Near-Symmetric Fission—Identification and Yield of Cd^{121}†

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(Received 5 March 1965)

Evidence was acquired for the formation of Cd^{121} in the thermal-neutron fission of U^{235}. Its half-life and cumulative fission yield are $12.8_{-0.3}^{+0.4}$ sec and $(6.4\pm0.5)\times10^{-3}\%$, respectively. From the cumulative fractional chain yield, a value of ≤ 48.45 was inferred for the most probable nuclear charge (Z_p) of the primary fission fragment formed in this chain. The addition of this value to a partially formulated empirical Z_p function suggests that the value of Z_p proceeds toward the point of symmetrical fission by a gentle rather than abrupt approach. The mass-121 chain yield was redetermined and the value obtained was $(1.2\pm0.1)\times10^{-2}$ %. This value is based on a measured cumulative yield of $(1.1\pm0.01)\times10^{-2}$ % for Sn^{121} (27.5 h) and a literature value of 0.08×10^{-2} % for Sn^{121m} (25 yr).

I. INTRODUCTION

A STUDY of the nuclear charge distribution in near-symmetric fission is in progress. As part of this work the formation of two In^{121} isomers (30 sec and 3.1 min) in the fission of U^{235} was examined.[1] The results of that study suggested that these nuclides were at least partly dependent for their formation upon short-lived fission precursor(s). This report describes the search for the immediate Cd^{121} precursor(s).

A rapid method was developed for the separation of cadmium from its descendants, indium (trivalent) and tin (di- and tetravalent). The procedure was based upon the solubility of a cadmium-ammine complex and the insolubility of tin and indium in an ammoniacal oxalate solution. Further decontamination from daughter elements was effected by filtration of the precipitated suspension through an adsorbent. With this method the quantity of descendant Sn^{121} (27.5 h) was measured in Cd samples isolated at various times after irradiation. The curve which resulted from a plot of Sn^{121} activity as a function of separation time constituted a decay curve for Cd^{121}.

Also, data were obtained on the mass-121 chain-fission yield in U^{235} thermal neutron fission. Only an unpublished value has previously been given for this yield.[2]

From the Cd^{121} yield the most probable nuclear charge (Z_p) for the mass-121 chain was calculated and compared with an empirical Z_p function.

II. EXPERIMENTAL

Procedure for the Determination of Cd^{121}

A 0.1 *N* HCl solution of enriched U^{235} (100 or 200 mg), cadmium carrier (28 mg Cd^{++}), and indium carrier (20 mg In^{3+}) was contained in a rabbit. A gold foil taped to the rabbit was used to monitor the neutron flux, a measurement from which the number of fissions was calculated.[1] Each rabbit was irradiated for 10 sec in the Vallecitos nuclear test reactor (NTR) in a flux of $\sim10^{12}$ neutrons sec^{-1} cm^{-2}. The cadmium ratio was determined to be 2.7 and is defined as the ratio of total activity to the activity of cadmium-covered gold (cadmium and gold thickness 20 mil and 0.5 mil, respectively). The rabbit was immediately returned to the laboratory area with the NRDL pneumatic rabbit transport facility.[3] At a predetermined time the irradiated solution was transferred to a vessel that contained 22 ml of hot precipitating solution (1.5 *N* NH_4OH and 0.05 *M* $(NH_4)_2C_2O_4$). The precipitated fission solution was immediately poured through a bed of Bentone 38 (primarily a hectorite clay, supplied by the National Lead Company, New York, N. Y.). The clay (0.5 g) was supported by a stainless-steel mesh (2-μ pore size), $2\frac{1}{2}$-in. diam. The filtration was completed in about 1 sec. The suction flask which collected the filtrate contained 2 ml of standardized tin carrier.

The filtrate was acidified with concentrated HCl and was diluted to 25.0 ml. A colorimetric procedure[4] was applied to an aliquot of this solution to determine the cadmium yield. Several hours after irradiation, the remainder of the diluted filtrate was treated first with 10 *N* NaOH until the appearance of permanent slight turbidity and then with 1.5 ml of concentrated HCl. After this pH adjustment Sn^{121} was determined by the radiochemical procedure of Cowan.[5] The decay of the beta-ray activity was measured on a gas-flow proportional beta-ray counter. After subtraction of a long-lived tail, the decay curve was resolved into components, one with a half-life of 27.5 h (Sn^{121}). The counting rate was corrected for decay, the chemical yield of tin and cadmium, and the aliquot size. The self-absorption correction for Sn^{121} was made with a factor derived for Zr^{95} which has a beta ray with E_{max} close to that of Sn^{121}. [The Zr^{95} counting efficiency was interpolated from an empirically determined curve which related counting

† This work was sponsored by the U. S. Atomic Energy Commission.

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[2] E. P. Steinberg, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Vol. 9, p. 913.

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efficiency (as evaluated from 4π beta-ray counting) to beta-ray energy.]

Procedure for the Mass-121 Chain-Fission Yield Determination

Two rabbits each with 100 mg of uranium solution were irradiated for 300 sec in the Vallecitos NTR. About 18 h after the irradiation, the solutions were diluted to 25.0 ml with water after the addition of standardized tin carrier. Two 10-ml aliquots from each sample were radiochemically analyzed for tin. Beta-decay measurements were made as before, and the data were resolved into components of 27.5 h (Sn^{121}), 9.4 day (Sn^{125}), and 125 day (Sn^{123}) by a computer program method. Mo^{99} radiochemical analyses[6] were performed in duplicate on 2-ml aliquots from each sample to estimate the total number of fissions.

III. RESULTS AND DISCUSSION

In the rapid separation procedure of cadmium from indium and tin, 40-50% of the cadmium was recovered in the filtrate. Preliminary experiments with appropriate radioactivities indicated that contamination by indium, divalent and quadrivalent tin in this solution was only $(0.06\pm0.03)\%$, $(0.72\pm0.09)\%$, and $(0.33\pm0.16)\%$, respectively. Therefore, even for later separations where the amount of Sn^{121} was high relative to Cd^{121} the quantity of Sn^{121} measured was reliably attributable to the decay of Cd^{121}.

The counting rate of Sn^{121} was normalized to 10^{12} fissions for 18 separations extending in time from 3–42 sec after fission and appears in Fig. 1. The time of separation was the difference in time between the end of irradiation and the end of filtration. Separations beyond 42 secs were precluded by low counting rate and relatively greater contribution of an unidentified longer-lived contaminant compared to Sn^{121}. The linear relationship between the logarithm of the counting rate and separation time is apparent. The half-life of Cd^{121} derived from these data by the method of least squares is $12.8_{-0.3}{}^{+0.4}$ sec. An empirically determined correction for the counting efficiency of a 0.38-MeV beta ray was applied to the counting rate of Sn^{121} to derive the disintegration rate of Cd^{121} at the end of the irradiation. Cd^{121} was corrected for its decay during and after the course of the irradiation by the expression

$$Y=N'(\lambda)/F(1-e^{-\lambda\tau})(e^{-\lambda t}),$$

where Y is the cumulative Cd^{121} yield, N' the number of atoms at t the time since the end of the irradiation, F the fission rate, τ the duration of the irradiation, and λ the decay constant for Cd^{121}. The cumulative fission yield of Cd^{121} thus was calculated as $(6.4\pm0.5)\times10^{-3}\%$. The cumulative fractional chain yield of

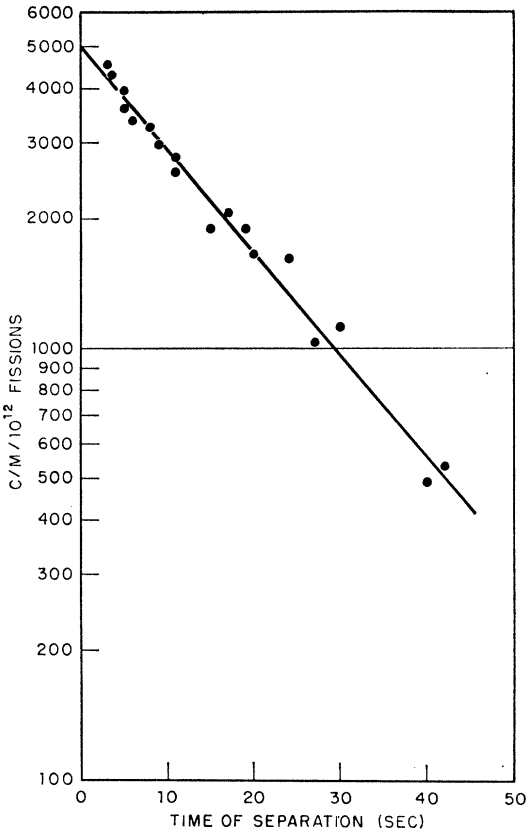

FIG. 1. The growth of Sn^{121} activity from Cd^{121}, in counts/min per 10^{12} fissions, at various times of Cd separation. (The time of separation is the difference in time between the end of the irradiation and the end of filtration.)

Cd^{121} (based on the total chain yield given below) is 0.53 ± 0.09.

From the yield for Cd^{121}, and the assumptions that the distribution of nuclear charge is Gaussian and that the width parameter[7] σ is 0.62, a value of 48.45 was computed for Z_p.

Experimental information about charge distribution for mass numbers in the region of near-symmetric fission was previously unavailable. Such information is of interest because of the observed large decrease in total kinetic energy of fission fragments in the region of near-symmetric fission.[8] It was convenient to compare Z_p for $A=121$ with the empirical Z_p function described by Wahl, Ferguson, Nethaway, Troutner, and Wolfsberg.[7] This function is shown in Fig. 2, in which complementary light (l) and heavy (h) primary fragments were matched on the abscissa according to their mass number (A') before neutron evaporation. The fragment mass number A' was interpolated from data on the number of neutrons emitted per fragment.[8] In the ordinate a

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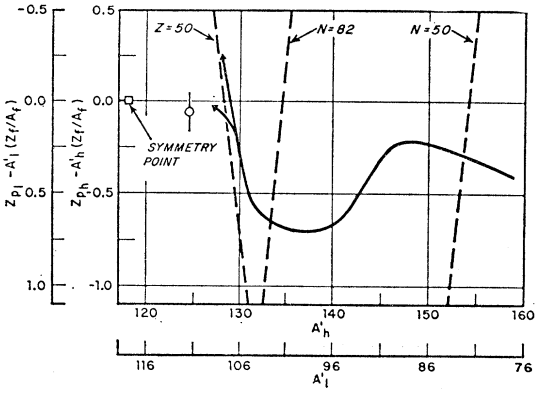

FIG. 2. Z_p value from this work and the empirical Z_p function of Wahl *et al.* (Ref. 7).

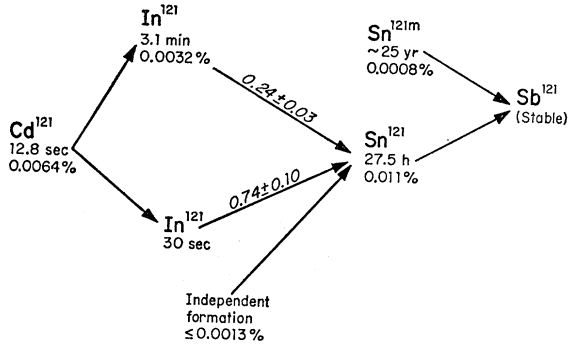

FIG. 3. Genetic relationships between fission products with mass 121 from thermal neutron fission of U^{235}.

multiple of the mass number is subtracted from Z_p. The multiple is (Z_f/A_f), the ratio of atomic to mass number for the fissioning nucleus U^{236}. The smooth and continuous line represents the Z_p function which best fits the experimental data. The bifurcation shown by two arrows displays alternatives in the Z_p function. The sharp rise in Z_p near the 50-proton shell edge was based on the small independent yield of I^{128}. With recognition that evidence for this relationship rested heavily on only a single value, the less radical alternative was also drawn by Wahl *et al.*[7] The experimental value for Z_p at $A=121$ ($A'=124.4$) favors the latter alternative [see Fig. 2]. This suggestion is supported by the value of Z_p (49.5) for $A=127$ derived from the yield of Sb^{127}.[9] Clearly, however, further information is required to characterize the nature of Z_p in the region of symmetric fission.

The effect of possible isomerism in Cd^{121} on the Z_p function was considered. The existence of two isomeric states for Cd^{121} is suggested by comparison of the nuclide with the characteristics of related neighboring nuclides that contain an odd neutron or proton, e.g., Cd^{115}, Cd^{117}, Cd^{119}, Sn^{123}, Te^{125}, and Xe^{127}. Were one of the isomeric states substantially shorter-lived than the several seconds required for separation, it would remain undetected. Such a circumstance would result in a value of Z_p greater than the actual value.

The mass-121 chain yield, as measured by 27.5 h Sn^{121}, was determined to be $(1.1\pm0.1)\times10^{-2}\%$. This value together with the fission yield of Sn^{121m} ($\sim$25 yr)[10] gives a total chain yield of $(1.2\pm0.1)\times10^{-2}\%$. The total chain yield value falls in line with neighboring values in the fission-yield curve.

The currently available information on the genetic relationships among the members of the mass-121 chain formed in the thermal neutron fission of U^{235} are shown in Fig. 3 together with half-lives, cumulative fission yields, and an upper limit for the independent yield of Sn^{121}. The value for the independent formation of Sn^{121}, and the fractional contribution of In isomers to Sn^{121} yield were given by Wahl and Nethaway[11] and the yield of Sn^{121m} was determined by Orth.[10]

It may be remarked that a search for the possible precursor Ag^{121} with a procedure which separated silver essentially quantitatively within 3 secs after fission failed to reveal its existence.[12]

ACKNOWLEDGMENTS

The author wishes to express sincere appreciation to N. E. Ballou for his encouragement and constructive suggestions in this work and to P. O. Strom for critical review of the manuscript. Thanks are also accorded to W. Reichert, M. Kay, and E. Roberts for assistance with the radiochemical analyses, J. A. Reichardt and F. Lybolt for electronic assistance, and R. P. Marquiss and J. R. Kielwasser for attention to health-physics monitoring. M. Sager and the reactor group at Vallecitos nuclear test reactor cooperated generously in scheduling the reactor time.

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