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MASTER

Fission Mass Yield Studies*

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1. Introduction

Fission yields from proton-induced fission of Th^{232} , photon-induced fission of U^{238} , alpha-particle induced fission of U^{238} , and deuteron-induced fission of natural uranium have previously been tested [1] for consistency with the two-mode-of-fission hypothesis [2]. Deviations were not statistically significant except for the yields of a few mass numbers at the highest excitation energies for photon-induced fission of U^{238} . Schmitt and Sugarman [3] have proposed that one mode of fission is predominantly symmetric and the other predominantly asymmetric with the symmetric mode being more prevalent at higher excitation energies. Ford [1] found no compelling reason in the data then available to restrict the mode dominant at high energies to a single-hump yield curve. However, more recently Britt et. al., [4] from considerations of fission fragment kinetic energies and velocities, have found support for the two-mode-of-fission hypothesis and have derived mass yield curves that are the single- and double-hump curves envisioned by Schmitt and Sugarman.

Recent mass yield data obtained by Levy et. al. [5] for the U^{236} compound nucleus generally agreed with the two-mode-of-fission hypothesis. However, it was disturbing that the yields from thermal-neutron-induced fission of U^{235} did not conform with these data within the framework of the two-mode-of-fission hypothesis. Continuation of this work [6] showed deviations at higher excitation energies for some mass numbers. These deviations gave rise to a second concern about mass yield tests of the two-mode-of-fission hypothesis, namely, that fissions following neutron emission perturb the results; the fission yield curves for the two

hypothetical modes of fission cannot be expected to be exactly the same for fission of different compound nuclei. However, yields from the original compound nucleus, uncontaminated by fissions of the others, may still be expected to conform to the hypothesis.

The present study concerns recent data and an analysis method obtained by Ford and Leachman [7] that are relevant to these two significant deviations from the two-mode-of-fission hypothesis.

For an initial U^{236} compound nucleus the emission of one or two neutrons followed by fission is possible at excitation energies of approximately 13.5 MeV and higher or 20.5 MeV and higher. Fission before the emission of a neutron is called first-chance fission, fission after the emission of one neutron is called second-chance fission, etc. The competition between neutron emission and fission is called the fission-chance split.

Our data are better adapted to consider the effect of the fission-chance split than the data of Hicks et. al. [6] in that the excitation energy of the original U^{236} compound nucleus is better known.

Our data analysis method is discussed in Section 2; it consists of removing the effect of the fission-chance split to obtain first-chance yields from measured yields. Also averages and standard deviations of excitation energy for second- and third-chance fission and probabilities for first-, second-, and third-chance fission, which are byproducts of the method, are given in Section 2. The test of the two-mode-of-fission hypothesis for the U^{236} initial compound nucleus is considered in Section 3 for both observed and first-chance fission yields. The test of the two-mode-of-fission hypothesis for observed mass yields from neutron-induced fission of Th^{232} and U^{238} are also considered in Section 3. The results of the test are discussed in Section 4.

2. Mass Yields for First-Chance Fission

In the following Y 's are fission mass yields and σ 's are fission cross sections. Superscripts (1), (2), and (3) refer to first-, second-, and third-chance fission of U^{236} , U^{235} , and U^{234} , respectively. The

absence of a superscript denotes a combination of available stages.

First-chance yields were estimated from observed yields as follows:

(1) In the region below 13.5-MeV excitation energy of exclusively first-chance fission of the U^{236} compound nucleus, $Y^{(1)}$ is the observed yield Y .

(2) In the region below 20.5-MeV initial excitation energy of first- and second-chance fission, the first chance yield is given by

$$Y^{(1)} = (Y\sigma - \langle Y^{(2)} \rangle_{av.} \sigma^{(2)}) / \sigma^{(1)}. \quad (1)$$

(3) Above 20.5-MeV initial excitation energy where first-, second-, and third-chance fission are possible, the first-chance yield is given by

$$Y^{(1)} = (Y\sigma - \langle Y^{(2)} \rangle_{av.} \sigma^{(2)} - \langle Y^{(3)} \rangle_{av.} \sigma^{(3)}) / \sigma^{(1)}. \quad (2)$$

In equations (1) and (2) the averages are over excitation energies below 13.5 MeV and 20.5 MeV, respectively. It was necessary to approximate $\langle Y^{(2)} \rangle_{av.}$ and $\langle Y^{(3)} \rangle_{av.}$ by $\langle Y^{(1)} \rangle_{av.}$ since $Y^{(1)}$ is the only mass yield known.

The probabilities of the various possible histories resulting from the U^{236} initial compound nucleus used in averaging $Y^{(1)}$ over the energies of second- and third-chance fission were obtained from Markov chain [8] theory which is applicable since the partial fission widths and partial neutron widths at any state of de-excitation are independent of the previous decay history. In our calculations the populations of 101 states of the system are given by a 101-dimensional vector. States 1 through 99 are nuclei in one of 99 equal excitation-energy intervals. The system is in state 100 if a nucleus in a previous stage has fissioned, and the system is in state 101 if both neutron emission and fission were energetically impossible for a nucleus in a previous stage. For the initial compound nucleus U^{236} at our fixed irradiation energies, this population vector $\underline{q}^{(1)} = (q_1^{(1)}, \dots, q_{101}^{(1)})$ is simply $\underline{q}^{(1)} = (1, 0, \dots, 0)$ but the corresponding population vectors $\underline{q}^{(2)}$ for U^{235} , $\underline{q}^{(3)}$ for U^{234} , and $\underline{q}^{(4)}$ for U^{233} are to be calculated. The probability

of a nucleus being in state j after the first neutron emission is $q_j^{(2)}$ and the conditioned probability $p_{jk}^{(2 \rightarrow 3)}$ is the probability of such a nucleus in state j contributing to the probability $q_k^{(3)}$ of state k by means of neutron emission (or fission, or neither). The conditional probabilities are matrix elements of transition matrices $P^{(1 \rightarrow 2)}$, $P^{(2 \rightarrow 3)}$, etc. In terms of these matrices

$$\begin{aligned} \underline{q}^{(2)} &= \underline{q}^{(1)} P^{(1 \rightarrow 2)} \\ \underline{q}^{(3)} &= \underline{q}^{(2)} P^{(2 \rightarrow 3)} = \underline{q}^{(1)} P^{(1 \rightarrow 2)} P^{(2 \rightarrow 3)} \\ \underline{q}^{(4)} &= \underline{q}^{(3)} P^{(3 \rightarrow 4)} = \underline{q}^{(1)} P^{(1 \rightarrow 2)} P^{(2 \rightarrow 3)} P^{(3 \rightarrow 4)}. \end{aligned}$$

The last equations result from the Chapman-Kolmogorov equation

$P^{(u \rightarrow w)} = P^{(u \rightarrow v)} P^{(v \rightarrow w)}$ for $u < v < w$. (Only the fission state 100 was needed in $\underline{q}^{(4)}$.) This detailed calculation of the decay chain was made at about 1-MeV intervals of initial U^{236} excitation energies over the span of energies under consideration. For each incident energy of the calculations the numbers $\underline{q}_1^{(2)} P_{1,100}^{(2 \rightarrow 3)}$ give the probability of second-chance fission for nuclei in the energy interval 1. There are similar expressions for third- and fourth-chance fission. The population vectors $\underline{q}^{(u)}$ are illustrated in Fig. 1 where probabilities have been converted to probability densities by dividing by the length of the energy interval of the state. Some of the transitions and conditional probabilities are also illustrated in Fig. 1. States 100 and 101 are not shown.

The conditional probabilities used in the transition matrices depend on the statistical formulation ($N = 1$ or 3 space coordinates for the phase spaces of the transition state or saddle point configuration for fission) and on the level density formula and its parameter. The Fermi-gas level density formula $\rho(E) = C_a \exp [2(aE)^{1/2}]$ and the constant-temperature level density formula $\rho(E) = C_T \exp (E/T)$ with the parameters $a_n = 23.4 \text{ MeV}^{-1}$ and $T_n = 0.827 \text{ MeV}$ respectively for neutron emission were used with both $N = 1$ and $N = 3$. The corresponding parameters a_f and T_f

for fission, the fission threshold for the U^{235} compound nucleus and the ratio K of C_a or C_T for neutron emission to C_a or C_T for fission were determined by fits to measured fission cross sections for 1- to 6-MeV neutron induced fission of U^{233} , [9], and U^{235} [10]. Optical model capture cross sections were used [11]. The combinations with the Fermi-gas level density formula for fission and the constant-temperature level density formula for neutron emission were ruled out on theoretical grounds [12]. The combinations with the constant-temperature level density formula for fission and the Fermi-gas level density formula for neutron emission were ruled out by extremely poor agreement between measured and calculated fission cross sections in the 1- to 6-MeV neutron-energy range.

First-chance yields for Ag^{113} , Pd^{112} , Ag^{111} , and Pd^{109} for our favored combination of statistical formulation, level density formula and parameter set is given as an example in Fig. 2. In this example (1) $a_n = 23.4 \text{ MeV}^{-1}$ and $a_f = 22.6 \text{ MeV}^{-1}$ in the Fermi-gas level density formula, $N = 3$, the fission threshold for the U^{235} compound nucleus is $B_f^{(2)} = 6.05 \text{ MeV}$ and $K = 10.5$. This and the other combinations of parameter sets that were used are given in Table I.

Although our primary concern is to calculate average yields to use in equations (1) and (2), energies were also averaged over the various possible histories resulting from the U^{236} initial compound nucleus for various initial excitation energies (at nearly 1 MeV intervals from 13.44 to 26.41 MeV) to give average energies for second-chance (U^{235}) and third-chance (U^{234}) fission. Similarly standard deviations were derived by averaging the square of energies over these probabilities. All of the combinations of statistical formulation, level density formula, and parameter sets listed above gave nearly the same average energies which are plotted in Fig. 3. The largest difference in average energy was 0.3 MeV for second-chance fission with an initial U^{236} excitation energy of 18.45 MeV. The standard deviations are also shown in Fig. 3. Extremes of the standard deviations (high for combination (3) and low for

combination (2) are shown for third-chance fission, otherwise they are nearly all the same for all of the combinations, (1) to (4) listed above.

The major difference in results for the different combinations of statistical formulations, level density formula, and parameter sets is in the fission-chance split. The probabilities for second- and third-chance fission for the various combinations are shown in Fig. 4. Further numerical details are available from the authors.

3. Test of the Two-Mode-of-Fission Hypothesis

It has been shown [1] that the two-mode-of-fission hypothesis implies that fission mass-yield curves (n-tuples of mass yields) lie in a two dimensional subspace of the vector space of n-tuples. This subspace inclusion also holds when the yields are not normalized so that their sum is 200%. A linear relation

$$x_{ji} = u_j a_i + w_j b_i, \quad (3)$$

where x_{ji} is the yield or mass number A_i in fission yield curve (or n-tuple of yields) j and u_j and w_j are two other yields from fission yield curve j , was derived from the subspace inclusion. With the added requirement that the fission mass-yield curves be normalized so that the sum of yields is 200% Levy et. al. [5] showed that the two-mode-of-fission hypothesis implies a linear relation

$$x_{ji} = c_i u_j + d_i \quad (4)$$

between two yields. By then relaxing the normalization requirement they arrive at an equation equivalent to (3) which they use [5,6] to test the two-mode-of-fission hypothesis. Our yields are purported to be normalized so we are able to use equation (4) in a graphical test of the two-mode-of-fission hypothesis. The graphs, which should be straight lines if the two-mode-of-fission hypothesis holds, are shown in Figs. 5 and 6. The points are labeled with the excitation energy to provide ready identification with entries in Table I of reference [7]. In every case the abscissa u_j is the yield of Ag^{113} . Both observed and first-chance yields are shown for the U^{236} initial compound nucleus. First chance yields for neutron

induced fission of U^{235} are sufficient to illustrate the situation above 19-MeV excitation energy; only these were calculated above 19-MeV excitation energy. First-chance yields above 13-MeV initial excitation energy where second- and third-chance fission are possible are not available for neutron induced fission of Th^{232} and U^{238} .

4. Discussion

None of the plots show any systematic or significant deviations from linearity except the plots of yields of Pd^{109} for the U^{236} initial compound nucleus.[†] For neutron induced fission of U^{238} the data for Ag^{111} and Pd^{112} exist in two clusters that may be connected by a line and do not provide a test of the two-mode-of-fission hypothesis. For neutron induced fission the Th^{232} and Pd^{109} data are better distributed and show no trend deviating from linearity such as is shown by Pd^{109} data for the U^{236} initial compound nucleus.

Our points for thermal neutron induced fission of U^{235} agree well with the two-mode-of-fission hypothesis in contrast with the results of Levy et. al.* [5].

For the U^{236} initial compound nucleus Hicks et. al. [6] found significant deviations from linearity for Zn^{72} , Pd^{112} , Cd^{115m} , Cs^{136} , Sm^{153} , and Tb^{161} . Of these we have no data for Zn^{72} , Sm^{153} , and Tb^{161} . Our data for Cs^{136} and Cd^{115m} are rather sparse and first-chance yields in the energy region where second- and third-chance fission are possible were not calculated. Our data for Pd^{112} show no significant deviations from linearity; our highest energies are not as high as those of Hicks et. al. [6]. Our first-chance yields for Pd^{109} do not so clearly deviate from linearity as the observed yields although some of the improvement

[†] Pd^{109} occurs on the Britt et. al. [4] curve for the symmetric mode at a point down from the maximum by approximately 0.75.

*A plot, using equation (3), of the data for neutron induced fission of Pu^{239} in the near-thermal region being presented in this session by Cowan, Bayhurst, Prestwood, Gilmore, and Knobeloch shows no significant deviation from the two-mode-of-fission hypothesis for thermal neutrons.

could be caused by the larger spreads and errors induced by correcting for second- and third-chance yields. The errors shown are the result of propagation of error from the errors in the observed yields assuming that the lower energy first-chance yields occurring in equations (1) and (2) are completely known. The replacement of $Y^{(2)}$ and $Y^{(3)}$ in the averages by $Y^{(1)}$ could not be taken account of in the error calculation. The remaining possible deviation of Pd^{109} first-chance yields from linearity for the U^{236} initial compound nucleus may possibly be attributed to this, or to errors in normalization. Normalization errors would be expected to affect the other plots too. Hicks et. al. [6] use the three-yield equation (3) that does not require normalization. Correction for second- and third-chance fission is feasible with our computer codes only when the yields are normalized. First-chance yields for combinations (2), (3), and (4) in Table I were slightly better than for combination (1) but not sufficiently better to warrant any one of them being chosen over combination (1).

In addition to the fission-chance split, the following possible causes for deviations at higher excitation energies of yields from the linear relations predicted by the two-mode-of-fission hypothesis have been considered [6]: (1) changes in shape of the mass yield curves for the hypothetical modes of fission, (2) neutron emission from the fission fragments, (3) angular momentum effects, (4) change in charge distribution. Of these (2) and (4) seem to be mechanisms for (1) and seem to be another way to say that the hypothesis does not hold. Hicks et. al. [6] noted that all of the deviations except for Tb^{161} are qualitatively in the direction to be expected from neutron emission either after fission or before fission (fission-chance split). They also noted that for any of the charge-distribution hypotheses considered (minimum potential energy, equal chain length, and constant charge to mass ratio) the relative deviations of Pd^{109} and Pd^{112} are in the wrong direction to be attributed to changes in charge distribution. Hicks et. al. [6] assumed that angular momentum only affects the fission-chance split and Ford and

Leachman [7] found reasons not to invoke any angular-momentum effect other than the effect on fission-chance split in their considerations of the effect of angular momentum on valley yields at excitation energies where third-chance fission is possible.

Table I. Parameter sets used. The parameters from fits were derived in reference [7] by fits of calculated to observed fission cross sections for neutron-induced fission of U^{233} and U^{235} in the 1- to 6-MeV neutron-energy region of exclusively first-chance fission.

Combination number and plotting symbol	Space coordinates, assumed N	Level parameter assumed ^a	Parameters from fits		
			Level parameter ^a	U^{235} fission threshold (MeV)	Level density coefficient K
1 —————	3	$a_n = 23.4$	$a_f = 22.6$	6.05	10.5
2 — — — — —	1	$a_n = 23.4$	$a_f = 27.0$	6.10	0.747
3 — — — — —	1	$T_n = 0.827$	$T_f = 0.80$	6.25	5.33
4 — — — — —	3	$T_n = 0.827$	$T_f = 0.80$	6.05	6.93

^a a_n and a_f are in units of MeV^{-1} ; T_n and T_f are in MeV.

Fig. 1. Population vectors $q^{(2)}$, $q^{(3)}$, and $q^{(4)}$ for an initial U^{236} compound nucleus excitation energy of 23.43 MeV. Probabilities have been converted to probability densities by dividing by the common length of the 99 energy intervals. States 100 (fission) and 101 (neither fission nor neutron emission) are not shown.

Fig. 2. First-chance fission yields from fission of the U^{236} compound nucleus. These yields were derived from observed yields by the method of Section 2 using combination (1) of statistical formulation and parameter sets.

Fig. 3. Average energies and standard deviations. The abscissa is the excitation energy of the original U^{236} compound nucleus. All of the combinations of statistical formulation and parameter sets of Table I gave nearly the same average energy which is shown here. The standard deviations were nearly all the same except the extremes shown for third-chance fission. Combination (3) is high and combination (2) is low.

Fig. 4. Probabilities of first-, second-, and third-chance fission. The plotting symbols are identified in Table I.

Fig. 5. Plot, using equation (4), to test the two-mode-of-fission hypothesis for the initial U^{236} compound nucleus for both observed and first-chance yields. The numbers are excitation energies in MeV of the initial U^{236} compound nucleus. Thermal neutron-induced fission of U^{236} has an initial excitation energy of 6.5 MeV.

Fig. 6. Plots, using equation (4), to test the two-mode-of-fission hypothesis for neutron-induced fission of Th^{232} and U^{238} for observed yields.

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