Nuclear-charge distribution for $A = 121$ from thermal-neutron-induced fission of ²³⁵U

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The fractional cumulative yield of ^{121}Ag and the fractional independent yields of ^{121}Cd , ^{121}In , and 121 Sn from thermal-neutron-induced fission of 235 U were determined radiochemically to be 0. 12 ± 0.05 , 0.61 ±0.09 , 0.24 ±0.08 , and 0.03 ±0.04 , respectively. The yield values were used to determine the nuclear-charge-distribution parameters $\sigma_Z = 0.55 \pm 0.10$ and $\Delta Z = 0.50 \pm 0.05$ for $\overline{A} = 121$. The σ_z for $\overline{A} = 121$ is close to $\overline{\sigma_z} = 0.52 \pm 0.02$ for high-yield fission products, and no evidence for an even-odd Z effect was found for $A = 121$. The positive ΔZ value, which corresponds to $Z_p = 48.15$, is similar to those for several higher mass numbers reported previously, and it is considerably greater than the negative values predicted by the scission-point theoretical model. The use of a separation distance between nascent fragments greater than 1.4 fm, the value used in the theoretical calculations, could reduce the discrepancy and could also account for the observed enhanced independent yields of tin fission products with Z_p near 50 ($A = 126 - 129$).

I. INTRODUCTION

The research¹ described in this article was undertaken to measure fission yields of members of the $A = 121$ decay chain from thermal-neutron-induced fission of ^{235}U . The research complements a previously reported investiga- μ concerning yields of indium and tin nuclides with $A = 121 - 128$, and the same experimental approach was used, i.e., solvent-extraction separation soon after a short irradiation of a portion of a continuously circulating solution.

Very little is known about nuclear-charge distribution for near symmetric fission, 4.5 and experimental information about nuclear-charge distribution for mass numbers near symmetry is needed. The scission-point theoretical model of Wilkins et al.⁶ predicts that the ΔZ function⁷ is negative for all heavy fragments, as observed experimentally for high-yield fission products ($A_H \ge 130$). The prediction differs from the observed sharp increase in ΔZ to positive values when Z_P is near 50 ($A_H = 126 - 129$). ^{2,4,8,9} There is evidence² that ΔZ remains positive as A_H decreases toward symmetry $(A'=118)$, but more evidence on this point is desirable because the trend disagrees with theory.

II. EXPERIMENTAL

Experiments to determine fission yields were preceded by solvent-extraction studies of Ag(l), Cd(II), In(III), and $\text{Sn}(IV)$ at tracer concentrations $(10^{-10} - 10^{-7} M)$ and of \sim 0.02M U(VI) solutions. Use was made of the SISAK system, 10,11 in which aqueous and organic phases were mixed in a static mixer and separated ~ 0.5 s later by centrifugal acceleration up to 30000g in an H-10 centrifuge. The measured extraction fractions for the two systems used for yield measurements are shown in Table I. Data for several other systems, which also gave satisfactory separation of the elements, are given in Ref. 1.

Neutron irradiations for fission-yield measurements were carried out in the thermal column of the Omega West 8-megawatt Research Reactor at the Los Alamos National Laboratory. The experimenta1 setup was similar to the one described in Refs. 2 and 3, except a plug (rather than a tank) target was used that consisted of a 0.32 cm

TABLE I. Single-step extraction fractions of tracer quantities of various elements separated from aqueous solution into organic solvent by the continuous SISAK (Refs. 10 and 11) method.

Organic solute	Ag(I)	Cd(II)	In(III)	Sn(IV)	U(VI)
TBA^a	0.995	0.0009	0.0010	0.0004	0.0010
TPAI^b	0.994	0.992	0.0005	0.0002	0.0004

^aOrganic phase: 0.5M tributylamine in CHCl₃. Aqueous phase: 0.5M H₂SO₄, 1.0M NaNO₃, 0.025M $UO_2(NO_3)_2$, 0.001*M* Br₂.

^bOrganic phase: 0.05M tetrapentylammonium iodide in CHCl₃. Aqueous phase: 0.1M HCl, 0.025M $UO_2(NO_3)_2$, 0.001*M* Br₂.

inside diameter polyethylene tube loop. Both arms of the loop extended through 120 cm of concrete shielding and 30.5 cm into the reactor thermal column. The polyethylene tubing was supported by a grooved rod (plastic and brass) in a stainless steel case. The thermal-neutron flux at the target position varied from $\sim 1.5 \times 10^{10}$ cm⁻² s⁻¹ near the end of the loop to $< \sim 10^9$ $\text{cm}^{-2} \text{s}^{-1}$ near the shielding wall. The total holdup time of solution in the 61 cm of target was 0.⁵ s, and 70% of the fissions occurred during 0.2 s in the central 25.5 cm of target. The target holdup time was taken to be 0.2 s, which is small compared to the shortest delay time (see the following) of 2.¹ s and is less than the estimated 0.4 s uncertainty in the delay time.

Two liters of aqueous phase containing 25.0 g of $UO_2(NO_3)_2.6H_2O$, the uranium being enriched to 93% in 235 U, were circulated through the target with a mean time period of 203 s. $Br₂(aq)$ was injected continuously into the irradiated solution to oxidize lower oxidation states of tin to Sn(IV). The irradiation time was usually 10.0 min. About seven liters of organic phase flowed from one storage tank through the mixer and centrifuge to another storage tank. The flow rate for both phases was 10.0 ml/s.

The main variable in the experiment was the delay time (τ) , the time required for passage of the aqueous phase from the target outlet $(0.1 \text{ s after mean irradiation time})$ to about the middle of the mixer-centrifuge assembly, where separation of the different elements in the decay chain occurred. The delay time was varied for different experiments from 2.¹ s to 27.8 s by changing the length and diameter of a connecting tube. Chemical separation was assumed to have occurred halfway between the start of mixing of phases and their separation in the centrifuge, which is believed to have occurred about midway through the centrifuge. The total time during which chemical separation could have occurred was 0.8 s, and the midpoint of this period was chosen as the separation time with an uncertainty of ± 0.4 s, which is then also the uncertainty in τ .

After an irradiation, ^{121}Ag , $^{121}\text{Cd}^m$, $^{121}\text{Cd}^g$, $^{121}\text{In}^m$, and

 21 In^g were allowed to decay to 27.0-h ¹²¹Sn, which, with added Sn(IV) carrier, was separated and purified from both the organic and aqueous phases using radiochemica procedures.^{3,12} Purification occurred \sim 30 h after irradiaion when 40-min ^{123}Sn , 59.3-min ^{128}Sn , and 2.1-h ^{127}Sn had decayed and their decay products could be removed. Metallic tin samples were mounted for beta activity measurements, and their decay was followed with beta-
proportional counters for \sim 5 weeks.

III. DATA REDUCTION

Decay data were resolved using a modification of the CLSQ least-squares program¹³ into three exponential components with half-lives of 27.0 h, 9.64 d, and 129.3 d associated with ^{121}Sn , ^{125}Sn , and ^{123}Sn , respectively. The program calculated counting-rate intercepts, I_i , at the end of irradiation for each component. The intercepts, corrected for chemical yield and counting efficiency, are functions of numbers of radioactive atoms and decay constants and are used, as described in the following, to calculate fission yields. The intercepts for the 129.3-d component were small (a few counts/min) and may have included trace impurities, so no attempt was made to derive yields from these intercepts.

For 121 Sn and 125 Sn, intercepts I^{org} and I^{wat} from organic- and water-phase samples, corrected for chemical yield and counting efficiency, were used to derive the fraction of activity found in the organic phase, I^{org}/I^{tot} $I^{tot} = I^{org} + I^{wat}$. Plots of these ratios for ¹²¹Sn from the Ag, Cd/In, Sn and Ag/Cd, In, Sn separations are shown as points in Figs. ¹ and 2, respectively. There are few data from the Ag/Cd, In, Sn separation because measurable amounts of 0.8 -s 121 Ag existed only at the shortest delay times. The heavy lines shown in Figs. ¹ and 2 are functions of fission yields and other quantities, discussed in the following, that best represent the data.

The $I^{\text{org}}/I^{\text{tot}}$ ratios for 125 Sn from the Ag, Cd/In, Sn separations decreased rapidly with delay time, τ , from ~ 0.2 for the shortest τ to ~ 0.05 for the larger τ values.

FIG. 1. Intercept ratio $I^{\text{org}}/I^{\text{tot}}$ vs delay time τ for $^{121}\text{Sn}^g$ from separation of Ag and Cd from In and Sn. Points are experimental. Curves are calculated using parameters from Table $II: \,$ - , selected; - - -, B3; - - --, B4.

FIG. 2. Intercept ratio I^{org}/I^{tot} vs delay time τ for $^{121}Sn^g$ from separation of Ag from Cd, In, and Sn. Points are experimental. The curve is calculated using selected parameters from Table II. The two data for $\tau=2.1$ are plotted at $\tau=2.05$ and 2.15 s for clarity.

The ratios varied considerably for similar τ 's, and they are much too large to be due to extraction of ~ 0.65 s (Ref. 14) ¹²⁵Cd because only 13% of ¹²⁵Sn is formed by beta decay of 2.3-s ¹²⁵In.² Even if all 2.3-s ¹²⁵In were formed by decay of \sim 0.65-s ¹²⁵Cd, the ratios should be \lt \sim 0.01.

The larger ratios observed for 125 Sn are believed to be due to chemical effects associated with the large fraction (0.87) of 125 Sn that is formed directly in fission.² These atoms formed in the target could react with unstable radiation-damage products, also formed in the target, to form organic soluble reaction products, some of which could decompose rapidly to account for the rapid decrease in $(I^{org}/I^{tot})₁₂₅$.

Essentially none of the 121 Sn atoms are formed directly in fission, 2 and they have relatively long-lived precursors. Therefore, ¹²¹Sn atoms were not formed in the target and passed through the target only during subsequent circulation of the water solution after ¹²¹Sn atoms had been oxidized to stable Sn(IV). Therefore, the behavior of 125 Sn is not a measure of radiation damage effects for ¹²¹Sn, so little or no correction for such effects should be applied to the 121 Sn data.

The uncertainty in the ^{121}Sn I^{org}/I^{tot} ratios was taken to be 5% of the ratio plus 0.05. The 5% error reflects errors associated with chemical-yield and efficiency corrections. The 0.05 includes unknown systematic errors, e.g., possibly some small radiation damage and/or other chemical effects. Other error estimates and data corrections were explored,¹ but these changed the calculated results only a little and did not affect the overall interpretation of the results, so are not discussed in this paper.

The variation of $(I^{org}/I^{tot})_{121}$ with τ was treated by the method of least squares with modifications of the ORGLs program¹⁵ using equations developed earlier^{2,3} for radioactive decay and growth during steady-state flow of a circulating solution with chemical separation occurring soon after irradiation. A minor modification was made by replacing production terms, $1/(1+\lambda_i T)$, for a tank target with the appropriate terms, $(1-e^{-\lambda_i T})/\lambda_i T$, for plug target used, λ_i representing decay constants and T the target holdup time (0.2 s).

The parameters and variables used in the analysis included the fractional independent yields (FI), half-lives, and branching fractions of the various members of the $A = 121$ decay chain, and single-step extraction fractions of the elements (Table I). The FI values were determined and are discussed in the next section. The half-lives and branching fractions were derived from the literature and are shown in Fig. 3.

IV. RESULTS

The data were treated in two ways. First, preliminary calculations were made using only data directly related to he yield calculated, e.g., the data from Ag/Cd, In, Sn separation experiments for calculation of the fractional cumulative yield (FC) of 121 Ag; then a global treatment of all data for all chain members was carried out to obtain yields that best represented charge dispersion for $A=121$, conserved yield balance $[\sum FI(Z)=1.00]$, and allowed consideration of isomer-yield ratios. The preliminary calculations are discussed in subsection A, and the global treatment of the data is discussed in subsection B.

A. Preliminary calculations

The data from experiments in which Ag was separated from Cd, In, and Sn were treated by equations representing a two-membered chain,

$$
\rightarrow^{121\mathrm{Ag}}_{0.8\pm0.1\,\mathrm{s}}(^{121}\mathrm{Cd}^{g,m},^{121}\mathrm{In}^{g,m},^{121}\mathrm{Sn}^{g})
$$

o obtain the fractional cumulative yield of ^{121}Ag , $FC(^{121}Ag)=0.135\pm0.052$. The error includes uncertainties from the least-squares calculation (σ_{LS}) and from τ , ± 0.4 s, determined as the average change (Δ_{τ}) in the yield when τ was changed by plus and by minus the uncertainty,

$$
\sigma = (\sigma_{LS}^2 + \Delta_\tau^2)^{1/2} = 0.052 \; .
$$

The data from experiments in which Ag and Cd were separated from In and Sn were treated by equations for a four-membered chain, 121 Inⁿ, 121 In^g, and 121 Sn^g being considered together as the last chain member. The yield of ²¹Ag determined above was used with the data to calculate yields of $^{121}Cd^{m}$ and $^{121}Cd^{g}$, and the combined yields of $^{121}In^{m}$, $^{121}In^{g}$, and $^{121}Sn^{g}$ were determined by difference. The results are given in the first line of Table II. Uncertainties in the yield values include uncertainties in τ and in $FC(^{121}Ag)$ as well as from the least-squares calculaions, as already described. The yield values are based on $FC(^{121}Sn^g) = 1.00$, which neglects the branching of $^{121}In^g$ to 121 Sn^m, and the yield of 121 Sn^m, factors which approximately cancel. (The data were treated exactly by the glo-

FIG. 3. $A = 121$ decay chain: (a) average of values from Refs. 16 and 17; (b) derived from decay scheme given in Ref. 16; (c) from Ref. 18; {d) average of values from Refs. ¹⁸—21; (e) derived from decay scheme given in Ref. 19; (f) derived from values given in Refs. 20, and ²²—24; {g) derived from values given in Refs. 20, 22, and 23; (h) from Ref. 25; (i) derived from decay scheme given in Ref. 25; (j) from Ref. 26; (k) from Ref. 12; (l) from Ref. 27.

Calculation			Fraction		Fraction	
method	$FC(^{121}Ag)$	FI ⁽¹²¹ Cd)	$^{121}Cd^m$	$FI(^{121}In)$	121 In ^m	$FI(^{121}Sn)$
$A^{\rm a}$	0.135 ± 0.052	0.667 ± 0.176 [<i>m</i>]	1.02 ± 0.21^b	0.212 ± 0.235 ^c		
		$-0.014\pm0.122[g]$				
B1 ^f	0.123 ± 0.025	0.658 ± 0.454 [<i>m</i>] ^d	1.01 ^b	0.208 ± 0.234 [<i>m</i>]	1.08 ^b	0.045 [<i>m</i>] ^e
		$-0.006\pm0.106[g]$		-0.015 ± 0.435 [g]		-0.013 ± 0.129 [g]
B2 ^f	0.123 ± 0.025	$0.651 + 0.269$ ^d	1.01 ± 0.16	$0.193 + 0.208$	$1.08 + 2.34$	$0.033 + 0.169$
B3 ^f	$0.123 + 0.023$	$0.650 + 0.047$ ^d	(1.00)	$0.200 + 0.034$	(1.00)	$0.028 + 0.022$
B4 ^f	$0.115 + 0.026$	0.560 ± 0.068 ^d	(0.67)	$0.314 + 0.053$	(0.33)	$0.010 + 0.033$
B5 ^g	0.117 ± 0.032	0.605 ± 0.073 ^d	(0.83)	0.244 ± 0.073	(0.67)	$0.033 + 0.029$
Ref. 2	$0.207 + 0.210$	0.585 ± 0.267	(1.00)	0.179 ± 0.103	(0.0)	$0.029 + 0.054$
Selected	0.12 ± 0.05	0.61 ± 0.09		0.24 ± 0.08		0.03 ± 0.04

TABLE II. Fractional fission yields for thermal-neutron-induced fission of ^{235}U . Yields of both isomers are included where appropriate. (): Values assumed and not varied; []: Isomeric state designation.

^aValues based on $FC(^{121}Sn^g) = 1.00$.

Calculated from isomer yields.

Combined yields of $^{121} \text{In}^m$, $^{121} \text{In}^g$, and $^{121} \text{Sn}^g$.

Calculated by difference.

^eCalculated from the reported cumulative yield of $(1.7 \pm 0.6)10^{-3}\%$ for $^{121}Sn^m$ (Refs. 2 and 29) minus the amount formed by beta decay of 121 In^g.

Errors are from the least-squares calculation.

^gErrors are calculated from $\sigma = [\sigma_{LS}^2 + \Delta_{\tau}^2 + \Delta_{\text{fract}}^2]^{1/2}$.

bal method discussed in subsection B.)

The small negative value of the 121Cd^g yield is zero within experimental error. The use of a longer half-life for $^{121}Cd^{m}$, e.g., 8.3 s, 19 resulted in even more negative values for the $^{121}Cd^{g}$ yield and gave a poorer fit of the data. Calculations to determine the half-life of 121 Cd^m gave 3.8 ± 0.8 s and $FI(^{121}Cd^m) = 0.75 \pm 0.11$ when FI (^{121}Cd g) was assumed to be zero and not varied. However, the literature value of 4.8 ± 0.4 s (Ref. 18) was used for all yield calculations reported in Table II.

B. Global treatment

The exact equation^{2,3} for a six-membered chain, Fig. 3, but without 12^1 Sn^m, was used in a program that allowed simultaneous analysis of the three sets of data for $A = 121$, the two described in this paper and data from the Ag, Cd,In/Sn-separation experiments described previously.² The errors for I^{org}/I^{tot} from the Ag, Cd, In/Sn separation experiments² were increased by 0.03 to achieve more uniform weighting factors $(1/\sigma^2)$ and to allow for possible small systematic errors, e.g., possible deviations from expected chemical behaviors of newly formed fission products.

The yields of all six members of the decay chain were determined, one by difference, and the yield of the seventh member, $^{121}Sn^m$, was included in the calculations to achieve proper normalization (\sum FI(Z) = 1.00). The results are shown as the second entry (Bl) of Table II. The large uncertainties and small negative yields, zero within the calculated error, indicate that the data are inadequate for determination of yields of individual isomers.

Therefore, the basic program² was modified to calculate fractional yields of the four elements. The fractions of the element yields for the metastable states, $^{121}Cd^{m}$ and 121 In^m, could also be calculated, or assumed and not

varied. Results of these calculations are shown in the third to sixth entries of Table II. The best fit of the data (B3), without negative yields, assigns no yield to either ²¹Cd^g or ¹²¹In^g, which is surprising, because isomer ratios, the ratio of FI values for high to low spin isomers, are usually in the range of $1-5$.²⁸ Entry B4 shows the effect on yields of assigning isomer ratios of 2.0 to both ¹²¹Cd and 121 In. The yield of 121 In is raised considerably at the expense of 121 Cd, but the values are certainly possible and do represent the data nearly as well as the function with parameters from B3, as shown in Fig. 1. (For the other two data sets, the curves calculated using the 83 or 84 parameters are very close to the curve derived from the selected parameters and are not shown in Figs. 2 and 4.)

A compromise was made by using intermediate yield fractions to metastable states, as shown in entry B5 of

FIG. 4. Intercept ratio I^{org}/I^{tot} vs delay time τ for $^{121}Sn^g$ from separation of Ag, Cd, and In from Sn. Points are experimental (Ref. 2). The curve is calculated using selected parameters from Table II.

FIG. 5. Charge dispersion for $A = 121$ from thermalneutron-induced fission of ^{235}U . Points are selected fractional yields from Table II: \circ independent; \bullet cumulative. Curves are calculated $[Z_P=48.15 \, (\Delta Z=0.50)$ and $\sigma_Z=0.55$]: --- independent; $- - -$ cumulative.

Table II. Errors given for the 85 parameters include the errors from the least-squares calculation, the uncertainty in τ , and an error, Δ_{fract} , associated with the uncertainty in isomer ratios. The last was taken to be the average change in parameter values resulting from variation of the yield fraction to 121 Cd^m from 0.83 to 0.67 or 1.00 and of the yield fraction to 121 In^m from 0.67 to 0.33 or 1.00. The values and errors selected for investigation of charge dispersion for $A = 121$ (last entry in Table II) are close to those of entry B5, but are rounded and have somewhat larger errors that bracket values from other methods of calculation.

V. DISCUSSION

The fractional cumulative yield of 121 Ag and the sum of yields for the 121 Cd and 121 In isomers based on data from Ag, Cd,In/Sn separation experiments, literature values for half-lives and for branching fractions, and some assumptions about yields have been reported previously.^{2,3} These values with their large uncertainties, listed in the next to last line of Table II, are consistent with the selected values (last line of Table II) deduced from the more comprehensive data sets discussed in this paper.

Before 121 Cd^m was known, the yield of 12.8-s 121 Cd was reported to be $FC=0.53\pm0.09$, ²¹ which is considerably larger than $FC({}^{121}Cd^g) \approx 0.2$ deduced from our data. However, a portion of the larger yield reported earlier²¹ must have been contributed by 4.8-s ¹²¹Cd^m, which has a large yield.

The selected values and uncertainties for ^{121}Ag , ^{121}Cd , ^{21}In , and ^{121}Sn yields listed in the last line of Table II were treated by the method of least squares, with a modification of the ORGLS computer program,¹⁵ to obtain parameters for a Gaussian representation of the data. The results are listed in the first line of Table III and shown in Fig. 5. Calculation of the even-odd proton factor (EOZ) gave a value of 0.91 \pm 0.10, so values of ΔZ and σ_Z were obtained with EOZ and EOX set equal to 1.0 and not varied. [As shown in the second line of Table III, EOZ and EON are normally > 1.0 , and the even-odd neutron factor (EON) is \langle EOZ.]

The width parameter, $\sigma_Z = 0.55$, is somewhat less than the 0.62 value reported previously,² and it is close to the average of 0.52 ± 0.02 (Ref. 4) derived from data for most fission products, those with relatively large yields $(A < 105, >129)$. The σ_Z for $A = 121$ is considerably larger than those for mass numbers with Z_p near 50 A_H \simeq 126–129) and their complements (A'_L = 105–107), for which $\sigma_Z = 0.35 \pm 0.03$.⁹ Thus, there is evidence that near symmetry the charge dispersion width is about normal and that even-odd effects are absent.

The positive value of ΔZ = 0.50 for $A = 121$ is somewhat larger than reported previously² (0.32) and confirms that the ΔZ function,⁷ which rises sharply near $Z = 50$ to positive values,^{2,8,9} remains positive as Z_p becomes appreciably less than 50, e.g., $Z_p = 48.15$ for $A = 121$. The ab-

	ΔZ	σ _z	EOZ	EON
121 ^a	$0.50 + 0.05$	0.55 ± 0.10	$(1.00)^{b}$	$(1.00)^{b}$
< 104 , > 130 $105 - 107$	-0.47 ± 0.02 ^c	0.52 ± 0.02	1.27 ± 0.03	1.08 ± 0.03
а $126 - 129$	-0.4 to > 0.0	0.35 ± 0.03	$(1.00)^{b}$	$(1.00)^{b}$

TABLE III. Parameters for the Z_p model.

'Errors were estimated from the variation of parameter values that resulted from calculations with data values changed within their error ranges, but summing to 1.00. Calculation of EOZ gave a value of 0.91 ± 0.10 .

"Assumed.

 ${}^{\circ}$ ΔZ is for A'_{H} = 140; $\partial \Delta Z / \partial A'_{H}$ = -0.010±0.005 (Ref. 4). Reference 9.

solute value of ΔZ depends on the neutron-emission function used,² but it is difficult to conceive of a function, consistent with experiment, that would lead to negative ΔZ values between Z = 46 and 50 (A'=118 to ~128), as predicted by the scission-point theory.

A possible cause of the discrepancy is the use of the separation distance (d) between the tips of the nascent fragments as a constant, $d=1.4$ fm, for the theoretical calculations. Wilkins et al ⁶ state that increasing the value of d would shift ΔZ toward the maximum-energy-release line (Q), and extrapolation of the Q line below $A'_H = 126$ indicates that ΔZ would become more positive in this mass-number region. Indeed, Wilkins and Steinberg³⁰ found in preliminary theoretical calculations with a larger d (\sim 2.4 fm) that small positive ΔZ values (0.0 to +0.2) were obtained for the $A' = 118$ to 126 region.

A larger separation distance could lead to less fragment distortion, which could account for the greater influence of the spherical 50-proton shell on fission yields observed experimentally² than is predicted theoretically.⁶ The ΔZ function just discussed is one way of describing the 50 proton shell effect; others are discussed in Ref. 2.

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