Nuclear Charge Distribution in Symmetric Fission of ^{235}U with Thermal Neutrons: Yields of ^{117}Ag, ^{118}Ag, and ^{118}Pd†

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^{118}Pd was identified; it decays with a half-life of 3.1±0.3 sec. Its cumulative fractional chain yield is 0.29±0.03. The half-lives of ^{117}Ag and ^{118}Ag were verified. Their independent fractional chain yields are 0.35±0.05 and 0.55±0.10, respectively. The width parameter σ in the 118-mass chain is 0.65±0.16, with the assumption that the distribution of nuclear charge is Gaussian. The most probable nuclear charge Z_p in the 117- and 118-mass chains is 46.22±0.17 and 46.88±0.12, respectively. These values of Z_p range close to those calculated for unchanged charge distribution.

INTRODUCTION

In a study on the distribution of nuclear charge in the region of symmetry for fission of ^{235}U with thermal neutrons, ^{118}Pd was identified and its cumulative fractional chain yield was measured. The reported half-lives of ^{117}Ag [1,2] and ^{118}Ag [3] were verified by a procedure different from that previously used, and their independent fractional chain yields were determined. From these data, and a previously reported value for the cumulative fractional chain yield of ^{117}Pd, [4] the most probable nuclear charge Z_p was computed for the two mass chains. A measure of the width parameter σ for Gaussian distribution of nuclear charge was obtained for the 118-mass chain.

EXPERIMENTAL

General Procedure

^{235}U (93.22% enrichment) was dissolved in a medium suitable for the separation of Ag or Pd from the other and from Cd. In order to preclude interference from isobars for Pd isolations the solution consisted essentially of concentrated HBr,[4] while, for Ag isolations, a complexing medium of sodium citrate and hexamethylenetetramine was used.[3]

Samples were irradiated for a definite time in the Vallecitos Nuclear Test Reactor, in a flux of about 5×10^{11} neutrons cm^{-2} sec^{-1}. They were transported into, and out of, the reactor with a rapid pneumatic system. For the separation of Ag or Pd, the sample carrier was impaled after irradiation upon a hypodermic needle, and the solution was withdrawn by suction at a specified time. The solution passed through copper powder and the desired element was retained by the solid substrate. Except for an experiment in which Cd daughter product was stripped from deposited Ag (see below), the Cu powder was dissolved with concentrated HNO_3. This solution was purified for the determination of the daughter Cd radioactivities.[3]

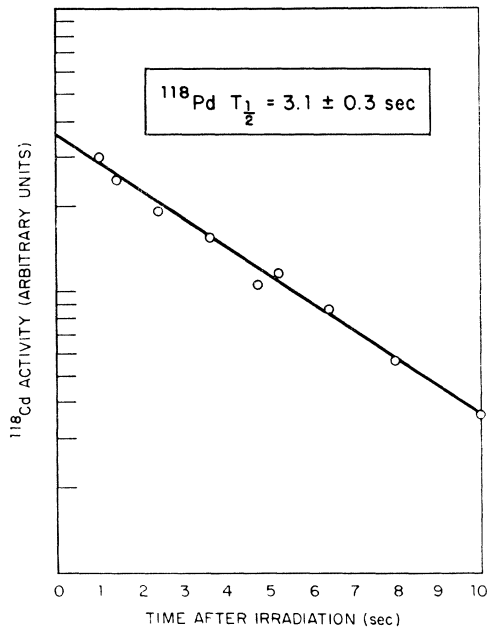

FIG. 1. Growth of ^{118}Cd activity from palladium separated at various times after fission.

Methods previously reported were used for counting ^{117}Cd [4] and ^{118}Cd,[3] for determining the Cd,[3] Ag,[3] and Pd [4] carrier yields, and for correcting minor variations in the neutron flux.[3]

^{118}Pd Half-Life

The ^{118}Pd half-life was determined the same way as was that of ^{117}Pd,[4] except that the ^{118}Cd granddaughter product was measured.

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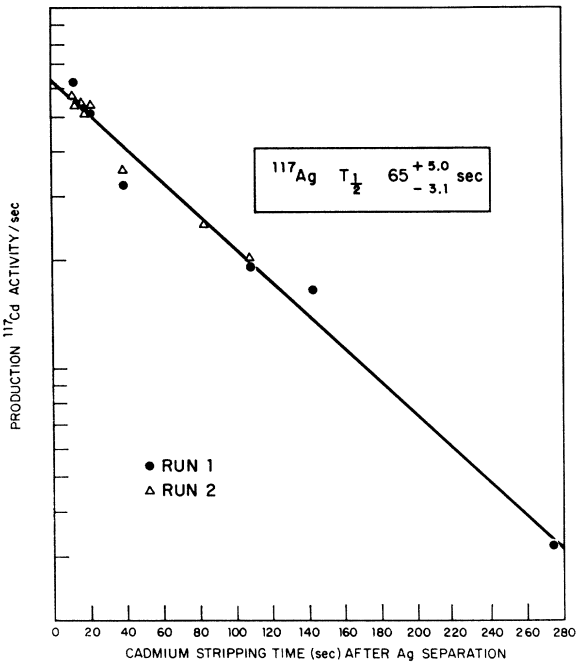

FIG. 2. Rate of production of ^{117}Cd from ^{117}Ag at various stripping times after Ag separation.

^{117}Ag and ^{118}Ag Half-Lives

For verification of the half-lives of ^{117}Ag and ^{118}Ag, the uranium solution was prepared in the following manner: Enriched uranium (4 g) in the form of uranyl nitrate was dissolved in 20 ml of saturated sodium citrate solution by heating. Small volumes of water were added to replenish evaporated solvent during the slow dissolution process. The uranyl citrate solution was diluted to 25.0 ml with distilled water after cooling. To this solution were added 1 μl of ^{110}Ag tracer solution, 9.6 ml of $3M$ hexamethylenetetramine, 0.16 ml of Pd carrier solution [39.4-mg $Pd(NO_3)_2$/ml in $1N$ HNO_3], and 5.0 ml of distilled water.

The uranium solution was irradiated for 20 (or 80) and 5 sec in the ^{117}Ag and ^{118}Ag half-life determinations, respectively. Within 1 sec after irradiation the solution was passed through 500 mg of copper powder that previously had been washed with $4N$ HNO_3. Within 1 sec the copper was washed with 16 ml of $0.1N$ HNO_3. At certain intervals after this initial wash, the Cd daughter products were quantitatively stripped[5] from the radiosilver deposited on the copper by washing 8-ml portions of $0.1N$ HNO_3 through the bed.

Each wash solution was collected in a separate vessel and was analyzed for ^{117}Cd and ^{118}Cd. These fractions were prepared for radiochemical purification and subsequent processing by the addition of 10 mg of Cd, 10 mg of Ag, and 50 mg of Cu carriers. The initial copper sulfide precipitate in the purification process was reserved for the determination of the quantity of silver that leached from the copper substrate with each washing. This determination was carried out by the isotopic dilution technique.[3] In addition to the usual corrections, the ^{117}Cd and ^{118}Cd activities were adjusted for the small amount of silver lost (0.3–0.5%) with each stripping.

TABLE I. Yields of Pd and Ag isotopes of mass 117 and 118 in thermal-neutron fission of ^{235}U.

Mass	Pd[a]	Ag[b]
117	0.67±0.07	0.35±0.05
118	0.29±0.03	0.55±0.10

[a] Cumulative fractional chain yields.
[b] Independent fractional chain yields.

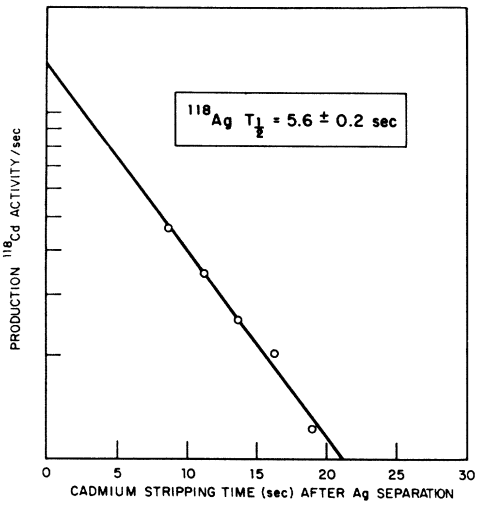

FIG. 3. Rate of production of ^{118}Cd from ^{118}Ag at various stripping times after Ag separation.

^{117}Ag Independent Fractional Chain Yield

A ^{235}U solution made up for the separation of Ag from Pd and Cd was irradiated for 3 sec. The silver was separated 1 sec after irradiation. After an 8-min delay to permit decay of ^{117}Ag to ^{117}Cd, Cd carrier was added and radiochemical purification of the daughter product was undertaken.

Total ^{118}Cd Activity Yield

^{235}U solutions of composition for Pd or Ag separation were irradiated in duplicate for 20 sec. The ^{118}Cd activity was measured on aliquots of the irradiated solutions which were purified according to a Cd radiochemical procedure developed for the determination of total ^{117}Cd activity.[4]

[5] H. V. Weiss, N. E. Ballou, J. L. Elzie, and J. M. Fresco (unpublished).

RESULTS

The corrected counting rate of ^{118}Cd for nine Pd separations, made 1–10 sec after fission, appears in Fig. 1. Separation time is defined as starting at the end of irradiation and ending at the midpoint of filtration of the fission solution through the Cu bed. The relationship between the logarithm of the counting rate and the separation time is linear. Analysis of the data by the method of least squares gives a half-life of 3.1±0.3 sec for ^{118}Pd.

For the calculation of the ^{118}Pd cumulative fission yield, a correction was made for the loss of ^{118}Ag daughter product from the copper bed during the washing interval.[4] Correction was also calculated for the decay of ^{118}Pd during the 3-sec irradiation period through

$$R_1=\lambda_1 A_1^0/[1-\exp(-\lambda_1\tau)],$$

where R_1 is the rate of formation of ^{118}Pd as reflected by ^{118}Cd activity, λ_1 is the decay constant of ^{118}Pd, A_1^0 is the ^{118}Cd activity from ^{118}Pd at the end of the irradiation, and τ is the irradiation time. R_1 was compared with the ^{118}Cd production rate for the total chain and a cumulative fractional chain yield of 0.29±0.03 was computed for ^{118}Pd.

The results of the experiments that were designed to verify the half-lives of ^{117}Ag and ^{118}Ag appear in Figs. 2 and 3, respectively. The ordinate in these figures represents the quantity of the particular Cd radioactivity produced per second; the abscissa refers to the midpoint in time between strippings. The last point in the ^{117}Ag experiment, however, is placed at the time at which the average rate of production (prior to the last stripping) occurs, with the assumption that the half-life of ^{117}Ag is 66 sec.

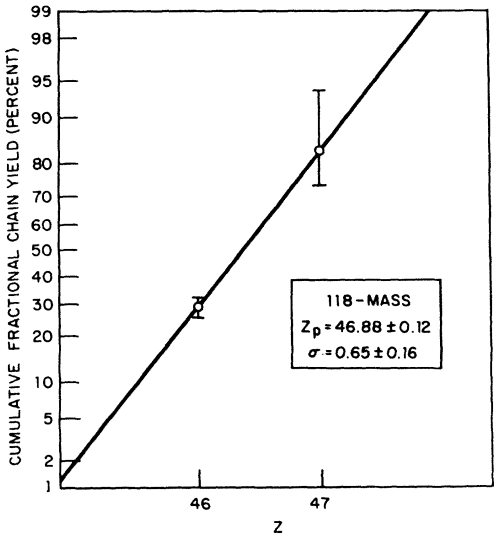

FIG. 4. Probability-scale plot of fractional cumulative yields measured in the 118-mass chain.

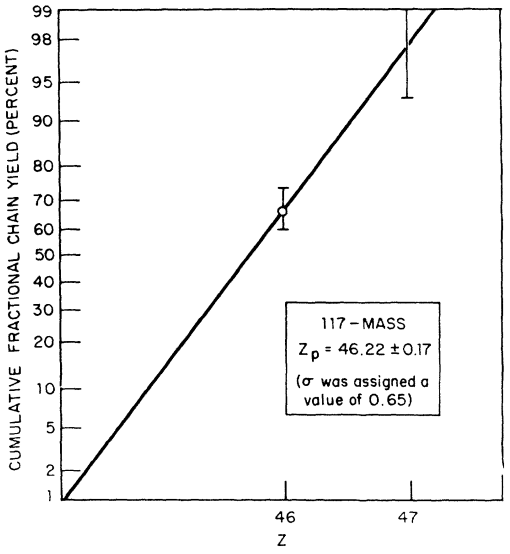

FIG. 5. Probability-scale plot of fractional cumulative yields measured in the 117-mass chain.

The half-life of ^{117}Ag, derived from the data of eight separations in each of two runs, is $65.0_{-3.1}^{+5.0}$ sec and the above-mentioned assumption is consistent with this half-life.

The half-life of ^{118}Ag, determined from the values of five strippings over a period of about two half-lives, is 5.6±0.2 sec.

The ^{118}Ag independent yield was calculated from the previously reported data,[3] which were obtained by the measurement of ^{118}Cd activity derived from the separation of Ag at different times after irradiation. The ^{118}Cd activity A_2 at a given Ag separation interval was corrected for decay from the end of the irradiation, and for growth from ^{118}Pd during this interval, by means of the equation

$$A_2^0=[\exp(-\lambda_2 t)]^{-1}\times\left(A_2-\frac{\lambda_1 A_1^0}{\lambda_2-\lambda_1}[\exp(-\lambda_1 t)-\exp(-\lambda_2 t)]\right),$$

where A_2^0 is the relative quantity of ^{118}Ag (reflected by ^{118}Cd activity) at the end ot the irradiation, λ_2 is the decay constant of ^{118}Ag, t is the time of separation with respect to the end of irradiation, and λ_1 and A_1^0 are as defined earlier.

The relative rate of ^{118}Ag production R_2 was calculated from the equation

$$R_2=\frac{\lambda_2}{1-\exp(\lambda_2\tau)}\left[A_2^0-R_1\times\left(\frac{1-\exp(-\lambda_2\tau)}{\lambda_2}-\frac{\exp(-\lambda_1\tau)-\exp(-\lambda_2\tau)}{\lambda_2-\lambda_1}\right)\right],$$

where τ is irradiation time, as defined earlier.

TABLE II. Values of Z_p, A', and $A'(92/236)$ for mass chains 117 and 118.

Mass	Z_p	A' [a]	$A'(92/236)$	$Z_p-A'(92/236)$
117	46.22±0.17	118.9	46.35	−0.13±0.17
118	46.88±0.12	119.8	46.70	0.18±0.12

[a] Fragment mass before neutron evaporation.

Comparison of R_2 with the ^{118}Cd production rate for the 118-chain resulted in a fractional independent yield of 0.55±0.10 for ^{118}Ag.

The ^{117}Ag data obtained from four separations of Ag made at 1 sec after fission were treated precisely as described above with the use of a cumulative fractional yield of 0.67±0.07 for ^{117}Pd.[4] The independent yield for ^{117}Ag was determined to be 0.35±0.05.

DISCUSSION

The value for the half-life of ^{117}Ag agrees with a value of 1.1 min as originally reported by Coryell and co-workers.[1] Their data were acquired by the solvent extraction of Cd from separated Ag at various intervals, and by the subsequent measurement of ^{117}Cd. Kiefer[2] determined the half-life of this radionuclide to be 42.4±3.2 sec. However, his procedure depended on γ-ray spectrometric measurement of an ^{117}In granddaughter product. This procedure is somewhat less direct than those which give the longer half-life, and thereby is subject to greater uncertainty.

The half-life found for ^{118}Ag agrees closely with the value 5.3±0.7 sec previously reported.[3] However, the greater precision of the present determination reflects the simplicity of the stripping technique, as compared with the usual method of multiple separations from the fission solution at different times after irradiation.

Table I shows the fission yields for palladium and silver in the 117- and 118-mass chains. From these yields, and with the assumption that the distribution of nuclear charge is Gaussian, a value for the most probable nuclear charge Z_p was estimated for each chain by the method of Wahl and co-workers.[6] A value for the width parameter σ was derived for the 118-mass chain.

Figures 4 and 5 show the probability-scale plot of the fractional cumulative yields for the 118- and 117-mass chains, respectively. From Fig. 4 the value of σ was determined to be 0.65±0.16, and that of Z_p to be 46.88±0.12. (The uncertainty in Z_p represents the deviations which were computed from the uncertainties in the two cumulative fractional chain yields).

In the 117-mass chain, because of the fact that the mean fractional cumulative yield for ^{117}Ag is 1.02, the value of σ is not determinable from the data. However, if σ is assumed to be 0.65, as in the 118-mass chain, then Z_p is 46.22±0.17. It may be remarked that the assumed width parameter is consistent with the lower limit of the silver cumulative yield (Fig. 5). (The uncertainty in Z_p in this case combines the uncertainty in the yield of ^{117}Pd and an assigned uncertainty of 25% in the value of σ.) Further, as was previously indicated,[4] Z_p is insensitive to variation of σ for this mass chain since ^{117}Pd is situated close to Z_p.

Experimental information about charge distribution for mass numbers in the region of symmetrical division was previously unavailable. It was of interest to compare Z_p of each chain with the charge of the fragment for an unchanged charge distribution. Therefore Z_p was considered as a function of A', the fragment mass number before neutron evaporation. The value of A' was estimated from the neutron-emission data reported by Apalin *et al.*[7]

Table II gives the values of experimentally determined Z_p, the value of A' for the respective chain, and $A'(92/236)$, the theoretical charge for a fragment of unchanged charge distribution formed from the fissioning nucleus. The difference between Z_p and $A'(92/236)$ is also shown in the table. In these two chains this difference is small by comparison with differences which exist in other regions of fission, and the difference is not necessarily regarded as significant.

Data obtained from thermal-neutron fission of ^{235}U indicate that there is a deviation from unchanged charge distribution of about 0.6 charge units in favor of the light fragment for mass numbers produced in high yield.[8] The interpretation of this deviation from unchanged charge distribution is that, at some time prior to scission, a redistribution of charge occurs so that the light fragment has a higher charge density than the heavy fragment.[9] It was pointed out that this effect was consistent with the "neck model" of fission in which the heavy- and light-fragment cores are built around closed shells of nucleons, and in which the density of charge in the heavy-fragment core is less than that of the light-fragment core.

The results for the 117-mass and 118-mass chains, in which no significant deviation is observed between Z_p and that expected from unchanged charge distribution, indicate that, in symmetric fission, there is no noticeable redistribution in charge and that the fragments do not manifest influence of closed shells of nucleons.

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[9] K. Wolfsberg, Phys. Rev. **137**, B929 (1965).