Charge distribution in the reactor-neutron-induced fission of ^{232}Th

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The independent yields of ^{82}Br, ^{86}Rb, ^{96}Nb, ^{98}Nb^{m}, ^{128}Sb^{g}, and ^{136}Cs were determined in the reactor-neutron-induced fission of ^{232}Th using radiochemical techniques. Results: $(2.3\pm2.3)\times10^{-4}\%$ for ^{82}Br, $<3.8\times10^{-4}\%$ for ^{86}Rb, $<4.2\times10^{-5}\%$ for ^{96}Nb, $(2.48\pm0.53)\times10^{-3}\%$ for ^{98}Nb^{m}, $(2.34\pm0.37)\times10^{-3}\%$ for ^{128}Sb^{g}, and $(1.70\pm0.13)\times10^{-4}\%$ for ^{136}Cs. Using the extended Zp model of Wahl with the yield data from this work and the literature the following parameters were obtained for the charge distribution in ^{232}Th fission: width of Gaussian dispersion $\bar{\sigma}_Z=0.52\pm0.01$, $\Delta Z_P\ (=Z_P-Z_{\rm UCD})=0.45\pm0.02$. The even-odd proton and neutron enhancement factors were found to be small. These parameters and systematics of even-odd proton and neutron effects in low energy fission are discussed.

[NUCLEAR REACTIONS, FISSION Radiochemical fission yields ^{232}Th(n,f), calculated charge dispersion parameters, and odd-even effects.]

I. INTRODUCTION

Only few experimental data are available about charge distribution in the reactor-neutron-induced fission of ^{232}Th.[1,2] Such information, as well as the magnitude of the even-odd proton and neutron effects, may contribute to the understanding of the low energy fission process when compared with similar data from heavier fissionable nuclides. In this study we have attempted to measure independent yields of some shielded nuclides as well as that of 9.1 h ^{128}Sb^{g} in the reactor-neutron-induced fission of ^{232}Th.

II. EXPERIMENTAL

A. Materials and irradiations

Fission sources each containing $\sim$60 mg of thorium nitrate (purissimum quality, FLUKA, Buchs, Switzerland) were irradiated in the swimming pool reactor SAPHIR at our institute. Irradiation times varied from one hour to ten hours depending on the half-life of the nuclide of interest. The fast flux was $\sim3.3\times10^{12}$ cm^{-2}s^{-1}. The sources were wrapped in aluminum foils ($\sim$4 mg cm^{-2}) and were placed in quartz ampules. The ampules were further placed in cadmium containers with 0.5 mm thick walls to reduce the formation of ^{233}Th.

B. Chemical separations

The irradiated samples and the aluminum wrappings were dissolved in 6*M* HNO_3 (+Hg) or 6*M* HCl containing carriers of the elements of interest. In order to eliminate ^{233}Pa, which was formed in a considerable amount, the samples were filtered twice through a stack of ten glass-fiber filters (no. 6, Schleicher and Schüll). In niobium determinations ^{233}Pa was separated by the solvent extraction procedure of Moore[3] using 6*M* H_2SO_4-6*M* HF and disobutylcarbinol. Modified radiochemical procedures[4] were used to isolate the element of interest as well as molybdenum. Sn-Sb separations were made about 2.5 h after the end of one hour irradiations. The sources were further purified, precipitated, and mounted for counting. The chemical yields, except for niobium, were determined by gravimetry.

C. Counting and treatment of data

The samples were counted using two calibrated Ge(Li) detectors with volumes of 45 and 54 cm^3. The source-to-detector distance was $\sim$2 cm. Coincidence summing losses were experimentally determined by counting the sources at various distances from the detector. The highest coincidences summing losses were about 7%. The samples were counted several times, and more than one gamma line was used in yield determinations, wherever possible. The half-lives and the γ-ray data of the fission products used in the yield determinations are given in Table I.

The gamma-ray spectra were analyzed by a modified GASPAN peak analysis program.[12] After correcting the measured intensities of the γ lines for detector efficiencies, the decay curves were analyzed. A modified CLSQ program[13] was used that also corrected for chemical yield. The activities at chemical separation time were corrected for absolute γ-ray emission intensities and genetic relationships. Finally they were extrapolated to the end of irradiation and corrected to saturation activities. The fission yields were determined relative to ^{99}Mo except those of ^{96}Nb and ^{98}Nb^{m}, where ^{97}Nb was used as the standard nuclide. The yield of ^{99}Mo (2.98 ± 0.15) %, and ^{97}Nb (4.52 ± 0.19) % have been taken from our recent measurements.[14]

III. RESULTS

The independent yields measured in this work and those from the literature are given in Table II. Yields from this work represent mean values of several determinations, except for ^{86}Rb and ^{96}Nb, where only upper limits could be determined. The errors include statistical errors as well as systematic errors due to uncertainties in the yields of the standard nuclides, in the detector efficiencies, and, for ^{128}Sb^{g}, in the contributions due to the precursors. The large error in ^{82}Br is due to the uncertain correction for the ^{81}Br(n,γ) reaction. This correction amounted to about 50% of the observed value and was estimated from the formation of ^{80}Br^{m} by the ^{79}Br(n,γ) reaction. Contributions from precursors had to be considered only in the case of ^{128}Sb^{g}. In recent measurements[9,15] it was found that ^{128}Sn decayed to ^{128}Sb^{m} only. This isomer was found to decay in 3.6% of the cases by isomeric transition (IT) to ^{128}Sb^{g}.[15] The measured values of ^{128}Sb^{g} were corrected using these genetic relationships, a cumulative yield of (0.17 ± 0.02) % for ^{128}Sn, and an isomer ratio, $Y_m/Y_g=1.38$, to estimate the independent yield of ^{128}Sb^{m}. The isomer ratio was calculated using the equations of Madland and England[16] and spins of 5 (^{128}Sb^{m}) and 8 (^{128}Sb^{g}).[15] The contribution of independently formed ^{128}Sb^{m} to the corrections was small.

The total independent yields of ^{98}Nb and ^{128}Sb were obtained from the experimental values of ^{98}Nb^{m} and ^{128}Sb^{g} given in Table II and calculated isomer ratios.[16] Spins of 5 and 1 were used for ^{98}Nb^{m} and ^{98}Nb^{g}, respectively.[5] According to these calculations ^{98}Nb^{m} and ^{128}Sb^{g} represent 78% and 42% of the total independent yields, respectively.

IV. DISCUSSION

Recently Wahl[18] extended the conventional Z_P model.[19] Using the method of least squares and the

TABLE I. Nuclear data of the fission products used in this work.

Nuclide	Half-life	E_γ (keV)	I_γ (%)	Reference
^{82}Br	35.3 h	554.3	70.7 $\pm$0.8	5
		619.1	43.0 $\pm$0.6	
		776.5	83.4 $\pm$0.9	
^{86}Rb	18.8 d	1076.6	8.79$\pm$0.9	5
^{96}Nb	23.5 h	778.2	96.8 $\pm$0.3	6
^{97}Nb	72.1 min	657.9	98.4 $\pm$0.1	7
^{98}Nb^{m}	51.3 min	787.4	93.2 $\pm$0.2	5
^{99}Mo	66.0 h	140.5	90.7 $\pm$0.044[a]	8
		181.1	6.08$\pm$0.16	
		739.4	12.14$\pm$0.22	
^{128}Sn	59.1 min			9
^{128}Sb^{m}	10.4 min			9
^{128}Sb^{g}	9.1 h	743.3	100	10
		754.0	100	
^{136}Cs	13.0 d	340.6	46.8 $\pm$0.5	11
		818.5	99.8 $\pm$0.3	
		1048.1	79.8 $\pm$0.8	

[a]In equilibrium with ^{99}Tc^{m}.

TABLE II. Yield data from this work and from the literature used in the determination of the charge distribution parameters in the reactor-neutron-induced fission of ^{232}Th.

Nuclide	Yield (%)[a]	Chain yield (%) Ref. 14[b]	Fractional yield	Reference
^{82}Br	$(2.3 \pm 2.3) \times 10^{-4}$	1.25 ± 0.13	$(1.8 \pm 1.8) \times 10^{-4}$	This work
^{86}Rb	$< 3.8 \times 10^{-4}$	6.60 ± 0.66	$< 5.7 \times 10^{-5}$	This work
^{91}Kr	5.69 ± 0.51	7.59 ± 0.42	0.750 ± 0.079	2
^{96}Nb	$< 4.2 \times 10^{-5}$	5.60 ± 0.56	$< 7.5 \times 10^{-6}$	This work
^{98m}Nb[c]	$(2.48 \pm 0.53) \times 10^{-3}$	3.85 ± 0.39	$(6.4 \pm 1.5) \times 10^{-4}$	This work
^{128g}Sb[d]	$(2.34 \pm 0.37) \times 10^{-3}$	0.17 ± 0.02	$(1.38 \pm 0.27) \times 10^{-2}$	This work
^{131}Sn	1.34 ± 0.38	1.80 ± 0.10	0.74 ± 0.22	2
^{131}Te	$(2.8 \pm 0.8) \times 10^{-2}$	1.80 ± 0.10	$(1.6 \pm 0.5) \times 10^{-2}$	17
^{132}Sn	1.52 ± 0.23	2.96 ± 0.12	0.514 ± 0.080	2
^{135}I	0.76 ± 0.15	6.14 ± 0.36	0.124 ± 0.025	1
^{136}Cs	$(1.7 \pm 0.13) \times 10^{-4}$	5.70 ± 0.57	$(2.98 \pm 0.38) \times 10^{-5}$	This work
^{140}Xe	6.63 ± 0.37	8.12 ± 0.45	0.817 ± 0.064	2

[a]Cumulative yields for ^{91}Kr, ^{131}Sn, and ^{140}Xe; independent yields for the other nuclei.
[b]Unmeasured chain yields were interpolated from the smooth yield-mass curve.
[c]Total independent yield; $(3.20 \pm 0.7) \times 10^{-3}$ %, see text.
[d]Total independent yield; $(5.6 \pm 1.4) \times 10^{-3}$ %, see text.

available yield data for light and heavy fission products, he obtained values of parameters that describe nuclear charge distribution for a number of fissioning systems.

The parameters that could be determined are the following: $\bar{\sigma}_Z$, the Gaussian dispersion width; $\overline{\text{EOZ}}$ and $\overline{\text{EON}}$, the even-odd proton and neutron yield enhancement factors; $\Delta Z(A'=140)$, the charge displacement at fragment mass 140; and its slope $\partial \Delta Z / \partial A'_H$. The last two parameters are related to $\Delta Z_P = |Z_P - Z_{\text{UCD}}|$, the deviation of the most probable charge, Z_P, from that using the unchanged charge distribution description, Z_{UCD}, by the relation

$$\Delta Z_P(A'_H) = \Delta Z(A'_H = 140) + \frac{\partial \Delta Z}{\partial A'_H}(A'_H - 140) ,$$

where A'_H is the heavy fragment mass.

The above mentioned parameters were determined in the reactor-neutron-induced fission of ^{232}Th, applying the general least-squares program ORGLSW of Busing and Levy[20] as modified by Wahl.[18] The number of neutrons emitted by heavy and light fission fragments ν_H and ν_L were calcu-

TABLE III. Nuclear charge distribution parameters derived for the reactor-neutron-induced fission of ^{232}Th.

$\bar{\sigma}_Z$	$\overline{\text{EOZ}}$	$\overline{\text{EON}}$	$\Delta Z(A'=140)$	$\frac{\partial \Delta Z}{\partial A'_H}$	Fit[a]
0.52 ± 0.01[b]	1.27 ± 0.10[b]	1.13 ± 0.09[b]	0.39 ± 0.02[b]	-0.012 ± 0.014[b]	1.23[b]
0.52 ± 0.01	1.07 ± 0.10	0.96 ± 0.09	0.46 ± 0.02	0.0018 ± 0.013	1.12
0.52 ± 0.01	1.08 ± 0.08	0.98 ± 0.07	0.45 ± 0.02	0[c]	0.98

[a]Fit $= (\chi^2/N)^{1/2}$, where N number of observations minus number of variable parameters.
[b]Including the yield of ^{132}Sn.
[c]Not varied.

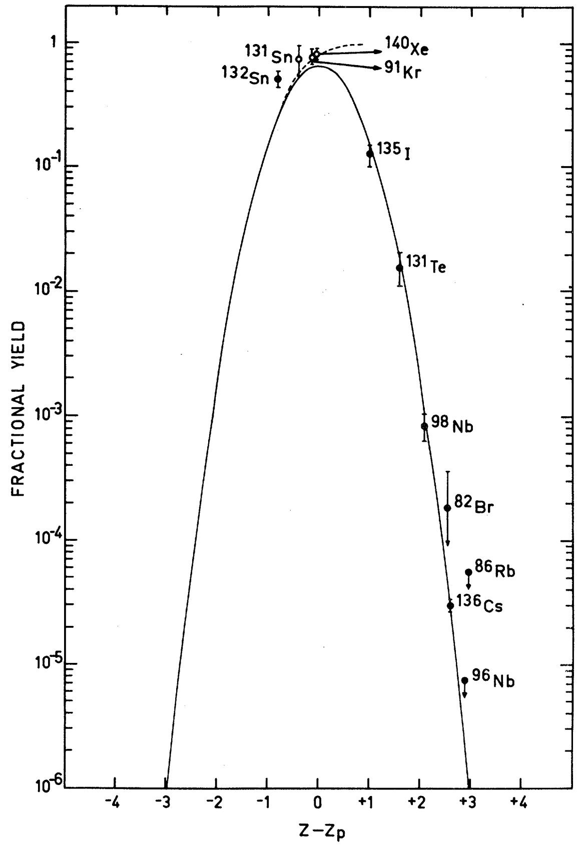

FIG. 1. Charge distribution curve in the reactor-neutron-induced fission of ^{232}Th ($A'_H \geq 130$). The Gaussian dispersion width parameter $\bar{\sigma}_Z$ was taken as 0.52 ± 0.1 and ΔZ_P as 0.45 ± 0.02. The broken line shows fractional cumulative yields. The solid points represent experimental fractional independent yields and the open points experimental fractional cumulative yields.

lated using the R_H ($=\nu_H/\nu_T$) function of Wahl,[18] and a value of $\nu_T=2.35$.[21] The value of R_H is not critical for the determination of the charge distribution parameters.[18] The results of the calculations are given in Table III. The experimental data given in Table II with the exception of the limits and ^{128}Sb were used. ^{128}Sb was omitted since Wahl[18] excludes products with $A'_H \leq 130$ because not enough is known about variations of the parameters for charge distribution in the symmetric region. Only in the first example of Table III the yield of the double-magic nulceus ^{132}Sn has been included for the calculation. This gave rise to a considerable increase in even-odd proton and neutron enhancement factors. As this may arise from 50 proton and/or 82 neutron shell effects, the results without the ^{132}Sn yield are considered to better represent the average charge distribution in ^{232}Th fission. The large even-odd proton effect of 1.30 ± 0.12 reported by Izak-Biran and Amiel[2] is only confirmed if the yield of ^{132}Sn is included in the calculations. The values for $\bar{\sigma}_Z$, $\overline{\text{EOZ}}$, $\overline{\text{EON}}$, and ΔZ in rows two and three of Table III differ only very little. The resulting charge distribution curve is shown in Fig. 1.

Even-odd proton and neutron enhancement factors $\overline{\text{EOZ}}$ and $\overline{\text{EON}}$ for various low energy fission processes are given in Table IV. They were calculated according to the extended Z_P model of Wahl.[18] It is observed that the even-odd proton effect is well established and pronounced in thermal-neutron-induced fission of ^{233}U and ^{235}U only. It is difficult to understand this with presently available fission theories and models. The even-odd neutron effect EON appears to be quite small in all cases.

TABLE IV. Comparison of even-odd proton and neutron enhancement factors $\overline{\text{EOZ}}$ and $\overline{\text{EON}}$ for various low energy fission processes ($A'_H \geq 130$).

Fission process	No. of data	$\overline{\text{EOZ}}$	$\overline{\text{EON}}$	Reference
^{232}Th(n_f,f)	9	1.08 ± 0.08	0.98 ± 0.07	This work
^{233}U(n_{th},f)	53	1.32 ± 0.03	1.06 ± 0.03	18
^{235}U(n_{th},f)	192	1.25 ± 0.02	1.08 ± 0.02	18
^{238}U(n_f,f)	25	1.02 ± 0.04	1.07 ± 0.04	18
^{239}Pu(n_{th},f)	59	1.09 ± 0.03	1.04 ± 0.03	18
^{241}Pu(n_{th},f)	20	1.01 ± 0.03	1.01 ± 0.03	22[a]
^{245}Cm(n_{th},f)	11	1.12 ± 0.05	1.02 ± 0.08	23,24[a]
^{249}Cf(n_{th},f)	25	1.11 ± 0.07	0.89 ± 0.05	25
^{252}Cf(s.f.)	37	0.97 ± 0.04	1.00 ± 0.04	18

[a]Calculations made by us using experimental yields of given references.

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