A METHOD FOR DETERMINING LOW LEVEL ALPHA ACTIVITY FROM AMERICIUM ^{241}, CURIUM ^{244}, AND CALIFORNIUM ^{252} IN BIOLOGICAL MATERIALS

Anne deG, Low-Beer and Troy L. Story, Jr.
Lawrence Radiation Laboratory
University of California
Berkeley 4, California

The nuclides are coprecipitated with lanthanum fluoride, Separation is effected on specially designed small columns, using a series of three different ion exchange resins. The final eluate is collected on platinum and counted directly by pulse height analysis.

The method is sensitive enough to detect activities as low as 0,3 dpm, and is applicable to both urine and tissues,

Volumes involved in the chromatographic steps are small, so that separation can be carried out in about two hours.

Identification of nuclides is made by pulse height analysis, so that the method is useful when a single nuclide is present or when two or more are present in combination.

DISCUSSION

The following procedures are applicable to any actinide element which is carried by $BiPO_4$ precipitation.* These include: actinium, thorium, neptunium, plutonium, americium, curium, and californium.

The method may be used for determinations in urine, feces, or tissues.

The activity is precipitated with $BiPO_4$ and then separated by ion exchange chromatography. The final eluate is electrodeposited on stainless steel. Preparations may be counted for gross activity on a low background proportional alpha counter; for identification of radionuclides by pulse height analysis; or they may be radioautographed for alpha track counting.

Recoveries vary somewhat for the several nuclides, but are in the range 75 - 90 ± 3%.

The method has a detection limit of 0.2 dpm at the 99% confidence level.

The procedures, exclusive of counting time, require about 14 hours after ashing of the sample is complete.

Urine and feces are prepared for processing by wet ashing with nitric acid. Tissues are dry ashed for 24 hours in an oven at 100° C followed by 24 hours in a muffle furnace at

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* Our own experience with this method is limited to Am^{241}, Cm^{244}, and Cf^{252}. These nuclides have been used separately and in combination.

500° C. In order to produce a pure white ash, one or two digestions with 10 ml portions of hot nitric acid may be necessary.

The white ash is taken up in 2N nitric acid and centrifuged to remove any undissolved material. The activity is then precipitated with $BiPO_4$ at pH 1.7 and 80 - 85° C with continual stirring for one hour (1). One ml of $Bi(NO_3)_3$ containing 10 mg Bi^{+++} per ml and 0.5 ml H_3PO_4 are used for the precipitation.

The precipitate is dissolved in 0.5 ml of 8N HCl and placed on an anion exchange column 3 mm in diameter and packed to a height of 8 cm with Dowex 1 x 8 200 - 400 mesh in the chloride form (2). The effluent is collected and the column washed three times with 0.5 ml portions of 8N HCl. The washings are added to the effluent. The combined effluent is evaporated to dryness and the residue is dissolved in 0.5 ml 0.05N HCl. This is put on a column 3 mm in diameter and packed to a height of 5 cm with Dowex 50 x 4 200 - 400 mesh resin in the hydrogen form. The column is equipped with a glass jacket (see Fig. 1), and is operated at a temperature of 87° C by means of boiling trichloroethylene in a flask attached to the jacket of the column and to a reflux condenser.

The effluent from this column operation is discarded, and the column is washed with 0.65 ml 2N HCl. The effluent from the wash is also discarded. The activity is then eluted with 6N HCl. The first three drops are discarded and the next 30 drops are collected in a 10 ml beaker.

The eluate is evaporated to incipient dryness without boiling, and 0.3 ml H_2SO_4 is added and heated to dense white fumes (3). After cooling, the contents of the beaker are transferred with copious rinsing with distilled water to an

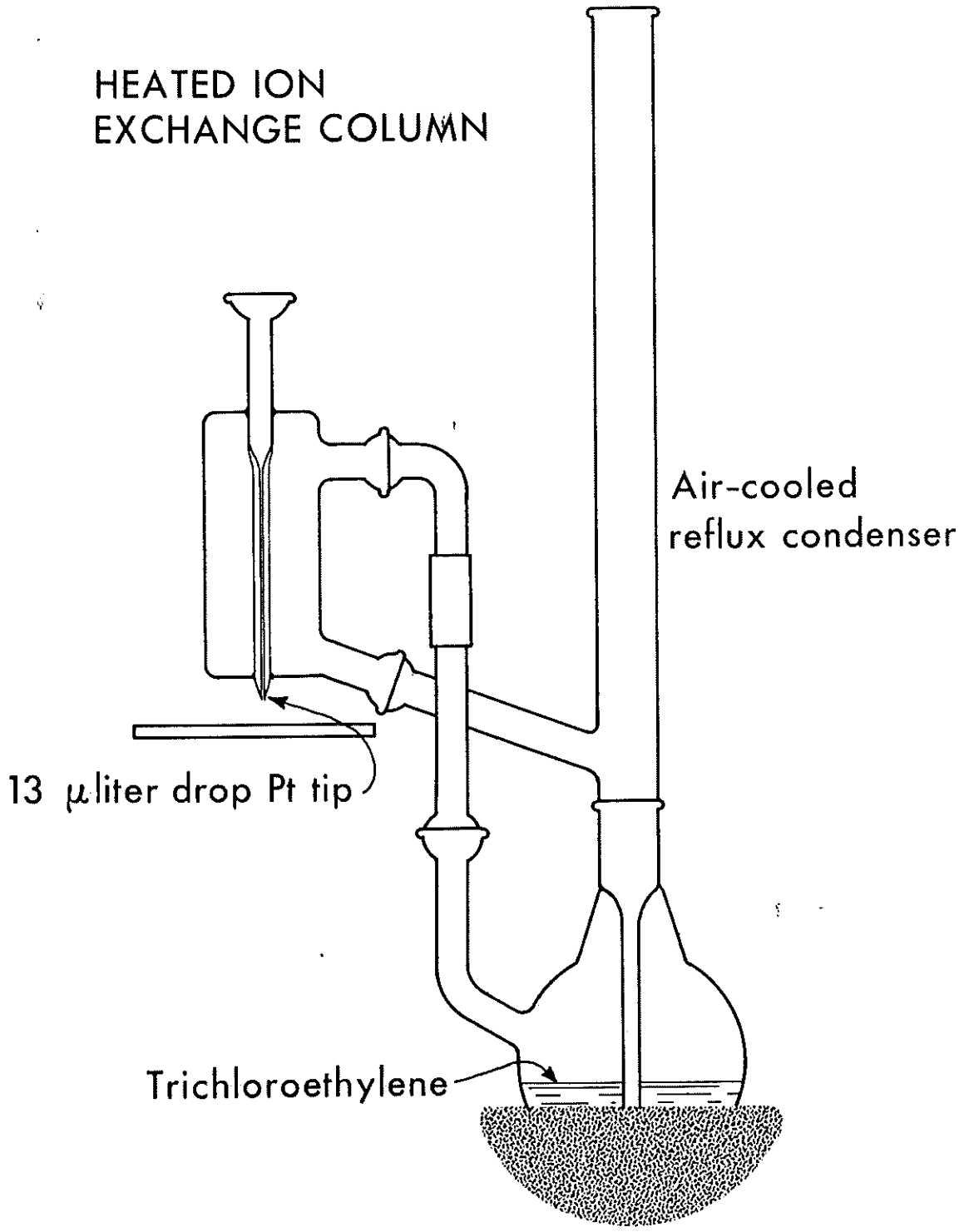
HEATED ION
EXCHANGE COLUMN
Air-cooled
reflux condenser
13 μliter drop Pt tip
Trichloroethylene

FIGURE 1

electrodeposition cell equipped with a 10 mil stainless steel disc and a platinum electrode (4). The volume in the cell is brought to 10 ml with distilled water and one drop of 1% methyl red is added. The sample is titrated with NH_4OH to the first permanent yellow, and is then titrated back to one drop on the red side with 1.5N H_2SO_4. Electrodeposition is carried on for 5 hours at 12 V and 180 ma. Before turning off the current, one drop of NH_4OH is added and electrodeposition is continued for one minute. The contents of the cell is decanted and the cell rinsed several times with distilled water. The stainless steel disc is removed and washed with distilled water and acetone and dried in air. It is then flamed to cherry red.

Gross alpha activity may be determined by counting in a low background proportional alpha counter. With low activities, counts should be made for at least 120 minutes.

Identification of nuclides is accomplished by means of a counter equipped with a multi-channel alpha energy analyzer. Peaks are identified by comparison with the peaks of two or more nuclides used as standards. In this work we have used an alpha grid chamber and an RIDL counter with a one hundred channel analyzer. In order to develop discernible peaks with low activities, it has been necessary to count the samples for at least 10 hours. We have found this system to have about the same geometry as our proportional alpha counter, i.e., approximately 50%. At the 99% confidence level our detection limit is 0.2 dpm, based on three times the standard error of

background counts. We are now about to experiment with semiconductor detectors in the hope that their superior resolving capacity and their insensitivity to background conditions will enable us to lower our detection limit and otherwise to increase counting efficiency.

In this work we have used activities of 0.5 dpm or less. Our recoveries have been: Am^{241}, 90 ± 3%; Cm^{244}, 75 ± 3%; Cf^{252}, 88 ± 3%. In the tissues of mice which had been given Am^{241} and Cf^{252} in combination (5), the Am^{241}/Cf^{252} ratios found by our method were in close agreement with those obtained by gamma counting.

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