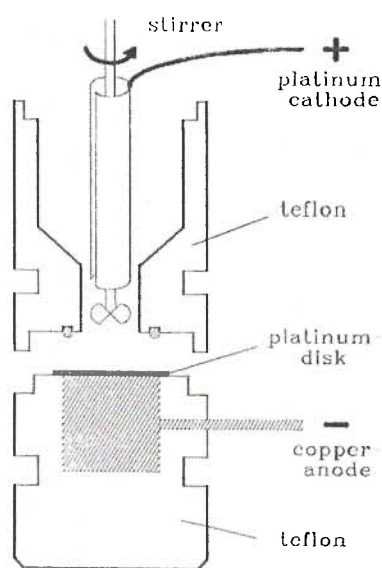


Fig. 8. Electroplating cell. The two parts are held together by a clamp. The inner diameter of the plating cell is 11 mm.

drops of methyl red were added and then NH_4OH until the solution was basic. The solution was then neutralized with 3 M HCl to the methyl red end point and electroplated on a platinum disk for 30 minutes in the plating cell. Before using the platinum disk was washed with water and ethanol and flamed. Plating conditions were 5 V and 2 amperes/cm². The solution was stirred during the plating process. Before the end of the electroplating the solution was made basic by adding 1 ml of conc. NH_4OH . The electrolyte was removed from the cell, the platinum disk rinsed with water and flamed [10]. Plating yields were > 90%.

2.6.2. Electroplating of uranium

The uranium fraction from section 2.5 (Fig. 7) was evaporated several times to dryness after adding 1 ml of conc. HCl. The residue was picked up in 3 ml of 0.01 M HCl and diluted with 3 ml of 4.2 M NH_4Cl . This solution was electroplated for 40 minutes at 10 V and 2.5 amperes/cm². At the end of the plating process 1 ml of conc. NH_4OH was added, the electrolyte was removed and the platinum disk rinsed with water and flamed. Plating yields were > 80%.

2.6.3. Electroplating of thorium

Interfering amounts of fluoride in the thorium fraction from section 2.3 (Fig. 5) were removed by adding 100 μl of conc. HClO_4 and evaporating the solution to less than 0.1 ml. The residue was picked up in 1 ml of 10 M HNO_3 and evaporated to dryness, dissolved in 1 ml of 10 M HNO_3

and passed through an anion exchange column (AG 1-X8, -400 mesh, 3 mm \times 60 mm) pretreated with 3 ml of 10 M HNO_3 . Thorium was eluted with 3 ml of 9 M HCl. This solution was evaporated to dryness and taken up with 1 ml of conc. HCl, evaporated to dryness and the residue picked up in 0.1 ml of conc. HCl. Four milliliters of 5 M NH_4Cl were added and the solution was electroplated under the same conditions as described for Pu and Np. Plating yields were > 90%.

2.7. General

Twenty to thirty minutes were required for the entire procedure (without the electroplating step) if three to four experimenters worked simultaneously on the separation, purification and preparation of the samples. The chemical yields of the separations were determined by the added yield tracers. Depending on the nuclides of interest suitable tracers had to be selected. The usual overall chemical yields for the investigated elements where: Am 95%, Pu 85%, Np 85%, U 50%, Pa 55%, Th 40%, Ac 50%, Ra 50%.

Acknowledgment

This work was supported in part by the U. S. Department of Energy under Contracts Nos. DE-AC03-76SF00098 and W-7405-ENG 36 and by the Swiss National Science Foundation.

References

1. LEE, D., VON GUNTEN, H. R., JACAK, B., NURMIA, M., LIU, Y. F., LUO, C., SEABORG, G. T., HOFFMAN, D. C.: *Phys. Rev. C* **25**, 286 (1982).
2. LEE, D., MOODY, K. J., NURMIA, M. J., SEABORG, G. T., VON GUNTEN, H. R., HOFFMAN, D. C.: *Phys. Rev. C* **27**, 2656 (1983).
3. HOFFMAN, D. C., FOWLER, M. M., DANIELS, W. R., VON GUNTEN, H. R., LEE, D., MOODY, K. J., GREGORICH, K. E., WELCH, R. B., SEABORG, G. T., BRÜCHLE, W., BRÜGGER, M., GÄGGELER, H. W., SCHÄDEL, M., SÜMMERER, K., WIRTH, G., BLAICH, TH., HERRMANN, G., HILDEBRAND, N., KRATZ, J. V., LERCH, M., TRAUTMANN, N.: *Phys. Rev. C* **31**, 1763 (1985).
4. GÄGGELER, H. W., BRÜCHLE, W., BRÜGGER, M., SCHÄDEL, M., SÜMMERER, K., WIRTH, G., KRATZ, J. V., LERCH, M., BLAICH, TH., HERRMANN, G., HILDEBRAND, N., TRAUTMANN, N., LEE, D., MOODY, K. J., GREGORICH, K. E., WELCH, R. B., SEABORG, G. T., HOFFMAN, D. C., DANIELS, W. R., FOWLER, M. M., VON GUNTEN, H. R.: *Phys. Rev. C* **33**, 1983 (1986).
5. FOWLER, M. M., DANIELS, W. R., VON GUNTEN, H. R., GÄGGELER, H. W., HOFFMAN, D. C., LEE, D., GREGORICH, K. E., MOODY, K. J., LERCH, M., HERRMANN, G., TRAUTMANN, N.: *Radiochim. Acta* **40**, 75 (1986).
6. GINDLER, J. E.: *NAS-NS 3050* (1962).
7. GREGORICH, K. E.: *LBL 20192* (1985).
8. FLYNN, K. F.: *ANL-75-24* (1975).
9. KEIL, R.: *Fresenius Z. Anal. Chem.* **292**, 13 (1978).
10. MITCHELL, R. F.: *Anal. Chem.* **32**, 326 (1960).