PHYSICAL REVIEW C VOLUME 17, NUMBER 1 JANUARY 1978

Charge distribution in nuclear fission: Determination of fractional cumulative yields of ^{134}Te and ^{135}I in the spontaneous fission of ^{252}Cf

S. B. Manohar, Tarun Datta, S. S. Rattan, Satya Prakash, and M. V. Ramaniah
Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Bombay 400085, India
(Received 8 February 1977)

The fractional cumulative yields of ^{134}Te and ^{135}I in the spontaneous fission of ^{252}Cf are determined by following the growth and decay of ^{134}I and ^{135}Xe^{g} activities, respectively, on a 60-cm^3 Ge(Li) detector. The values of the most probable charge (Zp) for mass chains 134 and 135 are calculated assuming Gaussian distribution. The values of fractional cumulative yield are 0.791 and 0.975 for ^{134}Te and ^{135}I, respectively, and corresponding Zp values are 52.03 ± 0.06 and 52.40 ± 0.06. The data are compared with other fractional cumulative yield values from literature in the spontaneous fission of ^{252}Cf to see the effect of fragment shells on the width of the charge distribution.

[RADIOACTIVITY, FISSION Spontaneous fission of ^{252}Cf: measured γ activities ^{134}I and ^{135}Xe^{g} using Ge(Li) detector and obtained fractional cumulative yields; Zp values for the mass chain 134 and 135 deduced.]

INTRODUCTION

Nuclear charge distribution in the spontaneous fission of ^{252}Cf has been studied by several workers using physical methods such as coincidence measurements of fission fragments either with x rays[1-3] or γ transitions from even-even[4] nuclei. There are a few reported yields of specific fission products determined by radiochemical techniques. The fractional cumulative yields of several isotopes of xenon were reported by Wahl *et al.*[5] Von-Gunten, Flynn, and Glendenin[6] have reported independent yields of several shielded nuclei such as ^{82}Br, ^{86}Rb, ^{136}Cs, and ^{150}Pm. Troutner, Eichor, and Pace[7] have reported fractional independent yields of ^{112}Ag, ^{134}I, and ^{138}Cs. Troutner and Runnalls[8] and Harbour and Troutner[9] have reported the fractional cumulative yields of ^{131}Sb and ^{131}Te^{m} and ^{99}Nb^{m} in the spontaneous fission of ^{252}Cf.

The fractional cumulative yields of ^{134}Te and ^{135}I in the spontaneous fission of ^{252}Cf are reported in this paper as a part of our program of the study of charge distribution. The values of the most probable charge Zp were calculated assuming a Gaussian distribution with $\sigma = 0.56 \pm 0.06$.[10]

EXPERIMENTAL

The decay chains for the isobars involved in this work are

$$^{134}\mathrm{Sb}_{0.85\ \mathrm{sec}} \xrightarrow{\beta^-} {}^{134}\mathrm{Te}_{41.8\ \mathrm{min}} \xrightarrow{\beta^-} {}^{134}\mathrm{I}_{52.0\ \mathrm{min}} \xrightarrow{\beta^-} {}^{134}\mathrm{Xe}_{\mathrm{Stable}},$$

$$^{135}\mathrm{Te}_{18\ \mathrm{sec}} \xrightarrow{\beta^-} {}^{135}\mathrm{I}_{6.59\ \mathrm{h}} \begin{array}{l} \xrightarrow{\beta^-} 8.5\%\ {}^{135}\mathrm{Xe}^{m}_{15.3\mathrm{min}} \xrightarrow{\beta^-} {}^{135}\mathrm{Cs}_{2\times10^{6}\ \mathrm{y}}\ . \\ \qquad\qquad\qquad\quad \downarrow \\ \xrightarrow{\beta^-} 91.5\%\ {}^{135}\mathrm{Xe}^{g}_{9.17\ \mathrm{h}} \end{array}$$

The half-lives of the radionuclides were taken from "*KARLSRUHER NUKLIDKARTE*" (1974).[11] The branching in the decay of ^{135}I to ^{135}Xe^{m} is 8.5% as taken from Hawkings, Edwards, and Olmstead.[12]

The californium source of about 1.7 μg (of ^{252}Cf) used in this experiment was prepared by electrodeposition on stainless steel plate. The 0.5-mm-thick aluminium catcher foils were kept at a distance of 1 mm from the source using a sample holder. The time of collection of fission fragments varied from 10 minutes to 3 hours depending on the nuclide of interest. After collection, the catcher foils were mounted on aluminium plates and were checked and found to be free of α contamination.

The catcher foils were counted on a 60-cm^3 Ge(Li) detector coupled with a 400-channel analyzer. The resolution of the Ge(Li) system was about 1.5 keV at 122 keV and 3.9 keV at 1332 keV. The counting was done in the live-time mode of the analyzer where the maximum dead time was of the order of 5%. The samples were counted for suitable durations at appropriate intervals. The area of the photopeaks of interest were calculated by subtracting Compton contribution, from the total counts under the peak.

CALCULATIONS

Since the half-lives of the precursors of ^{134}Sb and ^{135}Te in the two cases are very short compared to the time of fission fragments collection and interval up to the first γ counting, the 134 and 135 mass chains were assumed to consist of ^{134}Te and ^{134}I and ^{135}I, ^{135}Xe^m, and ^{135}Xe^g, respectively.

In the case of ^{134}Te-^{134}I the peak area (A) of 847-keV γ ray of ^{134}I was related to the initial number of atoms of tellurium (N^0_{Te}) and of iodine (N^0_{I}) formed at the end of the irradiation by Eq. (1):

$$Y = \mathcal{E} X N^0_{\mathrm{Te}} + \mathcal{E} N^0_{\mathrm{I}}, \tag{1}$$

where

$$Y = \frac{A}{(1-e^{-\lambda_2 t})e^{-\lambda_2 T}}$$

and

$$X = 1 + \frac{\lambda_2}{\lambda_2 - \lambda_1} \times \frac{(e^{-\lambda_2 t} - e^{-\lambda_1 t})e^{-\lambda_2 t} + (1-e^{-\lambda_1 t})(e^{-\lambda_1 T} - e^{-\lambda_2 T})}{(1-e^{-\lambda_2 t})e^{-\lambda_2 T}} . \tag{3}$$

$\mathcal{E}$ is the net counting efficiency of the detector. λ_1, λ_2 are the decay constants of ^{134}Te and ^{134}I, respectively, t the time of collection, and T the time interval between the end of collection and the mean time of counting. Five different measurements with t varying from 10 to 20 min were carried out. The values of X and Y in Eq. (1) were calculated for each of these measurements and were fitted to obtain straight lines with least-squares analysis.

The fractional cumulative yield (FCY) of ^{134}Te is defined as the ratio of number of atoms of ^{134}Te to the sum of the number of atoms of ^{134}Te and ^{134}I, formed independently at the end of the collection. Since ^{134}Sn and ^{134}Sb have short half-lives, N^0_{Te} gives cumulative formation up to ^{134}Te and N^0_{I} gives independent formation of ^{134}I. Since the number of ^{134}Xe atoms independently formed is very small compared to that of ^{134}I the FCY for ^{134}Te is expressed as

$$\mathfrak{F}(^{134}\mathrm{Te}) = \frac{N^0_{\mathrm{Te}}}{N^0_{\mathrm{Te}} + N^0_{\mathrm{I}}} \tag{4}$$

and according to Eq. (1)

$$\mathfrak{F}(^{134}\mathrm{Te}) = \frac{m}{m+c} , \tag{5}$$

where m and c are the slope and intercept of the line represented by Eq. (1). Table I shows the data on the measurements of FCY of ^{134}Te. The calculated values of slope and intercept for each measurement are shown along with number of determinations for each collection. A typical plot of Y against X is given in Fig. 1.

The partial decay of ^{135}I to ^{135}Xe^g is through ^{135}Xe^m. Therefore, the growth and decay of ^{135}Xe^g, during the collection and interval up to the counting time, depends on the branching ratio, relative independent yields of ^{135}Xe^m and ^{135}Xe^g as well as on the half-life of ^{135}Xe^m. On the basis of similar data in the fission of ^{235}U (Ref. 13) it was assumed that the independent yields of ^{135}Xe^m and ^{135}Xe^g are the same. Since 91.5% of ^{135}I decays to ^{135}Xe^g and the half-life of ^{135}Xe^m is shorter compared to the collection time and the interval between collection and counting time, the error in FCY of ^{135}I due to the above assumption is negligible.

The peak area A of ^{135}Xe^g (249.6-keV γ ray) is related to the number of atoms of ^{135}I (N^0_{I}) and ^{135}Xe^g (N^0_{Xe}) formed at the end of irradiation by Eq. (6)

$$Y = \mathcal{E} X N^0_{\mathrm{I}} + \mathcal{E} N^0_{\mathrm{Xe}} , \tag{6}$$

where

$$Y = \frac{A}{E_{13}} , \tag{7}$$

TABLE I. The data on fractional cumulative yield of ^{134}Te.

Time of collection in minutes t	Number of measurements	Calculated values Slope m	Intercept c	Fractional cumulative yields of ^{134}Te $(m/m+c)$
10	5	0.538 ± 0.009	0.140 ± 0.016	0.794
12	12	0.313 ± 0.014	0.898 ± 0.013	0.762
15	12	0.300 ± 0.010	0.092 ± 0.014	0.765
18	14	0.337 ± 0.007	0.079 ± 0.019	0.810
20	8	0.424 ± 0.012	0.093 ± 0.016	0.820

Mean FCY = 0.791 ± 0.026

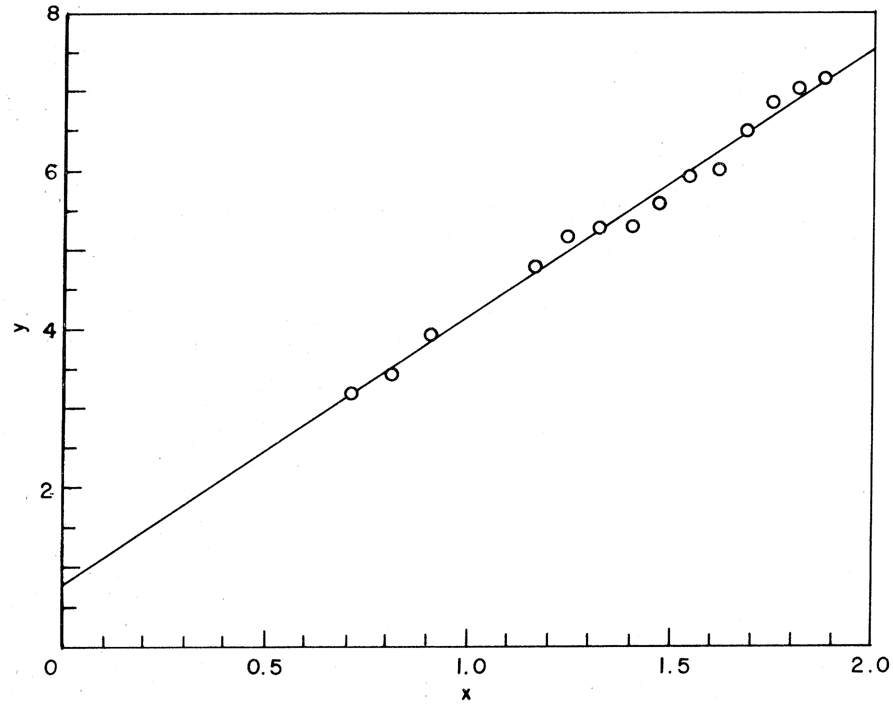

FIG. 1. Plot of x and y values for $t = 18$ min for the case ^{134}Te-^{134}I.

$$X = \frac{X'}{E_{13}}, \quad (8)$$

$$X' = [E_3 + E_1][Q \times b(1 - E_8) + E_8 \times Q] + Q[E_3 + E_1] + (1 - b)[E_8(1 - E)(E_4 + E_5) - E_5 \times E_{11}] + B(1 - E)E_4 . \quad (9)$$

λ_1, λ'_2, and λ_2 are the decay constants of ^{135}I, ^{135}Xe^m, and ^{135}Xe^g, respectively. t is the collection time and T is the time interval between the end of the collection and the mean time of counting. The different symbols in Eq. (7)–(9) are

$$E = e^{-\lambda_1 t}, \quad F = e^{-\lambda'_2 t}, \quad G = e^{-\lambda_2 t}, \quad H = e^{-\lambda_1 T},$$

$$P = e^{-\lambda'_2 T}, \quad Q = e^{-\lambda_2 T},$$

$$E_1 = \frac{(G - F)}{(\lambda_2 - \lambda'_2)}, \quad E_2 = \frac{(G - E)}{(\lambda_2 - \lambda_1)}, \quad E_3 = \frac{(1 - G)}{\lambda_2},$$

$$E_4 = \frac{(H - Q)}{(\lambda_2 - \lambda_1)}, \quad E_5 = \frac{(Q - P)}{(\lambda_2 - \lambda'_2)}, \quad E_6 = \frac{(1 - F)}{\lambda_2},$$

$$E_8 = \frac{\lambda'_2}{(\lambda'_2 - \lambda_1)}, \quad E_{12} = \lambda'_2 \times E_6 ,$$

$$E_{11} = E_{12} + E_8(F - E), \quad E_{13} = Q[2 \times E_3 + E_1) - E_{13} \times E_5 ,$$

$b = 0.915$,

the branching ratio of ^{135}I to ^{135}Xe^g, and A is the

TABLE II. The data on fractional cumulative yield of ^{135}I.

Time of collection in minutes t	Number of measurements	Calculated values Slope m	Intercept c	Fractional cumulative yields of ^{135}I $(m/m+c)$
40	12	30.53 ± 0.22	0.968 ± 0.10	0.969
60	11	7.50 ± 0.12	0.162 ± 0.06	0.979
120	10	27.43 ± 0.31	0.940 ± 0.014	0.967
150	9	27.60 ± 0.22	0.686 ± 0.10	0.979
180	10	30.24 ± 0.21	0.609 ± 0.10	0.980

Mean FCY $= 0.975 \pm 0.006$

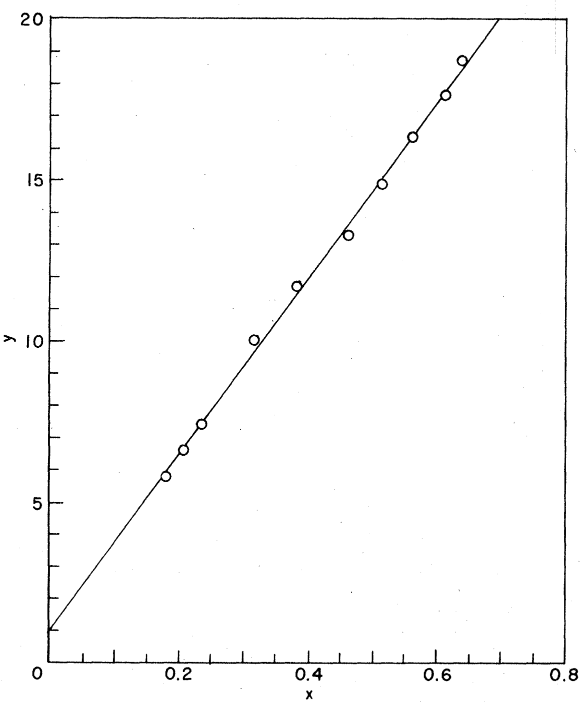

FIG. 2. Plot of x and y values for $t = 120$ min for the case ^{135}I-^{135}Xe.

peak area of $^{135}Xe^{g}$ (249.6 keV) peak.

The FCY of ^{135}I was obtained in a similar manner as in the case of ^{134}Te. Table II gives the data on FCY of ^{135}I for all the five sets of measurements and the values of slope and intercept along with number of determinations for each collection. A typical plot of Y against X is given in Fig. 2.

RESULTS

It is well established that the fractional cumulative yields for an isobaric chain are represented by a Gaussian distribution given by Eq. (10):

$$Y_Z^{\text{cum}} = \frac{1}{\sigma\sqrt{\pi}} \int_{-\infty}^{Z+0.5} \exp - \frac{(Z-Z_p)^2}{2\sigma^2}\, dZ \,, \tag{10}$$

where Z_p is the most probable charge for the fission product chain and σ is the standard deviation.

The fractional cumulative yields for ^{134}Te and ^{135}I were used to calculate the Z_p values by using Eq. (10) with $\sigma = (0.56 \pm 0.06)$. Table III shows a comparison of the values of Z_p obtained in this work with literature values from radiochemical data.[5]

It has been shown by Wahl *et al.*[10] that the difference between most probable charge (Z_p) and the one calculated on the basis of unchanged charge distribution (Z_{UCD}) is given by Eq. (11) for the heavy fission products

$$(Z_p - Z_{\text{UCD}}) = -0.45 \pm 0.10 \,. \tag{11}$$

Column 6 of Table III shows that the correlation following Eq. (11) is valid for ^{252}Cf fission also. Figure 3 shows these data along with most recent data of physical experiments. It can be seen that the difference in Z_{UCD} and Z_p values is constant and is about -0.45 charge unit.

DISCUSSION

The fractional cumulative yields ^{134}Te and ^{135}I obtained in the spontaneous fission of ^{252}Cf in the present investigation along with literature values

TABLE III. The fractional cumulative yields for different fission products and the comparison of Zp values obtained from this with those calculated on the basis of unchanged charge distribution (UCD).

Nuclide	Fractional cumulative yield	The most probable value of charge (Zp) assuming $\sigma = 0.56 \pm 0.06$	A' [a]	Z_{UCD} $A'\frac{98}{252}$	$Z_p - Z_{\text{UCD}}$	$Z - Z_{\text{UCD}}$
^{134}Te	0.74 ± 0.03	52.14	135.1	52.539	−0.399	−0.539
^{134}Te [b]	0.69 ± 0.03	52.03 ± 0.06	135.1	52.539	−0.509	−0.539
^{135}I [b]	0.975 ± 0.006	52.40 ± 0.06	136.2	52.967	−0.567	+0.033
^{136}Xe	0.995 ± 0.001	52.05	137.4	53.433	−0.383	+0.567
^{138}Xe	0.89 ± 0.03	53.83	139.6	54.288	−0.488	−0.288
^{139}Xe	0.67 ± 0.01	54.26	140.6	54.677	−0.419	−0.677
^{140}Xe	0.45 ± 0.01	54.57	141.7	55.105	−0.536	−1.105
^{141}Xe	0.172 ± 0.005	55.06	142.8	55.533	−0.473	−1.533

[a] $A' = A + \nu(A)$; values taken from Ref. 14.
[b] This work. Other values taken from Ref. 5.

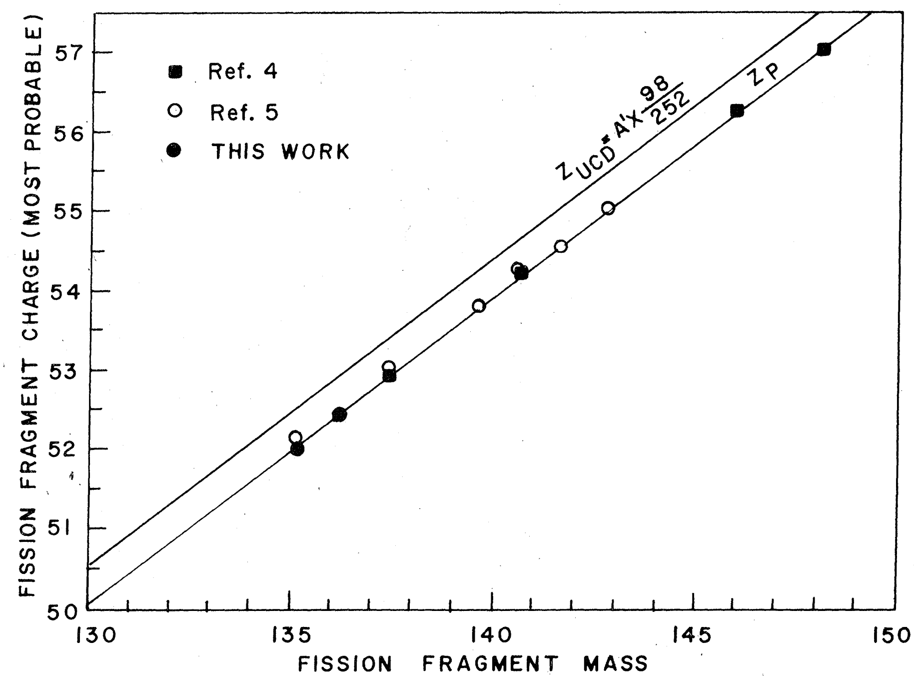

FIG. 3. A plot showing the comparison of Z_{UCD} and Zp in the spontaneous fission of ^{252}Cf.

for ^{136}Xe, ^{138}Xe, ^{139}Xe, ^{140}Xe, and ^{141}Xe are shown in Fig. 4 in which FCY's are plotted as a function of $Z - Z_{\mathrm{UCD}}$. Two straight lines corresponding to two values 0.50 and 0.62 for σ are drawn on the probability graph. It is seen that while four out of the five FCY's of Xe isotope are within the limits of $\sigma = 0.56 \pm 0.06$, the values of ^{134}Te and ^{135}I from the present work are on the lower side of the σ values, i.e., nearer $\sigma = 50$. The high FCY's for ^{134}Te and ^{135}I may be explained as being due to the odd-even effect and/or shell effect and both these are briefly discussed here.

Odd-even effect. In the context of odd-even effect, it is relevant to refer to two recent contributions. The first one is that of Amiel and Feldstein[14] who have analyzed the data on independent yields of fission products in the light and heavy mass regions in the thermal-neutron fission of ^{235}U and found that the probability of formation of fragments with even Z is 25% higher compared to normal distribution while that of fragments with odd Z is 25% lower. The second one pertains to the work of Siegert *et al.*[15] who have formulated an empirical equation for correcting for the odd-even effect; according to their formulation, the yield Y of the nuclear charge Z in the mass chain A is given by Eq. (12):

$$Y(A, Z) = Y_{\mathrm{Wahl}}^{(A, Z)}(1 + 0.19\delta p + 0.04\delta n) , \qquad (12)$$

where δp or δn is +1 or −1 for even or odd number of protons or neutrons as the case may be. This formulation was arrived at by them on the basis of experimental data obtained in mass region 79 to 100. The high FCY of ^{135}I obtained in the present work may be attributed to the odd-even effect, since the fission product mass chain 135 is due to neutron emission from fragment chain of even mass 136 ($A' = 136.2$).[16] Calculations were carried out to correct the normal yield value for odd-even effect for three values 0.50, 0.56, and 0.62 for σ using (a) the method of Amiel and Feldstein[14] and (b) the empirical equation of Siegert *et al.*[15] In both cases the corrected normal yield values compared well with the experimental value of 0.975 for $\sigma = 0.50$ as the contributions of neutron and proton pairing effects are in the same direction (total correction in both the cases is ~25%). Since the fission product mass chain 134 is due to neutron emission from fragments of odd mass chain 135 ($A' = 135.1$),[16] the formation of even-even or odd-odd fission fragments is ruled out and so the method of Amiel and Feldstein cannot be applied to explain the high FCY of ^{134}Te. Therefore, calculations were carried out to assess the odd-even effect in the case of ^{134}Te using the equation of Siegert *et al.* where the effects of proton and neutron pairings are brought out separately. It was found that for all the three values of σ, namely,

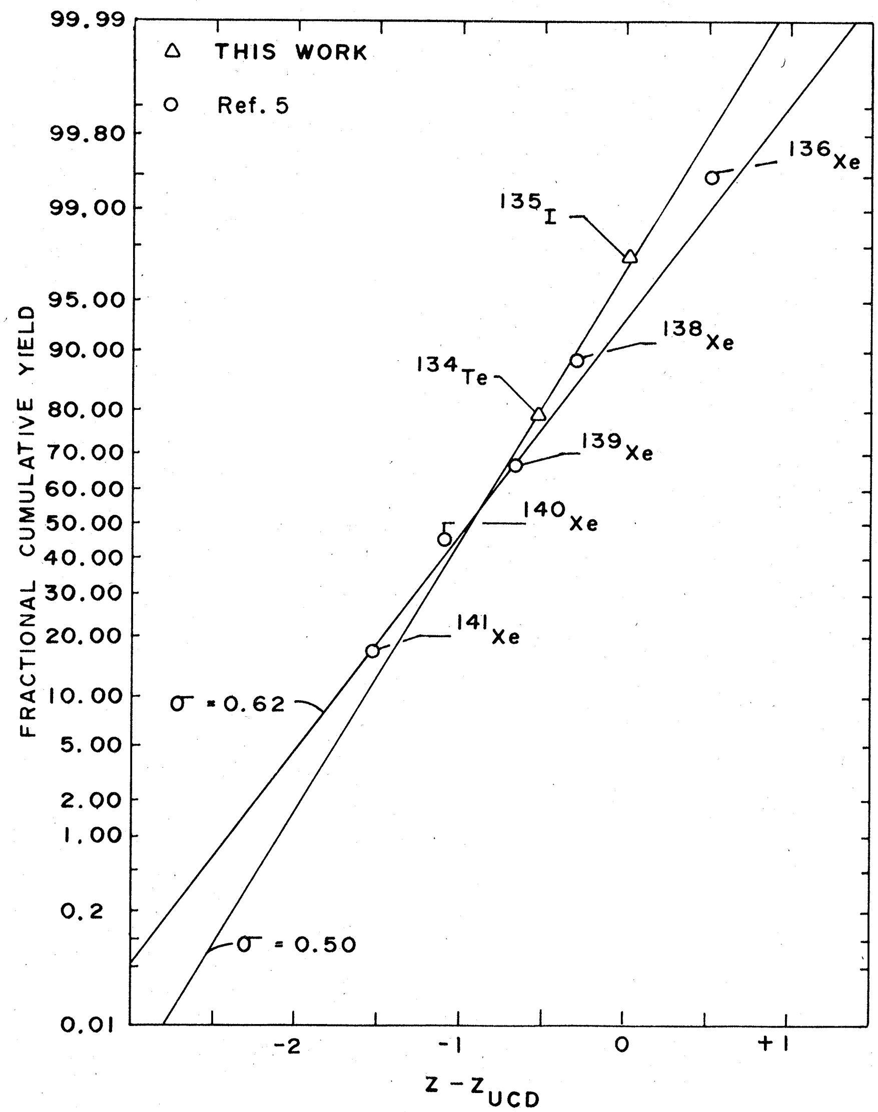

FIG. 4. A plot of Z-Z_{UCD} as a function of FCY for ^{252}Cf spontaneous fission.

0.50, 0.56, and 0.62, the corrected normal yield value is higher than the experimental value 0.791 making it difficult to say whether the same apportionment of proton and neutron pairing effects as observed in the light mass region 79 to 100 is valid for the heavy mass region 134–136 of the present work for assessing the odd-even effect.

Shell effect. For the mass chain 135, it was found that even after the correction for odd-even effect, the measured FCY corresponds to the lower value of σ (near 0.50 for 136 mass chain) indicating the possible influence of shells. Similarly the FCY of ^{134}Te indicates value of σ near 0.50, though the analysis carried out for odd-even effect was not conclusive. In this connection it is relevant to refer to the analysis by Notea,[17] of the data of Strom *et al.*[18] and Wahl *et al.*[10] on the charge distribution in the fission of ^{235}U which has shown that the values of σ for mass chains 131, 132, 133, and 136 are 0.68, 0.54, 0.45, and 0.32, respectively, indicating a lower σ value in the mass region 132–136 in the fission of ^{235}U. Extending this trend to the spontaneous fission of ^{252}Cf, one can attribute the high FCY's of ^{134}Te and ^{135}I of the present work to shell effects. Further work in this region would be very interesting.

[1]L. E. Glendenin and J. P. Unik Phys. Rev. 140, B1301 (1965).
[2]S. S. Kapoor, H. R. Bowman, and S. G. Thompson, Phys. Rev. 140, 1310 (1965).
[3]S. R. Dolce, W. M. Gibson, and T. D. Thomas, Bull. Am. Phys. Soc. 11, 335 (1966).
[4]E. Cheiftez, J. B. Wilhelmy, R. C. Jared, and S. G. Thompson, Phys. Rev. C 4, 1913 (1961).
[5]A. C. Wahl, R. L. Furguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. 126, 1112 (1962).
[6]H. R. Von-Gunten, K. F. Flynn, and L. E. Glendenin, J. Inorg. Nucl. Chem. 31, 3357 (1969).
[7]D. E. Troutner, M. Eichor, and C. Pace, Phys. Rev. C 1, 1044 (1970).
[8]D. E. Troutner and N. G. Runnalls, J. Inorg. Nucl. Chem. 33, 2271 (1971).
[9]R. M. Harbour and D. E. Troutner, J. Inorg. Nucl. Chem. 33, 1 (1971).
[10]A. C. Wahl, A. E. Norris, R. A. Rouse, in *Proceedings of the Second International Atomic Symposium on the Physics and Chemistry of Fission, Vienna, 1969* (IAEA, Vienna, 1969), paper SM-122/116, p. 813.
[11]Chart of Nuclides, prepared by W. Seelmann-Eggebert, G. Pfennig, and H. Munzel, Institut fur Radiochemie, Kernforschungszentrum, Karlsruhe, 1974.
[12]R. C. Hawkings, W. J. Edwards, and W. J. Olmstead, Can. J. Phys. 44, 237 (1966).
[13]M. E. Meek and B. F. Rider, Compilation of Fission Product Yields (NEDO-12151-1), Vallecitos Nuclear Center, 1974 (unpublished).
[14]S. Amiel and H. Felstein, in *Proceedings of the Third International Atomic Energy Symposium on the Physics and Chemistry of Fission, Rochester, 1973* (IAEA, Vienna, 1974), paper SM-174/25, p. 76.
[15]G. Siegert, H. Wollnik, J. Freif, R. Decker, G. Fielder, and B. Pfeiffer, Phys. Rev. C 14, 1864 (1976).
[16]V. F. Apalin, Yu. N. Gritsyuk, I. E. Kutikev, V. I. Lebedev, and L. A. Makaelian, Nucl. Phys. 71, 535 (1965).
[17]A. Notea, Phys. Rev. 182, 1331 (1969).
[18]P. O. Strom, D. L. Love, A. E. Greendale, A. A. Delucchi, D. Sam, and N. E. Ballou, Phys. Rev. 144, 984 (1966).