PHYSICAL REVIEW C VOLUME 16, NUMBER 1 JULY 1977

Independent yields of fast-neutron fission of ^{232}Th: Observation of proton odd-even effect and neutron shell effect on the yields

Talma Izak-Biran and Saadia Amiel
Nuclear Chemistry Department, Soreq Nuclear Research Centre, Yavne, Isreal

(Received 10 November 1976)

Elemental yields in the fission of ^{232}Th induced by reactor neutrons were measured and found to disclose a pronounced odd-even effect. Independent and cumulative yields of isotopes of Kr, Xe, Sn, and Sb were measured and the independent yields of isotopes of Rb, Cs, and Sb were subsequently derived. A comparison of the yield values obtained with the calculated "normal" yields reveals a constant enhancement of products with an even number of protons, relative to those with an odd number. An average amplitude of 30(±12)% in the fluctuation of proton pairing was established for all isotopes measured, except for the doubly magic ^{132}Sn, which shows an extreme effect, apparently due to its closed neutron shell. The odd-even effect is discussed in connection with the mass distribution, excitation, and total kinetic energies. The neutron shell effect on the yields is discussed with regard to the primary mass split and the neutron emission probabilities.

[NUCLEAR REACTIONS, FISSION Fission yields ^{232}Th(n,f), calculated proton odd-even effect, calculated neutron closed shell effect.]

I. INTRODUCTION

Detailed cumulative and independent yields are available only for thermal neutron fission of ^{235}U, which were found to disclose a substantial odd-even effect[1,2] (viz., enhancement of paired nuclear configurations). As there is a paucity of information on elemental yields of other even-Z fission nuclides, an attempt was made to measure the independent yields in neutron-induced fission of ^{232}Th. Most of the available experimental data on ^{232}Th deal with chain yields, while very little is available on the yields of individual species. The only independent yields that have been measured so far are of ^{134}I and ^{135}I.[3]

Thorium, a light fissionable nuclide, is of interest since comparison of its fission yield distribution and the extent of odd-even fluctuation with similar data from heavier fissionable nuclides may contribute to the understanding of factors affecting the mass and charge distribution in low energy fission of even-Z nuclides.

II. EXPERIMENTAL

Samples of 2 g of ^{232}Th in the form of thorium nitrate were irradiated for periods of 10–40 sec in the Israel research reactor IRR-1 at a fast neutron flux of 4×10^{11} neutrons/cm^2 sec. The plastic capsule containing the sample was covered with a Cd shield to minimize thermal neutron capture in thorium which results in a very radioactive sample due to 23 min ^{233}Th.

The fission-produced Kr and Xe were separated from the other elements by sweeping with a stream of helium. Sweeping started 5 sec after the end of irradiation and lasted for 1 sec. The γ ray spectra of the noble gases and their daughters were followed from 20 sec after the end of irradiation to up to a few hours. From these γ ray activities and the known absolute γ ray abundances, the corresponding cumulative and independent yields were calculated. The yields of the isotopes of Cs and Rb were derived from the measured yields of the corresponding isobaric members Xe and Kr and from the respective chain yields. More details are given elsewhere.[4]

Fission-produced tin and antimony were chemically separated as hydrides (using NaBH$_4$) from the other fission products in a separation time of 10 sec, which started 10 sec after the end of irradiation. Their γ ray spectra were followed for several hours, starting 30 sec after the end of irradiation. The independent yield of ^{132}Sn was calculated from the γ activities, peak energies, and the known absolute γ ray intensities. The yield of ^{131}Sn cannot be determined in the same way, since the absolute γ ray intensities of ^{131}Sn are unknown. Therefore, tin was also isolated from thermal fission of ^{235}U under the same conditions, and from the two measured γ spectra of tin from ^{232}Th and from ^{235}U and the known ^{130,131}Sn yields in ^{235}U, the independent yield of ^{131}Sn from ^{232}Th fission was obtained. The independent fission yields of ^{131,132}Sb were derived from the measured yields of the corresponding isobaric members ^{131,132}Sn and from the respective chain yields. The independent yield of ^{132}Sb was also obtained directly from the growth and decay of the measured spectra of this isotope. More details are given elsewhere.[5]

The above procedures were verified by remeasuring the yields of the corresponding isotopes from thermal fission of ^{235}U and good agreement with published values was obtained.[4,5]

III. RESULTS

The fractional independent yields were calculated by dividing the measured independent yields by the respective chain yields.[6-16] (All the data available for each mass chain were averaged.) Whenever the cumulative yields were measured and the parent contributions were relatively low, the independent yields were derived by subtraction and iteration of the parent contributions. The odd-even effect in the yield distributions of the isotopes studied here was revealed by comparing the measured fractional independent yields (FIY) with the calculated "normal" yields.[17] The normal FIY were calculated from the Z_p, the most probable charge of the isobaric chains, and a constant width of the charge distributions. As the widths of the charge distribution are the same for all fission nuclides in low energy fission[18] the width of this Gaussian was taken to be 0.56 ±0.06 as was established for ^{235}U.[17] The width for the thermal fission of ^{235}U has been shown[19] to be Z dependent but since the main variation of the width in the nuclear charge distribution is due to the proton pairing[19] (i.e., the width variation and the odd-even fluctuation both result from even-proton yield enhancement) a constant width has to be taken in order to establish the proton odd-even effect. The Z_p of each isobaric chain in the fission of Th was calculated from the equation[20,21]:

$$Z_p = Z_p^{(\mathrm{ref})} + \Delta Z_p ,$$

where $Z_p^{(\mathrm{ref})}$ is the Z_p of ^{235}U.[17] ΔZ_p of ^{232}Th was obtained semiempirically,[20,21] using the equation of Coryell, Kaplan, and Fink[22]:

$$\Delta Z_p = a(Z_c - 92) + b(A_c - 236) + C(E^* - 6.52) ,$$

where Z_c and A_c are the nuclear charge and mass of thorium and E^* is the energy of the compound nucleus ^{233}Th. It seems more accurate to use different ΔZ_p values for products in the light and heavy peaks of the mass yield curve than to take the same value, since the shift of the respective light and heavy mass peaks is incoherent. This was done by Nethaway and Barton[20] while Wolfsberg[21] used a constant ΔZ_p. We calculated two sets of "normal" yields, one based on a variable ΔZ_p and the other based on a constant ΔZ_p. The "normal" FIY, the measured FIY, and the odd-even fluctuations in the yields in fast fission of ^{232}Th can be seen in Table I. The odd-even fluctuations for the various fission products are shown graphically in Figs. 1(a) and 1(b). These average odd-even effects are seen to be about 30% and do not depend on the distance from Z_p, within the examined interval of plus or minus one charge unit. Though the statistical dispersion is rather broad (one standard deviation from the average is marked by a broken line), the effect is clearly demonstrated and the mean value is easily derived. An enhancement of +28(±10)% in the fission yields of products with an even number of protons, and a corresponding decrease of −32(±12)% for products with an odd number of protons was observed by comparing the experimental results with the set of calculated data, based on a variable ΔZ_p [Fig. 1(a)]. Using a constant ΔZ_p, an effect of +29(±13)% and −35(±13)% was obtained [Fig. 1(b)]. A weighted mean effect of ±30(±12)% can be established for the reactor neutron fission of ^{232}Th, using either or both approaches.

The systematic enhancement of the yields of nuclides with paired proton numbers is clearly demonstrated in Fig. 2, which shows the isotopic distribution of several elements. A proton-pairing effect of ~30% is apparent, as the measured yield fits well with the curves obtained by multiplying the "normal" distribution by a factor of 1.3 or 0.7 for even- and odd-proton numbers, respectively. Figure 3 presents the charge dispersion in several isobaric chains where the experimental yields show a sawtooth structure superimposed on the "normal" distribution.

A high effect of +71(±26)% (when ΔZ_p is variable) or +91(±30)% (when ΔZ is constant) was obtained for the doubly "magic" nucleus ^{132}Sn. It was mentioned above, that it seems to be more accurate to use different ΔZ_p values for products in the light and heavy peaks of the mass yield curve. Since the effects obtained for the two sets of normal data are quite different, it is preferable to take the lower value based on a variable ΔZ_p. The net shell effect is therefore: 71(±26)% − 30(±12)% = 41(±28)%. The other $N = 82$ isotones do not show any significant deviation from the common odd-even effect.

IV. DISCUSSION

A. Odd-even effect

The odd-even effect found in reactor neutron fission of ^{232}Th indicates that some of the proton-paired structure is preserved, since the even fragments are considerably more abundant than the unpaired configurations. The proton-pairing energy at the saddle configuration is 2.6 MeV[23] and if energy in excess of this pairing gap is available by excitation, particle-hole excited states may be filled by single nucleons. Then, one can assume that there will be an equal probability for formation of odd- or even-proton number fragments, due to random pairing of nucleons upon scission.

^{232}Th, as shown in Fig. 4, can fission only with neutrons which possess energies above 1.4 MeV. The energy distribution of the neutrons responsible for the fission reactions was obtained by multiplying the fission neutron spectrum $\phi(E)$, described by the equation[24] $\phi(E) = 0.77Ee^{-0.776E}$, by the fission cross section (σ) of ^{232}Th.[25] It can be seen (in Fig. 4) that 22% of the fission reactions occur with more energy that is needed to reach the particle-hole state. Consequently, only fis-

TABLE I. Fractional independent yields (FIY) and odd-even effect in fast fission of ^{232}Th.

				FIY "normal"		Δ(%)[a]	
Mass	Chain yields	Element	FIY measured	ΔZ_p (Ref. 20)	ΔZ_p (Ref. 21)	ΔZ_p (Ref. 20)	ΔZ_p (Ref. 21)
90	7.20 ± 0.21%	Kr	0.762 ± 0.114[b]	0.567	0.550	+35.8% ±20.8%	+37.4% ±20.7%
91	6.80 ± 0.30%	Kr	0.740 ± 0.064[b]	0.619	0.624	+20.6% ±7.7%	+17.5% ±7.4%
		Rb	0.163 ± 0.066[c]	0.230	0.216	−27.0% ±24.2%	−25.9% ±22.5%
131	1.73 ± 0.08%	Sn	0.710 ± 0.202[b]	0.546	0.512	+30.7% ±37.1%	+38.2% ±38.6%
		Sb	0.221 ± 0.202[d] 0.230 ± 0.206[d]	0.368	0.410	−39.8% ±52.7%	−43.9% ±48.0%
132	2.87 ± 0.14%	Sn	0.529 ± 0.085[b] 0.550 ± 0.139[d] 0.524 ± 0.140[d]	0.315	0.275	+67.8% ±27.4% +74.6% ±43.6% Mean +70.8% ±25.7%	+92.0% ±31.4% +90.5% ±50.2% Mean +91.3% ±29.6%
		Sb	0.330 ± 0.132[b] 0.345 ± 0.096[d] 0.319 ± 0.097[d]	0.582	0.604	−43.3% ±22.7% −40.7% ±16.5% Mean −42.0% ±14.8%	−45.3% ±21.8% −47.1% ±16.4% Mean −46.2% ±14.4%
134	5.28 ± 0.10%	I	0.03 ± 0.03[e]	0.066	0.084	−54.0% ±29.0%	−64.0% ±24.0%
		Te	0.722 ± 0.074[f] 0.688 ± 0.078[f]	0.575	0.546	+27.0% ±12.8%	+25.9% ±11.6%
135	5.09 ± 0.31%	I	0.15 ± 0.03[e]	0.231	0.269	−35.0% ±13.0%	−44.0% ±11.0%
		Te	0.754 ± 0.048[f] 0.760 ± 0.045[f]	0.620	0.606	+21.7% ±8.3%	±25.4% ±8.4%
139	6.99 ± 0.27%	Xe	0.755 ± 0.078[b]	0.575	0.598	+31.8% ±13.9%	+26.1% ±12.6%
140	7.76 ± 0.36%	Xe	0.773 ± 0.056[b]	0.615	0.598	+24.3% ±9.1%	+31.0% ±9.3%
		Cs	0.146 ± 0.068[c]	0.247	0.286	−39.8% ±27.8%	−48.8% ±23.7%

[a] $\Delta(\%) = \dfrac{\mathrm{FIY}_{\mathrm{measured}} - \mathrm{FIY}_{\mathrm{normal}}}{\mathrm{FIY}_{\mathrm{normal}}} \cdot 100.$

[b] This work.

[c] This work, derived from the measured FIY of the parent.

[d] Derived from the measured FIY of the parent, but the daughter contributes a few percent and its yield, estimated as the (1.0 ± 0.3) normal yield (Refs. 20 and 21) was subtracted, respectively.

[e] Denschlag and Qaim (Ref. 3).

[f] Denschlag and Qaim as per footnote d.

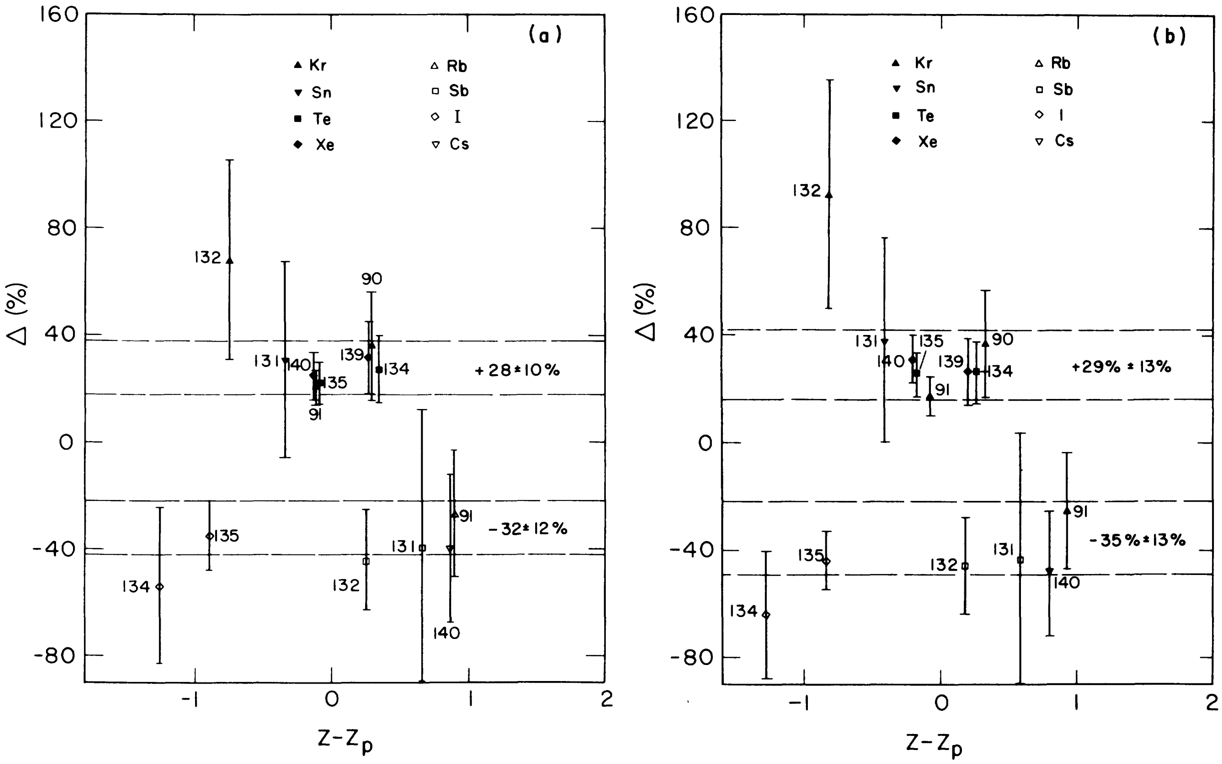

FIG. 1. (a) Odd-even effect as a function of the distance from Z_p [ΔZ_p variable (Ref. 20)]. One standard deviation from the average is marked by a broken line. (b) Odd-even effect as a function of the distance from Z_p [ΔZ_p constant (Ref. 21)]. One standard deviation from the average is marked by a broken line.

sion reactions taking place at excitation below the particle-hole state (i.e., 78% of the reactions) are responsible for preservation of proton pairs in the fission of the even-Z thorium. If the thorium fission would have been induced by neutrons of energy lower than required to exceed the particle-hole state, one can assume that the odd-even effect would be higher: $[30(\pm 12)\%] \times \frac{100}{78} = 38(\pm 13)\%$. The validity of this treatment can be further substantiated by the observed drop in the experimentally determined odd-even effect of the fast-neutron fission of ^{235}U, in which 35% of the fission reactions possess energies above the particle-hole state, as compared with the thermal fission of the same nuclide. The calculated odd-even effect of the fast-neutron fission of ^{235}U (based on that of the thermal neutron fission[2] is $[22(\pm 7)\%]\frac{65}{100} = 14.5(\pm 10)\%$, while the value based on measured yields is $10(\pm 10)\%$.[26] Despite the high errors, the trend and the extent of drop in the odd-even effect can be adequately derived.

If only the excitation energy at the saddle point determined the magnitude of the odd-even effect, one would expect to observe 89% even product nuclides corresponding to an odd-even effect of

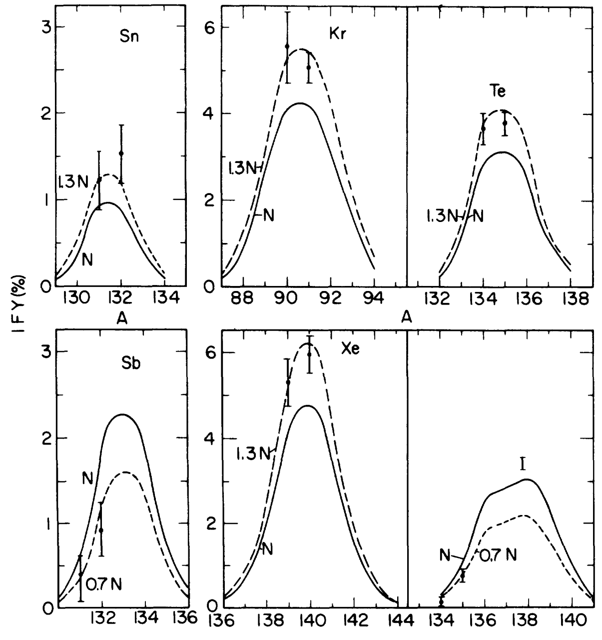

FIG. 2. Isotopic yield distributions in fast fission of ^{232}Th.

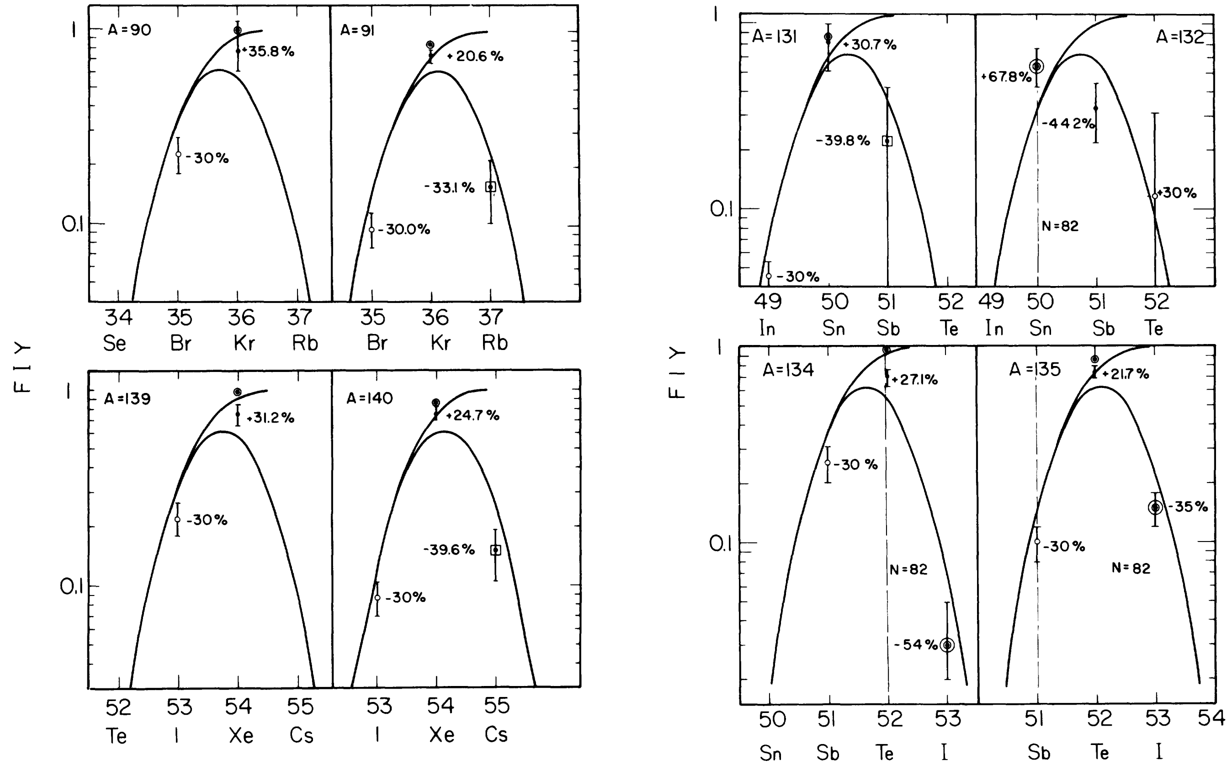

FIG. 3. Fractional independent yields of isobars in fast fission of ^{232}Th [normal based on ΔZ_p (Ref. 20)]. ◎, FIY measured directly; ◉, FCY measured directly; ●, FIY derived from the measured FCY; ◻, FIY derived from the measured FCY of its isobar; ○, "normal" FIY multiplied by 1.30 for even-Z elements and by 0.7 for odd-Z elements.

78%. Since the effects in ^{232}Th and in other fission nuclides[26] are much smaller, another excitation process must be considered. One possibility is the conversion of the kinetic energy of the deforming system before scission. This energy may be partly converted into single particle excitations if the mass flow of the deformed nucleus is not completely adiabatic. Direct experimental evidence in support of this assumption is found in the observed fluctuations in the total kinetic energy (TKE) as a function of mass, in fast-neutron fission of ^{232}Th.[27] At the lowest neutron energy which induces fission (1.38 MeV) the fine structure is very pronounced, but even in fission by 5.6 MeV neutrons, some fine structure is still observed. If the energy required to break a proton pair, i.e., filling the particle-hole excited states by single nucleons, comes exclusively at the expense of TKE, then one would expect the even-Z fragments to possess ~2.6 MeV more than odd-Z fragments. The fluctuations of the fine structure in the TKE as a function of mass are in the range of 0.5-1 MeV. This indicates that a substantial part of the

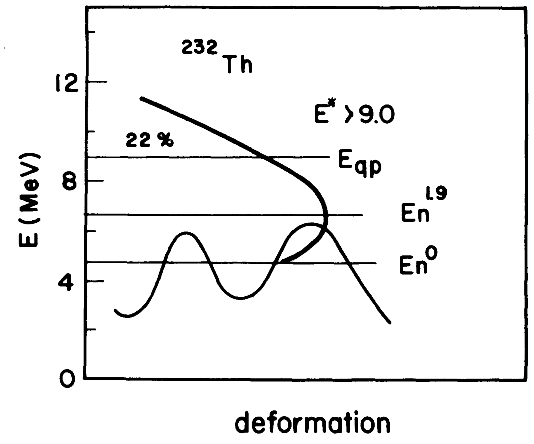

FIG. 4. Fission barrier and particle-hole state of ^{232}Th. $E_n^{0,1.9}$ is the energy induced by the absorbed neutron, thermal, and mean neutron energy 1.9 MeV, respectively. E_{qp} is the energy of the particle-hole state. ——: The fission barrier. The height was taken from Vandenbosch and Huizenga (Ref. 31). ——: ϕ (Ref. 24) × σ (Ref. 25).

energy required to fill the particle-hole states originates from conversion of TKE into internal degrees of freedom by sort of partially viscous flow of the rapidly deforming nucleus before scission.

Additional proton-pair breakings can result from either a reduction in the pairing gap or even level crossings at deformations between saddle and scission points. These can reduce the excitation needed to populate the particle-hole states and possibly explain the quantitative disagreement between the odd-even fluctuation in the TKE and the energy required (viz. 2.6 MeV) for proton-pair breaking at the saddle point.

A great deal of fine structure was already found to exist in the mass distribution curve of ^{232}Th, based on the individual chain yields.[6-16] One can observe good agreement between the average product masses of the even-Z elements Kr, Xe, Te, and Sr (complementary to Te) measured in this work, and the masses corresponding to the peaks in the fine structure of the mass distribution curve. This is consistent with the observation in the fission of another thorium isotope, ^{229}Th[28] in which a very pronounced fine structure indicates a substantial enhancement of even configurations. One can expect that the amplitude of the elemental fission yields in thermal fission of different Th nuclides would be almost identical or very similar as is seen in the various uranium fission reactions.[26] It should be noted however, that the magnitude of the amplitude in the odd-even fluctuation cannot be calculated quantitatively from mass measurements, primarily because of the unknown relative contribution to each mass of the even- and odd-proton configurations and because of poor mass resolution.

B. Closed shell effect

The influence of the closed shell in the fission of ^{232}Th (by fission spectrum neutrons) is very pronounced in ^{132}Sn; its yield is enhanced by ~70%, i.e., the net shell effect is ~40%. Since the other measured isotope with the same closed proton shell (^{131}Sn) does not reveal an enhanced effect, the high yield of ^{132}Sn may be due to the closed shell of neutrons $N = 82$. In the fissioning system of ^{232}Th, which has an odd number of neutrons, every fragment with an odd number of neutrons is complemented by a fragment with an even number of neutrons. Therefore, the enhancement of the ^{132}Sn yield cannot result only from the stability of primary paired neutron configurations. On the other hand, the exceptionally high yield of ^{132}Sn can result from the contributions of the tin isotopes with $N = 83$ and 84, which have substantially high prompt neutron emission probabilities, unlike ^{132}Sn, and relatively quite high "normal" yields (^{133}Sn = 0.418% and ^{134}Sn = 0.102%) compared with the ^{132}Sn yield (0.904%). An indication of the validity of this explanation can be found from the measured yield of ^{134}Te in thermal fission of ^{235}U as a function of kinetic energy.[29] The yield of ^{134}Te was observed to decrease with increasing kinetic energy, i.e., the lower excitation energy makes prompt neutron emission less likely and therefore further contribution of ^{135,136}Te to the ^{134}Te yield diminishes.

The other isotones ^{134}Te and ^{135}I do not have such a pronounced neutron shell effect. Since the yield of ^{136}I is low compared with that of ^{135}I ($N = 82$), the ^{136}I cannot contribute significantly to the yield of the ^{135}I and the expected effect should be low, which is indeed the case. On the other hand, one would expect a high shell effect for ^{134}Te, since the isotope with an additional neutron has a high yield. It might be that the independent yield of ^{134}Te is slightly increased, but this yield was only derived from the measured cumulative yield and carries a significant error, which eventually masks the shell effect.

In the thermal neutron fission of ^{235}U, the relative yield of ^{133}Sn compared with that of ^{132}Sn is low and the yield of ^{134}Sn is negligible. In this case one would expect only a low net shell effect which results mainly from the primary neutron odd-even effect in the ^{236}U even neutron system and indeed an effect of only ~12% was observed experimentally.[30] A careful examination of the net neutron shell effect of ^{133}Sb, ^{134}Te, and ^{135}I in thermal fission of ^{233,235}U[5] reveals that the effect is high when the yields of the corresponding isotopes with $N = 83$ and 84 are relatively high, viz., when they are near the top of the isobaric dispersion.

---

[1] S. Amiel and H. Feldstein, in *Proceedings of the Third IAEA Symposium on Physics and Chemistry of Fission, 1973* (IAEA, Vienna, 1974), paper No. IAEA-SM-174/25.

[2] S. Amiel and H. Feldstein, Phys. Rev. C 11, 845 (1975).

[3] H. O. Denschlag and S. M. Qaim, J. Inorg. Nucl. Chem. 33, 3649 (1971).

[4] S. Amiel and T. Izak-Biran (unpublished).

[5] S. Amiel and T. Izak-Biran (unpublished).

[6] T. J. Kennet and H. G. Thode, Can. J. Phys. 35, 969 (1957).

[7] R. H. Iyer, C. K. Mathews, N. Ravidndran, K. Rangen, D. V. Singh, M. V. Ramaniah, and H. D. Sharma, J.

Inorg. Nucl. Chem. 25, 465 (1963).

[8] K. M. Broom, Phys. Rev. 133, B874 (1964).

[9] A. Wyttenbach, H. R. Von Gunten, and H. Dulakas, Radiochim. Acta 3, 118 (1964).

[10] M. Bresesti, G. Burei, P. Farrari, and L. Moretto, J. Inorg. Nucl. Chem. 29, 1189 (1967).

[11] I. F. Croall, Atomic Energy Research Establishment Report No. AERE-R-5086, 1967 (unpublished).

[12] J. W. Harvey, W. B. Clarke, D. J. Gorman, and R. H. Tomlinson, Can. J. Chem. 46, 2911 (1968).

[13] S. J. Lyle, G. R. Martin, and M. D. M. Rahman, Radiochem. Acta 9, 90 (1968).

[14] D. L. Swindale, D. T. Moore, J. N. Beck, and P. K. Kuroda, J. Inorg. Nucl. Chem. 33, 3643 (1971).

[15] C. Chauvin, Ph.D. thesis, University of Grenoble, 1973 (unpublished).

[16] M. Lammer and O. J. Eder, in *Proceedings of the Symposium on Applications of Nuclear Data and Technology, 1973* (IAEA, Vienna 1973), Vol. I, paper No. IAEA-SM-170/13.

[17] A. C. Wahl, A. E. Norris, R. A. Rouse, and J. C. Williams, in *Proceedings of the Second IAEA Symposium on Physics and Chemistry of Fission, Vienna, 1969* (IAEA, Vienna, 1969), paper No. IAEA-122/116.

[18] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973), p. 332.

[19] G. Siegert, H. Wollnik, J. Greif, R. Decker, G. Fiedler, and B. Pfeiffer, Phys. Rev. C 14, 1864 (1976).

[20] D. R. Nethaway and A. W. Barton, UCRL Report No. UCRL-51458, 1973 (unpublished).

[21] K. Wolfsberg (private communication).

[22] C. D. Coryell, M. Kaplan, and R. D. Fink, Can. J. Chem. 39, 646 (1961).

[23] H. C. Britt, W. R. Gibbs, J. J. Griffin, and R. H. Stokes, Phys. Rev. 139, B354 (1965).

[24] R. C. Barrall and W. N. McElroy, in *Symposium on Personnel Dosimetry for Accidental High-Level Exposure to External and Internal Radiation, 1965* (IAEA, Vienna, 1965), paper No. IAEA-SM-56/3.

[25] D. W. Muir and L. R. Veeser, in *Proceedings of the Third Conference on Neutron Cross Sections and Technology, Oak Ridge, 1971* (National Technical Information Center, Springfield, Virginia, 1971), Vol. I, p. 292.

[26] S. Amiel, H. Feldstein, and T. Izak-Biran, Phys. Rev. C 15, 2119 (1977).

[27] A. I. Sergachev, V. G. Voroleva, B. D. Kuzminov, V. B. Mikhailov, and M. Z. Tarasko, Yad. Fiz. 7, 778 (1968) [Sov. J. Nucl. Phys. 7, 475 (1968)].

[28] J. P. Unik, J. E. Gindler, L. E. Glendenin, K. F. Flynn, A. Gorski, and R. K. Sjoblom, in *Proceedings of the Third IAEA Symposium on Physics and Chemistry of Fission* (see Ref. 1), paper No. IAEA-SM-174/209.

[29] W. Fauble and H. O. Denschlag, in Annual Report, Institute Für Kernchimie der Universitat, Mainz, Germany, 1975 (unpublished), p. 103.

[30] R. Naeumann, H. Folger, and H. O. Denschlag, J. Inorg. Nucl. Chem. 34, 1785 (1972).

[31] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (see Ref. 18), p. 255.