Nuclear Charge Distribution in Fission: Independent Yields of ¹¹²Ag, ¹³⁴I, and ¹³⁸Cs from Spontaneous Fission of ²⁵²Cf*

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(Received 17 September 1969)

Independent yields of ¹¹²Ag, ¹³⁴I, and ¹³⁸Cs from the spontaneous fission of ²⁵²Cf have been measured. These nuclides were separated from metal foils which were used as collectors for fission fragments recoiling from a one-microgram ²⁵²Cf source and purified radiochemically. Independent yields were calculated from a knowledge of the activities of the nuclides as a function of separation time. The yields, expressed as fractional independent yields, are 0.009±0.004, 0.26±0.03, and 0.11±0.03, for ¹¹²Ag, ¹³⁴I, and ¹³⁹Cs, respectively. Corresponding values of Z_p are 45.18, 52.14, and 53.82. These values differ from unchanged charge distribution by ~ 0.45 charge units in agreement with previously measured fractional cumulative yields from ²⁵²Cf fission.

INTRODUCTION

HERE have been several reports¹⁻³ of measure-L ments of distribution of nuclear charge in fission of ²⁵²Cf by simultaneous measurement of the masses and characteristic K x-ray energies of the fission fragments. The values of the most probable nuclear charge Z_p for fission fragments as a function of mass determined by this method by different workers are in reasonably good agreement. For masses near 135, however, the values of Z_p given by Glendenin and Unik¹ show large uncertainties and are not in good agreement with those reported by Kapoor et al.² Watson et al.⁴ have shown that in this mass region there is a much greater x-ray yield from odd-Z fragments than from even-Z fragments and point out that Z_p values determined by x-ray measurements in this region may represent poorly the true charge distribution.

This mass region is also of interest for any fissioning system since the fragments are near the 50-proton and 82-neutron shells. Wing and Fong⁵ have suggested that for ²⁵²Cf fission the value of Z_p for fission fragments in this mass region falls close to or even slightly above the value A'(98/252), where A' is the mass number before neutron emission, and consider this to be a fine structure in Z_p due to shell effects. Armbruster⁶ also predicts a similar fine structure. Most of the results

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from radiochemical experiments for ²³⁵U fission,⁷⁻⁹ however, do not support the existence of fine structure in Z_p . The results of radiochemical experiments for ²³³U fission^{10,11} also provide little evidence for fine structure.

Unfortunately, there are few data available for fractional cumulative or independent yields of products of ²⁵²Cf fission. Wahl et al.⁷ report fractional cumulative yields of ^{139,140,141}Xe and a fractional independent yield for a shielded nuclide ¹³⁶Cs. In order to provide more information about charge distribution in fission of ²⁵²Cf, we have undertaken a series of experiments to measure independent and fractional cumulative yields of products of that fissioning system. We report here the results of measurements of the independent yields of ¹¹²Ag, ¹³⁴I, and ¹³⁸Cs.

EXPERIMENTAL

Fragment Collection

Fission fragments recoiling from a thin ²⁵²Cf source were collected in aluminum or magnesium catcher foils in a manner similar to that described by Nervik.¹² The source was $\sim 1 \ \mu g$ of ²⁵²Cf deposited on a platinum foil over an area of $\sim 1 \text{ cm}^2$. The source was placed in a borated-paraffin holder as shown in Fig. 1. A thin film of Formvar ($\sim 20 \, \mu \text{g/cm}^2$) was placed between the source and the catcher foil to prevent self-transfer of ²⁵²Cf to the foil. The Formvar foils were replaced after about one week of use.

^{*} Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation. ¹L. E. Glendenin and J. P. Unik, Phys. Rev. **140**, B1301

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³ S. R. Dolce, W. M. Gibson, and T. D. Thomas, Bull. Am. Phys. Soc. 11, 335 (1966).
⁴ R. L. Watson, H. R. Bowman, and S. G. Thompson, Phys. Rev. 162, 1169 (1967).
⁵ I. Wing and P. Fong, Phys. Rev. 157, 1038 (1967).

 ⁶ J. Wing and P. Fong, Phys. Rev. 157, 1038 (1967).
 ⁶ P. Armbruster, in *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. I, p. 103.

⁷ A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962). ⁸ P. O. Strom, D. L. Love, A. E. Greendale, A. A. Delucchi, D. Sam, and N. E. Ballou, Phys. Rev. **144**, 984 (1966).

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¹² W. E. Nervik, Phys. Rev. 119, 1685 (1960).



FIG. 1. Source and holder.

Approximately 10⁷ fission fragments were collected each minute. Duration of exposure ranged from a few minutes to a few hours for this series of experiments. A small tab of transparent tape on the upper surface of each catcher foil facilitated its placement and removal with the use of tweezers. The beginning and end of each exposure was timed with the aid of electric stop watches. It is estimated that the uncertainty in the length of each exposure is less than ± 5 sec. Each foil was monitored for α activity after each exposure and if the activity was greater than 200 dis/min as a result of self-transfer of ²⁵²Cf, the foil was discarded.

Chemical Separations

The fission-product β -decay chains of interest in this work are shown in Fig. 2. For each chain, a member of the next-higher mass chain was used as a fission monitor in order that counting data for all experiments with a given nuclide could be normalized to the same number of fissions. This was necessary, since the rings holding the Formvar foils were not cut to tolerances such that geometry was reproducible for each run, and since the strength of the source changed as a result of decay and self-transfer.

The nuclide of interest in each experiment and its monitor were separated chemically from precursors at known times after exposure. They were then purified radiochemically and prepared for counting as solid samples mounted on flat aluminum planchets. Initial chemical separations as well as purifications were based on standard methods for each nuclide and are described in more detail in the Appendix.

Counting

The radioactivity of samples containing ¹¹²Ag and ¹¹³Ag was measured with a low-background gas-flow counter operated in the proportional region. Samples were counted through an \sim 500-mg/cm² (\sim 650-mg/ cm² for runs 95 and 106) aluminum absorber in order to reduce the observed ¹¹³Ag activity to a level such

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Runª	A_{112} (cpm)	A_{113} (cpm)	T (min)	t (min)	$\epsilon_{113}R_{113}$ [atoms ¹¹³ Ag /(min exposure)]	A_{112}^{0} [(A_{112}) /($\epsilon_{113}R_{113}$)]	J	JA_{112}^{0}	K
79	541	571	100.0	232.0	3913	0.1382	5.266	0.728	0.134
79G	168	178	100.0	232.0	1217	0.1377	5.266	0.725	0.134
81	221	662	100.0	105.0	3456	0.0639	3.298	0.211	0.032
81G	68.2	216	100.0	105.0	1128	0.0605	3.298	0.199	0.032
95	38.6	203	50.0	53.0	2000	0.0193	5.977	0.115	0.016
106	395	210	100.0	411.0	2121	0.1864	10.225	1.906	0.367
117	210	609	100.0	103.0	3153	0.0668	3.263	0.218	0.031
123	775	407	100.0	415.0	4142	0.1871	10.379	1.942	0.374
126	86.9	408	50.0	54.0	4035	0.0215	6.002	0.129	0.016

TABLE I. Data from measurements of independent yield of ¹¹³Ag.

⁸ Runs 79 and 81 were counted in a γ spectrometer as well as on a β counter. Data for γ counting are listed as 79G and 81G. Runs 95 and 106 were counted through a thicker absorber than that used for the other runs.

Raw counting data for 79G, 81G, 95 and 106 have been corrected for differences in relative counting efficiencies of ^{112}Ag and ^{113}Ag .

that decay curves could be resolved for ¹¹²Ag and ¹¹³Ag components. Samples from runs 79 and 81 were also counted on a γ -ray spectrometer.

Radioactivity of samples containing ¹³⁴I and ¹³⁵I was measured with a NaI γ spectrometer set to measure radiations in the energy band 750–950 KeV. This band includes the principal γ ray from ¹³⁴I as well as one from ¹³⁵I. Small amounts of activity from the decay of ¹³²I and ¹³³I were also detected.

Activities of ¹³⁸Cs and ¹³⁹Ba were measured with a low-background gas-flow counter operated in the Geiger region.

Decay of all samples was followed for more than 10 half-lives of the nuclide of interest and for 5 to 10 half-lives of the monitor. Decay curves were resolved using a least-squares computer program.¹³



1001	6 sec	14 min	32,2 min	
		139Xe 43 sec	139 _{Cs} 9.5 min	139 _{Ba} 83 min

FIG. 2. Beta-decay chains of interest in this work. Half-lives are taken from Lederer *et al.* (Ref. 24) except as noted: (a) this work, (b) Ref. 17, (c) Ref. 18, (d) Ref. 25.

CALCULATIONS

Half-lives of the early members of the chains considered here are so short that all chains can be considered to consist of only two members, the member for which the yield is being measured and its immediate precursor. For a two-member decay chain, the activity of member 2 separated at time t after the beginning of the exposure is related to the rate of production of 1 and 2 by the expression

$$JA_2 = K\epsilon_2 R_1 + \epsilon_2 R_2, \tag{1}$$



FIG. 3. Plot of JA_{112}^0 as a function of K. Units are discussed in text.

¹³ P. C. Rogers, Massachusetts Institute of Technology Progress Report No. MIT-LNS 76, 1962 (unpublished).

Run	$A_{134}{}^{\mathbf{a}}$ (cpm)	A ₁₃₅ (cpm)	T (min)	t (min)	$\frac{\epsilon_{135}R_{135}}{[(\text{atoms }^{135}\text{I})}/(\text{min exposure})]$	A_{134}^{0} [(A_{134}) /($\epsilon_{135}R_{135}$)]	J	$JA_{134}{}^{0}$	K
3A	2500	248	10	15.5	14 550	0.1718	8.616	1,480	0.168
3 B	3375	281	10	61.5	17 890	0.1887	15.907	3.002	0.852
49	1830	153	10	64.0	9 763	0.1874	16.447	3.082	0.886
51	1098	136	10	19.5	8 047	0.1364	9.085	1.239	0.232
238	975	97.4	40	137.0	1 729	0.5639	8.817	4.972	1.590
240	1144	112	40	44.0	1 692	0.6761	2.551	1.725	0.351
242	665	69.0	40	133.0	1 214	0.5478	8.360	4.580	1.544
244	905	84.3	40	47.0	1 280	0.7070	2.657	1.878	0.397

TABLE II. Data from measurements of independent yield of ¹³⁴I.

^a Samples from runs 3A, 3B, 49, and 51 were counted with one γ -ray spectrometer, those from the other runs with another. Raw counting data

were corrected for the difference in the relative counting efficiency for $^{134}\mathrm{I}$ and $^{136}\mathrm{I}.$

where

 A_2 = observed activity of 2 at time t,

$$J = \exp(\lambda_2 t) / [\exp(\lambda_2 T) - 1]$$

$$K = \{ [\lambda_1 / (\lambda_1 - \lambda_2)] - [\lambda_2 / (\lambda_1 - \lambda_2)] \\ \times \{ [\exp(\lambda_1 T) - 1] / [\exp(\lambda_2 T) - 1] \} \\ \times [\exp(\lambda_2 t) / \exp(\lambda_1 t)],$$

 R_1 = rate of production of 1,

 R_2 = rate of production of 2,

 $\epsilon_2 = \text{counting efficiency for } 2$,

T = time interval from beginning to end of exposure, t = time interval from beginning of exposure to separa-

 $\lambda_1 = \text{decay constant of 1, and}$ $\lambda_2 = \text{decay constant of 2.}$

If values of A_2 from all runs are normalized to a constant fission rate through use of a fission monitor and all samples are counted in the same way so that ϵ_2 is a constant, Eq. (1) can be rewritten as the linear equation

$$JA_{2^{0}} = \epsilon_{2}R_{1}K + \epsilon_{2}R_{2}, \qquad (2)$$

tion of 2 from 1,

where A_{2^0} is A_2 normalized to a constant rate of production of the monitor and $\epsilon_2 R_1$ and $\epsilon_2 R_2$ are the slope and intercept. Since the fractional independent yield of 2 is defined as $R_2/(R_1+R_2)$, it is equal to the slope of the line obtained from a plot of JA_{2^0} as a function of K divided by the sum of the slope and intercept of the line.

The normalization described above can be accomplished by dividing A_2 by the rate of production of another fission product for which the observed activity at time of separation is not a function of the fractional yields within the chain of which it is a member. This condition is met if $R_1 \gg R_2$ or if $\lambda_1 \gg \lambda_2$ and t is much longer than the half-life of 1. Monitors chosen for these experiments were ¹¹³Ag, ¹³⁵I, and ¹³⁹Ba for ¹¹²Ag, ¹³⁴I, and ¹³⁸Cs, respectively. Since ¹¹²Ag and ¹³⁴I were isotopic

with their monitors, only one chemical purification was necessary and no correction for chemical yield was required. Purifications for both ¹³⁸Cs and ¹³⁹Ba were, of course, required and activities were corrected for chemical yields.

RESULTS

Data from all runs are given in Tables I–III. Figures 3–5 are plots of JA_2^0 as a function of K for each chain. Error limits shown on individual points are estimated from the standard deviations of activities at the time of separation resulting from computer resolution of the decay curves.

In some runs, counting conditions were varied slightly, so that ϵ_2 also changed for both the nuclide of interest and its monitor. In every case in which this was true, two runs, one with a short separation time and the other with a long, were made. The activity ratio of the nuclide to monitor for the run with the longer separation time was multiplied by a factor which made the ratio consistent with that for other runs with long separation times. The activity ratio for the run with the shorter separation time was then multiplied by the same factor. This normalization resulted, of course, in very small deviations from the lines shown in Figs. 3–5 for the points for the longer separation times. Therefore, these points were not included in calculating the standard deviations of the slope and intercept of the lines, so that the standard deviations reported are not biased by the normalization procedure.

Results of the measurements are shown in Table IV. The error limits shown for each value of ϵR are the standard deviations from least-squares analyses of the points shown in Figs. 3–5. Error limits shown for the calculated value of each yield are also standard deviations calculated from the standard deviations of $\epsilon_2 R_1$ and $\epsilon_2 R_2$. Error limits for the preferred values include estimates of uncertainties due to uncertainties in halflives and separation times as well as the standard deviations. These total uncertainties were calculated by _

Run	$A_{138}^{\mathbf{a}}$ (cpm)	$A_{139}^{\mathbf{b}}$ (cpm)	T(min)	t° (min)	<i>t</i> ′ ^d (min)	$\epsilon_{139}R_{139}^{ m e}$	A_{138}^{0} [(A_{138}) / $(\epsilon_{139}R_{139})$]	J	JA_{138}^{0}	K
336	1824	1280	100.0	134.1	147.7	3010	0.6063	2.359	1.430	1.437
340	2076	1667	150.0	184.1	192.5	2977	0.6972	2.171	1.514	1.461
342	302.5	175.9	10.0	15.1	75.0	3381	0.0895	5.766	0.516	0.425
345	128.5	93.8	5.0	10.1	70.3	3546	0.0362	10.943	0.396	0.337
347	1394	898.3	50.0	84.1	91.3	3365	0.4142	3.163	1.310	1.358
349	486.4	298.5	15.0	20.1	54.0	3277	0.1484	4.047	0.601	0.503

TABLE III. Data for measurement of independent yield of ¹³⁸Cs.

 $^{\rm a}$ Activity expressed as cpm $^{138}{\rm Cs}$ per mg of recovered cesium tetraphenylboron.

 $^{\rm b}$ Activity expressed as cpm $^{129}{\rm Ba}$ per mg of recovered barium chromate.

^o Time at which xenon was separated from cesium.

squaring each uncertainty, adding these values, and then taking the square root of this sum. Major uncertainties for each yield are given in footnotes to Table IV. The calculated values for the yields of ¹¹²Ag and ¹³⁸Cs were especially sensitive to the half-lives used for ¹¹³Ag and ¹³⁸Xe, respectively. Experiments to measure the half-lives of these two nuclides are described in the Appendix.

DISCUSSION

Wahl *et al.*⁷ have assumed the charge distribution for direct formation of fission fragments to be Gaussian. For such a distribution, the fractional cumulative yield



 $^{\rm e}$ Expressed as atoms of $^{\rm 139}{\rm Ba}$ per minute exposure per mg of recovered barium chromate.

of any member Z_i of a chain is given by

$$Y_{Z_i}^{\text{cum}} = (2\pi)^{-1/2} \sigma^{-1} \int_{-\infty}^{Z_i + 1/2} \exp\left(-\frac{(Z - Z_p)^2}{2\sigma^2}\right) dZ, \quad (3)$$

where Z_p is the most probable charge for the chain, and σ is the standard deviation of the distribution. They found that a value of 0.62 ± 0.06 gave good agreement with most of the yields reported for fission of ²³⁵U. Later work by Norris and Wahl¹⁴ resulted in a new value for σ of 0.59 ± 0.06 . That value is in general



FIG. 4. Plot of JA_{134}^{0} as a function of K. Units are discussed in text.



FIG. 5. Plot of JA_{138}^0 as a function of K. Units are discussed in text.

¹⁴ A. E. Norris and A. C. Wahl, Phys. Rev. 146, 926 (1966).

Nuclide	$\epsilon_2 R_1$	$\epsilon_2 R_2$	Calculated yield	Preferred ^a yield	
¹¹² Ag	5.073 ± 0.030	0.0458 ± 0.0045	0.0089 ± 0.0009	0.009 ± 0.004^{b}	
134 I	2.495 ± 0.116	0.865 ± 0.109	$0.258 {\pm} 0.026$	0.26±0.03°	
138Cs	$0.923 {\pm} 0.035$	$0.112 {\pm} 0.037$	$0.108{\pm}0.032$	$0.11{\pm}0.03^{d}$	

TABLE IV. Fractional independent yields of ¹¹²Ag, ¹³⁴I, and ¹³⁸Cs.

^a Values for preferred yields include uncertainties due to uncertainties in half-lives and separation times as well as the uncertainties shown in the calculated values. Total uncertainties were calculated by taking the square root of the sum of the squares of all the individual uncertainties. Major sources of error are listed below for each vield.

⁹ Uncertainties in the yield due to uncertainties of ± 3 min in the half-life of ¹¹³Ag, ± 1 min in the half-life of ¹¹²Ag, and ± 1 min in *t*, are ± 0.0038 ,

use for interpreting the results of experiments such as these. The value of Z_p for a chain for which only one yield has been measured is calculated using $\sigma = 0.59$, and limits of error are calculated to include uncertainties in the yield as well as the ± 0.06 uncertainty in the value of σ .

Wahl et al.⁷ have also compared values of Z_p for several chains by calculating the value of Z_p - $A'(Z_F/A_F)$ for each chain, where A' is the fragment mass number before neutron emission, Z_F is the charge of the fissioning nucleus, and A_F is the mass number of that nucleus. The value $A'Z_F/A_F$ represents a charge distribution equal to that of the fissioning nucleus and is referred to as unchanged charge distribution. The value of this term was found to be ${\sim}{-}0.6$ for the mass region studied here and ${\sim}{+}0.6$ for the complementary mass region. Wolfsberg10 reports similar results for fission of ²³³U and ²³⁹Pu.

In a recent reevaluation of the available data for fission of ²³⁵U, Wahl¹⁵ concludes that the best value for σ is 0.56±0.06. That value is used in interpreting the results of this work. He also finds that Z_p is ~ 0.45 charge units less than unchanged charge distribution for the heavy-mass peak.

Values of Z_p calculated from the results of this work and from cumulative yields of xenon isotopes reported by Wahl et al.⁷ are shown in Table V. Values of A' are estimated using the neutron-emission results of Bowman et al.¹⁶ Yield values are shown as fractional cumulative yields. The value reported for the yield of ¹³⁴Te may be high by ~ 0.01 , since the independent yield of ¹³⁴Xe, which may be that high, has been neglected in the calculations. In order to show the part of the uncertainty in Z_p due to uncertainties in the yield and the part due to uncertainties in σ , we have calculated Z_p for $\sigma = 0.56$ and also for $\sigma = 0.56 \pm 0.06$. Values of Z_p -A'(98/252) are shown in the last column.

 ± 0.0007 , and ± 0.0008 , respectively.

^c Uncertainties in the yield due to uncertainties of ± 1 min in the half-life of 134 I, ± 1 min in the half-life of 134 Te, and ± 0.25 min in t are ± 0.011 , ± 0.005 , and ± 0.003 , respectively.

^d Uncertainties in the yield due to uncertainties of ± 0.2 min in the halflife of ¹³⁸Xe and ± 0.125 min in t are ± 0.007 and ± 0.005 , respectively.

There is no direct evidence that the value of σ for ²³⁵U fission is satisfactory for interpreting yields from ²⁵²Cf fission. However, Z_p calculated for A = 136, assuming that ¹³⁶Sb is nearly complementary to ¹¹²Ag, is in good agreement with those calculated from the yields of $^{\bar{1}34}$ I and 138 Cs, indicating that σ for 252 Cf fission must be similar to that for ²³⁵U fission.

Strom *et al.*⁸ have suggested that σ decreases with mass in the region A = 131-133 in ²³⁵U fission and that the same effect might be expected in the mass-134 region. This suggestion was based on measurements of yields of ¹³¹Sn, ¹³¹Sb, ¹³¹Te, ¹³²Sn, ¹³²Sb, ¹³²Te, ¹³³Sb, ¹³³Te, and ¹³³I. When yields of ¹³¹I and ¹³²I reported more recently by Greendale and Delucchi17 are considered, however, the values of σ for A = 131, 132, and 133 are consistent, within the limits of error reported for the vields, with the limits of error chosen for σ in this work. Delucchi et al.¹⁸ do estimate a significantly smaller value for σ for the A = 134 chain from fractional cumulative yields of ¹³⁴Sb, ¹³⁴Te, and ¹³⁴Xe. However, since ¹³⁴Sb has both 51 protons and 83 neutrons, it is not surprising that its yield is very low. When only ¹³⁴Te and ¹³⁴Xe are considered, Delucchi et al.¹⁸ estimate σ to be 0.55 for that chain. We believe, therefore, that the limits for σ chosen here are reasonable ones.

Furthermore, the fractional cumulative yields for ¹³⁴Te, ¹³⁹Xe, and ¹⁴⁰Xe are such that the values of Z_p calculated from them are relatively independent of the values chosen for σ . An uncertainty of ± 0.06 in σ represents an uncertainty of less than ± 0.05 in Z_p as shown in Table V. The same uncertainty in σ results in an uncertainty of less than ± 0.10 in Z_p values calculated from the yields of ¹³⁸Xe and ¹⁴¹Xe. Only the Z_p value calculated from the fractional cumulative yield of ¹¹²Pd is a sensitive function of the value chosen for σ.

¹⁵ A. C. Wahl (private communication). ¹⁶ H. R. Bowman, J. C. D. Milton, S. G. Thompson, and W. J. Swiatecki, Phys. Rev. **129**, 2133 (1963).

¹⁷ A. E. Greendale and A. A. Delucchi, U.S. Naval Radiological Defense Laboratory, Laboratory Report No. USNRDL-TR-69-11, 1968 (unpublished)

¹⁸ A. A. Delucchi, A. E. Greendale, and P. O. Strom, Phys. Rev. 173, 1159 (1969).

			Fractional cumulative	Unc	ertainty Z_p		
 A	Α'	Ζ	yield	$\sigma = 0.56$	$\sigma = 0.56 \pm 0.06$	$Z_p - A'(98/252)$	
112	114.9	46	$0.991{\pm}0.004^{a}$	$45.18_{-0.11}^{+0.08}$	$45.18_{-0.27}^{+0.20}$	$+0.50_{-0.27}$ $+0.20$	
134	135.0	52	$0.74{\pm}0.03^{a}$	52.14 ± 0.05	52.14 ± 0.09	-0.36 ± 0.09	
138	139.5	54	0.89±0.03ª	$53.82_{-0.10}^{+0.08}$	$53.82_{-0.19}^{+0.14}$	$-0.43_{-0.19}^{+0.14}$	
139	140.6	54	0.67±0.01 ^b	54.25 ± 0.02	54.25 ± 0.04	-0.43 ± 0.04	
140	141.6	54	$0.45{\pm}0.01^{b}$	54.57 ± 0.01	54.57 ± 0.02	-0.50 ± 0.02	
141	142.7	54	0.172 ± 0.005^{b}	55.03 ± 0.01	55.03 ± 0.07	-0.46 ± 0.07	

TABLE V. Values of Z_p and $Z_p - A'(98/252)$ calculated from fractional cumulative yields

^a This work.

It is apparent from Table V that Z_p is ~0.45 charge units lower than A'(98/252) for the heavy chains and ~0.45 charge units higher than A'(98/252) for the one light chain. This is similar to the results mentioned above for fission of ²³³U, ²³⁵U, and ²³⁹Pu with the exception that Z_p 's for ²⁵²Cf fission seem to fall somewhat closer to unchanged charge distribution than they do for ²³³U and ²³⁹Pu fission. These results provide no evidence for any peak or fine structure in the Z_p curve in this mass region.

Values of $Z_p - A'(98/252)$ calculated from the results of radiochemical experiments and from the x-ray measurements of Glendenin and Unik¹ and of Kapoor *et al.*² are shown in Table VI. The results of Glendenin and Unik are taken directly from their paper. The results of Kapoor *et al.* are estimated from Fig. 5 of their

TABLE VI. Comparison of $Z_p - A'(98/252)$ values from radiochemical and x-ray measurements.

	Z_{1}	-A'(98/252)	
		X-1	ray
A'	Radiochemical	Glendenin and Unik	Kapoor et al.
114.9	+0.50 _{-0.27} +0.20 a		
114.7		$+0.6\pm0.2$	
115.2			$+0.7 \pm 0.1$
135.0	-0.36 ± 0.09^{a}		
134.9		$+0.2\pm1.0$	
135.4			-0.8 ± 0.1
139.5	-0.43 _{-0.19} +0.14 a		
138.2		-0.6 ± 0.2	
138.8			-1.1 ± 0.1
140.6	-0.43 ± 0.04^{b}		
141.3		-0.2 ± 0.2	
140.5			-0.8 ± 0.1
141.6	$-0.50{\pm}0.02^{b}$		
142.2			-0.6 ± 0.1
142.7	-0.46 ± 0.07^{b}		

^a This work.

^b Reference 7.

^b Reference 7.

paper. The results of Glendenin and Unik are in general agreement with those from the radiochemical experiments but show a very large uncertainty near A'=135. The results of Kapoor *et al.* differ from those from radiochemical experiments by ~ 0.3 charge units on the average. There is better internal agreement for the results of the radiochemical experiments than for either of the x-ray experiments.

Watson *et al.*⁴ have shown that x-ray yields from even-Z fragments in this mass region are much smaller than yields from odd-Z fragments for ²⁵²Cf fission. They conclude from these results that true charge distribution in this region may be represented poorly by values of Z_p calculated from the results of x-ray measurements. The results of this work and previously published radiochemical results for ²⁵²Cf fission support this conclusion.

ACKNOWLEDGMENTS

Most of this work was carried out during 1967–1968 when D.E.T. was a visiting scientist at Oak Ridge National Laboratory and M.E. and C.P. were enabled to make short visits to the Laboratory by travel grants from Oak Ridge Associated Universities. We wish to thank Dr. Robert Harbour for help with many of the exposures and Mr. Loren Berge for analyzing decay curves. We are especially grateful to Dr. O. L. Keller, Dr. C. E. Bemis, Jr., and Dr. Robert L. Ferguson for their generous assistance during our stay in Oak Ridge. Both the ²⁵²Cf source and the borated-paraffin shield were loaned to us by Dr. Bemis.

APPENDIX

Purification of ¹¹²Ag and ¹¹³Ag

Magnesium catcher foils were dissolved in 1 M HNO₃ containing 20 mg Ag⁺ carrier. Silver was precipitated as AgCl and the precipitate separated from the solution by centrifugation. The end of centrifugation was taken as the time of separation of silver from palladium. The silver was purified by repeated precipitations as

AgCl and Ag₂S using the method of Glendenin.¹⁹ The final product was collected as AgCl on tared filterpaper circles, dried, weighed, mounted on flat aluminum planchets, and covered with 6.5-mg/cm² polyester tape.

Purification of ¹³⁴I and ¹³⁵I

Separation of iodine was based on a method described by Wahl.20 Aluminum foils were dissolved in concentrated HCl containing 20 mg of I⁻ carrier. After a foil was dissolved, 10 ml of H₂O and 20 ml of CCl₄ were added and the solution stirred vigorously. As the solution was being stirred, 1 ml of $3\%~\mathrm{H_2O_2}$ was added to oxidize the I^- to $\mathrm{I}_2.$ Stirring was continued for 15 sec, then stopped, and the phases separated. The end of stirring was taken as the time at which separation of iodine from tellurium occurred. Iodine was purified by a series of steps consisting of reduction to I- with HSO_3^- , back-extraction into water, oxidation to I_2 with NaNO₂, and extraction into CCl₄. The product was collected as AgI and mounted as above. For runs 242 and 244, foils were dissolved in NaOH containing KI carrier. The I was oxidized to IO_4^- by heating with NaClO and then reduced to I⁻ before separation. This procedure was followed in order to force exchange between radio-iodine and iodine carrier. Since the ¹³⁴I/¹³⁵I ratios were not significantly different than those found in runs with similar separation times, namely, runs 238 and 240, we conclude that satisfactory exchange was achieved using the normal procedure.

Purification of ¹³⁸Cs and ¹³⁹Ba

The method described by Wolfsberg²¹ was used to separate ¹³⁸Cs from ¹³⁸Xe. Magnesium foils were dissolved in 20 ml of hot (95°C) 1 M HCl containing 20 mg Ba^{++} and 10 mg Cs^{+} carriers. The solution was in an 80-ml fritted glass funnel through which helium was flowing. As soon as the foil had dissolved ($\sim 15 \text{ sec}$) the gas flow was stopped and the solution transferred to a flask. The mid-dissolving time was taken as the time of separation of cesium from xenon. After about 1 h (to allow most of the 9.5-min ¹³⁹Cs to decay), barium was separated from cesium by precipitation as BaCO₃. This time was taken as the time of separation of cesium from barium. The cesium was purified using the method of Handley and Burros²² which calls for repeated precipitations as cesium tetraphenylboron and was collected as that compound, weighed, and mounted as above. After about 2 h (to allow decay of short-lived barium isotopes) barium was purified by repeated precipitations of BaCl₂ from ether-HCl solutions using the method of Minkkinen.23 The final product was collected as BaCrO₄, weighed, and mounted as above.

Half-Life of ¹¹³Ag

The only value given by Lederer et al.²⁴ for the halflife of ¹¹³Ag is 5.3 h (318 min). Roche²⁵ reports a value of 315 ± 3 min. Resolution of decay curves for our runs 81, 117, and 126, runs for which the amount of 187-min ¹¹²Ag was small compared to the amount of ¹¹³Ag, gave a half-life of \sim 322 min for ¹¹³Ag. Since the calculated value of the ¹¹²Ag yield was a very sensitive function of the value used for the half-life of ¹¹³Ag, we remeasured that half-life.

Natural uranium was irradiated in the University of Missouri Research Reactor for a few seconds and silver was separated less than 5 min after the end of the irradiation by a palladium dimethylglyoxime scavenge followed by the silver purifications described above. Roche²⁵ has shown that the independent yield of ¹¹²Ag from fission of $^{235}\mathrm{U}$ is negligible, so the only $^{112}\mathrm{Ag}$ in the samples was the very small amount growing from 21-h ¹¹²Pd. Three such runs were made. The product from one was counted on an end-window Geiger tube, from another on a gas-flow end-window detector operated in the Geiger region, and from the third on a gas-flow pancake counter also operated in the Geiger region. Initial count rates were ~ 600 cpm (counts/min) so that the corrections for coincidence losses were small. Each sample was counted continuously, i.e., it was not removed from beneath the detector and counting was stopped only long enough to record times and counts, for three days. After that period samples were counted at less frequent intervals for about 30 days or until the count rate was less than 1 cpm above background.

The decay curve for each sample was resolved into a 7.5-day component due to ¹¹¹Ag and into two unknown components, one representing ¹¹³Ag and the other a possible impurity. The half-lives of ¹¹³Ag from the three experiments were 320.8 ± 2.3 min, 321.7 + 2.9 min, and 322.4 ± 2.4 min. In only one, the third, was an impurity greater than 1 cpm found. That one had \sim 7 cpm of an impurity with a half-life of \sim 5000 min. On the basis of these experiments we have used 322 ± 3 min as the halflife of ¹¹³Ag.

²² T. H. Handley and C. L. Burros, quoted by H. L. Finston and M. T. Kinsley, *The Radiochemistry of Cesium* (National Academy of Sciences—National Research Council, Washington, D.C., 1961).

²³ C. O. Minkkinen, quoted by D. N. Sunderman and C. W. Townley, The Radiochemistry of Barium, Calcium, and Strontium (National Academy of Sciences-National Research Council, Washington, D.C., 1960).

 ²⁴ C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (John Wiley & Sons, Inc., New York, 1967).
 ²⁵ M. F. Roche, Ph.D. thesis, University of Missouri, 1968

⁽unpublished).

Half-Life of ¹³⁸Xe

Lederer et al.²⁴ list both 17 min and 14 min as halflives of ¹³⁸Xe. In order to determine which of the two values was correct we remeasured the independent yield of ¹³⁸Cs from fission of ²³⁵U using our technique and compared that value to the one reported by Wolfsberg.²¹ The precision of his measurement was not sensitive to the half-life, since he separated cesium from xenon within a few seconds after fission. Our measurements, however, were extremely sensitive to the halflife of ¹³⁸Xe since separations were not made until five or more minutes after fission. Using 17 min as the halflife, we calculated a value of 0.12 for the independent

yield of ¹³⁸Cs from fission of ²³⁵U, a value in gross disagreement with that of 0.047 reported by Wolfsberg. We found, however, that our calculated yield of ¹³⁸Cs was consistent with that of Wolfsberg if we used 13.9 min as the half-life of ¹³⁸Xe. This value is in agreement with previously reported values of 14.0 ± 0.2 min by Clarke and Thode,²⁶ 14.5±0.5 min by Patzelt and Herrmann,²⁷ and 14.1±0.8 min by Archer and Keech.²⁸ We have therefore used 14 min as the half-life of ¹³⁸Xe.

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VOLUME 1, NUMBER 3

MARCH 1970

Level Structure of Ce^{141} from the $Ce^{140}(n, \gamma)Ce^{141}$ Reaction*

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The level scheme of the $_{58}$ Ce $_{83}$ ¹⁴¹ nucleus was studied by observing the γ rays from the Ce¹⁴⁰ (n_{th}, γ) reaction at the Brookhaven high flux beam reactor. γ -ray singles and coincidence spectra were obtained with a 20-cm³ Ge(Li) detector and a Ge(Li)-Ge(Li) detector combination. Thirty-two γ rays were assigned to Ce^{141} and all but three of these γ rays were included in the Ce^{141} level scheme. We have identified nine of these transitions as primary transitions from the neutron capture state to low-lving levels in Ce¹⁴¹. These primary transitions have the following energies (in keV) and relative intensities [I(662) = 100]; 4766.6± $0.5(48.0), 4291.4 \pm 0.6(22.1), 3619.7 \pm 0.6(4.0), 3435.0 \pm 0.6(2.0), 3239.0 \pm 1.5(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.0), 3239.0 \pm 0.5(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.0), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.7), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.7), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.7), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.7), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3435.0 \pm 0.6(2.7), 3239.0 \pm 0.6(2.7), 3092.5 \pm 0.5(2.8), 3092.5 \pm 0.5(2.8),$ $3017.1 \pm 0.7(4.7)$, $3003.3 \pm 0.6(3.9)$, and $2905.9 \pm 0.5(3.0)$. Our data, together with the information available from earlier charged particle reaction and radioactive decay studies, establish levels in Ce¹⁴¹ with energies 662.0 ± 0.1 , 1137.0 ± 0.3 , 1497.3 ± 0.4 , 1808.7 ± 0.5 , 1994.0 ± 0.6 , 2189.6 ± 0.5 , 2336.3 ± 1.0 , 2410.8 ± 0.5 , 2336.3 ± 1.0 , 2410.8 ± 0.5 , 2336.3 ± 1.0 , 2410.8 ± 0.5 , 2336.3 ± 0.5 , 2336.3 ± 0.5 , 2410.8 ± 0.5 , 2336.3 ± 0.5 , 2410.8 ± 0.5 , 2410.80.6, 2425.6 ± 0.8 , and 2522.9 ± 0.6 keV. The measured neutron separation energy is 5428.6 ± 0.6 keV. All of these levels may be identified with levels observed earlier with lower accuracy in the $\operatorname{Ce}^{140}(d, p)\operatorname{Ce}^{141}$ reaction. As in the neighboring N=83 nuclei Ba¹³⁹ and Nd¹⁴³, there appears to be a correlation between the strengths of excitation of the $p_{3/2}$ and $p_{1/2}$ levels in the (n, γ) and (d, p) reactions. The 1808.7-keV state, shown here to have $\frac{3}{2}$ - spin and parity and not $\frac{5}{2}$ - as previously believed, exhibits properties typical of a core excitation state formed from the coupling of an $f_{7/2}$ neutron to the first excited 2+ state of the semimagic Ce¹⁴⁰ core. In particular, an E2 transition proceeds from this state to the $\frac{7}{2}$ - ground state in competition with M1 transitions to the $\frac{3}{2}$ - and $\frac{1}{2}$ - first and second excited states. Accordingly, a calculation of the properties of the low-lying excited states in Ce¹⁴¹ was carried out on the basis of the weakcoupling model. The results obtained are in reasonable agreement with the measured properties of the levels of Ce141. In particular, it is shown that the measured branching ratios of the transitions from the 1497.3- and 1808.7-keV levels are correctly predicted by this model. The level scheme of Ce¹⁴¹ is compared with the level schemes of the other even-Z, N = 83 nuclei.

I. INTRODUCTION

THE Ce¹⁴¹ nucleus has eight protons outside the \blacksquare closed shell at Z=50 and one neutron outside the N = 82 closed shell. The low-lying states of this nucleus are expected to have the relatively simple character associated with the single particle outside the closed shell. Information on the properties of the energy levels of this nucleus is thus of great interest for comparison with the predictions of nuclear models.

Our present limited knowledge of the properties of the energy levels of the Ce¹⁴¹ nucleus is derived from studies of the La¹⁴¹ β^- decay, the Ce¹⁴⁰(d, p) reaction, and the elastic scattering of protons from Ce¹⁴⁰.

The decay of 3.8-h La¹⁴¹ to levels in Ce¹⁴¹ has been studied by Hahn and Strassmann,¹ Katcoff,² Duffield and Langer,3 and Schuman, Turk, and Heath.4 Their

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^{*} Work performed under the auspices of the U.S. Atomic Energy Commission.

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