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Use of Alumina as an Ion Exchanger in the Separation of Carrier-Free ^{144}Pr from ^{144}Ce

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NOTE

Use of Alumina as an Ion Exchanger in the Separation of Carrier-Free ^{144}Pr from ^{144}Ce

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Abstract

A very simple procedure for separation of carrier-free ^{144}Pr and continuous milking of ^{144}Pr from ^{144}Ce in a alumina column has been developed. Cerous cerium was oxidized and adsorbed as ceric iodate in the column, and the praseodymium was eluted out unadsorbed. Study of the β-decay half-life of ^{144}Pr proved that the separated product is of high radiochemical purity. The separation process is rapid and the yield is quantitative.

INTRODUCTION

Separation of adjacent rare earths such as cerium and praseodymium has been carried out by several investigators using methods such as liquid–liquid extraction, precipitation, and ion exchange chromatography. Use of di(2-ethylhexyl) phosphoric acid (*1*), tributylphosphate (*2*), acetylacetone (*3*), and methyl isobutyl ketone (*4*) as solvents; and allyl triphosphate (*5*) or bis(2-ethylhexyl) orthophosphoric acid supported in silica gel (*6*), cellulose phosphate (*7*), and insoluble inorganic salts such as zirconium phosphate (*7, 8*), stannic phosphate (*9*), and amorphous stannic molybdate (*10*) as ion exchangers have been described for this separation. Werner (*11*) separated Ce–Pr and also a few other adjacent rare earths with dinonylnaphthalene sulfonic acid (HDNNS) treated paper as the stationary phase and 0.12 *M* α-hydroxyisobutyric acid and 0.2 *M* lactic acid as eluents at different pH's. Strain et al. (*12*) have separated Ce–Pr and a few other parent–daughter mixtures by electrochromatography. Peppard, Mason, and Molin (*13*) have claimed the separation of highly purified cerium target material from ^{143}Pr and ^{144}Pr daughter activities (by a factor of 10^7) in less than 10 min by using 10 M nitric acid and 0.75 M or 0.3 M HDHEP in heptane. The ^{144}Pr obtained in the procedures described above is however not radiochemically pure, the processes are time-consuming and they involve a number of steps. A method similar to ascending paper chromatography using various ion exchangers in the form of narrow zones on paper strips has been reported (*14*) for the separation of ^{144}Ce–^{144}Pr and others but do not give quantitative yield. Radiochemical separation of this pair was also described (*15*) by a ring oven method. Radiochemical purification and continuous milking of carrier free ^{144}Pr from ^{144}Ce on an MnO_2 column has been recently claimed by certain authors (*16*). Separation of ^{144}Ce by immobilization of Ce^{+4} on a column of HDEHP supported on Hostaflon C_2 and the continuous milking of the ^{144}Pr daughter was described (*17*), but the method is unsatisfactory as regards yields and purity.

Cerous cerium has the advantage of being easily oxidized to well-known +4 oxidation state by strong oxidizing agents such as BrO_3^- in acid medium. Precipitation of insoluble ceric iodate has been mostly applied for the separation of cerium from other rare earths (*18–20*).

In the present investigation an inorganic ion exchanger like alumina is utilized for a very simple and rapid separation of carrier-free ^{144}Pr from ^{144}Ce. Separation of several parent–daughter radioisotope pairs such as ^{99}Mo–^{99}Tc(m), ^{210}Pb–^{210}Bi, ^{140}Ba–^{140}La, and ^{90}Sr–^{90}Y through alumina columns have been described (*21–24*) where it was argued that alumina as a powerful adsorbent, adsorbs either one or both of the pair, and subsequently the desired element could be preferentially eluted out from the column by suitable reagent. A study was therefore undertaken with the view that ^{144}Ce, similar to its macrobehavior, would form insoluble ceric iodate and be adsorbed in the alumina column, and ^{144}Pr in the +3 state might pass out unadsorbed.

EXPERIMENTAL

^{144}Ce (as cerous chloride) in equilibrium with its daughter ^{144}Pr was supplied by BARC Trombay, India. Chromatographic aluminium oxide was of G.R.E. Merck quality. All other reagents were of analytical grade.

About 10 ml of the dilute activity solution (4000 cpm) was evaporated three times with nitric acid and then taken up with a little water. To this 0.5 g of boric acid, 5 ml conc HNO_3, and 5 ml of 10% $NaBrO_3$ solution were added. The mixture was kept on a steam bath for ~10 min, cooled, and 10 ml of 1% HIO_3 was added with constant stirring. This was then cooled in an ice atmosphere for some time and fed into the glass column (30 × 1 cm) filled with ~17 g of aluminum oxide. Size and arrangement of the glass column was kept the same as for RaD–RaE separation (*22*). The eluate was collected in a conical flask connected to a suction pump which

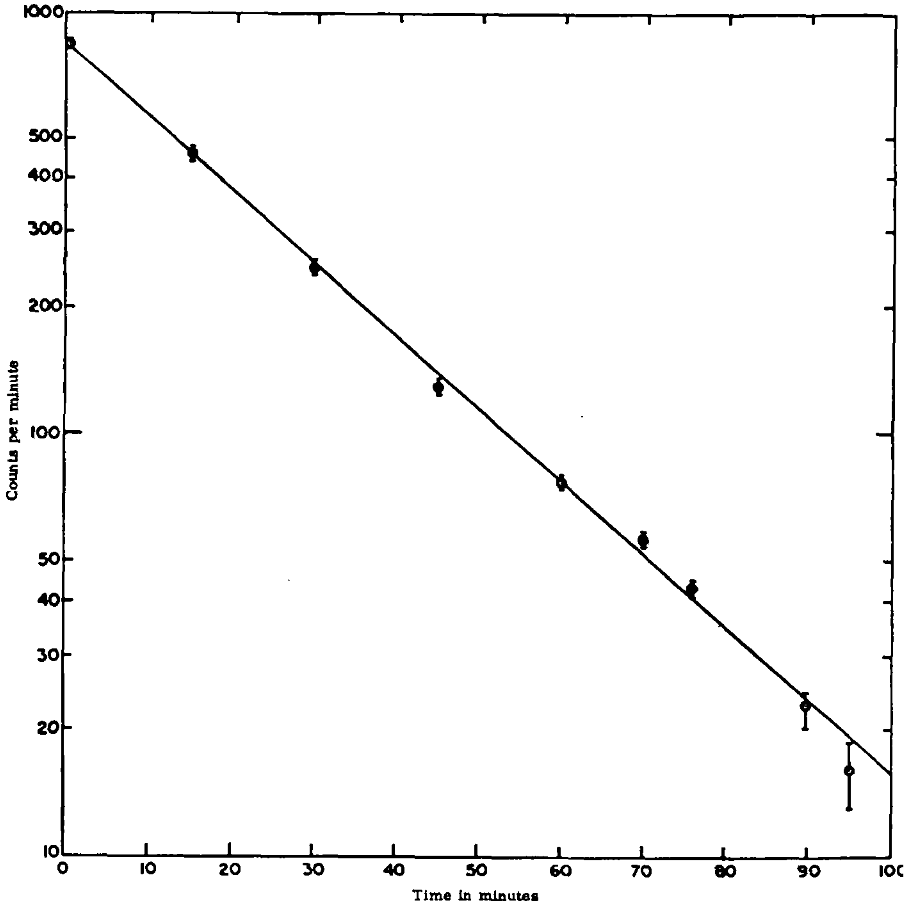

FIG. 1. Decay curve of ^{144}Pr separated from ^{144}Ce.

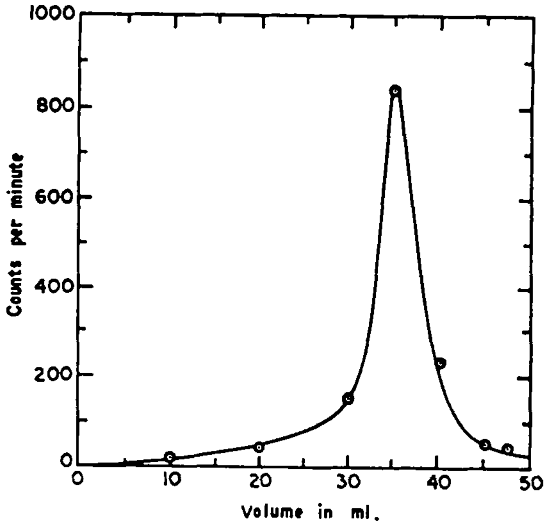

FIG. 2. Elution curve for ^{144}Pr separated from ^{144}Ce.

regulated the flow of solution to a rate of 14 to 15 drops/min. The column was next washed with 20 ml of 1 % HIO_3 solution and then with a further 10 ml of water. All the washings were mixed together, and 10 ml of this solution was counted with a Philips G.M. β-type liquid counter (mica window thickness, 2.5 to 3.5 mg/cm^2, diam, 27.8 mm). The results, shown in Fig. 1, indicate that the collected solution contains only ^{144}Pr and no ^{144}Ce. The remaining portion of the solution was evaporated to a small volume and filtered from the minute quantity of the solid matter that separated. These fine solid particles (probably of alumina) were washed with water, dried, and counted in a counting tray which showed no activity.

In order to investigate the volume of the eluant required to wash down the maximum amount of ^{144}Pr, a separate experiment was carried out by taking and counting successive 10-ml portions of the eluate collected in the way already described, and an elution curve was obtained (Fig. 2). The curve shows that only 40 ml of the eluant is required to separate almost all of the ^{144}Pr activity. The experiment was repeated and the results confirmed.

^{144}Ce was thus found to be adsorbed in the alumina column, which was kept for sufficient time to allow for fresh growth of the ^{144}Pr activity.

The column was then washed with 1% HIO_3 and the washings counted as before. This study showed that almost all of the ^{144}Pr daughter which separated from ^{144}Ce was eluted, and found to be followed continuously for a few days without any change in the elution pattern.

DISCUSSION

A glance at the β-decay curve in Fig. 1, shows that the separated ^{144}Pr product is of high radiochemical purity. Cerous cerium was oxidized by sodium bromate to the ceric state which was adsorbed on the alumina column, probably as insoluble $Ce(IO_3)_4$, and the unchanged praseodymium passed down unadsorbed. Oxidation of cerous cerium by bromate is very important, but simple oxidation to the ceric state does not allow clean separation of ^{144}Pr through adsorption of ^{144}Ce by alumina. The formation of insoluble $Ce(IO_3)_4$ was thus proved important, and it was also observed that the application of the same amount of KIO_3 instead of HIO_3 does not effect the efficiency of separation in any way. Pr^{+3} does not change to the Pr^{+4} state and therefore elutes out, fixing ^{144}Ce in the Ce^{+4} state over the alumina column. The total process takes 30 to 35 min.

The preferential adsorption character of alumina and the formation of a fixed "^{144}Ce-cow" in its surface for continuous milking of ^{144}Pr indicates that this type of alumina column can be used for other similar chemical separation problems.

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