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RAPID ISOLATION OF RADIOSILVER AND CERTAIN OTHER RADIOELEMENTS FROM SOLUTION

Application to a Search for Ag^{121} in U^{235} Fission

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ABSTRACT

Rapid separation of silver from cadmium, indium, and tin was effected by filtration through a thin bed of copper powder. This method was applied to a search for Ag^{121} in the thermal neutron fission of U^{235}. None was detected and this result indicated that either the half-life of Ag^{121} is substantially shorter than the post-fission separation time of 3.5 sec or its yield is inordinately low.

The study was extended to examine the behavior of an additional number of elements in the separation. Of the 28 elements studied, only palladium, silver, gold, and mercury were removed from solution to the extent of 70 % or more. Selenium, ruthenium, tellurium, and iodine were partly retained by the copper to the extent of 5-33 %. Of the remaining 20 metals, with the exception of zirconium and arsenic, appreciably less than 0.1 % of the element adhered to the filter bed. This selectivity may allow wider application of the method.

INTRODUCTION

A previous investigation related to a general study of the nuclear charge distribution in near-symmetric fission suggested the possibility that short-lived Ag^{121} forms in the thermal neutron irradiation of U^{235}.[1] For exploring this possibility a rapid procedure for the separation of silver from a freshly irradiated solution was required. Evaluation of the existence of Ag^{121} would depend upon the measurement of the quantity of Sn^{121} (half-life of 27.5 hours) growing from separated silver for various intervals between the time of irradiation and the time of silver separation. Therefore, the procedure demanded the separation of silver from tin as well as from the intervening decay product elements, cadmium and indium.

This note describes a very rapid method for the separation of silver from the descendant chain elements. It is based upon the well-known reduction of ionic silver by metallic copper. The results of the search for Ag^{121} are reported. Also the behavior of a variety of other elements in this separation is given. The latter information may provide the basis for wider application of the separation procedure.

EXPERIMENTAL

Chemicals and Radioactivities

Copper powder, purified grade, was supplied by the J. T. Baker Chemical Company (Lot No. 29,981).

Other chemicals used were of reagent grade quality.

Radioactive tracers were obtained from the Oak Ridge National Laboratory and Nuclear Science and Engineering Corporation. Their purity was established by gamma-ray spectroscopy.

Filtration Apparatus

The filtration apparatus was adapted from a commercially available filter tower (Tracerlab Division of Laboratory for Electronics). A disc (7/8 in. diameter) of 10-micron pore size stainless steel mesh (Chas. Lowe Co., San Francisco) replaced the existing porous steel disc. The wire mesh supported 1 gram of copper powder. The powder was washed several minutes before use with 10 ml 1.6 N HNO_3. The tower was connected to a suction flask and the filtration rate was 30 ml/sec.

Procedure

A preliminary study of the distribution of silver, cadmium, indium, and tin between copper bed and filtrate was carried out under conditions that approximated those which were to exist in the irradiation experiments. Fifteen milliliters of 4 N HNO_3 containing 100 mg of uranium and 10^6 c/m each of Ag^{110}, Cd^{109}, In^{114}, or Sn^{113} were rapidly filtered through a copper bed which was then immediately washed with 5 ml 1.6 N HNO_3. The copper was dissolved in conc. HNO_3 and then brought to a definite volume. One day after separation the gamma-ray activity of this solution was compared with that of a similarly prepared standard (including the same amount of copper).

In the Ag^{121} experiments, a pneumatically driven sample carrier, (rabbit) loaded with 100 mg U^{235} in 0.6 ml 4 N HNO_3, was irradiated at the Vallecitos Nuclear Test Reactor (VTR) for 10 seconds in a flux of 10^{12} neutrons/cm^2/sec. At the end of the irradiation the rabbit was pneumatically transferred in about 1 sec a distance of 50 ft. to the mobile laboratory. The irradiated solution was transferred by suction to a tube containing 10 ml of 4 N HNO_3, and the rabbit was washed with 5 ml 4 N HNO_3. The combined solution was passed through the copper powder in the filtration apparatus at a definite time with reference to the end of the irradiation. Passage of the solution through the copper was accomplished in about 0.5 sec. The copper bed was washed immediately with 5 ml 1.6 N HNO_3.

Copper and supporting stainless steel mesh were transferred to a centrifuge tube which contained a known volume of standardized tin carrier. Several hours after irradiation concentrated HNO_3 was added dropwise to dissolve the copper and 10-ml saturated NH_4Cl were added to the solution. The solution was adjusted to pH 8 with NH_4OH, and the precipitate which formed was collected by centrifugation. To the precipitate of tin hydroxide were added 10 ml of conc. NH_4OH, and the mixture was stirred thoroughly to solubilize residual copper precipitate. The mixture was centrifuged and after the excess supernatant liquid was decanted, 1 ml of conc. HCl was added to the precipitate. Analysis of the resultant solution for Sn^{121} by the radiochemical procedure of Cowan[2] was begun several hours after the irradiation.

TABLE 1

Quantity of Radioactivity Retained by Copper Bed

Radioactivity	Retained on Copper (%)	Radioactivity	Retained on Copper (%)
Ag^{110}	98.3 ± 1.2*	In^{114}	0.06 ± 0.02
Pd^{109}	85.0 ± 6.5	Tc^{99}	0.05 ± 0.02
Hg^{203}	74.4 ± 5.6	Sr^{85}	0.04 ± 0.01
Au^{195}	72.5 ± 3.0	Zn^{65}	0.04 ± 0.02
$I^{131}(I^-)$	33.3 ± 1.0	Sn^{113}(IV)	0.03 ± 0.01
Te^{135}(IV)	7.2 ± 0.08	Ge^{68}	0.02 ± 0.00
Ru^{106}(III)	6.8 ± 0.2	Mo^{99}	0.01 ± 0.00
Se^{75}(IV)	5.3 ± 1.3	Rb^{86}	0.01 ± 0.00
Zr^{95}	0.68 ± 0.18	Ba^{133}	< 0.01
As^{77}(III)	0.60 ± 0.00	Co^{60}(II)	< 0.01
Nb^{95}	0.09 ± 0.01	Ce^{144}(III)	< 0.01
Ti^{44}	0.08 ± 0.07	Mn^{54}(II)	< 0.01
Sb^{125}(III)	0.08 ± 0.03	Y^{91}	< 0.01
Cd^{109}	0.06 ± 0.02	Tl^{204}(I)	< 0.01

*For silver, cadmium, indium and tin the standard deviations are based upon at least four determinations. The errors for the other radioelements are the average deviation of two determinations from the mean.

For the other radioelements studied (see Table 1), 10^6 c/m of the respective radioactivity in 10 ml of 4 N HNO_3 was filtered through the bed, and the bed was then immediately washed with 20 ml 1.6 N HNO_3. The bed was dissolved and radiochemically analyzed as described above in the preliminary study.

RESULTS AND DISCUSSION

Preliminary experiments indicated that silver was essentially quantitatively reduced and deposited on the copper bed; the retention of cadmium, indium, and tin was less than 0.1 % (Table 1). The procedure could therefore be used reliably to study the formation of Ag^{121} in fission.

In the search for Ag^{121} twelve irradiations were performed and separations were made in the interval of 3.5 to 60 seconds after the end of fission. Sn^{121} was undetected even in the shortest time of separation. Therefore, either the half-life of Ag^{121} is considerably less than several seconds or its fission yield is inordinately low.

The retention at the tracer level of various other radionuclides by the copper bed is shown in Table 1. Of the 28 elements studied, only palladium, silver, gold and mercury were deposited in excess of 70 %. Selenium, ruthenium, tellurium and iodine were partly retained by copper to the extent of 5-33 %. Of the remaining 20 metals, with two exceptions substantially less than 0.1 % of the radionuclide adhered to the filter. The selectivity inherent in the process may provide for the isolation of certain few elements to the exclusion of a significant number of other elements. It seems probable that even greater selectivity may be achieved by modification of the pH and by the action of complexing agents.

Comparison of the oxidation potentials for the electrode half-reactions of these elements with the half-reaction for copper[3] clearly accounts for the results obtained. Where the oxidation potential for the element is more negative than the potential for copper, appreciable reduction and removal from solution occurs. When the potential is more positive only a negligible fraction of the element is retained by the filter.

It is noteworthy that the kinetics of the oxidation-reduction reaction are extremely rapid. For silver, for example, the reaction is complete within the time required for the solution to pass through the copper filter bed (about 1/2 sec.).

Current investigation is concerned with the use of this process to remove silver and palladium separately from solution. Because of its inherent speed such a procedure should afford characterization of the half-lives and yields of short-lived species of these two elements in the region of symmetric fission.

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