where E and L refer to 10-min and 70-h separations of ^{77}As. A_{Ga} is the ^{77}As counting rate at separation time after a Ga isolation, A is the ^{77}As counting rate at separation time with no separation of As precursors, f_1 is the fraction of ^{77}Ga decaying to ^{77m}Ge, f_4 is the fraction of ^{77m}Ge decaying to ^{77}As, $\lambda_1, \lambda_3, \lambda_4$ are the decay

constants of ^{77}Ga, ^{77g}Ge, and ^{77}As, respectively, t_i is the length of irradiation time, and t_d is the time between the end of irradiation and Ga separation. The last term of the numerator is the correction factor for the ^{77}Ga decay during the irradiation time and during the time between the irradiation and the Ga separation.

Identification of 5.3-sec ^{118}Ag as a Product of ^{235}U Fission*

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A search for ^{118}Ag was carried out as part of a study on the distribution of nuclear charge in symmetric fission. A method was developed for the separation of silver from its palladium precursor and cadmium descendant, based upon the rapid reduction and retention of silver by finely divided copper. Silver was separated at various times after thermal-neutron fission of ^{235}U. The copper powder with affixed silver was dissolved in concentrated nitric acid and radiochemically purified for the analysis of ^{118}Cd derived from its silver precursor. The linear relationship between the logarithm of ^{118}Cd activity and separation time led to the identification of ^{118}Ag. The half-life of this radionuclide was determined to be $5.3_{-0.7}^{+0.9}$ sec.

I. INTRODUCTION

A SEARCH for ^{118}Ag was carried out as part of a study on the distribution of nuclear charge in symmetric fission. Other members of the mass-118 chain already identified when this study began were ^{118}Cd (49 min) and its ^{118}In daughter (5.1 sec).[1]

A method was developed for the separation of silver from its palladium precursor and cadmium descendant, based upon the rapid reduction of silver by finely divided copper.[2] The irradiated uranium solution was washed through copper powder and the nonreducible constituents separated from the depositing silver. Palladium, whose oxidation potential is more negative than that of silver, was made nonreducible by complexation with hexamethylenetetramine (tren) in a citrate solution. Cadmium, whose oxidation potential is positive with respect to copper, remained in solution in the oxidized state.

By this procedure silver was separated from thermal-neutron-irradiated ^{235}U at various times after fission. The copper powder and affixed silver were dissolved in concentrated nitric acid and radiochemically purified for the analysis of daughter ^{118}Cd. A plot of ^{118}Cd activity as a function of separation time represented the radioactive decay of the ^{118}Ag precursor.

It was anticipated that the half-life of ^{118}Ag would be relatively short. Therefore, rapid transfer of solutions after the irradiation was necessary. An automatic

system was developed which afforded the separation of silver from the fission solution in less than 1 sec after irradiation.

II. EXPERIMENTAL

Preparation of Uranium Solution

The uranium solution was prepared as follows: One gram of ^{235}U metal (93.22% isotopic enrichment) was dissolved in concentrated nitric acid. For elimination of the excess acid, the solution was evaporated to dryness, the residue redissolved in water, and again evaporated to dryness. The uranyl nitrate was dissolved with warming in 5 ml of recrystallized saturated sodium citrate. This solution was added to one composed of 40 μl of palladium carrier [(39.4 mg $Pd(NO_3)_2$)/(ml 1N HNO$_3$)], 100 μl of silver carrier [(162 mg AgNO$_3$)/(ml 1N HNO$_3$)], and 2.4 ml of 3M tren solution. After warming under the hot-water tap to complete the dissolution, an additional 2.5 ml of water were added.

Separation Apparatus

The chemical-separation apparatus (Fig. 1) consisted of a hypodermic needle, wash reservoir, chemical-separation chamber, and suction flask. These parts were interconnected by rubber tubing in which continuity of flow could be interrupted by the pinch of separately controlled solenoid valves.

The chemical-separation chamber consisted of a glass cylinder ($\frac{1}{2}$ in. long) interposed between two socket joints (size 18/9). Between the cylinder and lower socket joint was a 10-μ pore stainless-steel mesh, $\frac{7}{8}$ in. diam, which supported 500 mg of copper powder. The

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[1] C. E. Gleit and C. D. Coryell, Phys. Rev. **122**, 229 (1961).
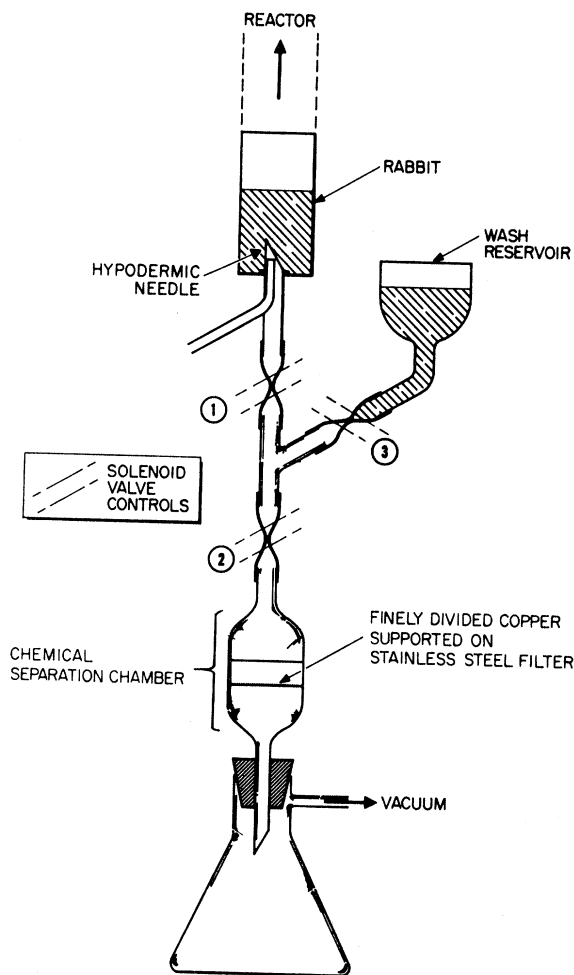[2] H. V. Weiss and W. H. Reichert, Anal. Chim. Acta **34**, 199 (1966).

FIG. 1. Chemical separation apparatus.

chemical-separation chamber was secured by a ball-and-socket clamp and then the spaces between cylinder and socket joints were sealed with a coating of paraffin to ensure maintenance of vacuum during operation. The powder was washed several minutes before use, first with 10 ml of $1.6N$ nitric acid and then with a similar volume of distilled water.

Irradiation and Separation Procedure

One ml of the uranium solution was placed in each of a number of pneumatically driven sample carriers (rabbit). Each was irradiated 3 sec in the Vallecitos Nuclear Test Reactor in a flux of approximately 10^{12} neutrons cm^{-2} sec^{-1}. A gold foil taped to the rabbit served as a monitor of the number of fissions as before.[3]

At the end of the irradiation the rabbit was pneumatically driven onto the hypodermic needle in the laboratory (12 ft away) in about 0.25 sec. Coincident

[3] H. V. Weiss and N. E. Ballou, J. Inorg. Nucl. Chem. 27, 1917 (1965).

with ejection of the rabbit from the reactor a preset interval timer graduated in 0.1 sec was actuated for control of the start of the chemical separation. Upon completion of the run of the timer an impulse was transmitted to a programmer (Beattie-Coleman, Inc., Anaheim, Calif.) to initiate and control the chemical separation. The program was started at various times in the interval 0.3–60 sec after the end of the irradiation.

The programmer sent signals of definite duration and sequence to the three solenoid valves, which operated the chemical-separation apparatus. Solenoid valves 1 and 2 (Fig. 1) were opened simultaneously for 0.32 sec to transfer the uranium solution from the rabbit into the evacuated system and through the copper bed. Valves 2 and 3 were opened over the next 0.64 sec to permit water in the reservoir to wash through the system. At the conclusion of this operation the support disk with the copper-silver powder was removed from the apparatus for the ^{118}Cd analysis.

Radiochemical Determination of ^{118}Cd

Standardized cadmium carrier [(10 mg Cd)/(ml $1N$ HCl)] was added to the isolated sample, after which the copper-silver powder was dissolved in 3 ml of concentrated nitric acid. The solution was separated from the support disk by decantation. The volume of the solution was raised to 50 ml with $3N$ hydrochloric acid. About 8 min after the irradiation, hydrogen sulfide gas was bubbled through the solution for 2–3 min. The copper and silver sulfides precipitated and were collected quantitatively on a fine-sintered-glass funnel and reserved for silver carrier yield determination.

The cadmium-containing filtrate was alkalinized by the addition of sodium hydroxide pellets. Cadmium sulfide precipitated and was collected on sintered glass, and the filtrate was discarded. The cadmium salt was dissolved in 5–10 drops of concentrated hydrochloric acid and the solution sucked through the filter; the filter was further washed with water. The volume of this solution was brought to approximately 20 ml. Two drops of a solution of 10% sodium nitrite and 5–10 mg of iron carrier were added. Ferric hydroxide was precipitated by the addition of ammonium hydroxide and then separated from the solution by filtration. The filtrate was heated to a boil and about 10 pellets of sodium hydroxide were added to precipitate cadmium hydroxide. The precipitate was collected on sintered glass and then dissolved in 10 ml of $2N$ hydrochloric acid. Thirty ml of water were added to the dissolved cadmium, and hydrogen sulfide gas was passed through this solution for 3 min. The precipitated cadmium sulfide was collected on a filter paper disk, washed with water and ethanol, mounted on a brass planchet, and then covered with pliofilm. Samples were usually ready for counting within about 40 min after the end of the irradiation.

The β radiations were measured through a 200-mg/cm^2 aluminum absorber in a gas proportional counter for a minimum of 20 h. The ^{118}Cd count rate was corrected for ^{117}Cd contribution (as discussed below), decay from the time of irradiation, cadmium carrier yield, and silver carrier yield in the initial separation and finally was normalized for 10^{11} fissions.

Silver and Cadmium Carrier Yield Determinations

Silver and cadmium carrier yield were determined through atomic absorption spectrometry. Samples were prepared for analysis as follows.

Cadmium determination: Upon completion of the β count the filter-paper disk and associated pliofilm covering were removed from the planchet and transferred to a test tube. Cadmium sulfide was dissolved in 10 ml of 6N hydrochloric acid; the solution was heated to evolve hydrogen sulfide, the solids were filtered out on sintered glass. The solids were thoroughly washed with 0.1N hydrochloric acid, and the wash was combined with filtrate. Each step was carefully performed to ensure quantitative recovery. The solution was diluted to a definite volume with water, and a small aliquot was diluted with 0.1N hydrochloric acid to bring the cad-

mium concentration within the range of measurement. The quantity of cadmium in this solution was determined by comparison of the absorptivity, at 228.8 mμ, with that of a cadmium standard also prepared in 0.1N hydrochloric acid.

Silver determination: The copper sulfide-silver sulfide fraction was quantitatively dissolved from the sintered glass with about 100 ml of aqua regia. A known quantity of ^{110}Ag activity was added, and the sample was evaporated to dryness. The residue was baked over a bunsen burner to evolve free sulfur. About 30 ml of concentrated hydrochloric acid, and as much concentrated nitric acid as needed to induce complete dissolution, were added, and the solution was again evaporated to dryness. The residue was dissolved in 10–20 ml of concentrated hydrochloric acid; the solution was reduced in volume to 2–3 ml by evaporation. About 0.5 ml of concentrated nitric acid and 30 ml of water were added. The precipitated silver chloride was separated by centrifugation, dissolved in 100 μl of 10% sodium cyanide and reprecipitated by the addition of water. The precipitate was again separated and dissolved, as above. The ^{110}Ag in solution was counted in a sodium iodide (thallium activated) γ-well counter and compared with a standard to correct for the loss of carrier in the
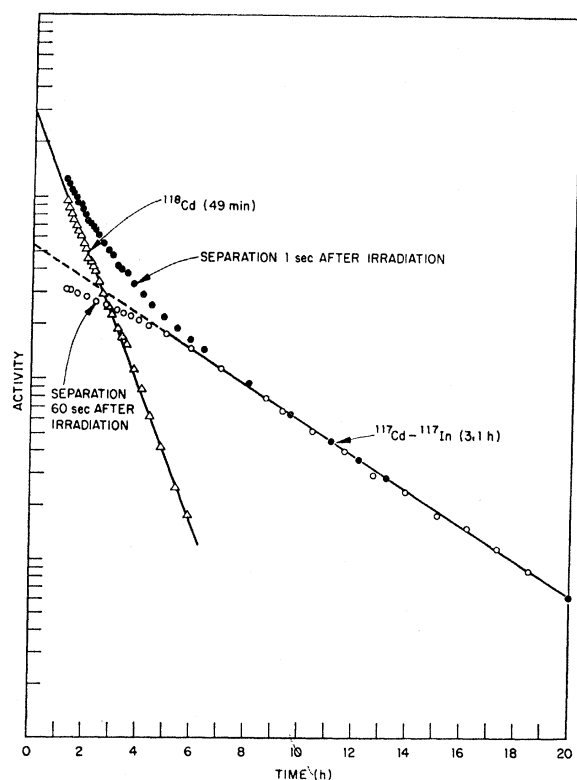

FIG. 2. Cd radioactivity derived from fission-formed Ag separated 1 and 60 sec after irradiation. (The β radiations were measured through a 200-mg/cm^2 aluminum absorber in a gas proportional counter.)
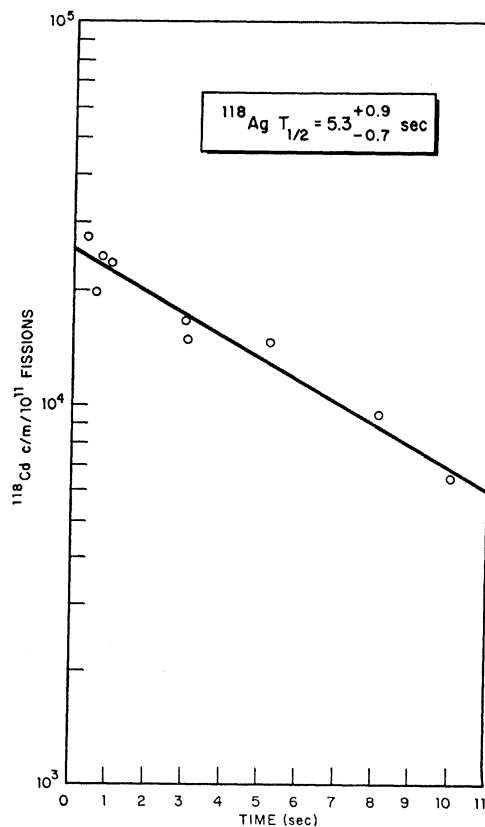

FIG. 3. Growth of ^{118}Cd from ^{118}Ag at various times of Ag separation. (The time of separation is the difference in time between the end of irradiation and the initiation of filtration.)

purification. The solution was then diluted to an appropriate volume, and its absorptivity was compared with that of silver standards of similar cyanide composition at a wavelength of 328.1 mμ.

The error associated with both the silver and cadmium yield determinations was about 2%.

III. RESULTS AND DISCUSSION

In the rapid separation of silver from palladium and cadmium, 40–50% of the silver was reduced and retained by the copper bed. Experiments with radioactive tracers and carriers had indicated that contamination by cadmium and palladium was only 0.15 and 0.7%, respectively.

Typical decay curves for the radioactive cadmium, derived from silver separated 1 and 60 sec after fission, are shown in Fig. 2. The curves were normalized so that the straight-line segments of both coincided. The linear portions appear at later times, and are ascribable to ^{117}Cd and ^{117}In isomers derived from the ^{117}Ag (1.1 min) precursor. These components decay with an apparent half-life of 3.1 h. The silver sample isolated 1 sec after irradiation shows an initial rapid decay attributable to ^{118}Cd. The half-life of ^{118}Ag is such that only a negligible quantity exists at 60 sec. Thus, the curve for the later separation time shows only the decay of ^{117}Cd isomers into their decaying ^{117}In daughter products. (Interference from ^{115}Cd, also present in the radiochemically purified cadmium fraction, was eliminated by absorption of its relatively weak β radiations with aluminum.) With the use of the empirically determined contribution of ^{117}Cd-^{117}In, the ^{118}Cd component was reliably resolved.

The normalized counting rate of ^{118}Cd for nine silver separations extending in time from 0.3 to 10 sec after fission appears in Fig. 3. The time of separation was the time between the end of irradiation and the initiation of filtration through the copper bed. The relationship between the logarithm of the counting rate and the separation time is linear. Analysis by the method of least squares gives a half-life of $5.3_{-0.7}^{+0.9}$ sec for the ^{118}Ag.

Calculation of the independent yield of ^{118}Ag depends upon characterization of its precursor. The existence of the ^{118}Pd precursor has already been demonstrated[4]; this nuclide decays with a half-life of approximately 2 sec. Refinement of the ^{118}Pd half-life value, determination of the total 118-mass chain yield, and calculation of the ^{118}Pd cumulative fractional yield are yet to be accomplished. This information will afford an accurate measurement of the ^{118}Ag independent yield and is currently being acquired. The yield value is needed for further evaluation of the distribution of nuclear charge in this region of fission.

ACKNOWLEDGMENTS

[4] H. V. Weiss and J. L. Elzie (to be published).

Identification and Yield of 5.0-sec ^{117}Pd in the Thermal-Neutron Fission of ^{235}U†

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As part of a study of nuclear charge distribution in the region of symmetric fission, evidence for the existence and cumulative fission yield of ^{117}Pd was sought. Palladium was isolated free from silver and cadmium at different times after fission, and the quantity of descendant ^{117}Cd activity produced was measured. A half-life for ^{117}Pd of $5.0_{0.7}^{+0.5}$ sec was derived from these data. Through a comparison of ^{117}Cd activity produced by ^{117}Pd with the total ^{117}Cd activity produced by all isobars, a cumulative fractional chain yield of 0.669 was computed for ^{117}Pd. From this yield the most probable nuclear charge Z_p for the 117-mass chain was estimated.

I. INTRODUCTION

AS an extension of a study on the distribution of nuclear charge in symmetric fission of ^{235}U, ^{117}Pd was identified and its cumulative fractional chain yield measured.

Palladium was separated from its descendants, silver and cadmium, at various intervals after irradiation by a procedure previously described.[1] The quantity of descendant ^{117}Cd was measured in the palladium sample isolated. A plot of ^{117}Cd activity as a function of time

[1] H. V. Weiss and W. L. Reichert, J. Inorg. Nucl. Chem. 28, 2067 (1966).