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Odd-even systematics in neutron fission yields of ^{233}U and ^{235}U

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An analysis of the distribution of independent yields in neutron induced fission of ^{233}U and ^{235}U revealed a constant enhancement of products with an even number of protons, relative to those with an odd number. This odd-even effect in the proton pairing, related to calculated "normal" yields of elements in fission, constitutes a sawtooth structure with an amplitude of $(22 \pm 7)\%$ for both ^{233}U and ^{235}U thermal neutron induced fission. The residual neutron pairing effect evident after the emission of prompt neutrons, while it is insignificant (< 3%) in the light mass peaks, is partly preserved in the heavy mass peaks of both ^{235}U and ^{233}U thermal neutron fission products. Its average magnitude is about $(8 \pm 5)\%$ relative to the "normal" distribution of isotonic yields. In the fission of ^{235}U with fission spectrum neutrons, the proton pairing effect drops to $(8 \pm 4)\%$. The odd-even effect is discussed in view of the various mass splits, the excitation energy and potential energy surfaces in the descent from the saddle to the scission configuration.

NUCLEAR REACTIONS, FISSION $^{235}U(n_{th}, f)$, $^{233}U(n_{th}, f)$, and $^{235}U(n_{fast}, f)$. Reevaluation of independent fission yields, deduced odd-even yield systematics.

INTRODUCTION

Some characteristics of the low-energy fission of actinides, such as asymmetric mass distributions and relatively low fission yields at the symmetric split, have been satisfactorily described using Strutinsky's method of calculating deformed shell potential energy surfaces.[1] By means of this method, the most probable mass splits in fission and peak-to-valley ratios have been calculated for several fissioning nuclides.[2-8] Those calculations were further extended by Wilkins and Steinberg,[2] who, using semiempirical masses, calculated the mass distribution of fission fragments and charge dispersion in isobaric chains. However, since the calculations are based on approximate mass values, only the general form of mass distribution in fission was reconstructed, while the structure and the actual fragment yields, found experimentally,[9-13] could not be calculated to a satisfactory agreement.

The enhanced yield of elements with an even number of protons, relative to those with an odd proton number (which is a characteristic feature of thermal neutron fission of ^{235}U), could not be calculated by the above method, due to inadequate knowledge of the fission fragment masses. This odd-even effect has been established experimentally[14,15] by measuring the independent fission yields of mass-separated noble gases, alkalis, and other elements obtained by radiochemical methods.

In a previous publication[16] it was observed that a plot of independent elemental yields as a function of z actually results in two curves, the upper one representing the yield of elements with an even number of protons (Fig. 1). The average of those two curves fitted well the "normal" curve calculated according to Wahl's method of predicting independent fission yields, based on a single Gaussian which represents the isobaric distributions in fission in the various mass chains.[17] The enhanced yields of elements with an even number of protons are not influenced by neutron emission, and therefore the observed proton pairing effect represents the primary charge division in the fissioning nucleus.

On the other hand, the neutron pairing effect which might be expected to be similar to the proton pairing effect was found to be not as pronounced, probably due to prompt neutron evaporation. However, this expected similarity is partly preserved in the heavy mass peak and was observed in the heavier isotopes of mass-separated cesium and rubidium.[18]

The odd-even effect described for thermal neutron fission of ^{235}U [16] should also be found in other fissioning nuclides, with similar excitation energy. In this work a comparison is made between the thermal neutron fission of ^{235}U and ^{233}U, which have approximately the same excitation energies but different complementary pairs of fission fragments. We may deduce from this comparison whether the effect observed in ^{235}U is actually due

TABLE I. Thermal neutron fission yields of ^{235}U.

Mass	Chain yield [a] (%)	Element	Corrected FIY [b]	Normal [c] FIY [d]	Δ [e] (%)
			a. Light mass peak		
84	0.986 ± 0.023	Ge	0.03	0.0279	
		As	0.37 ± 0.06 [f]	0.4222	−[12.4 ± 14.2]
		Se	0.57 ± 0.06 [f]	0.5014	+[14.0 ± 12.0]
		Br	0.03 ± 0.02 [f]	0.0481	−[37.5 ± 41.6]
85	1.32 ± 0.03	As	0.13 ± 0.05	0.1916	−[33.2 ± 26.1]
		Se	0.69 ± 0.07	0.6278	+[9.9 ± 11.0]
		Br	0.18 ± 0.07	0.1732	+[3.9 ± 10.4]
86	1.95 ± 0.05	As	0.04 ± 0.01 [g]	0.0537	−[25.5 ± 18.6]
		Se	0.62 ± 0.1 [g]	0.5169	+[19.9 ± 19.3]
		Br	0.31 ± 0.10 [g]	0.4044	−[23.3 ± 24.7]
		Kr	0.03 ± 0.01	0.0246	+[21.9 ± 40.6]
87	2.55 ± 0.07	As	0.01	0.0092	···
		Se	0.39 ± 0.07 [f]	0.2746	+[42.0 ± 25.5]
		Br	0.44 ± 0.07	0.6038	−[27.1 ± 11.6]
		Kr	0.15 ± 0.01	0.1110	+[36.9 ± 11.7]
88	3.62 ± 0.07	Se	0.11 ± 0.01	0.0893	+[22.4 ± 19.5]
		Br	0.51 ± 0.03	0.5821	−[12.4 ± 5.1]
		Kr	0.37 ± 0.03	0.3148	+[18.1 ± 8.9]
		Rb	0.01	0.0128	···
89	4.80 ± 0.10	Se	0.023 ± 0.010	0.0191	+[20.4 ± 52.4]
		Br	0.26 ± 0.04	0.3684	−[29.4 ± 10.8]
		Kr	0.67 ± 0.04	0.5456	+[22.8 ± 7.3]
		Rb	0.047 ± 0.016	0.0663	−[29.1 ± 24.1]
90	5.89 ± 0.11	Br	0.11 ± 0.025	0.1560	−[30.2 ± 16.4]
		Kr	0.76 ± 0.03	0.6253	+[21.7 ± 5.1]
		Rb	0.13 ± 0.02	0.2110	−[38.4 ± 9.5]
91	5.93 ± 0.11	Br	0.04 ± 0.016	0.0430	−[7.0 ± 37.2]
		Kr	0.55 ± 0.015	0.4852	+[13.3 ± 3.9]
		Rb	0.38 ± 0.03	0.4399	−[13.6 ± 7.3]
		Sr	0.03 ± 0.03	0.0315	
92	5.97 ± 0.07	Kr	0.31 ± 0.01	0.2526	+[22.7 ± 4.0]
		Rb	0.55 ± 0.03	0.6133	−[10.3 ± 4.9]
		Sr	0.14 ± 0.03	0.1248	+[12.2 ± 24.8]
93	6.40 ± 0.07	Kr	0.08 ± 0.01	0.0838	
		Rb	0.49 ± 0.03	0.5748	−[14.7 ± 5.2]
		Sr	0.41 ± 0.03	0.3266	+[25.5 ± 9.2]
		Y	0.016 ± 0.002	0.0140	+[14.3 ± 4.3]
94	6.44 ± 0.07	Kr	0.027 ± 0.005	0.0183	
		Rb	0.25 ± 0.015	0.3624	−[32.1 ± 4.1]
		Sr	0.66 ± 0.025	0.5501	+[21.2 ± 4.5]
		Y	0.06 ± 0.02	0.0686	−[12.5 ± 29.1]
95	6.50 ± 0.09	Rb	0.10 ± 0.005	0.1478	−[33.7 ± 3.4]
		Sr	0.77 ± 0.06	0.6232	+[23.9 ± 9.6]
		Y	0.13 ± 0.06	0.2213	−[42.3 ± 27.1]
96	6.28 ± 0.07	Rb	0.021 ± 0.001	0.0414	−[49.3 ± 2.4]
97	6.03 ± 0.10	Rb	0.006 ± 0.001	0.0072	−[22.2 ± 9.7]

TABLE I *(Continued)*

Mass	Chain yield [a] (%)	Element	Corrected FIY [b]	Normal [c] FIY [d]	Δ [e] (%)
		b. Heavy mass peak			
131	2.82 ±0.07	In	0.01 ±0.05	0.0101	
		Sn	0.39 ±0.08	0.2859	+[36.5±28.0]
		Sb	0.48 ±0.08	0.5983	−[20.4±13.4]
		Te	0.12 ±0.01	0.1044	+[18.8±13.4]
132	4.2 ±0.09	Sn	0.14 ±0.04	0.1013	+[34.2±39.5]
		Sb	0.49 ±0.01	0.5953	−[18.2± 1.7]
		Te	0.37 ±0.015	0.2917	+[35.7± 5.1]
133	6.75 ±0.16	Sn	0.02	0.0199	
		Sb	0.31 ±0.07	0.3744	−[17.2±18.7]
		Te	0.64 ±0.07	0.5411	+[19.0±12.6]
		I	0.026±0.002	0.0640	−[59.4± 3.1]
134	7.65 ±0.17	Sb	0.04 ±0.004	0.1322	−[70.5± 3.0]
		Te	0.83 ±0.01	0.6172	+[35.1± 1.8]
		I	0.12 ±0.01	0.2418	−[51.6± 4.1]
		Xe	0.01	0.0068	
135	6.60 ±0.16	Sb	0.02 ±0.01	0.0291	
		Te	0.49 ±0.03	0.4281	+[14.5± 7.0]
		I	0.45 ±0.03	0.4960	−[9.3± 6.0]
		Xe	0.04 ±0.01	0.0463	
136	6.18 ±0.14	Te	0.25 ±0.05	0.1916	+[30.5±26.1]
		I	0.51 ±0.11	0.6278	−[18.8±17.5]
		Xe	0.24 ±0.04	0.1732	+[38.6±23.1]
137	6.26 ±0.16	Te	0.08 ±0.03	0.0556	+[43.8±53.9]
		I	0.43 ±0.06	0.5219	−[16.8±11.5]
		Xe	0.47 ±0.06	0.3984	+[17.0±14.1]
		Cs	0.02 ±0.005	0.0236	
138	6.80 ±0.17	Te	0.03 ±0.01	0.0106	
		I	0.18 ±0.08	0.2917	−[39.0±27.4]
		Xe	0.74 ±0.08	0.5953	+[26.1±13.4]
		Cs	0.047±0.002	0.1013	−[53.6± 2.0]
139	6.50 ±0.12	I	0.09 ±0.10	0.1077	−[13.6±25.6]
		Xe	0.68 ±0.10	0.6011	+[13.3±17.1]
		Cs	0.21 ±0.03	0.2803	−[26.5±10.7]
		Ba	0.011±0.004	0.0096	+[14.6±41.7]
140	6.36 ±0.06	I	0.02 ±0.01	0.0257	
		Xe	0.58 ±0.04	0.4104	+[41.3± 9.7]
		Cs	0.33 ±0.03	0.5118	−[35.5± 5.9]
		Ba	0.07 ±0.03	0.0518	+[35.1±58.0]
141	5.82 ±0.06	Xe	0.20 ±0.02	0.1869	+[9.7± 9.7]
		Cs	0.54 ±0.04	0.6280	−[13.1± 6.4]
		Ba	0.26 ±0.05	0.1777	+[46.3±28.1]
142	5.87 ±0.06	Xe	0.107±0.015	0.0556	+[93.2±27.0]
		Cs	0.41 ±0.05	0.5219	−[21.4± 9.6]
		Ba	0.46 ±0.05	0.3984	+[16.7±13.0]
		La	0.017±0.004	0.0236	−[28.1±16.9]
143	5.95 ±0.08	Xe	0.008±0.001	0.0111	
		Cs	0.25 ±0.03	0.2974	−[15.9±10.1]
		Ba	0.62 ±0.07	0.5922	+[4.7±11.8]
144	5.39 ±0.06	Cs	0.052±0.016	0.1110	−[53.1±14.4]
		Ba	0.73 ±0.06	0.6038	+[20.4± 9.9]
		La	0.20 ±0.05	0.2746	−[28.3±18.2]

TABLE I *(Continued)*

[a] Reference 29.
[b] Reference 16.
[c] Numerical values of the "normal" distribution were calculated by Wolfsberg (Ref. 32) using Wahl's constants (Ref. 17).
[d] Reference 32.
[e] [(Corrected FIY − normal FIY)/normal FIY]× 100.
[f] Reference 28.
[g] D. R. Nethaway and G. W. Barton, UCRL Report No. UCRL-51458, 1973 (unpublished).

to proton pairing or to the combination of fission fragments pairs. The effect of closed neutron shells on the distribution of fission product yields may be better evaluated too, by comparing the two fissioning nuclei.

The influence of the excitation energy of the fissioning nucleus on the proton pairing effect was checked, since we expected the effect to be energy dependent as in the case of fine structures in mass and kinetic energy distributions.[19-27]

The purpose of this work is to elucidate the odd-even systematics by a rigorous treatment of the available experimental data and to establish quantitatively the magnitude of the pairing and shell effects in the over-all distribution of nuclides in the low energy neutron fission of ^{235}U and ^{233}U. Fission with 14 MeV neutrons will not be discussed here because of the complexity and inhomogeneity of the fissioning system, due to second and third chance fission.

EVALUATION AND PRESENTATION OF EXPERIMENTAL FISSION YIELDS

A compilation of experimental yields of ^{235}U thermal neutron fission products was published in our recent discussion of the odd-even effect in ^{235}U.[16] Table I presents the corrected fractional independent fission yields (FIY) from Ref. 16 and some recently published experimental results.[28] The chain yields are the recommended values from Ref. 29.

Yields of ^{233}U thermal neutron fission products were taken from a recent compilation by Walker[29] and from the current literature (see footnote of Table II). Yields of delayed neutron precursors were calculated from updated P_n values[30] and delayed neutron yields of the various groups.[31] The data is summarized in Table II and compared with the normal distribution for ^{233}U thermal neutron fission.[32] The relative deviation from the normal value indicates the magnitude of the odd-even effect. The "corrected" fractional independent yields are either the experimental values or those calculated from fractional yields of other members of the isobaric chain, whenever possible. The corrected values were considered best for the calculation of the odd-even effect and for the following discussion.

Table III and Figs. 1 and 2 present the independent elemental yields in the thermal neutron fission of ^{235}U and ^{233}U, compared with the normal ones. The yields were obtained by multiplying the corrected fractional independent yields by the chain yields from Ref. 29. The yields of isotopes, which have not yet been determined experimentally, were calculated according to the prevailing odd-even systematics, provided they contributed only a small part to the elemental yield. Figs. 3 and 4, which present the independent isotonic yields, were constructed similarly.

The systematic enhancement of the yields of fission fragments with paired proton numbers is clearly demonstrated in Figs. 5 and 6, presenting the isotopic distribution of several elements in the thermal neutron fission of ^{235}U and ^{233}U, respectively, and in Figs. 7 and 8, presenting a charge dispersion in several isobaric chains where the experimental yields constitute a saw-tooth structure on a "normal" distribution.

The magnitude of the odd-even effect for the various fission products in thermal neutron fission is presented graphically in Figs. 9 and 10 for ^{233}U and ^{235}U, respectively. As may be seen in those figures, the average odd-even effect is about 25% and within the examined interval (±2 charge units) does not depend on the distance from Zp, the most probable charge. Though the statistical dispersion is rather broad (one standard deviation from the average is marked by a broken line), the effect is clearly demonstrated. Table IV presents the experimental yields of ^{235}U fission products by fission spectrum neutrons (average energy 1.9 MeV). Even though these data are rather scarce, it seems that the proton pairing effect drops to about 35% of its value for thermal neutron fission, or more precisely it is (8 ±4)%. The experimental data of fast fission yields were taken from Wolfsberg.[33] The fission yields in fast neutron fission of ^{235}U are widely dispersed, with large errors, and therefore in Table IV we find a few deviations from the expected enhanced yields of even-z elements.

TABLE II. Fission yields in ^{233}U thermal neutron fission. FCY, fractional cumulative yield; FIY, fractional independent yield; Δ (%), [(FIY − "normal" FIY/"normal" FIY]×100.

Mass A	Chain yield[a] (%)		Fission yields $_{33}$As	$_{34}$Se	$_{35}$Br	$_{36}$Kr	$_{37}$Rb
82	0.6 ±0.12	FCY					
		FIY			0.002± 0.000 25[b]		
		Corrected FIY					
		Normal FIY[c]	0.604	0.275	0.009		
		Δ (%)					
84	1.68±0.04	FCY					
		FIY					
		Corrected FIY	0.13[d]				
		Normal FIY[c]	0.182	0.628	0.182	0.004	
		Δ (%)					
85	2.19±0.05	FCY					
		FIY					
		Corrected FIY	0.034[d]				
		Normal FIY[c]	0.052	0.512	0.410	0.026	
		Δ (%)					
86	2.84±0.06	FCY					
		FIY					
		Corrected FIY	0.007[d]				
		Normal FIY[c]	0.009	0.27	0.606	0.114	0.001
		Δ (%)					
			Fission yields $_{34}$Se	$_{35}$Br	$_{36}$Kr	$_{37}$Rb	$_{38}$Sr
87	4.00±0.09	FCY	0.19 ± 0.04[e]	0.58 ± 0.06[d]			
		FIY		0.37 ± 0.05[e]			
		Corrected FIY	0.15 ± 0.06[d]	0.43 ± 0.09[d]	0.41 ± 0.07[d]		
		Normal FIY[c]	0.090	0.582	0.315	0.013	
		Δ (%)		−[26.1 ±15.5]	−[30.1 ±22.2]		
88	5.52±0.09	FCY		0.47 ± 0.06[e]			
		FIY	0.036[f]				
		Corrected FIY	0.02 ± 0.005	0.27 ± 0.08[d]	0.66 ± 0.11	0.05 ± 0.02	
		Normal FIY[c]	0.0175	0.355	0.554	0.071	
		Δ (%)		−[24.5 ±22.2]	+[19.1 ±19.8]		
89	6.33±0.10	FCY		0.19 ± 0.09[e]	0.86 ± 0.05[g]		
		FIY					
		Corrected FIY		0.11 ± 0.027[d]	0.75 ± 0.06	0.14 ± 0.05	
		Normal FIY[c]	0.002	0.144	0.622	0.222	0.006
		Δ (%)			+[20.6 ± 9.6]	−[38.0 ±22.0]	
90	6.83±0.10	FCY			0.67 ± 0.01[g]		
		FIY		0.1 ± 0.03[e]			
		Corrected FIY		0.029 ± 0.01[d]	0.64 ± 0.02	0.29 ± 0.02	0.04 ± 0.01

TABLE II (Continued)

Mass A	Chain yield [a] (%)		Fission yields $_{34}Se$	$_{35}Br$	$_{36}Kr$	$_{37}Rb$	$_{38}Sr$
90		Normal FIY [c]		0.038	0.468	0.457	0.035
		Δ (%)			+[36.7 ± 4.3]	−[36.5 ± 4.4]	
			Fission yields $_{36}Kr$	$_{37}Rb$	$_{38}Sr$	$_{39}Y$	
91	6.51 ± 0.08	FCY	0.33 ± 0.01 [g]				
		FIY				0.012 ± 0.006 [h]	
		Corrected FIY	0.32 ± 0.01	0.52 ± 0.10	0.15 ± 0.10		
		Normal FIY [c]	0.237	0.620	0.136	0.002	
		Δ (%)	+[35.0 ± 4.2]	−[16.1 ± 16.1]			
92	6.64 ± 0.09	FCY	0.13 ± 0.005 [g]				
		FIY					
		Corrected FIY	0.13 ± 0.005				
		Normal FIY [c]	0.078	0.567	0.338	0.015	
		Δ (%)	+[66.7 ± 6.4]				
93	7.04 ± 0.09	FCY	0.023 ± 0.001 [g]				
		FIY					
		Corrected FIY	0.023 ± 0.001	0.326 ± 0.075 [d]			
		Normal FIY [c]	0.016	0.344	0.563	0.076	
		Δ (%)	+[43.7 ± 6.9]	−[5.2 ± 21.8]			
94	6.79 ± 0.09	FCY					
		FIY					
		Corrected FIY		0.104 ± 0.024 [d]			
		Normal FIY [c]	0.002	0.140	0.620	0.231	
		Δ (%)		−[25.7 ± 17.1]			
			Fission yields $_{50}Sn$	$_{51}Sb$	$_{52}Te$	$_{53}I$	$_{54}Xe$
131	3.52 ± 0.08	FCY		0.66 [i]	0.98 ± 0.002 [j]		
		FIY		0.52 ± 0.03 [k]	0.469 ± 0.03 [k]	0.014 ± 0.003 [k]	
		Corrected FIY	0.14 ± 0.03	0.52 ± 0.03	0.33 ± 0.03	0.014 ± 0.003	
		Normal FIY	0.0952	0.589	0.303	0.0116	
		Δ (%)	+[47.0 ± 31.5]	−[11.7 ± 5.1]	+[8.9 ± 9.9]		
132	4.82 ± 0.11	FCY					
		FIY	0.025 ± 0.005 [l]	0.27 ± 0.03 [l]	0.64 ± 0.04 [l]	0.06 ± 0.05 [l]	
		Corrected FIY	0.025 ± 0.005	0.27 ± 0.03	0.64 ± 0.04	0.06 ± 0.05	
		Normal FIY	0.0207	0.380	0.535	0.062	
		Δ (%)	+[20.8 ± 24.1]	−[28.9 ± 7.9]	+[19.6 ± 7.5]	−[3.2 ± 80.6]	
133	6.02 ± 0.21	FCY		0.08 ± 0.04 [k]	0.76 ± 0.03 [k]		
		FIY				{0.21 ± 0.01 [m] 0.14 ± 0.01 [n]	

TABLE II (*Continued*)

Mass A	Chain yield[a] (%)		Fission yields $_{50}$Sn	$_{51}$Sb	$_{52}$Te	$_{53}$I	$_{54}$Xe
133		Corrected FIY				0.17 ± 0.03	
		Normal FIY	0.002	0.148	0.623	0.222	0.006
		Δ (%)				−[23.4 ±13.5]	
134	6.13±0.14	FCY			0.62 ± 0.03 [j]		
		FIY				0.33 ± 0.02 [n] 0.37 ± 0.013 [m]	
		Corrected FIY			0.62 ± 0.04	0.35 ± 0.02	
		Normal FIY		0.030	0.434	0.491	0.045
		Δ (%)			+[42.8 ± 9.2]	−[28.7 ± 4.0]	
135	6.24±0.15	FCY		0.003 [d]		0.77 ± 0.007 [o]	
		FIY				0.606 ± 0.024 [n]	0.218 ± 0.005 [p] 0.195 ± 0.014 [q]
		Corrected FIY		0.003	0.184 ± 0.04	0.606 ± 0.024	0.21 ± 0.02
		Normal FIY		0.0039	0.188	0.631	0.179
		Δ (%)		−23.0	−[2.1 ±21.3]	−[4.0 ± 3.8]	+[17.3 ±11.2]

Mass A	Chain yield[a] (%)		Fission yields $_{52}$Te	$_{53}$I	$_{54}$Xe	$_{55}$Cs	$_{56}$Ba
136	6.87±0.16	FCY					
		FIY		0.27 ± 0.05 [r]		0.016 ± 0.003 [s]	
		Corrected FIY	0.065 ± 0.026 [d]			0.016 ± 0.003	
		Normal FIY	0.052	0.512	0.410	0.0255	
		Δ (%)	+[20 ± 8]			−[37.2 ±11.8]	
137	6.80±0.16	FCY	0.01 [d]	0.25 ± 0.09 [d]	0.896 ± 0.005 [g] 0.93 ± 0.04		
		FIY				0.07 ± 0.04 [f]	
		Corrected FIY	0.011	0.24 ± 0.09	0.68 ± 0.1		
		Normal FIY	0.009	0.275	0.604	0.111	0.001
		Δ (%)		−[13.3 ±32.5]	+[12.6 ±16.5]		
138	5.92±0.16	FCY			0.83 ± 0.01 [g]		
		FIY					
		Corrected FIY		0.086 ± 0.034 [d]	0.744 ± 0.04	0.17 ± 0.01	
		Normal FIY	0.001	0.099	0.599	0.301	0.011
		Δ (%)		−[13.1 ±34.3]	+[24.2 ± 6.7]	−[43.5 ± 3.3]	
139	6.40±0.20	FCY			0.48 ± 0.01 [g]		
		FIY		0.016 ± 0.006 [d]			0.084 ± 0.007 [t]
		Corrected FIY		0.016 ± 0.006	0.464 ± 0.02	0.436 ± 0.01	0.084 ± 0.017
		Normal FIY		0.023	0.392	0.526	0.058
		Δ (%)		−[30.4 ±26.1]	+[18.4 ± 5.1]	−[17.1 ± 1.9]	+[50.0 ±12.1]

TABLE II (Continued)

Mass A	Chain yield[a] (%)		Fission yields $_{53}$I	$_{54}$Xe	$_{55}$Cs	$_{56}$Ba	$_{57}$La
140	6.43 ± 0.12	FCY		0.23 ± 0.01[g]			
		FIY	0.0025 ± 0.001[d]			0.27 ± 0.04[u]	
		Corrected FIY		0.23 ± 0.01	0.50 ± 0.04	0.27 ± 0.04	
		Normal FIY	0.0033	0.174	0.632	0.192	0.0041
		Δ (%)	$-[24.2 \pm 30.3]$	$+[32.2 \pm 5.7]$	$-[20.9 \pm 6.3]$	$+[40.6 \pm 20.8]$	
141	6.60 ± 0.50	FCY		0.051 ± 0.003[g]			
		FIY					
		Corrected FIY		0.051 ± 0.003			
		Normal FIY	0.0003	0.050	0.507	0.416	0.0268
		Δ (%)		$+[2 \pm 6]$			
142	6.61 ± 0.12	FCY		$0.0098^{+0.0008}_{-0.0005}$[g]			
		FIY					
		Corrected FIY		0.0098 ± 0.0007			
		Normal FIY		0.0092	0.275	0.604	0.111
		Δ (%)		$+[6.2 \pm 7.6]$			
143	5.86 ± 0.10	FCY					
		FIY					
		Corrected FIY					
		Normal FIY		0.0011	0.101	0.595	0.292
		Δ (%)					

[a] Reference 29.
[b] B. L. Tracy and H. G. Thode, Can. J. Phys. 48, 1708 (1970).
[c] Normal FIY from Ref. 32 based on Wahl's constants (Ref. 17).
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TABLE III. Independent yields of complementary elements (Z, $92-Z$).

Elements	$^{233}U+n_{th}$ Yield[a] (%)	Normal yield[b]	Δ[c] (%)	$^{235}U+n_{th}$ Yield (%)	Normal yield[b]	Δ[c] (%)
$_{33}As-_{59}Pr$		1.5		1.2 ± 0.1	1.4	−(14.3 ± 7.1)
$_{34}Se-_{58}Ce$		4.1		4.3 ± 0.3	3.6	+(19.4 ± 8.3)
$_{35}Br-_{57}La$		8.5		5.9 ± 0.4	7.6	−(22.4 ± 5.3)
$_{36}Kr-_{56}Ba$	17.87 ± 0.8	14.0	+(27.7 ± 5.6)	15.2 ± 0.3	12.7	+(20.0 ± 2.4)
$_{37}Rb-_{55}Cs$	12.5 ± 1.2	16.4	−(23.8 ± 7.3)	11.7 ± 0.5	15.1	−(22.5 ± 3.3)
$_{38}Sr-_{54}Xe$	18.9 ± 1.1	16.2	+(16.7 ± 7.1)	19.8 ± 1.0	16.0	+(23.7 ± 6.2)
$_{39}Y-_{53}I$	12.2 ± 1.0	14.4	−(15.5 ± 6.9)	11.9 ± 1.0	15.8	−(24.7 ± 6.4)
$_{40}Zr-_{52}Te$	14.9 ± 1.3	12.3	+(21.0 ± 10.4)	18.2 ± 0.6	14.7	+(23.8 ± 4.1)
$_{41}Nb-_{51}Sb$		7.9			9.1	
$_{42}Mo-_{50}Sn$		3.1			2.9	
Average			21 ± 7.5			22 ± 5.4

[a] Yields taken here are based on the "corrected" values taken from Tables I and II.
[b] Reference 32.
[c] Δ (%) = [(corrected fission yield − normal fission yield)/normal fission yield] × 100.

DISCUSSION

Mass combinations of fission fragments

The proton pairing effect in the yields of elements in thermal neutron fission of ^{233}U and ^{235}U was found in both cases to be (22 ± 7)% relative to the normal distribution. In both cases a pair of fission fragments consists of the same elements but the mass combinations differ by two mass units.

The equality of the odd-even effect for the two fissile nuclei which have approximately equal excitation energies (Table V) proves that proton pairing rather than mass combinations is responsible for the observed effect. The pairing energy for a pair of even-z elements is about 2.7 MeV.[34] Therefore, the excitation energy required to produce odd-z fission fragments has to be higher by 2.7 MeV than that required for the formation of even-z fragments. Consequently, the magnitude of the odd-even effect is a function of the pairing energy and the available excitation.[35]

The various combinations of masses undoubtedly have an influence on the significant dispersion of the pairing effect of individual fragments relative to the mean value (Figs. 9 and 10), but since the masses, as calculated by various mass formulas,[36] are not known with sufficient precision, it cannot be evaluated.

Closed shells

In both ^{235}U and ^{233}U thermal neutron fission, the magnitude of the odd-even effect is higher than the average for products with 82 neutrons (^{132}Sn; ^{133}Sb; ^{134}Te; ^{135}I and ^{136}Xe) and lower than average for

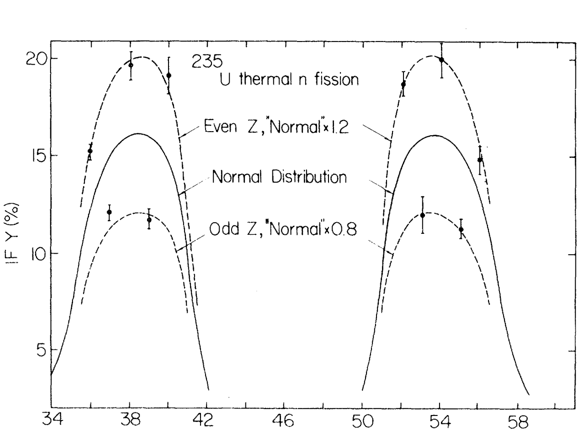

FIG. 1. Element yield distribution in thermal neutron fission of ^{235}U.

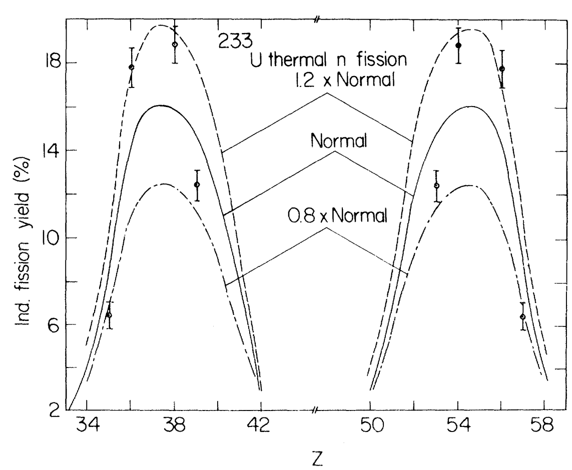

FIG. 2. Element yield distribution in thermal neutron fission of ^{233}U.

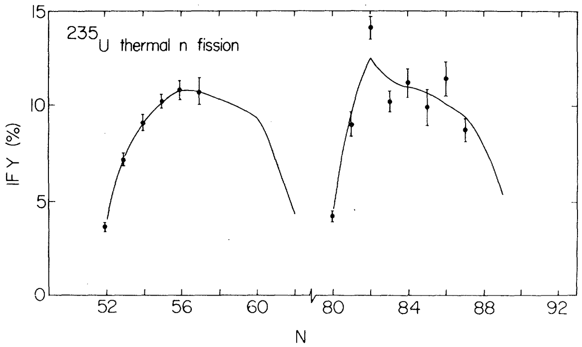

FIG. 3. Isotonic yields in thermal-neutron-induced fission of ^{235}U: ——, Wahl's "normal" curve; ●, experimental points.

the neighboring isobars, as may be seen from Figs. 9 and 10. In both cases, the influence of a closed neutron shell, though clearly discernible, does not exceed one standard deviation (~ 7%) of the odd-even proton effect. Even the enhanced yield of mass 134 in the thermal neutron fission of ^{235}U, due mainly (~83%) to the contribution of ^{134}Te, cannot be attributed only to the 82-neutron shell, since there is no parallel enhancement in the yield of ^{134}Te in the thermal neutron fission of ^{233}U. It was, therefore, suggested[37] that the effect was due to the mass combination ^{102}Zr-^{134}Te which may be energetically preferred to the combination ^{100}Zr-^{134}Te. The contributions of the 50-neutron and 50-proton closed shells to the odd-even effect is still smaller.

Neutron pairing

The neutron pairing effect in fission fragments cannot be established quantitatively in this work, due to prompt neutron evaporation. However, there is a preference for neutron pairing in the

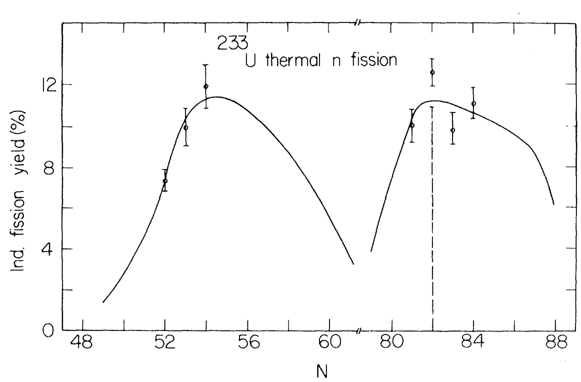

FIG. 4. Isotonic yields in thermal-neutron-induced fission of ^{233}U: ——, Wahl's "normal" curve; O, experimental points.

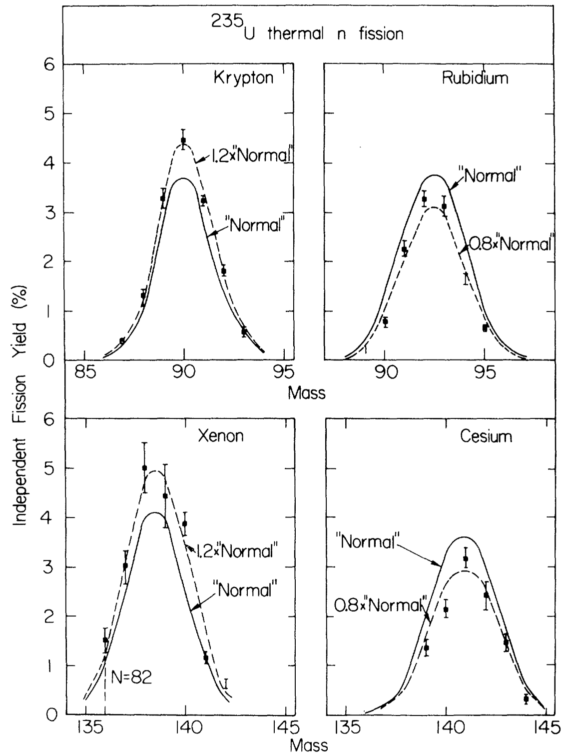

FIG. 5. Isotopic yield distributions in thermal neutron fission of ^{235}U.

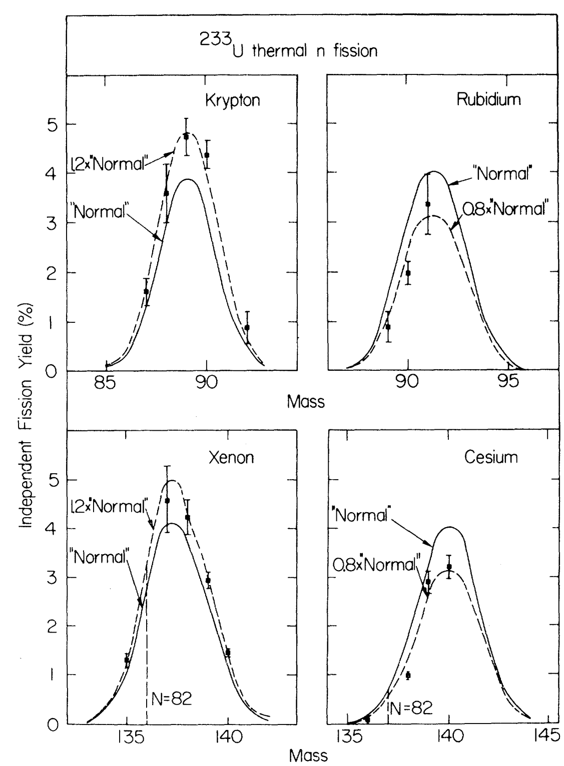

FIG. 6. Isotopic yield distributions in thermal neutron fission of ^{233}U.

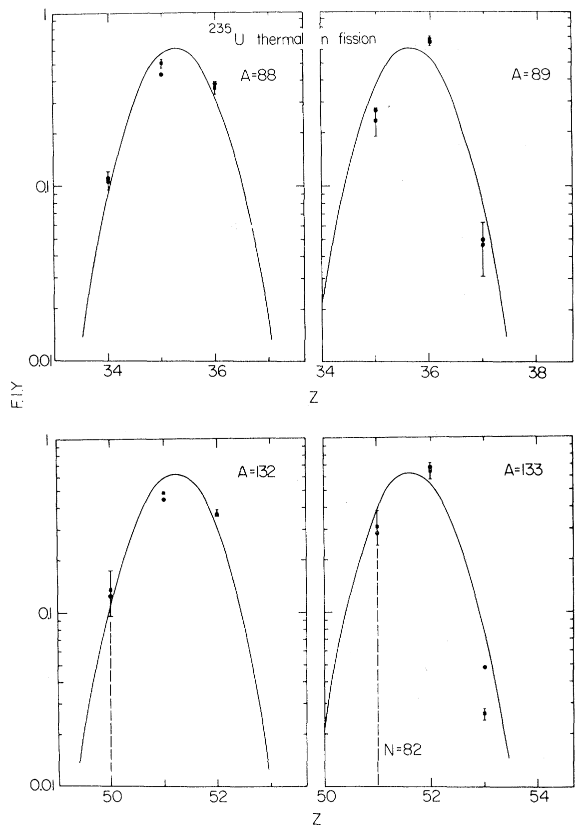

FIG. 7. Fractional independent yields of isobars in thermal neutron fission of ^{235}U: ———, Wahl's "normal" distribution; ×, experimental "corrected" values; ●, "normal" values multiplied by 1.22 for even-Z elements, and by 0.78 for odd-Z elements.

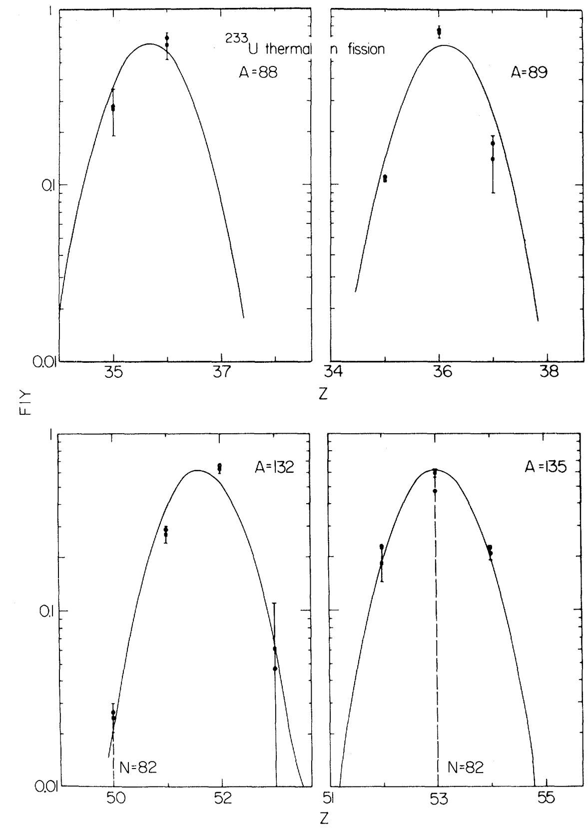

FIG. 8. Fractional independent yields of isobars in thermal neutron fission of ^{233}U: ———, Wahl's "normal" distribution; ×, experimental "corrected" values; ●, normal values multiplied by 1.22 for even-Z elements and by 0.78 for odd-Z elements.

post-neutron emission products, in the heavy mass peak. The yields of about $\frac{2}{3}$ of the fission products with even neutron numbers are higher than predicted by the odd-even systematics. This effect is better demonstrated in the isotonic yields (Figs. 3 and 4), where an average effect of $(8 \pm 5)\%$ is found.

The neutron pairing effect does not indicate whether it is due to neutron evaporation or to a residual primary effect. In an analogy to the proton pairing, we may assume neutron pairing in fission fragments as well. A careful analysis of independent fission yields of mass separated cesium and rubidium isotopes actually indicates a neutron pairing effect of about 15% in the heavy isotopes[18] where the neutron emission does not affect significantly the primary structure due to a rapid drop in yields with increasing mass. The above experimental value for neutron pairing which is close to the $22(\pm 7)\%$ found for proton pairing substantiates the assumption that there

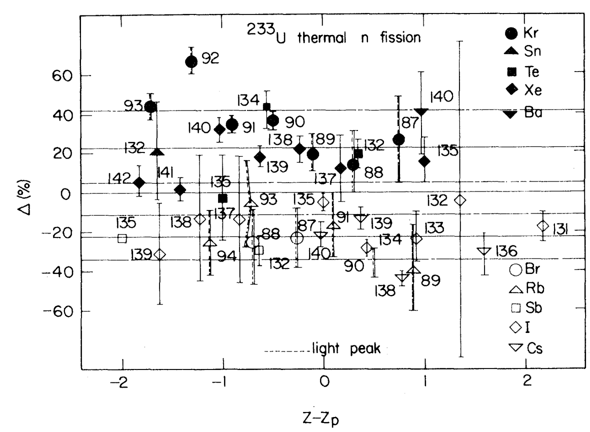

FIG. 9. Odd-even effect as a function of the distance from Zp, the most probable charge, in thermal neutron fission of ^{233}U: Δ (%), [(corrected fractional independent yield - "normal" FIY)/"normal" FIY] × 100; ———, average odd-even effect; - - -, one standard deviation from the average Δ.

TABLE IV. Independent yields in fast fission of ^{235}U (fission spectrum neutrons). Δ average is (8 ± 4)%.

A	Chain yield [a] (%)		Yields $_{34}$Se	$_{35}$Br	$_{36}$Kr	$_{37}$Rb	$_{38}$Sr
87	2.55	FCY		0.87 ± 0.17 [b]			
		FIY					
		Normal FIY	0.253	0.613	0.125	0.002	
		Corrected FIY			0.13 ± 0.17		
		Δ (%)					
89	4.60	FCY			0.935 ± 0.006 [c]		
		FIY				0.065 ± 0.006	
		Normal FIY	0.016	0.344	0.563	0.076	
		Corrected FIY				0.065 ± 0.006	
		Δ (%)				−[14.4 ± 7.9]	
91	5.40	FCY			0.52 ± 0.02 [c]		
		FIY					
		Normal FIY	0.0002	0.037	0.463	0.463	0.037
		Corrected FIY		0.033 ± 0.01	0.49 ± 0.03	0.44 ± 0.03	0.04 ± 0.01
		Δ (%)			+[5.8 ± 6.5]	−[5.0 ± 6.5]	
92	5.69	FCY			$0.204^{+0.03}_{-0.02}$ [c]		
		FIY					
		Normal FIY		0.006	0.231	0.620	0.14
		Corrected FIY			$0.20^{+0.03}_{-0.02}$		
		Δ (%)			−[11.7 ± 10.8]		
			Yields $_{53}$I	$_{54}$Xe	$_{55}$Cs	$_{56}$Ba	$_{57}$La
139	6.38	FCY		0.72 ± 0.01 [c]			
		FIY					
		Normal FIY	0.095	0.589	0.303	0.012	
		Corrected FIY	0.086 ± 0.01	0.634 ± 0.02	0.265 ± 0.02	0.015 ± 0.005	
		Δ (%)		+[7.6 ± 3.4]	−[11.9 ± 6.6]		
140	5.91	FCY		0.47 ± 0.01 [c]			
		FIY					
		Normal FIY	0.022	0.386	0.532	0.06	
		Corrected FIY	0.02 ± 0.01	0.45 ± 0.02	0.464 ± 0.03	0.066 ± 0.01	
		Δ (%)		+[16.6 ± 5.2]	−[12.8 ± 5.6]		
141	5.97	FCY		$0.137^{+0.02}_{-0.01}$ [c]			
		FIY					
		Normal FIY	0.003	0.169	0.627	0.196	0.004
		Corrected FIY		$0.137^{+0.02}_{-0.01}$			
		Δ (%)		$-[18.9^{+11.8}_{-5.9}]$			

[a] Reference 41.
[b] Reference 31.
[c] Reference 33.

TABLE V. Excitation and fission barrier energies.

Fissioning nuclide	Neutron energy (MeV)	Excitation energy [a] (MeV)	Symmetric fission barrier [b] I	II	Asymmetric fission barrier [c] (MeV)	Prompt neutrons emitted $\bar{\nu}$ [d]
^{234}U	0	6.84	4.9	8.1	6.0	2.48 ± 0.007
^{236}U	0	6.54	5.6	8.9	6.0	2.41 ± 0.007
	~1.9	8.44	5.6	8.9	6.0	2.50 ± 0.02

[a] Reference 35.
[b] Reference 38.
[c] Reference 3.
[d] J. P. Unik and G. E. Gindler, ANL Report No. ANL-7748, 1971 (unpublished).

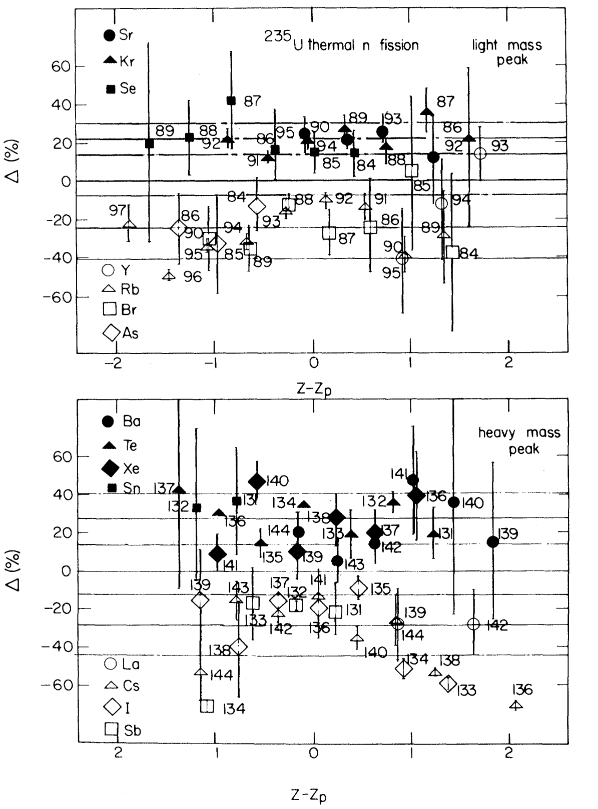

FIG. 10. Odd-even effect as a function of the distance from Zp, the most probable charge, in thermal neutron fission of ^{235}U: Δ (%), [(corrected fractional independent yield – "normal" FIY)/"normal" FIY] × 100; ●, even-Z elements; ○, odd-Z elements; ——, average odd-even effect; -----, one standard deviation from the average Δ.

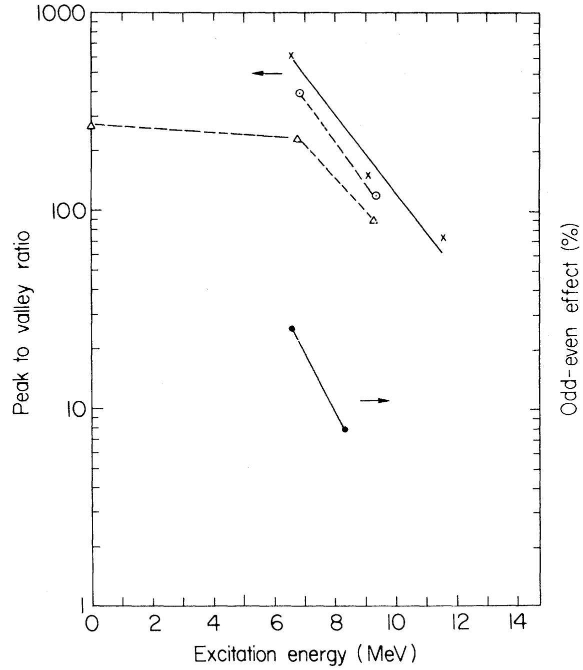

FIG. 11. Peak-to-valley ratio and odd-even effect as functions of excitation energy: ×, ^{236}U; ○, ^{234}U; Δ, ^{240}Pu, peak-to-valley ratio; ●, ^{236}U odd-even effect.

should be a neutron pairing effect, approximately equal to that of protons, in the primary distribution of yields of fission fragments. Thus the neutron effect in the isotonic yields, though observed only in the heavy mass peaks of both ^{235}U and ^{233}U (Figs. 3 and 4), may be a residual primary effect.

Excitation energy

Fission of ^{235}U with fission spectrum neutrons (average energy 1.9 MeV) results in an odd-even effect of only $(8 \pm 4)\%$ (Table IV), i.e., a decrease by a factor of (2.7 ± 2). Due to the paucity of experimental data, this figure should be considered to be preliminary, but it indicates clearly the tendency to "wash out" the structure with increasing excitation energy. Similarly, the peak-to-valley ratio between thermal and fast fission in ^{235}U and ^{233}U decreases by factors of 2.4 and 2.7, respectively (Fig. 11). This similarity might be expected since the proton pairing energy of 2.7 MeV[34] is comparable to the energy difference between symmetric and asymmetric splits, as calculated by Tsang and Wilhelmy.[3] The energy barriers for symmetric fission are between 8.1[38] and 9.1 MeV for ^{234}U and 8.5 and 9.7 MeV for ^{236}U, while the asymmetric saddle point energy is 6 MeV for both nuclei (Table V). (The differences in the energies are due to different methods of calculation). Since the energy difference between symmetric and asymmetric fission is about 2–3 MeV, the odd-even effect should be energy dependent in the same way as the peak-to-valley ratio.

In thermal neutron fission the excitation energy is just about that required to cross the saddle point. Fission, therefore, will proceed through the energetically most favorable path, preserving proton pairs which lower the potential energy surface by the pairing energy. At higher excitation energies, there is more available energy to split the nucleonic pairs and consequently the pairing effect drops.

Comparison of present work with other studies

Since the odd-even effect in thermal neutron fission of ^{233}U and ^{235}U is one of the parameters determining the mass split, it is interesting to check its existence in the fission yields of other, doubly even, fissioning nuclei. Elemental yields in thermal neutron fission as determined by K x-ray measurements of ^{233}U, ^{235}U, and ^{239}Pu and spontaneous fission of ^{252}Cf did not reveal any enhancement of even-charge elements,[11] probably because of the sensitivity of K x-ray yields to mass and charge and subsequent difficulties in quantitative interpretation. Likewise, independent fission yields of doubly even fission products from spontaneous fission of ^{252}Cf, as determined by Cheifetz by measuring the $+2 \rightarrow +0$ prompt γ transitions, did not reveal any odd-even effect.[39] In a later discussion, however, the possibility of the existence of about 25% odd-even effect was not excluded.[40]

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