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CHEMICAL SEPARATION OF RUTHERFORDIUM*

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Introduction

As current evidence (1,2,3) supports the original prediction that element 103 is the last member of the actinide series (4), element 104 would be expected to fall into the group IVB elements of the periodic table, i.e., to be eka-hafnium. The element is predicted to have a valence and ionic radius similar to Zr and Hf (5,6) and to exhibit similar chemical properties.

Russian researchers have concentrated their efforts to separate element 104 from the actinides on rapid gas phase techniques based on the predicted large differences in the relative volatilities of the chlorides (7,8). There is, however, some uncertainty in the assignment of the spontaneous fission activity ($T_{1/2} \sim 0.5$ sec.) used in these experiments (8,9,10).

The recent discovery of an isotope of element 104, ^{261}Rf (11,12), with half-life of ~ 70 seconds and alpha particle energies of 8.25-8.40 MeV, has made it possible to carry out more conventional aqueous chemical separations using ion-exchange techniques. Previous studies with actinide

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elements (13,14) show that cation exchange columns using chelating agents as eluants can provide rapid chemical separations on one atom at a time and yield sources suitable for alpha energy analysis. The experiments described in this work represent a further development of this method to test the above predictions.

Experimental

The ^{261}Rf was produced by irradiation of 47 micrograms of ^{248}Cm, which was electrodeposited over 0.2 cm^2 area onto a 0.0005-inch thick Be foil, with 92-MeV ^{18}O ions from the Berkeley Heavy Ion Linear Accelerator. Details of the bombardment assembly, fast sample transfer system and other experimental procedures have been described elsewhere (1,15,16).

In each chemical experiment, the rutherfordium recoil atoms were washed from an NH_4Cl-coated, platinum catcher foil with ~ 50 microliters of ammonium alpha hydroxyisobutyrate (0.1 M, pH 4.0) onto the top of a 2 mm. diameter by 2 cm. long, heated (~80°C) column of Dowex 50 x 12 cation resin. This solution was forced into the resin and, after adding more eluant, the washing was continued. The first two drops (free column volume) contained little or no activity and were discarded. The next four drops (taken in two drop fractions) were collected on platinum discs, evaporated to dryness and heated to ~500°C to burn off any carbon residue. The discs were placed active side down directly over Si(Au) alpha particle detectors. The number, energy and time distribution of alpha particles with energies between 6 and 12 MeV emitted by the sources were recorded in a PDP-9 computer. A large part of the chemical system was automated so that the average time from beam off to counting was reduced to ~ 60 seconds. Approximately 100 atoms of ^{261}Rf were produced for study in several hundred experiments, however, only about 1/10 of this number of events were observed after chemistry due to decay, counting geometry and chemical losses. At regular intervals, the platinum catcher foils were counted directly for yield measurements.

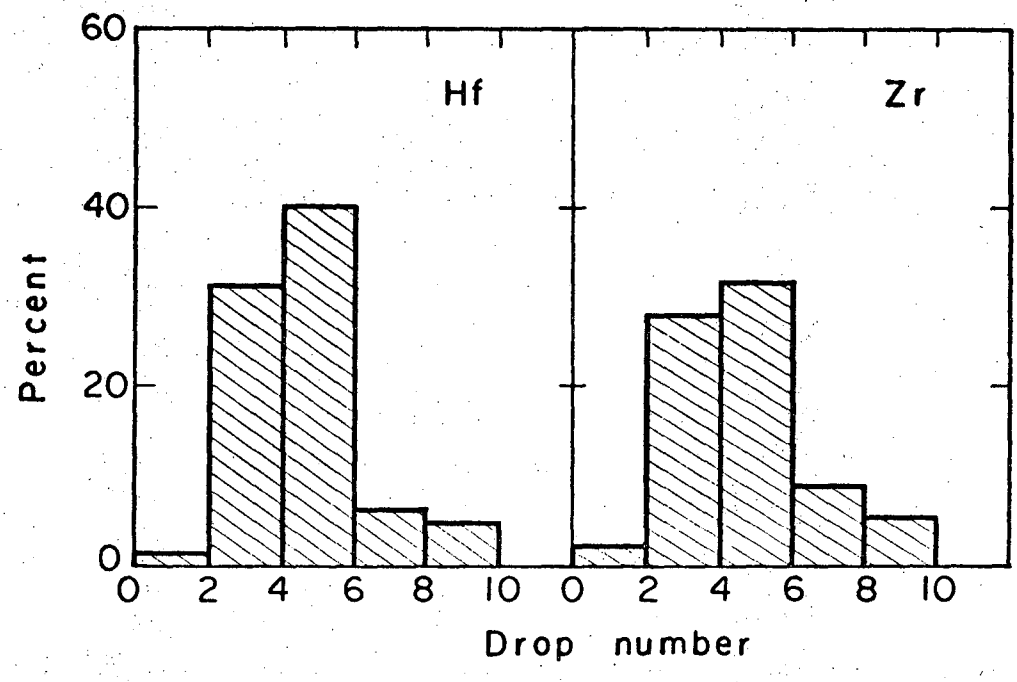

FIG. 1

Elution of Zr and Hf tracer from a Dowex 50 x 12 cation exchange resin column using Ammonium Alpha Hydroxyisobutyrate.

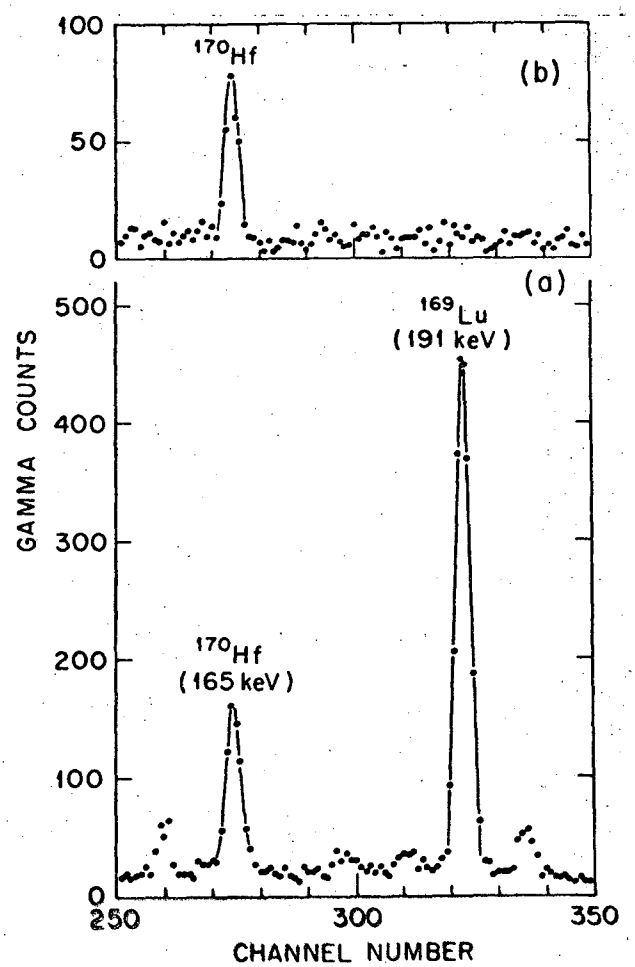

FIG. 2

Gamma-ray spectra taken with Ge(Li) detector before (a) and after (b) chemical separation and shows typical separation of ^{170}Hf from ^{169}Lu.

Results and Discussion

Under experimental column conditions described above, over 50% of tracer quantities of Zr and Hf were washed through the cation exchange column with the first few column volumes of eluant as shown in Fig. 1. In contrast, trace quantities of the 3+ ions Tm, Cf and Cm did not elute in over 100 column volumes. One would expect Lr to be the first 3+ actinide to be removed from the column and to elute in about the same position as Tm (13, 17). Divalent nobelium, as well as the alkaline earth elements, are retained even more strongly by the resin than the 3+ actinides (14). Alpha particle analysis of the same early column fractions using trace quantities of a number of other elements in this region of the periodic table showed the following amounts present: Bi < 3%, Ra < 1%, Fr < 1%, Th < 1%, Np < 5%, Pu ~ 50%, and Pa ~ 50%. Atoms of ^{170}Hf and ^{169}Lu, produced by the irradiation of a 1 mg/cm^2 target of ^{159}Tb with 67- MeV ^{14}N ions from the Oak Ridge Isochronous Cyclotron, were subjected to the identical chemical separations as described for ^{261}Rf. Fig. 2 shows γ-ray spectra taken with a 40 cc. Ge(li) detector both before (a) and after separation (b) and indicates typical separation of the Hf from Lu. These data indicated that only about 50% of the ^{170}Hf produced was recovered by this procedure.

Fig. 3 shows a summary of data accumulated over a 3 week period for ^{261}Rf. Although the actual time of occurrence as well as the energy of each alpha particle event was recorded, the time distributions given in Fig. 3 are four sequential one-minute decay intervals plus a summation of the four individual energy spectra. Fig. 3a shows energy spectra accumulated directly from platinum catcher foils from which the production rates of ^{261}Rf and the indicated impurity activities were determined. Fig. 3b shows the energy spectra accumulated from sources after chemical separation, which contained seventeen alpha particle events in the energy region 8.2 to 8.4 MeV. Approximately one half of these events are probably due to the decay of ^{257}No ($T_{1/2}$ = 26 sec., E_α's = 8.22 (55%), 8.27 (26%) and 8.32 (19%) MeV), the daughter of the separated ^{261}Rf. In two experiments, two alpha decay events

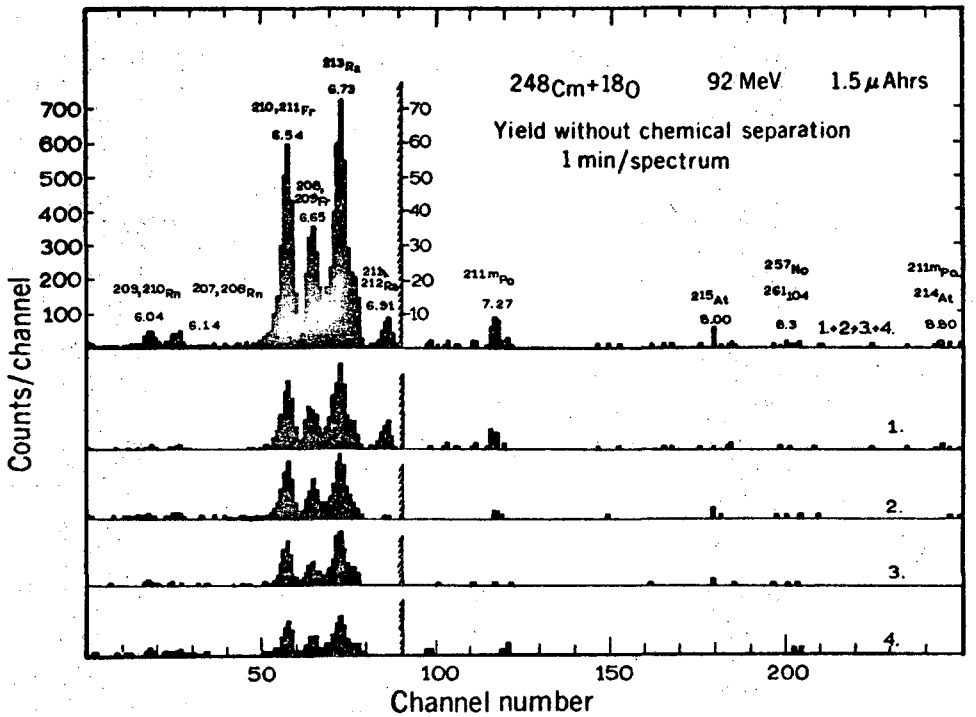

FIG. 3.a

Alpha particle energy spectrum accummulated from sources without chemical separation.

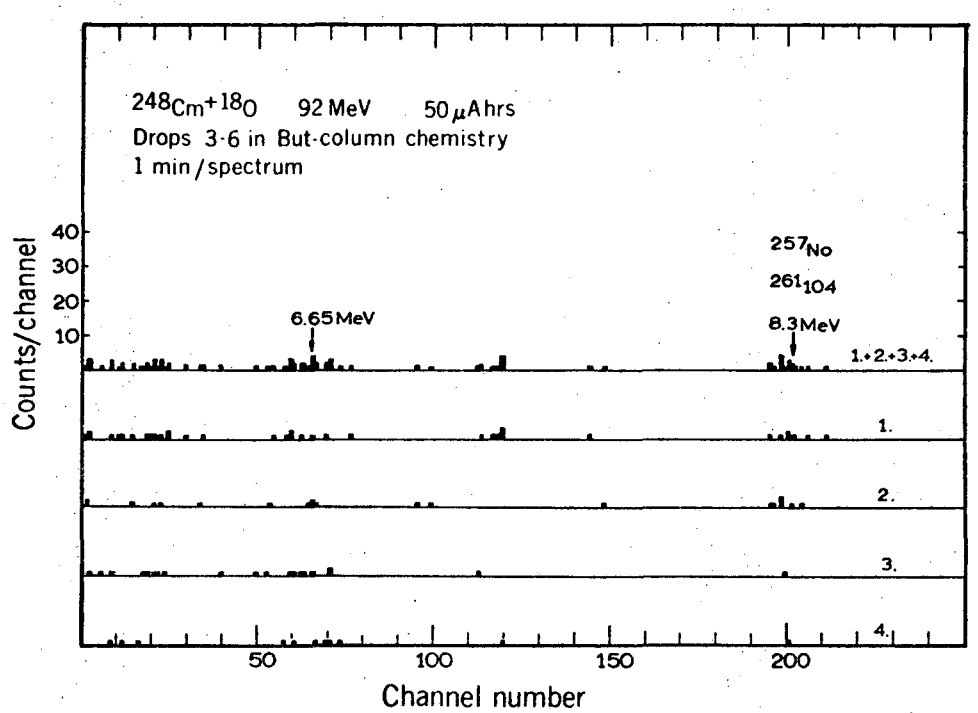

FIG. 3.b

Alpha particle energy spectrum accummulated from sources after chemical spearation.

occurred in the 8.2 to 8.4 MeV region within a time interval of one minute, most probably the first one due to ^{261}Rf and the second due to the daughter ^{257}No. This is consistent with the number of pair events expected in the counting system that was used. Decontamination factors of from 100 to 1000 of the Rf from the impurity activities were obtained.

The data show that, for the particular cation exchange conditions used in these experiments, the behavior of the activity assigned to element 104 with mass 261 is entirely different from trivalent and divalent actinide elements but is similar to Hf and Zr as one would predict for the next member of the periodic system following the actinide series of elements.

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