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Fission Product Mass Yield Curves and their Energy Dependence

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Abstract

It has been found that the mass yield curves for ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu neutron fission can be fitted, with an accuracy of better than 20%, by the superposition of two pairs of asymmetric gaussian curves and a single symmetric gaussian curve. The parameters of the fit have been investigated as a function of the nuclear temperature at the saddle point of the fissioning compound nucleus, and the widths and positions are found to vary linearly with this temperature. In addition, broad peaks are found in the weights of the gaussians, the weights being related to partial fission cross sections. This empirical analysis has been compared with the predictions of the Nix (1969) model of fission and deficiencies in the existing theory are discussed.

Introduction

In a recent review, Specht (1974) discussed the long-standing puzzle as to why the yield curves for spontaneous and neutron fission should be so asymmetric. No quantitative theory has yet explained this satisfactorily nor been able to predict the detailed shape of the yield curves. In an earlier model, Nix (1969) argued that the shape of the fissioning nucleus can be described in terms of normal coordinates which oscillate with time. The probability distributions for the initial position and momentum coordinates are gaussian in shape for such oscillators. Nix assumed that the transformation equations which take the nucleus to the saddle point are linear, and therefore the resultant distributions for the masses and energies at infinity must also be gaussian in shape. However, he neglected the well-known fact that for such nuclei (Specht 1974) the scission point takes place at a much later time than the saddle point, and in such a time the mass distributions between the two halves of the nucleus could change appreciably. Nix was well aware of these deficiencies in his model; furthermore, he only obtained symmetric fission with the model. It seems likely, therefore, that an empirical investigation of the yield curves could throw some light on the details of what a comprehensive theory should predict in relation to the magnitude of parameters and their energy dependence. Such information is of value in reactor physics design studies, in which the energy dependence of yield curves is usually neglected, and for which no suitable information is available for neutron energies between 2 and 14 MeV, over which range the curves change appreciably.

Delayed radiochemically determined yield curves for neutron fission of ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{239}Pu were fitted by E. A. C. Crouch (personal communication) who assumed that the prompt fragment yield curve consisted of a superposition of two gaussians and that their counterparts reflected about half the fissioning mass. He then fitted the delayed yield curves by generating a set of neutron emission probabilities from a Poisson distribution and determined the prompt yield gaussian parameters. Although he obtained an energy dependence for these parameters he did not investigate the laws involved. Furthermore, he assumed that the symmetric contribution is always flat.

Mass Yield Curve

Following the assumptions of Crouch (personal communication) and the conclusions of the Nix (1969) theory, one can see by inspection that the delayed yields evaluated by Flynn and Glendenin (1970) and Meek and Rider (1974) should fit reasonably well to five gaussian curves. Accordingly we put

$$Y(A) = 100 \sum_{i=1}^{5} W_i\{(2\pi)^{\frac{1}{2}}\sigma_i\}^{-1} \exp\{-(A-A_i)^2/2\sigma_i^2\}, \qquad (1)$$

where $Y(A)$ is the mass yield of a given chain of nuclides with mass number A, σ_i^2 the variance of the ith gaussian, A_i the peak of the ith gaussian and W_i the weight of the ith gaussian. Each of these quantities is a function of the nuclear excitation energy E. Integrating equation (1) over A, we find the condition on the weights

$$W_1 + \tfrac{1}{2} \sum_{i=2}^{5} W_i = 1, \qquad (2)$$

where the first gaussian is chosen to be symmetric about the symmetric delayed fission mass

$$A_1 = \tfrac{1}{2}(A_{\mathrm{f}} - \bar{\nu}), \qquad (3)$$

with A_{f} the mass of the nucleus undergoing fission and $\bar{\nu}$ the average number of neutrons released per fission.

We now postulate that neutron emission from the prompt fragments does not have much effect upon the symmetry of the mass yield curve and we put

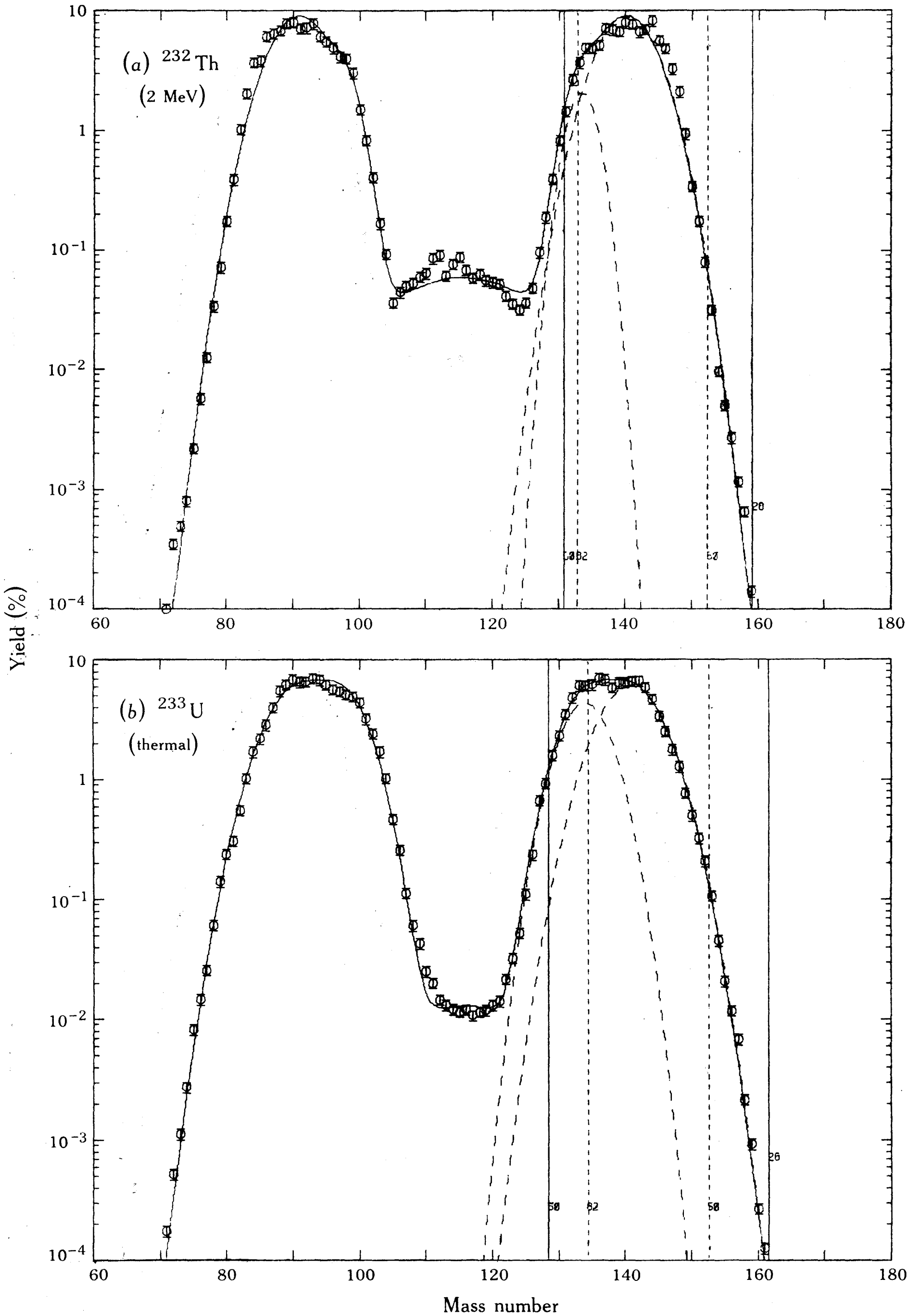
$$\left.\begin{array}{lll} A_4 = 2A_1 - A_2, & \sigma_4 = \sigma_2, & W_4 = W_2, \\ A_3 = 2A_1 - A_5, & \sigma_3 = \sigma_5, & W_3 = W_5, \end{array}\right\} \qquad (4)$$

so that the condition (2) becomes

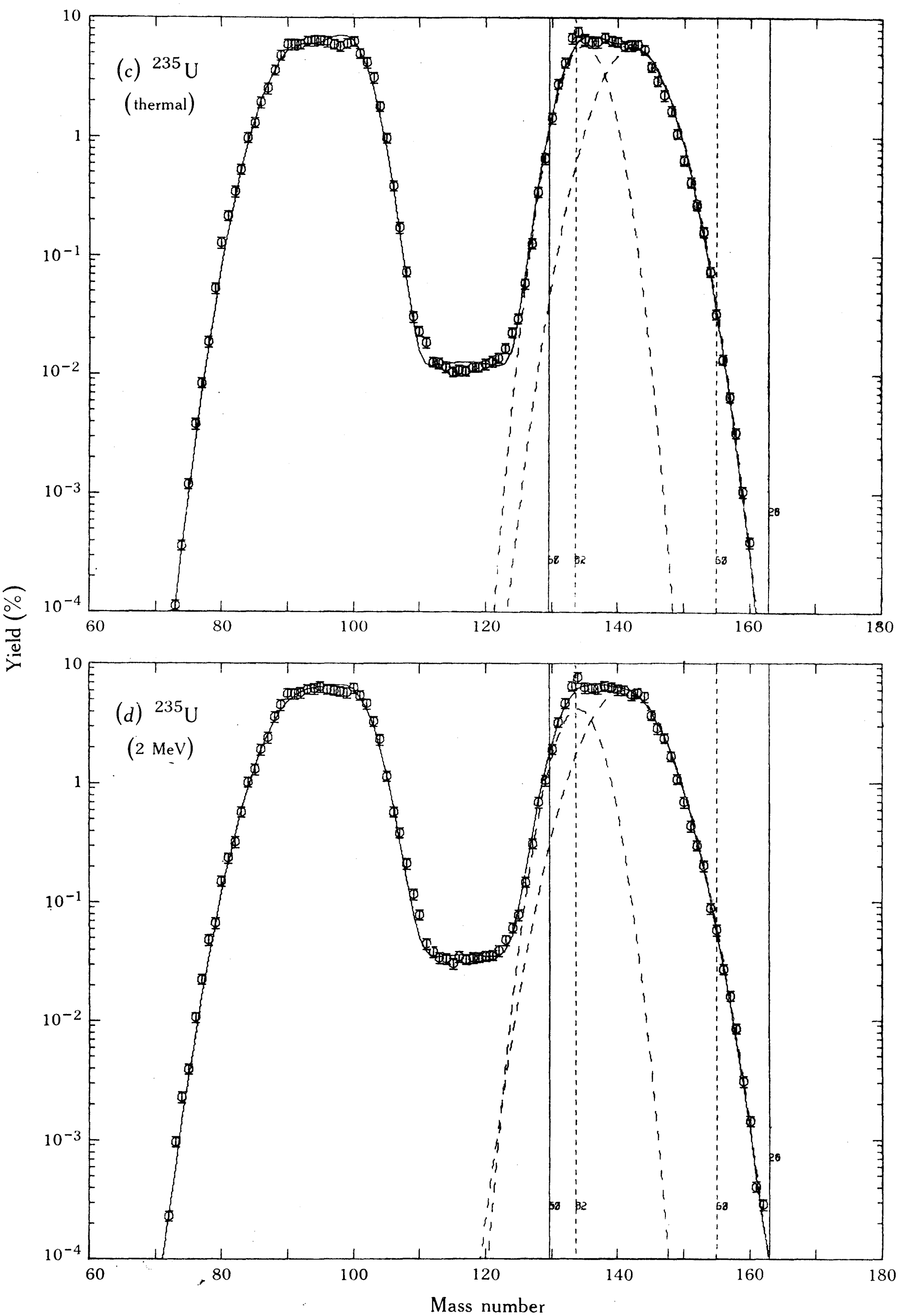
$$\sum_{i=1}^{3} W_i = 1. \qquad (5)$$

This yields eight parameters, which have in excess of 100 points and vary over five orders of magnitude, to be fitted to mass yield curves.

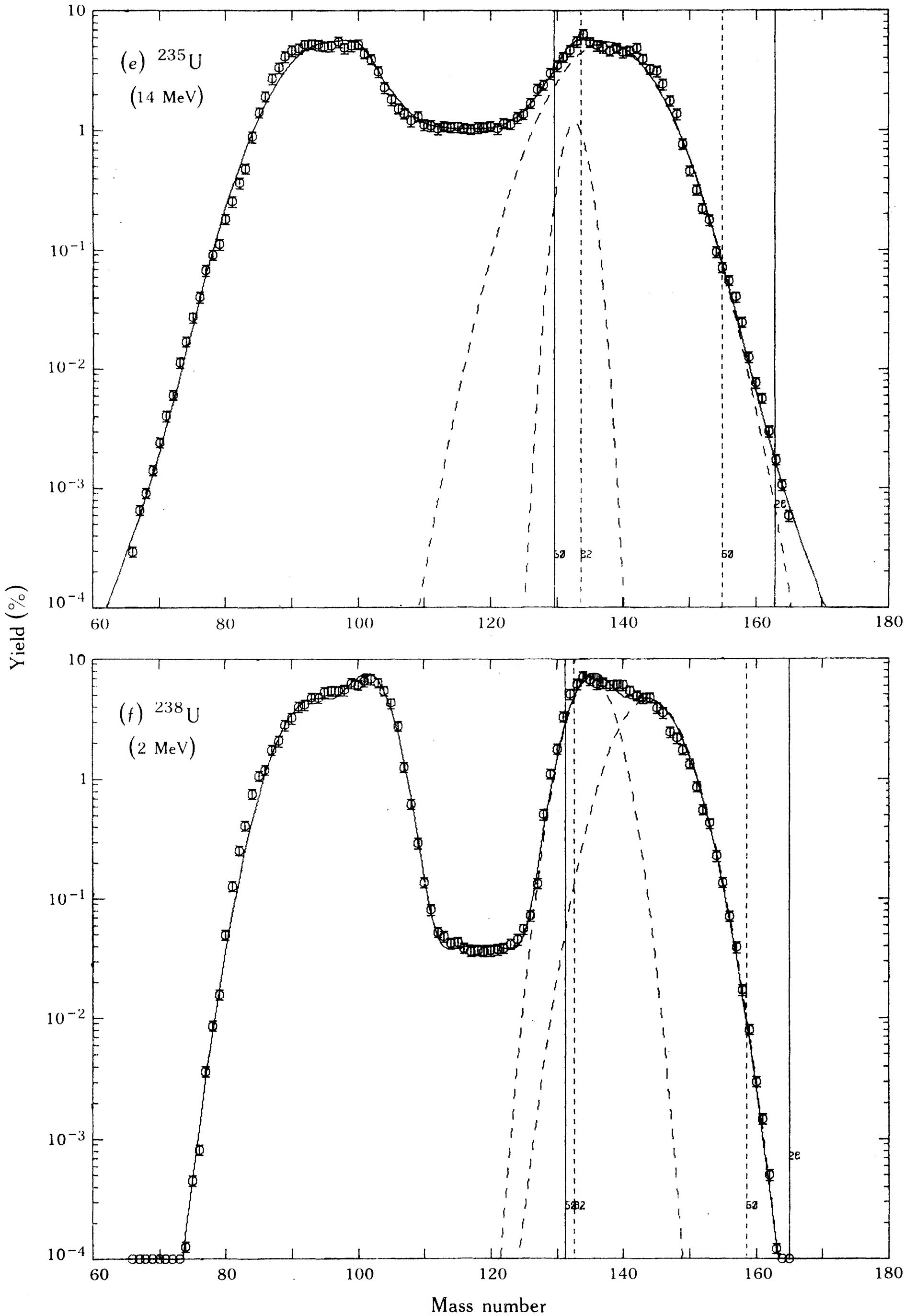
Figs 1*a*–1*i* (pp. 127–31). Fits to mass yield curves from neutron fission:
(*a*) 2 MeV neutrons on ^{232}Th, (*b*) thermal neutrons on ^{233}U, (*c*) thermal neutrons on ^{235}U, (*d*) 2 MeV neutrons on ^{235}U, (*e*) 14 MeV neutrons on ^{235}U, (*f*) 2 MeV neutrons on ^{238}U, (*g*) 14 MeV neutrons on ^{238}U, (*h*) thermal neutrons on ^{239}Pu, (*i*) 2 MeV neutrons on ^{239}Pu.



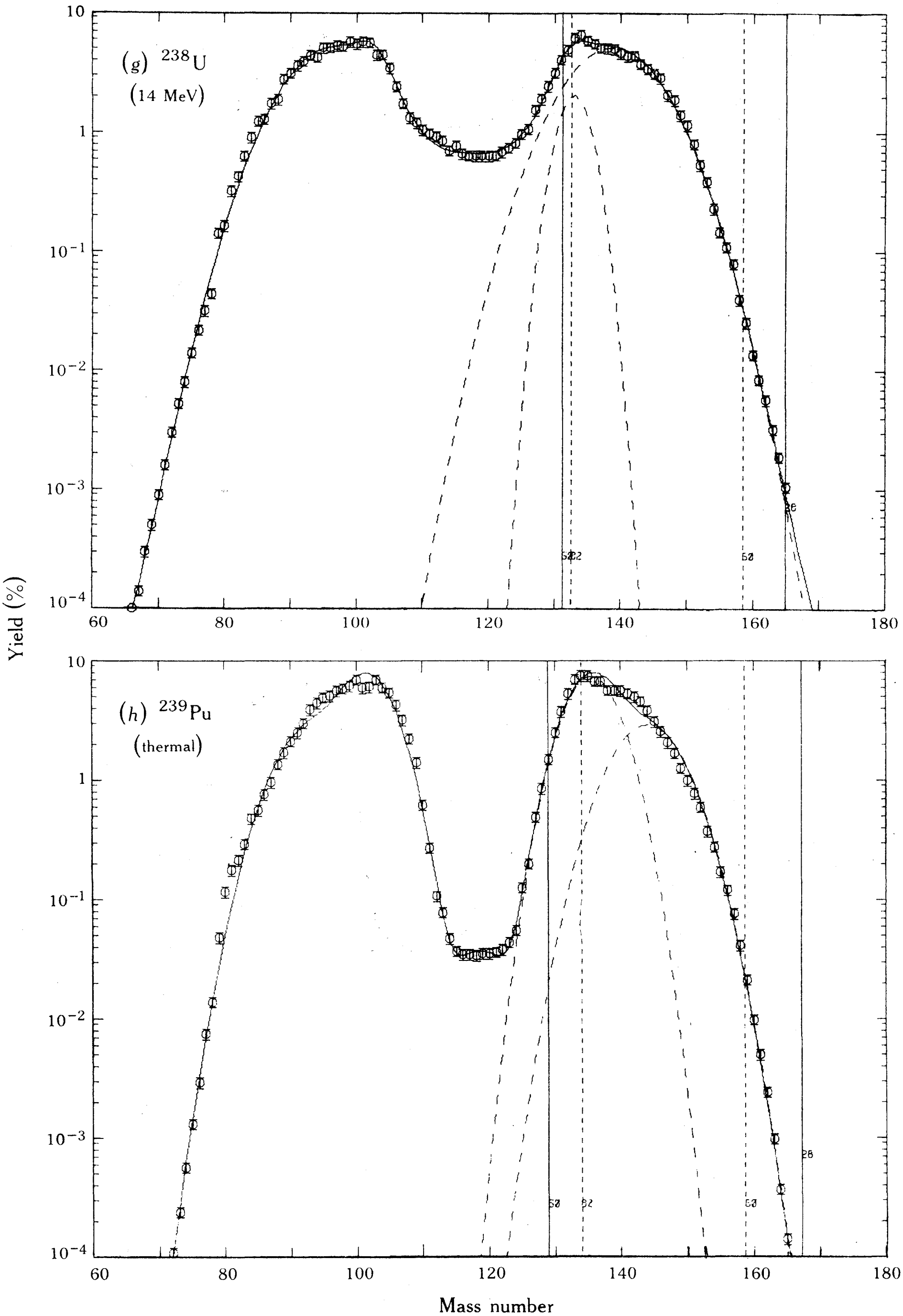
Figs 1a and 1b



Figs 1*c* and 1*d*



Figs 1*e* and 1*f*



Figs 1*g* and 1*h*

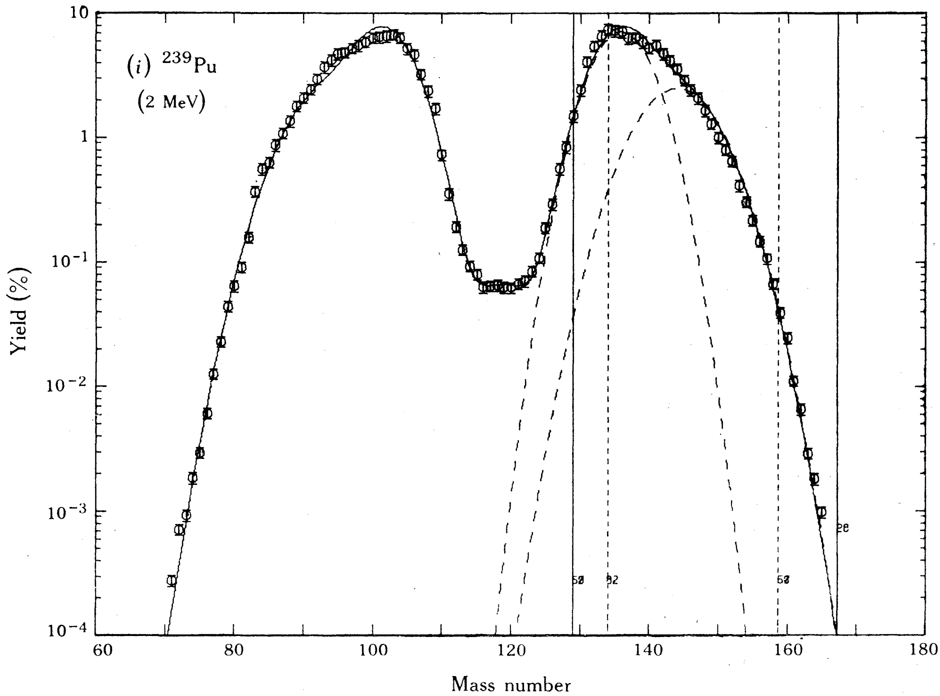


Fig. 1*i*

Using a least-squares analysis, the following fits were carried out:

^{232}Th. Neutron fission at incident energies of 2 and 14 MeV.

^{233}U. Neutron fission at incident energies of thermal and 2 and 14·8 MeV.

^{235}U. Neutron fission at incident energies of thermal and 2 and 14 MeV.

^{238}U. Neutron fission at incident energies of 2 and 14 MeV.

^{239}Pu. Spontaneous fission of ^{240}Pu; neutron fission at energies of thermal and 2 and 14 MeV.

The 14·8 MeV ^{233}U and 14 MeV ^{239}Pu data are very poor, with only a limited number of points. The fits where at least 100 points were measured are shown in Figs 1*a*–1*i*. The r.m.s. deviation from the fits nowhere exceeded 20% and improved as the excitation energy became greater. This deviation came almost entirely from fine structure which is in part understood (Musgrove *et al.* 1973). Let us now consider the energy dependence of the parameters obtained from the fits.

Energy Dependence of Gaussian Variances

Following Lang and Le Couteur (1954) and Gilbert and Cameron (1965), the nuclear temperature Θ of the fissioning nucleus at the saddle point, where the fission barrier of energy E_f has to be overcome, is related to the excitation energy E by the approximate equation

$$\Theta a^{\frac{1}{2}} \approx (E-E_f)^{\frac{1}{2}}, \qquad (6)$$

Table 1. Fits to standard deviations

Energy (MeV)	σ_1 (data)	σ_1 (fit)	σ_2 (data)	σ_2 (fit)	σ_3 (data)	σ_3 (fit)
^{232}Th ($E_f = 6\cdot15$ MeV)						
6·8	11·4±1·1	11·4	3·32±0·4	3·32	2·80±0·07	2·80
18·8	12·66±0·06	12·66	4·53	4·53	2·2±0·3	2·2
$\sigma_i(0)$:	11·1±1·4		2·38±0·06		3·0±0·1	
Slope:	0·4±0·4		0·49±0·03		−0·22±0·12	
^{233}U ($E_f = 5\cdot08$ MeV)						
6·8	9·9±0·3	10·1	4·28±0·03	4·24	3·33±0·07	3·32
8·8	13·9±0·1	13·9	4·42±0·02	4·44	1·5±0·1	1·51
21·6[A]	10±5	27	6·1±0·3	5·1	8±4	5
$\sigma_i(0)$:	2·2±1·0		3·81±0·09		7·2±0·3	
Slope:	6·1±0·5		0·33±0·05		−2·0±0·2	
^{235}U ($E_f = 6\cdot10$ MeV)						
6·5	10·68±0·03	10·66	4·03±0·02	4·06	2·90±0·05	2·93
8·5	11·01±0·06	11·10	4·64±0·03	4·55	2·97±0·09	2·83
20·5	12·60±0·04	12·60	6·03±0·04	6·06	1·73±0·42	2·5
$\sigma_i(0)$:	9·86±0·05		3·24±0·05		3·1±0·2	
Slope:	0·70±0·02		0·72±0·02		−0·16±0·11	
^{238}U ($E_f = 6\cdot12$ MeV)						
6·8	9·88±0·3	9·88	4·22±0·02	4·22	2·91±0·04	2·91
18·8	11·98±0·08	11·98	6·21±0·02	6·21	2·2±0·3	2·2
$\sigma_i(0)$:	0·04±0·05		3·43±0·04		3·2±0·1	
Slope:	0·81±0·03		0·77±0·02		−0·25±0·11	
^{239}Pu ($E_f = 4\cdot80$ MeV)						
0	2·2±1·0	2·2	3·9±0·3	3·8	1·9±0·2	2·4
6·5	4·5±0·7	4·5	4·74±0·03	4·73	3·59±0·06	3·50
8·5	5·5±1·1	5·5	5·16±0·04	5·17	3·84±0·08	3·97
20·5	8·8±4·4	9·1	11·8±2·0	6·6	5±1	5·6
$\sigma_i(0)$:	2·2±0·2		3·8±0·1		2·4±0·1	
Slope:	1·7±0·4		0·70±0·07		0·80±0·1	

[A] The data for 21·6 MeV were poor.

where a is the level density parameter of Gilbert and Cameron, which is given by

$$a = A_f 0\cdot00917\, S + 0\cdot142, \tag{7a}$$

with

$$S = S(Z) + S(N) \qquad (Z+N = A_f); \tag{7b}$$

Gilbert and Cameron provided tables of $S(Z)$ and $S(N)$. We used the recent measurements of the fission barriers for heavy nuclides of Back *et al.* (1973*a*, 1973*b*) for the values of E_f, the value taken being the largest of the two barriers in the now established double humped fission barrier theory.

We further postulate that the variances of the gaussians change slowly with nuclear temperature and so the Taylor expansion

$$\sigma_i(E) \approx \sigma_i(E_f) + \left(\frac{\mathrm{d}\sigma_i(E_f)}{\mathrm{d}\Theta}\right)\Theta + O(\Theta^2) \tag{8}$$

can be truncated at the second term. If this assumption is justified, the standard deviations of the gaussians should vary linearly with $(E-E_f)^{\frac{1}{2}}$.

Table 1 gives the results of linear least squares analysis of the three variances for the five nuclides investigated. The linear least squares error analyses of the slope and intercept of the straight line fit for each of ^{233}U and ^{235}U, where there are three points, and ^{239}Pu, where there are four points, give an indication of the acceptability of the law (equation 8). In each case, the errors were small and the fits can be considered to be good, especially in the case of ^{235}U.

The only theory that so far predicts the energy dependence of these variances is that of Nix (1969). In the quantized version of his theory each mode of oscillation in the liquid drop model contributes an amount

$$\sigma_{i\lambda}^2 = \sigma_{i\lambda}^2(0)\coth(h\omega_\lambda/2\Theta), \tag{9}$$

where ω_λ is the characteristic frequency of the λth mode, in the usual liquid drop model. For small Θ or large ω_λ

$$\sigma_{i\lambda}^2 \approx \sigma_{i\lambda}^2(0), \tag{10a}$$

while for large Θ or small ω_λ

$$\sigma_{i\lambda}^2 \approx \sigma_{i\lambda}^2(0)\,(2\Theta/h\omega_\lambda). \tag{10b}$$

We see, for low energy oscillators, that equation (10b) predicts a linear dependence upon Θ for $\sigma_{i\lambda}^2$. Thus if

$$\sigma_i^2(E) = \sum_{\lambda=2}^{\infty} \sigma_{i\lambda}^2(E) = \sigma_{i2}^2(E) + \sum_{\lambda=3}^{\infty} \sigma_{i\lambda}^2(E),$$

then

$$\sigma_i(E) = \left(\sigma_{i2}^2(E) + \sum_{\lambda=3}^{\infty} \sigma_{i\lambda}^2(E)\right)^{\frac{1}{2}}$$

$$\approx \left(\sum_{\lambda=3}^{\infty} \sigma_{i\lambda}^2(E_f)\right)^{\frac{1}{2}} + \sigma_{i2}^2(E_f)\left(\sum_{\lambda=3}^{\infty} \sigma_{i\lambda}^2(E_f)\right)^{-\frac{1}{2}} \frac{\Theta}{h\omega_2} + \ldots, \tag{11}$$

where the remaining terms in the Taylor expansion are small. One can see that under conditions where ω_2 is small, while ω_3 and the other frequencies are large, the standard deviations will be approximately linear with Θ.

Results for calculated values of a direct fit to equation (8) are shown in Table 2 and the frequencies obtained are compared with the predictions of the liquid drop model and the Nix (1969) model. One can see that the frequencies obtained from the experiments are of the same order as the predictions of the Nix theory, but one cannot specify which mode contributes most to each gaussian, as one would expect the frequencies at scission to have changed appreciably from the frequencies at the saddle point. Note also that the rule (9) predicts a monotonically increasing function of E for $\sigma_3(E)$, while the fits in Table 1 show clearly that it is a monotonically decreasing function of E. Clearly, the liquid drop theories are inadequate in explaining either the asymmetry or the energy dependence.

The third peak has one interesting feature. For each mass number of a product, Wahl *et al.* (1962) have shown that there is a dispersion in charge Z which has a gaussian distribution about a mean value $Z_p(A)$. From these values of Z_p we find that the position of the small third gaussian occurs for $A-Z_p = 82$, a magic number in the shell model. Thus, a shell structure in the daughter nuclides contributes appreciably to the shape of the inside slopes of the mass yield curves. No such correlation exists for the position of the large outside gaussians.

Table 2. Fits to density of states of oscillators

The frequencies ω_λ obtained from the fits are compared with the predictions of the liquid drop and Nix (1969) models

Nuclide	x^A	Calculated values $\sigma_1^2(0)$, $\sigma_2^2(0)$	Calculated values $\hbar\omega_1$ (MeV), $\hbar\omega_2$ (MeV)	Liquid drop $\hbar\omega_2$ (MeV), $\hbar\omega_3$ (MeV)	Nix $\hbar\omega_2$ (MeV), $\hbar\omega_3$ (MeV)
^{232}Th	0·7527	123, 8·5	1·01, 0·72	1·2, 2·8	0·60, 1·50
^{233}U	0·7739	111, 17·42	1·72, 0·928	1·2, 2·8	0·65, 1·55
^{235}U	0·7717	114·8, 15·32	1·22, 0·581	1·2, 2·8	0·65, 1·55
^{238}U	0·7686	81·7, 11·76	1·00, 0·558	1·2, 2·8	0·63, 1·58
^{239}Pu	0·7897	5, 20·2	0·1, 0·645	1·2, 2·8	0·70, 1·60

[A] x is Nix's fissionability parameter.

Energy Dependence of Positions

Owing to the symmetry of the mass yield curve about $\frac{1}{2}(A_f - \bar{\nu})$, we need consider only A_1, A_2 and A_3 in determining the energy dependence of the positions of the peaks. From equation (3) and the relation (Templin 1961)

$$\bar{\nu}(E) = \bar{\nu}(0) + BE, \tag{12}$$

where B is a constant, we find that

$$A_1(E) = \tfrac{1}{2}\big(A_f - \bar{\nu}(0)\big) - \tfrac{1}{2}BE \tag{13}$$

gives the energy dependence of the centre. This is indeed observed to be the case for all examples, as shown in Table 3. The values of $\bar{\nu}(0)$ obtained by us are within the experimental errors tabulated by Hyde (1971). The other two heavy peaks show a weaker dependence upon energy and so, once again, we surmise that the Taylor expansions

$$A_2(E) = A_2(E_f) + a^{-\frac{1}{2}}\big(\mathrm{d}A_2(E_f)/\mathrm{d}\Theta\big)\big(E - E_f\big)^{\frac{1}{2}} + \dots, \tag{14a}$$

$$A_3(E) = A_3(E_f) + a^{-\frac{1}{2}}\big(\mathrm{d}A_3(E_f)/\mathrm{d}\Theta\big)\big(E - E_f\big)^{\frac{1}{2}} + \dots \tag{14b}$$

might suffice. This conjecture proves to be correct in all cases, as shown in Table 3.

The Nix (1969) theory leads only to symmetric fission, so it makes no statement about the possible positions of asymmetric peaks.

Table 3. Energy dependence of positions of peaks

Energy (MeV)	Centre (data)	Centre (fit)	A_2 (data)	A_2 (fit)	A_3 (data)	A_3 (fit)
^{232}Th						
6·8	115·7±0·1	115·7	142·7±0·2	142·7	135·8±0·2	135·8
18·8	114·7±0·1	114·7	139·5±0·3	139·5	134·4±0·3	134·4
$A_i(E_f)$:	116·3±0·5 (E = 0)		143·7±0·3		136·3±0·3	
Slope:	−0·086±0·004		−1·18±0·13		−0·53±0·15	
^{233}U						
6·8	115·8±0·2	115·9	140·7±0·2	140·6	133·8±0·3	133·8
8·8	116·0±0·2	115·8	139·6±0·1	139·6	132·9±0·2	132·9
21·6	115·1±0·7	115·3	136·8±0·9	136·1	135 ±10	130
$A_i(E_f)$:	116·2±0·1 (E = 0)		142·8±0·5		135·9±0·9	
Slope:	−0·041±0·005		−1·64±0·25		−1·6±0·5	
^{235}U						
6·5	117·0±0·1	117·0	142·1±0·1	142·1	134·8±0·2	134·7
8·5	116·9±0·1	116·9	140·8±0·2	140·9	133·9±0·2	134·1
20·5	116·3±0·1	116·3	137·2±0·2	137·2	132·5±0·4	132·4
$A_i(E_f)$:	117·3±0·1 (E = 0)		144·1±0·2		135·7±0·3	
Slope:	−0·051±0·03		−1·77±0·08		−0·86±0·17	
^{238}U						
6·8	118·4±0·1	118·4	143·7±0·1	143·7	135·1±0·1	135·1
18·8	117·7±0·1	117·7	138·8±0·2	138·8	132·9±0·3	133·0
$A_i(E_f)$:	118·8±0·0 (E = 0)		145·6±0·2		136·0±0·2	
Slope:	−0·061±0·003		−1·89±0·10		−0·84±0·13	
^{239}Pu						
0	119·1±0·2	119·0	140·6±2·1	141·7	133·6±0·7	134·3
6·5	118·7±0·1	118·8	143·9±0·2	143·5	135·5±0·2	135·4
8·5	118·8±0·1	118·7	143·9±0·3	144·4	135·9±0·2	135·9
20·5	117·9±0·4	118·2	137·5±0·7	147·2	136·0±0·9	137·5
$A_i(E_f)$:	119·0±0·1 (E = 0)		141·7±0·5		134·3±0·3	
Slope:	−0·040±0·006		1·4±0·3		0·80±0·18	

Energy Dependence of Weights

The weights defined in equation (1) are related to the cross section for the formation of fragments in each mode, i.e.

$$\sigma_{\mathrm{f}i}(E) = W_i(E)\,\sigma_{\mathrm{f}}(E)\,, \tag{15}$$

where $\sigma_{\mathrm{f}}(E)$ is the fission cross section for the formation of all modes. Since the largest gaussians are evidently related to the collective modes of oscillations, we parameterized the three weights in terms of production phase shifts θ_1 and θ_2 such that

$$W_1 = \sin^2\theta_1\,, \qquad W_2 = \sin^2\theta_1\cos^2\theta_2\,, \qquad W_3 = \sin^2\theta_1\sin^2\theta_1\,. \tag{16}$$

We also tried parameterizing the production phase shifts by means of the relations

$$\tan\theta_1 = 2(E-E_1)/\Gamma_1\,, \qquad \tan\theta_2 = 2(E-E_2)/\Gamma_2\,, \tag{17}$$

where E_1 and E_2 are resonance energies and Γ_1 and Γ_2 are total widths. The values for these parameters are listed in Table 4. Plots of $\tan\theta_1$ and $\tan\theta_2$ gave straight lines in all cases, with a value for E_2 very close to 14 MeV and the second chance fission threshold. It appears likely that this peak is related to the (n, nf) fission cross section threshold, since it occurs just before the large enhancement in the fission cross section due to this process. In ^{239}Pu, a plot of $\cos\theta_2$ rather than the tangents gave a straight line, indicating that the character of the resonance had changed for that nuclide to a maximum in the inner peak.

Table 4. Resonance parameters for gaussian weights

Nuclide	E_1 (MeV)	E_2 (MeV)	Γ_1 (MeV)	Γ_2 (MeV)
^{232}Th	$4\cdot2\pm0\cdot2$	$14\cdot4\pm1\cdot3$	$59\cdot7\pm1\cdot1$	$20\cdot3\pm1\cdot4$
^{233}U	$5\cdot7\pm0\cdot2$	$14\cdot6\pm2\cdot1$	$53\cdot9\pm1\cdot2$	$29\cdot8\pm3\cdot6$
^{235}U	$4\cdot6\pm0\cdot2$	$17\cdot3\pm1\cdot0$	$95\cdot9\pm2\cdot0$	$25\cdot4\pm1\cdot2$
^{238}U	$3\cdot4\pm0\cdot2$	$15\cdot6\pm0\cdot7$	$94\cdot3\pm2\cdot9$	$17\cdot0\pm0\cdot6$
^{239}Pu	$1\cdot5\pm2\cdot6$	$21\cdot5\pm7\cdot0$	$2\cdot5\pm8\cdot0$	$39\cdot7\pm12\cdot2$

As a test of these laws, the eight yields quoted by Flynn and Glendenin (1970) are compared in the following tabulation with the predictions for ^{235}U bombarded by 8 MeV neutrons; the agreement is seen to be satisfactory.

Mass number	99	144	147	149	153	154	159	161
Measured yield (%)	5·4	3·6	2·05	1·25	0·185	0·035	0·0063	0·002
Calculated yield (%)	5·3	3·7	1·93	1·17	0·192	0·051	0·0058	0·0021

Conclusions

In this work, empirical laws for the energy dependence of gaussian parameters for fission product mass yield curves have been tested successfully. A comparison of the predictions of models for the fission process has shown that the Nix (1969) theory correctly forecasts the Gaussian shape of each mode and the variation of some of the variances. However, this theory does not predict the energy dependence of the peak positions or the gaussian weights, but nor does any subsequent theory.

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