Mass and Nuclear Charge Distributions from the Spontaneous Fission of ^{254}Fm†

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(Received 7 July 1972; revised manuscript received 28 June 1973)

The relative fission yields of 20 mass chains from ^{254}Fm spontaneous fission were measured by direct high-resolution γ-ray spectroscopy of the gross fission products. Reflected yields were calculated based on a prompt-neutron fission yield of 3.94; the absolute yields were determined by requiring the sum of all yields to be 200%. A peak-to-valley ratio for the mass-yield curve of $\geq$18 was determined radiochemically by measuring the cumulative yields of ^{140}Ba and ^{127}Sb.

^{254}Fm was produced by irradiating ^{253}Es in a high-flux reactor and then chemically separating the 3.24-h ^{254}Fm from its 39.3-h ^{254}Es^{m} parent. An aluminum "catcher" foil was used to collect the ^{254}Fm fission products. Nuclear charge distribution studies included measurements of the independent yields of ^{140}La, ^{135}Xe, ^{134}I, and ^{132}I relative to their cumulative yields from direct γ-ray-spectroscopy data.

INTRODUCTION

The fragment mass distributions for ^{257}Fm spontaneous fission (SF)[1,2] and for ^{257}Fm thermal-neutron-induced fission (n_{th}, f)[2] derived from kinetic-energy measurements are markedly more symmetric than any previously reported mass distributions for low-energy fission of actinide nuclides. The radiochemical determination of the ^{256}Fm(SF) mass-yield curve[3] showed definite asymmetric fission, but with a peak-to-valley ratio of only 12. Previous kinetic-energy studies[4] of the fragment mass distribution for ^{254}Fm(SF) showed asymmetric fission. Flynn *et al.*[3] are now investigating the mass distribution for ^{257}Fm(n_{th}, f) by the radiochemical method.

Radiochemical measurements of fission-product mass distributions eliminate both the uncertainty in mass resolution and the need for a mass-dependent correction for neutrons emitted by the fragments that are inherent in mass distributions derived from kinetic-energy measurements on coincident fission fragments. In this work, the fission-product mass distribution for ^{254}Fm(SF) was determined by direct high-resolution γ-ray spectroscopy of the gross fission products[5-7] to continue the study of trends in mass-distribution parameters for fissionable fermium isotopes. The ratio of the mass yield of ^{140}Ba to ^{127}Sb was measured radiochemically to estimate the peak-to-valley ratio of the ^{254}Fm(SF) mass-yield curve. Nuclear charge distribution studies included measurements of the independent yields of ^{140}La, ^{135}Xe, ^{134}I, and ^{132}I relative to their cumulative yields from direct γ-ray-spectroscopy data for the gross fission products as a function of time after fission.

EXPERIMENTAL AND CALCULATIONS

About 15 μg of ^{253}Es was irradiated in a quartz vial in the Savannah River Plant high flux reactor for 36 h. After the ^{254}Fm had grown into equilibrium with its 39.3-h ^{254}Es^{m} parent, fermium was separated from einsteinium by cation-exchange elution development chromatography with ammonium α hydroxyisobutyrate.[8] The fermium fraction was collected on a platinum plate, dried, and flamed. An aluminum-catcher-foil technique[9] was used to collect the fission fragments from the 3.24-h ^{254}Fm source. Two catcher foils were exposed, the first for direct γ-ray analysis and the second for radiochemical analysis.

The first catcher foil, exposed for 4.48 h, was analyzed for γ activities as a function of time after exposure by a 45-cm^3 Canberra Ge(Li) detector with a resolution of 2.6 keV (full width at half maximum) at 1332.5 keV. With the sample in the same position, γ spectra were recorded from a few minutes to a few months after exposure over an energy range of 0 to 1.5 MeV. Areas of photopeaks were determined by visually subtracting the Compton background from the total counts. The photopeak-area data for each nuclide were consistent with each nuclide's appropriate half-life. For the ^{144}Ce and ^{106}Ru measurements, a Compton suppression spectrometer consisting of a 35-cm^3 Ge(Li) detector and a 12.7- × 15.2-cm shield was used. The photopeak-to-Compton ratio was 190 for the 661.6-keV ^{137}Cs γ ray.

Because a standard fission system was not used as a reference, it was necessary to determine the detection efficiency of the Ge(Li) detector as a function of energy. The detector was calibrated

with the following standard sources: ^{241}Am, ^{139}Ce, ^{109}Cd, ^{203}Hg, ^{22}Na, ^{85}Kr, ^{137}Cs, ^{54}Mn, ^{88}Y, and ^{94}Nb. Relative yields could then be calculated because the abundances of the γ rays analyzed were known (see Table I).[10]

Consider a fission-product decay chain

$$F \rightarrow 1 \rightarrow 2, \tag{1}$$

where F is the ^{254}Fm spontaneous fission source, 1 is the effective first member of the chain and includes all precursors of 2, and 2 is the member of the chain whose γ radioactivity is measured. For the chains considered here, half-lives of precursors of 1 are very short so that atoms of 1 are formed at the time of fission. Under these conditions

$$N_1^T = f\lambda_{F1}N_F^\circ(e^{-\lambda_F T} - e^{-\lambda_1 T})/(\lambda_1 - \lambda_F) \tag{2}$$

and

$$N_2^T = f\lambda_{F1}\lambda_1 N_F^\circ\{e^{-\lambda_F T}/[(\lambda_1 - \lambda_F)(\lambda_2 - \lambda_F)] + e^{-\lambda_1 T}/[(\lambda_F - \lambda_1)(\lambda_2 - \lambda_1)] + e^{-\lambda_2 T}/[\lambda_F - \lambda_2)(\lambda_1 - \lambda_2)]\}$$
$$+ f\lambda_{F2}N_F^\circ(e^{-\lambda_F T} - e^{-\lambda_2 T})/(\lambda_2 - \lambda_F), \tag{3}$$

TABLE I. γ rays analyzed.

Nuclide	Energy (keV)	Abundance
2.8-h ^{88}Kr	196.1	0.35 ± 0.04 [a,b]
65-day ^{95}Zr–35-day ^{95}Nb	765.8 (^{95}Nb)	0.99 ± 0.03 [a]
17-h ^{97}Zr–72-min ^{97}Nb	743.4; 658.1	0.94 ± 0.05; 0.99 ± 0.03 [a,c]
67-h ^{99}Mo–6-h ^{99}Tc^m	140.3 (^{99}Tc^m)	0.90 ± 0.05 [a,d]
39.6-day ^{103}Ru	497.1	0.90 ± 0.05 [a,e]
4.44-h ^{105}Ru	724.2	0.48 ± 0.05 [f,g]
367-day ^{106}Ru	621.8	0.098 ± 0.006 [a,f]
3.2-h ^{112}Ag	617.4	0.41 ± 0.02 [a,h]
8.05-day ^{131}I	364.5	0.82 ± 0.05 [a,f]
78-h ^{132}Te–2.3-h ^{132}I	228.2 (^{132}Te)	0.88 ± 0.09 [f]
21-h ^{133}I	529.9	0.90 ± 0.05 [f,i]
6.7-h ^{135}I–9.2-h ^{135}Xe	1260.5 (^{135}I)	0.35 ± 0.02 [a,f]
9.5-min ^{139}Cs–82.9-min ^{139}Ba	165.8 (^{139}Ba)	0.22 ± 0.02 [f]
12.8-day ^{140}Ba–40.2-h ^{140}La	487.1 (^{140}La)	0.465 ± 0.014 [f,j]
33-day ^{141}Ce	145.4	0.48 ± 0.03 [a,f,k]
11-min ^{142}Ba–92.4-min ^{142}La	641.2 (^{142}La)	0.48 ± 0.04 [f,l]
33-h ^{143}Ce	293.2	0.46 ± 0.04 [f,k,m]
284-day ^{144}Ce	133.5	0.11 ± 0.01 [a,f,k,n,o]
11.1-day ^{147}Nd	91.0	0.28 ± 0.02 [a,f,k]
1.8-h ^{149}Nd	211.3	0.27 ± 0.03 [a,k]

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where N_F° is the number of atoms of F at the beginning of exposure; N_1^T, N_2^T are the atoms of 1 and 2 at the end of exposure; T is the duration of exposure; λ_F, λ_1, and λ_2 are decay constants of nuclides F, 1, and 2; λ_{F1} and λ_{F2} are partial decay constants for fission of F leading to nuclide 1 and 2; f is the fraction of fission products collected in the catcher foil.

The number of counts of 2, A_2, observed (corrected for dead time) during some counting period after the removal of the catcher foil from the source is

$$A_2/\epsilon a = N_1^T[\lambda_2(e^{-\lambda_1\tau-\lambda_1 t}-e^{-\lambda_1\tau})-\lambda_1(e^{-\lambda_2\tau-\lambda_2 t}-e^{-\lambda_2\tau})]/(\lambda_1-\lambda_2)-N_2^T(e^{-\lambda_2\tau-\lambda_2 t}-e^{-\lambda_2\tau}), \quad (4)$$

where ϵ is the counting efficiency, a is the fraction of decays leading to the observed γ ray, τ is the time interval between the end of exposure and the beginning of the count, and t is the duration of the count. Expressions for N_1^T and N_2^T from Eqs. (2) and (3) can be substituted into Eq. (4) so that A_2 is a function of $(f\lambda_{F1}N_F^\circ)$ and $(f\lambda_{F2}N_F^\circ)$.

For some β-decay mass chains ($A=132$, 134, 135, and 140), counts were taken while there were still significant amounts of both 1 and 2 present. The ratio N_2^T/N_1^T for a given chain was calculated from Eq. (4) using results from several γ counts. This ratio was then used with Eqs. (2) and (3) to calculate the value of $\lambda_{F2}/\lambda_{F1}$ as in Ref. 11. Because λ_{F1} is proportional to the rate of formation of all precursors of and including 1, and λ_{F2} is proportional to the independent rate of formation of 2, the fractional independent yield of 2 is $\lambda_{F2}/(\lambda_{F1}+\lambda_{F2})$. For $A=132$, 134, and 135, a correction has also been made based on estimates of the independent yields for the third member of the chain (see Results and Discussion).

For other chains, values of τ were such that decay of 1 was essentially complete or 2 was in transient equilibrium with 1. For those cases, the ratio $\lambda_{F1}/\lambda_{F2}$ was estimated (see Results and Discussion), and the value $f(\lambda_{F1}+\lambda_{F2})N_F^\circ$ was calculated for each chain. Because f and N_F° were not measured, results are expressed as the total number of atoms collected during the exposure.

A second catcher foil was exposed for the next 4.25 h and dissolved in 6 M HNO_3. Barium, with standard carrier present, was purified, isolated as $BaCrO_4$,[12] and β counted with a stilbene counter to determine ^{140}Ba. Antimony, with standard carrier present, was purified,[13] isolated as Sb_2S_3, and β counted with a low-background scintillation counter to determine ^{127}Sb. An identical procedure was followed for a ^{235}U(n_{th}, f) standard reference system for which cumulative yields of ^{140}Ba and ^{127}Sb are known.[14] The ratio of ^{140}Ba to ^{127}Sb mass yields for ^{254}Fm(SF) was calculated from

$$\frac{^{140}Y_{Fm}}{^{127}Y_{Fm}} = \frac{^{140}A_{Fm}\,^{127}A_U\,^{140}Y_U}{^{127}A_{Fm}\,^{140}A_U\,^{127}Y_U}, \quad (5)$$

where Y represents cumulative yield, and A represents specific activity in counts per minute per milligram of sample.

TABLE II. Fractional independent yields of ^{132}I, ^{134}I, ^{135}Xe, and ^{140}La relative to their cumulative yields.

Nuclide	N_2^T/N_1^T	$\lambda_{F2}/\lambda_{F1}$	Independent yield $\lambda_{F2}/(\lambda_{F1}+\lambda_{F2})$
^{132}I	0.14 ± 0.01	0.25 ± 0.02	0.20 ± 0.02
^{134}I	3.14 ± 0.25	1.38 ± 0.20	$0.58^{+0.03}_{-0.04}$
^{135}Xe	0.68 ± 0.01	0.39 ± 0.01	0.28 ± 0.01
^{140}La	0.028 ± 0.009	0.023 ± 0.009	0.023 ± 0.009

RESULTS AND DISCUSSION

The nuclear charge distribution results are presented in Table II. Errors shown for values of N_2^T/N_1^T are standard deviations for measurements for ^{132}I, ^{135}Xe, and ^{140}La. The calculation for ^{134}I is sensitive to the values of the half-lives of ^{134}I and ^{134}Te because they differ by only 10 min.[11] The additional uncertainty introduced in the value of N_2^T/N_1^T by an uncertainty of ±1 min in each of the half-lives is ±0.15. The total uncertainty is taken to be the square root of the sum of the squares of these uncertainties and the standard deviation, ±0.12, of the measurement. The uncertainties in the values of $\lambda_{F2}/\lambda_{F1}$ and $\lambda_{F2}/(\lambda_{F1}+\lambda_{F2})$ are calculated from those shown for N_2^T/N_1^T. For all of these measurements, identification of the nuclides is based entirely on the energy of the λ ray measured. Neither half-lives nor chemical separations could be used to identify the γ peaks.

The large values of the ratio $\lambda_{F2}/(\lambda_{F1}+\lambda_{F2})$ for ^{132}I, ^{134}I, and ^{135}Xe suggest that there may be significant independent yields of the next members of the chains, ^{132}Xe, ^{134}Xe, and ^{135}Cs. Assuming the Gaussian width parameter σ for fission of ^{254}Fm is 0.56 charge unit, as given by Wahl *et al.*[14] for ^{235}U fission, the fractional independent yields of ^{132}Xe, ^{134}Xe, and ^{135}Cs estimated from the results of this work are ~0.005, ~0.06, and ~0.01, respectively. If the odd-even effect proposed by those workers applies to these chains, the yields of ^{132}Xe and ^{134}Xe may be higher and the yield of ^{135}Cs lower. The existence of independent yields of these nuclides would also lower the calculated

fractional cumulative yields of ^{132}Te, ^{134}Te, and ^{135}I. Using the results of this work given in the last column of Table II, and the estimated independent yields above, the fractional cumulative yields of ^{132}Te, ^{134}Te, ^{135}I, and ^{140}Ba are found to be 0.80 ± 0.02, 0.40 ± 0.04, 0.71 ± 0.01, and 0.977 ± 0.009, respectively.

Values of Z_p, the most probable charge for a fission product for a given mass chain, were calculated using the method of Wahl *et al.*[14] and the fractional cumulative yields given above. The value of σ was taken as 0.56 charge unit. The values of Z_p for $A = 132$, 134, 135, and 140 are 1.0, 0.85, 0.70, and 1.0, respectively, greater than the values for the same chains calculated from yields of the same nuclides from ^{235}U fission.[14] (The fractional cumulative yield of ^{140}Xe from ^{235}U was used as a reference rather than that of ^{140}Ba because the latter appears to be inconsistent with yields of other isotopes of barium.) The average difference is 0.9 charge unit. About 0.5 unit of this difference is due to the difference in charge-to-mass ratios, Z_f/A_f, of the fissioning nuclei. The rest may be due to a greater number of neutrons emitted per fragment for ^{254}Fm fission, or Z_p may fall closer to unchanged charge distribution (UCD) for ^{254}Fm-(SF). The value of 0.9 was used with reported fractional yields from ^{235}U fission to predict those fractional yields which were necessary for chain yield calculations for $A = 139$ and 142. The small correction for $A = 97$ was made by assuming that Z_p for complementary mass $A = 153$ was 60.8, a value 0.9 charge unit higher than the one given by

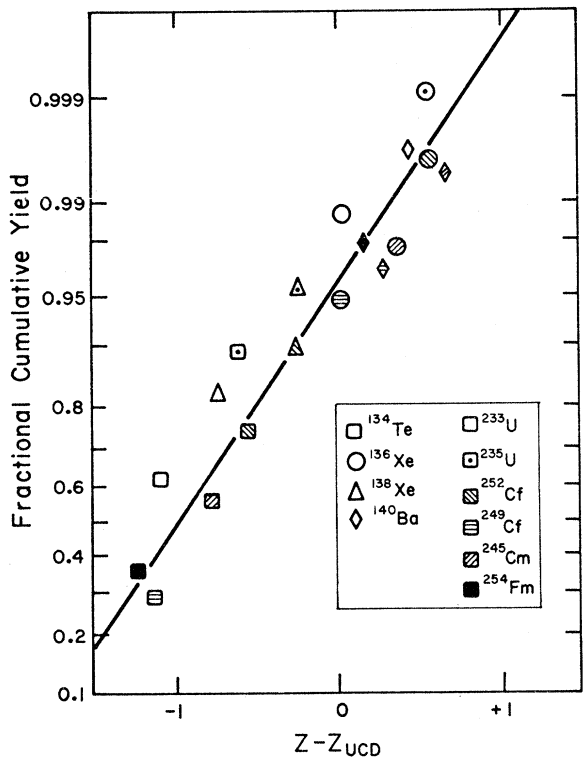

FIG. 1. Probability plot of fractional cumulative yields of ^{134}Te, ^{136}Xe, ^{138}Xe, and ^{140}Ba as a function of $(Z-Z_{UCD})$.

TABLE III. Summary of mass yields requiring independent-yield correction.

Nuclide	Fractional independent yield	Relative yield, 10^7 atoms [a] Uncorrected for independent yield	Corrected for independent yield	Normalized yield [b]
^{97}Nb	0.01 [c]	1.99 ± 0.07	2.01 ± 0.07	1.19 ± 0.06
^{112}Ag	0.10 [c]	9.75 ± 0.21	10.82 ± 0.23	6.42 ± 0.36
^{132}Te	0.79 [d,e]	3.95 ± 0.07	4.94 ± 0.09	2.93 ± 0.32
^{135}Xe	0.28 [d]	5.20 ± 0.09	7.25 ± 0.08	4.30 ± 0.26
^{139}Ba	0.25 [c]	6.47 ± 0.06	8.64 ± 0.10	5.13 ± 0.49
^{140}La	0.023 [d]	8.24 ± 0.65	8.43 ± 0.65	5.00 ± 0.42
^{142}La	0.30 [c]	6.07 ± 0.06	8.67 ± 0.10	5.14 ± 0.44

[a] Total number of atoms collected during exposure.
[b] Uncertainty represents the square root of the sum of the squares of γ-ray abundance, efficiency, and counting uncertainties.
[c] Estimated value.
[d] Measured value.
[e] Fractional cumulative yield.

TABLE IV. Summary of mass yields requiring no independent-yield corrections.

Nuclide	Number of determinations	Relative yield (10^7 atoms) [a]	Normalized yield (%) [b]
^{88}Kr	3	0.12 ± 0.01	0.08 ± 0.08
^{95}Nb	2	0.87 ± 0.03	0.52 ± 0.02
^{99}Tc^m	7	2.60 ± 0.05	1.54 ± 0.11
^{103}Ru	5	5.41 ± 0.20	3.21 ± 0.22
^{105}Ru	7	8.27 ± 0.17	4.91 ± 0.53
^{106}Ru	1	7.89	4.68 ± 0.56
^{127}Sb	1	0.45	0.27 ± 0.06 [c]
^{131}I [d]	3	3.92 ± 0.13	2.33 ± 0.17
^{133}I [e]	3	8.00 ± 0.26	4.74 ± 0.32
^{141}Ce	7	8.20 ± 0.20	4.87 ± 0.38
^{143}Ce	11	7.75 ± 0.16	4.60 ± 0.46
^{144}Ce	1	7.21	4.28 ± 0.60
^{147}Nd	2	6.00 ± 0.33	3.56 ± 0.35
^{149}Nd	5	5.03 ± 0.43	2.98 ± 0.43

[a] Total number of atoms collected during exposure.
[b] Uncertainty represents the square root of the sum of the squares of γ-ray abundance, efficiency, and counting uncertainties.
[c] Uncertainty estimated on basis of radiochemical analysis of ^{127}Sb (see text).
[d] Fraction of chain passing through ^{131}Te^m estimated to be 0.35.
[e] Fraction of chain passing through ^{133}Te^m estimated to be 0.35.

Wahl *et al.*[14] for $^{235}U(n_{th}, f)$. The corresponding Z_p for $A = 97$ is 39.2. The correction for $A = 112$ was estimated from the previously measured fractional yield of ^{112}Ag from $^{252}Cf(SF)$.[9]

Figure 1 is a probability plot of fractional cumulative yields as a function of $(Z - Z_{UCD})$. Selected data[11, 15, 16] were available for ^{134}Te, ^{136}Xe, ^{138}Xe, and ^{140}Ba from thermal-neutron-induced fission of ^{233}U, ^{235}U, ^{245}Cm, and ^{249}Cf, and from spontaneous fission of ^{252}Cf and ^{254}Fm. Values of A', the average mass number of the fragments which decay by prompt-neutron emission to the product of mass number A, were estimated for published values of A' as a function of A[17, 18] for ^{235}U and ^{252}Cf and $\bar{\nu}_p$, the average number of prompt neutrons emitted per fragment.[19-21] The increase in $\bar{\nu}_p$ compared to $^{252}Cf(SF)$ was equally divided between light and heavy fragments. The line shown was proposed by Wahl *et al.*,[14] for which $Z_p = A'(Z_f/A_f) - 0.45$ for heavy peak products and for which the width parameter, σ, of the curve is 0.56 charge unit. The points for ^{254}Fm and ^{252}Cf fission appear to be in closer agreement with those from ^{245}Cm and ^{249}Cf fission than with those from ^{233}U and ^{235}U fission.

The mass distribution results are listed in Tables III and IV. Uncertainties in the relative yields are standard deviations of multiple determinations and reflect the precision of the γ-ray spectroscopy. If uncertainties in γ-ray detection efficiencies and γ-ray abundances are included, the errors range from ±5 to ±15%. The ratio $^{140}Y_{Fm}/^{127}Y_{Fm}$ [see Eq. (5)] was found to be 18. Reflected yields were calculated based on $\bar{\nu}_p$ for $^{254}Fm(SF)$ of 3.94.[20]

The relative numbers of atoms in Tables III and IV were converted to absolute yields by requiring the sum of all yields to be 200%. Unmeasured yields were determined by interpolation on a smooth curve through the measured points. The radiochemical results of Flynn *et al.*[3] for $^{256}Fm(SF)$ are plotted for comparison with this work in Fig. 2. The shapes of the heavy-mass peaks are nearly identical. The light-mass peak for ^{256}Fm is shifted towards heavier masses to conserve mass.

Values of ν_p for any given chain may differ from $\bar{\nu}_p$. Bowman *et al.*[18] have shown that ν_p near the wings of the mass distribution curve may be as much as one neutron higher than the average. If this is also true for ^{254}Fm, the heavy-mass points reflected from yields for $A = 88$, 95, 97, and 99 would be shifted to the left accordingly. This would have little effect on the shape of the curve or the normalization to 200% because these yields are small compared to those in the peaks. The upper limit for the peak-to-valley ratio for $^{254}Fm(SF)$ (~18) is greater than that for $^{256}Fm(SF)$ (~12) but is low compared to that for $^{252}Cf(SF)$ (~750).[22] These results support the evidence for lower peak-to-valley ratios as the mass of the fissionable fermium isotopes increases. However, for $^{254}Fm(SF)$ and $^{256}Fm(SF)$, the fission-product mass distribution is definitely asymmetric, and the lowering of the peak-to-valley ratio appears to be the result of the light-mass peak of about constant width shifting towards heavier masses rather than the onset of a symmetric mode of fission.

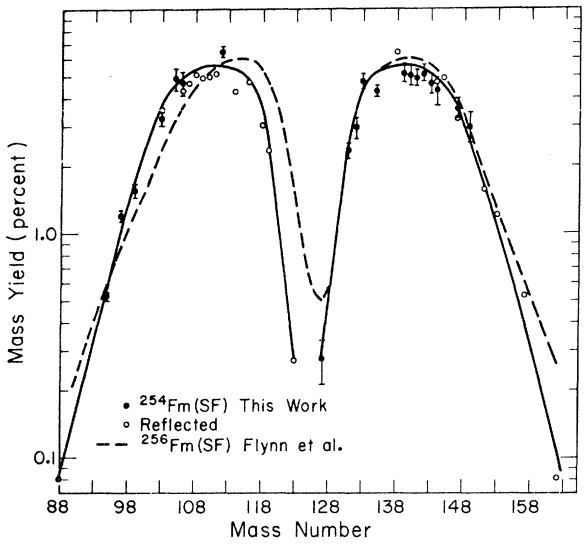

FIG. 2. $^{254}Fm(SF)$ and $^{256}Fm(SF)$ radiochemical mass-yield curves.

ACKNOWLEDGMENT

The authors wish to thank A. L. Boni for his assistance in counting the ^{144}Ce and ^{106}Ru γ activities.

†The information contained in this article was developed during the course of work under contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

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