

ISOLATION AND ELECTROPLATING OF ^{210}Po

E. Ordoñez-Regil, J.L. Iturbe G.

Instituto Nacional de Investigaciones Nucleares,
Apdo. Postal 18-1027, México, D.F., C.P. 11870, Mexico

Received 19 October 1992

Accepted 2 November 1992

A method for isolation and electroplating of ^{210}Po is described. Natural samples of autunite, after pretreatment, were dissolved in dilute hydrochloric acid and the solution was passed through a chromatographic partition column with di(2-ethylhexyl)phosphoric acid as stationary phase. Uranium and its daughters were strongly retained in the column. The ^{210}Po fraction was eluted with citric acid solution, then evaporated almost to dryness. Next, electroplating was carried out for 18 h, at 5-10 mA current, using an electrolyte bath of oxalic acid solution. The yield of recovery of radiochemically pure ^{210}Po was about 95% of the initial ^{210}Po activity.

INTRODUCTION

When dealing with the determination of low concentrations of uranium and its radioactive daughters in natural samples, it is very important to obtain information about the respective proportions of various isotopes of each of the elements present in the sample. α -Particle

spectrometry is a very precise and sensitive quantitative technique for α -emitters, which allows to determine the isotopic distribution of some actinides, in particular, the $^{234}\text{U}/^{238}\text{U}$ concentration ratio. However, some difficulties may often arise due to energy interferences between various α -emitters. To avoid the presence of such interferences in the energy spectra, chemical separation of uranium from its daughters is usually necessary.

Among the methods published to separate uranium from other elements, solvent extraction has been extensively used. In particular, Bogdanov¹ has described a partition chromatographic column, using di(2-ethylhexyl)phosphoric acid (DEHPA) as stationary phase, to extract uranium from dilute hydrochloric acid solution. The radioactive daughters are also extracted. Although this technique was originally developed to study the separation of uranium from natural samples, the method can be extended to any specified uranium daughter by determining the specific eluting conditions from the column. In this context, electroplating over metallic disks appears as a promising technique to complete the selective isolation of the chosen uranium daughters in the form of a thin and uniform deposit.

Uranium and its daughters represents valuable natural tracers for studying atmospheric, oceanic, and terrestrial processes². Furthermore, there is a strong biological interest in the $^{210}\text{Pb} \rightarrow ^{210}\text{Bi} \rightarrow ^{210}\text{Po}$ decay series³. For these reasons, it seemed useful to develop a method to isolate one of these daughters from the point of view of quantitative analysis. This paper, therefore, reports on the search for appropriate conditions to achieve uniform and reproducible electroplated thin foils of ^{210}Po for α -spectrometric measurements.

To this end, various experiments were run to assess the efficiency of different eluting solutions, the effect of acid concentration on eluting and electroplating and of the nature of electrolyte on the recovery yield.

EXPERIMENTAL

Reagents

All chemical reagents used were A.R. grade. Uranium, in radioactive equilibrium with its daughters, was electroplated from crystalline autunite from Strasbourg, France (Fig. 1). For trial experiments, some ^{210}Po was produced by thermal neutron irradiation of ^{209}Bi in the Triga Mark III reactor of Instituto Nacional de Investigaciones Nucleares in México.

Chromatographic separation

Uranium separation has been achieved over a chromatographic column packed with teflon beads impregnated with DEHPA as stationary phase. The autunite matrix was dissolved with concentrated HCl and the solution was diluted to 0.1M before passing through the chromatographic system. Most of the radioisotopes were retained in the chromatographic column. The ^{210}Po elution was achieved using a citric acid solution. The ^{210}Po fraction was evaporated almost to dryness and the remaining organic matter was mineralized with 1 cm³ concentrated sulfuric acid at about 450 K until the volume of the solution was greatly reduced, 3 cm³ of hydrogen peroxide (30%) was used to clear up the acid extract.

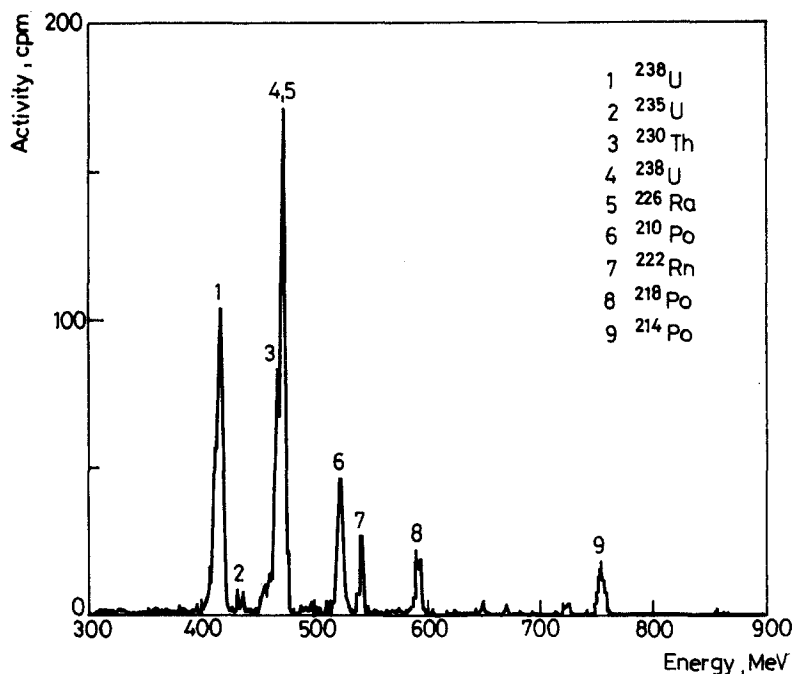


Fig. 1. α -Spectrum of natural uranium

Electroplating

The sulfuric acid solution obtained as described above was mixed with 10-20 cm^3 of acidic aqueous solution, then transferred to an electrolytic cell. The cathode used was made of stainless steel polished disks, with 1.5 cm diameter, and the anode was a platinum wire placed 1 cm in front of the cathode, the current intensity was 5-10 mA applied for 18 h.

α -Counting

After electroplating, the disks were washed with acetone and placed in a vacuum chamber for α -counting

using a 400 mm^2 surface-barrier detector, linked to a 2048 channel analyzer. The energy resolution was 18 keV (fwhm) for the 5.155 MeV ^{239}Pu peak.

RESULTS AND DISCUSSION

An untreated sample of autunite was electroplated to determine the activity ratio of uranium and its daughters, to prove the presence of ^{210}Po in the mineral sample (Fig. 1). For an efficient retention of ^{210}Po on the column, a chemical pretreatment of the sample was necessary. To this end, the samples were dissolved in various mineral acids, H_2SO_4 , HNO_3 , HCl and H_3PO_4 . However, only digestion using sulfuric acid led to a chemical form which was strongly retained. The digestion solution was evaporated almost to dryness and the residue was dissolved in 50 cm^3 of 0.1M HCl solution, and then passed through the column. The influence of the HCl concentration on the ^{210}Po retention is shown in Table 1 (left).

It may be seen that the best conditions to retain ^{210}Po are met with 0.1M HCl . In a next step, we sought the conditions for recovering the polonium fraction fixed in the column. Various organic (oxalic, lactic, citric) and mineral (HCl , HNO_3 , H_3PO_4) acids were used and it appeared that the best results were obtained with citric acid. Finally, efficient and specific elution of ^{210}Po was obtained using a 0.2M citric acid solution, as is illustrated in Table 1 (Right). No radionuclides other than ^{210}Po could be detected in the eluted solution (Fig. 2).

Finally, electroplating of ^{210}Po from the latter solution was attempted using various electrolyte baths:

TABLE 1

HCl (M)	^{210}Po retained (%)	Citric acid (M)	Activity eluted (%)
0.05	75	0.05	10
0.1	≥ 95	0.10	50
0.5	70	0.20	≥ 95
1.0	40	0.30	40
2.0	10	0.40	0
3.0	5		

Left: Fraction of ^{210}Po retained in the column as a function of HCl concentration in the initial uranium solution.

Right: ^{210}Po elution yield as a function of citric acid concentration.

aqueous solutions of NaF (0.1M), and acetic (0.2M) and oxalic (0.1, 0.5 and 1.0M) acid. The volume of baths was varied between 10 and 20 cm³ with little influence on the results. However, below 10 cm³, the evolution of H₂ on the cathode becomes too important, decreasing the active surface of the disk, the volume of electrolyte bath is reduced also. The best conditions for the highest recovery of ^{210}Po in the form of a uniform and thin film α -source were met with 0.5M oxalic acid solution.

CONCLUSION

^{210}Po was separated from a uranium bulk material by means of DEHPA liquid/liquid extraction. The proper purification of the natural samples and pretreatment gives a ^{210}Po . This element was electroplated over stainless steel polished disks. The best electroplating

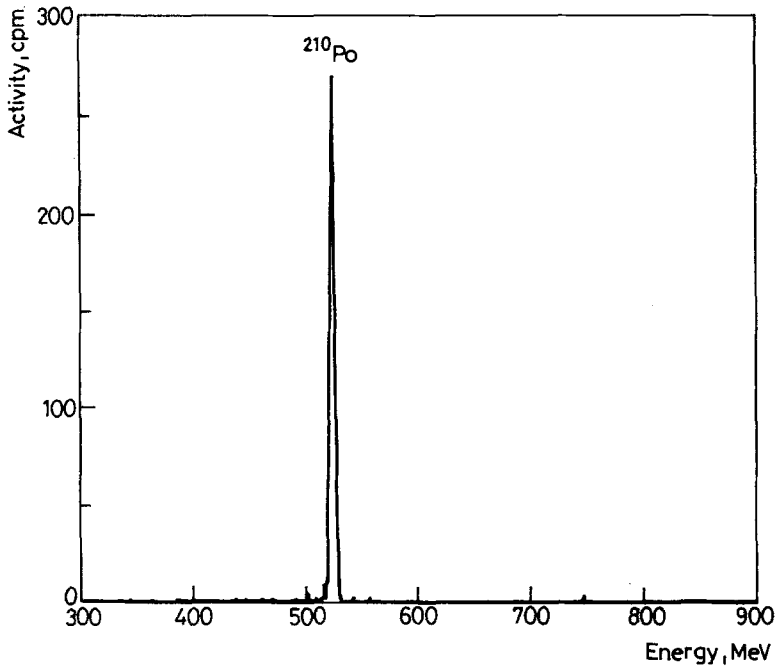


Fig. 2. α -Spectrum of pure ^{210}Po

bath was oxalic acid solution. A very low current density supply was necessary to provide a yield of nearly 95%.

*

The authors wish to thank Mr. G. DuPlâtre of C.N.R.S. Strasbourg, France, for his comments about the work.

REFERENCES

1. R.V. Bogdanov, S.A. Mokhnatkin, Soviet Radiochem., 10 (1978) 617.
2. A.E. Nevissi, J. Radioanal. Nucl. Chem., 148 (1991) 121.
3. S.C. Ehinger, R.A. Pacer, F.L. Romines, J. Radioanal. Nucl. Chem., 98 (1986) 39.