

ARTICLE TEMPLATE

Energy Dependent Calculations of Fission Product, Prompt, and Delayed Neutron Yields for Neutron Induced Fission on ^{235}U , ^{238}U , and ^{239}Pu

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ABSTRACT

We perform energy dependent calculations of independent and cumulative fission product yields for ^{235}U , ^{238}U , and ^{239}Pu in the first chance fission region. Starting with the primary fission fragment distributions taken from available experimental data and analytical functions based on assumptions for the excitation energy and spin-parity distributions, the Hauser-Feshbach statistical decay treatment for fission fragment de-excitation is applied to more than 1,000 fission fragments for the incident neutron energies up to 5 MeV. The calculated independent fission product yields are then used as an input of β -decay to produce the cumulative yield, and summation calculations are performed. Model parameters in these procedures are adjusted by applying the Bayesian technique at the thermal energy for ^{235}U and ^{239}Pu and in the fast energy range for ^{238}U . The calculated fission observable quantities, such as the energy-dependent fission yields, and prompt and delayed neutron yields, are compared with available experimental data. We also study a possible impact of the second chance fission opening on the energy dependence of the delayed neutron yield by extrapolating the calculation.

KEYWORDS

Fission Product Yield, Hauser-Feshbach Statistical Decay, Prompt Neutron Multiplicity, β Decay, Delayed Neutron Yield

1. Introduction

Nuclear systems that involve the nuclear fission process often require very high accuracy of both prompt and delayed neutron multiplicity data, $\bar{\nu}_p$ and $\bar{\nu}_d$, albeit model predictions for these quantities are not yet at the satisfactory level. For major fissioning systems, such as the neutron-induced reaction on ^{235}U , more than 99% neutrons are the prompt fission neutrons, which are emitted from highly excited two fission fragments formed just after fission. Typically there are more than 1000 fission fragments, and 2–3 prompt neutrons per fission are emitted. Whereas a small fraction ($\sim 1\%$) in the total neutron yield is produced during the β -decay chain of fission products, and approximately 270 nuclides have been identified as precursors for the delayed neutron emission [1,2]. Ideally we can calculate $\bar{\nu}_p$ and $\bar{\nu}_d$ by summing up all the decaying compound nuclei weighted by the fission yields, which is the so-called summation calculation (*e.g.* Ref. [3]). This method, however, requires a lot of well-tuned model inputs. This was partly done in our previous study [4] for the prompt neutron emission.

Since the discovery of delayed neutron by Roberts *et al.* shortly after the discovery of nuclear fission in 1939 [5], despite its tiny fraction, the delayed neutron has attracted people in various scientific communities, as its quite important role in keeping the thermal reactors critical, as well as the reactor systems containing high burn-up fuel, and transmutation of minor actinides. The delayed neutron yield has been measured [6–9] and repeatedly evaluated [1,10–12] for various fissioning systems at several incident neutron energies. Some models for predicting the time-dependent delayed neutron yield have been proposed [13–15], and these studies pointed out the importance of fission yield data to perform these model calculations.

When an incident neutron energy goes higher, it is natural that $\bar{\nu}_p$ also increases monotonously, since the formed compound nucleus has larger available total energy. However, in contrast to $\bar{\nu}_p$, $\bar{\nu}_d$ shows totally different behavior, depending on how the

delayed neutron precursors are produced. There still exists challenges to understand peculiar energy-dependence of $\bar{\nu}_d$, *i.e.*, a slight increase in the yield from thermal to 3 MeV and a steep decrease above 4 MeV as seen in $^{235,238}\text{U}$. To account for the abrupt changes, the evaluated $\bar{\nu}_d$ data in nuclear data libraries, JENDL-4.0 [16] and ENDF/B-VIII [17], include a very crude piecewise linear function to represent experimental data.

Such the energy-dependent behavior has not yet been explained theoretically. Alexander *et al.* [18] first interpreted the energy-dependence in $\bar{\nu}_d$ by taking into account the odd-even effect of fission products. Ohsawa *et al.* [19,20] introduced the multimodal random neck-rupture model [21] and fission mode fluctuations [22] to explain the energy-dependence. Minato [23] proposed a model to reproduce the energy-dependence of $\bar{\nu}_d$ based on the fission yield using Katakura’s systematics [24]. Although an explicit statistical decay was not performed in Minato’s model — hence the calculated $\bar{\nu}_p$ and $\bar{\nu}_d$ are independent of one another — it also supports Alexander’s observation: the odd-even effect in the the charge distribution is important. Recently the odd-even effect was explained by applying the microscopic number projection method [25].

By extending the Hauser-Feshbach Fission Fragment Decay (HF³D) model [4] to the β -decay process, consistency among the independent and cumulative fission yields $Y_I(Z, A)$ and $Y_C(Z, A)$, and neutron multiplicities $\bar{\nu}_p$ and $\bar{\nu}_d$, is automatically guaranteed. In this model, we start with the fission fragment distribution $Y(Z, A, E_{ex}, J, \Pi)$ characterized by the distributions of mass and charge, excitation energy, and spin/parity. We perform the Hauser-Feshbach statistical decay for the excited fission fragments to calculate the independent fission yields $Y_I(Z, A)$ and $\bar{\nu}_p$. A successive β -decay calculation gives the cumulative yields $Y_C(Z, A)$ and $\bar{\nu}_d$. The model parameters are adjusted to reproduce experimental data at thermal by applying the Bayesian technique, and we extrapolate the calculation to the second chance fission threshold. In this paper, we limit ourselves mainly to first-chance fission, because more uncertain parameters will be involved in the multi-chance fission case. Although we study the multi-chance fission case elsewhere [26], here, we briefly explore a possible impact of the second-chance opening with a particular focus on $\bar{\nu}_d$.

2. Methods

2.1. *Hauser-Feshbach statistical decay and β -decay calculations*

2.1.1. *Sources of energy-dependence*

The energy dependence of the independent and cumulative fission product yields (FPY) arises from properties of some model parameters. The primary fission fragment distribution $Y_P(Z, A)$, often approximated by a few Gaussian forms, gradually changes the shape as the incident neutron energy increases. When the excitation energy of the fissioning compound system increases, the fission path after the second barrier spreads along the most probable path, hence the asymmetric terms will have wider width, and the peaks of distributions will be lower to satisfy the normalization condition.

The energy dependence of total kinetic energy (TKE) is also the one of the related physical observables of predicting energy-dependent FPY. We often see that the experimental data of TKE decrease monotonously for some major fissioning nuclides such as $^{235,238}\text{U}$ and ^{239}Pu [27,28,49], except at very low energies [29,30].

The anisothermal parameter R_T , which changes the number of prompt neutrons removed from the fission fragments, often needs to be larger than unity to reproduce the neutron multiplicity distribution as a function of fragment mass number, $\nu(A)$. The reason of this is still unclear. It might be natural to assume $R_T = 1$ by the phase-space argument, where the total excitation energy would be shared by the two fragments according to the number of available states. The odd-even effect in the charge distribution of Wahl's Z_p model [31,32] might decrease at higher excitation energies, where a particular nuclear structure effect no longer persists. Since the original Wahl systematics does not consider any energy dependence of the odd-even effect, we incorporate the energy dependencies of these parameters, yet phenomenological parameterization is applied.

2.1.2. Generation of the fission fragment distribution

The primary fission fragment distributions are the key ingredient in the prompt neutron emission calculation. While this is a complicated multi-dimensional distribution, including energy, spin, parity, etc., we demonstrated that the numerical integration over all these distributions is feasible by the Hauser-Feshbach Fission Fragment Decay (HF³D) model. The model produces various fission observables simultaneously, *e.g.*, the prompt neutron multiplicity $\bar{\nu}_p$, independent FPY $Y_I(Z, A)$, and isomeric ratio (IR) [4]. Since the method and relevant equations are explained elsewhere [4], a brief description as well as newly developed components will be given here.

The primary fission fragment yield $Y_P(Z, A)$ is constructed by five (or seven if needed) Gaussians fitted to experimental primary fission fragment mass distributions of neutron induced reaction on ²³⁵U, ²³⁸U, and ²³⁹Pu. A charge distribution for a given mass number is generated by the Z_p model [32] of Wahl's systematics [31] implemented in the HF³D model.

TKE as a function of primary fission fragment mass TKE(A) is also generated based on the experimental data, which yields the average excitation energy of each fragment. An A -average of TKE(A) gives a TKE value at a given neutron incident energy TKE(E), and the variance of TKE(A) gives the excitation energy distribution. By combining with the distributions of excitation energy E_{ex} , spin J , and parity Π described in the previous work [4], an initial configuration of fission fragment compound nucleus $Y_P(Z, A, E_{ex}, J, \Pi)$ is fully characterized. The Hauser-Feshbach theory is applied to the statistical decay of generated $Y_P(Z, A, E_{ex}, J, \Pi)$. The experimental data sets used in this study are listed in Tables 1, 2, and 3.

The functional forms for TKE(A) and TKE(E) are given in our former work [4], and the parameters of these functions for ²³⁵U are the same as before. Those for ²³⁹Pu were taken from the CGMF code [33]. Because there is no primary fission fragment data for ²³⁸U at thermal, the parameters in $Y_P(Z, A)$ and TKE(A) are determined in the 1.1 – 1.3 MeV region. The obtained $Y_P(Z, A)$ is given later, and TKE(A) is

$$\text{TKE}(A_h) = (348.371 - 1.274A_h) \left\{ 1 - 0.1800 \exp \left(-\frac{(A_h - A_m)^2}{59.199} \right) \right\} \text{ MeV}, \quad (1)$$

and $\text{TKE}(E)$ is

$$\text{TKE}(E) = 171.11 - 0.320E_n \quad \text{MeV} , \quad (2)$$

where the incident energy E_n is in MeV.

Table 1. Experimental data of mass distributions included in the parameter fitting of $Y_P(A, E)$.

Nuclide	Energy (MeV)	Author & Reference
^{235}U	2.53×10^{-8}	Baba <i>et al.</i> [34]
	2.53×10^{-8}	Hambsch [35]
	2.53×10^{-8}	Pleasanton <i>et al.</i> [36]
	2.53×10^{-8}	Simon <i>et al.</i> [37]
	2.53×10^{-8}	Straede <i>et al.</i> [38]
	2.53×10^{-8}	Zeynalov <i>et al.</i> [39]
	$2.53 \times 10^{-8} - 7$	D'yachenko <i>et al.</i> [40]
^{238}U	1.11, 1.25	Goverdovskiy <i>et al.</i> [41]
	1.2 - 5.8	Vives <i>et al.</i> [42]
^{239}Pu	$2.53 \times 10^{-8} - 4.48$	Akimov <i>et al.</i> [43]
	2.53×10^{-8}	Surin <i>et al.</i> [44]
	2.53×10^{-8}	Wagemans <i>et al.</i> [45]
	2.53×10^{-8}	Schillebeeckx <i>et al.</i> [46]
	2.53×10^{-8}	Nishio <i>et al.</i> [47]
	2.53×10^{-8}	Tsuchiya <i>et al.</i> [48]

Table 2. Experimental data included in the parameter fitting of $\text{TKE}(A)$.

Nuclide	Energy (MeV)	Author & Reference
^{235}U	2.53×10^{-8}	Baba <i>et al.</i> [34]
	2.53×10^{-8}	Hambsch [35]
	2.53×10^{-8}	Simon <i>et al.</i> [37]
	2.53×10^{-8}	Zeynalov <i>et al.</i> [39]
	2.53×10^{-8}	D'yachenko <i>et al.</i> [40]
^{238}U	1.2	Vives <i>et al.</i> [42]
^{239}Pu	2.53×10^{-8}	Surin <i>et al.</i> [44]
	2.53×10^{-8}	Wagemans <i>et al.</i> [45]
	2.53×10^{-8}	Nishio <i>et al.</i> [47]
	2.53×10^{-8}	Tsuchiya <i>et al.</i> [48]

2.1.3. Model Parameters

The Gaussian terms for $Y_P(A)$ are parameterized as

$$Y_P(A) = \sum_{i=1}^5 \frac{F_i}{\sqrt{2\pi}\sigma_i} \exp \left\{ -\frac{(A - A_m + \Delta_i)^2}{2\sigma_i^2} \right\} , \quad (3)$$

Table 3. Experimental data included in the parameter fitting of $\text{TKE}(E)$.

Nuclide	Energy (MeV)	Author & Reference
^{235}U	0.18 – 8.83	Meadows and Budtz-Jørgensen [27]
	2.53×10^{-8} – 35.5	Duke [29]
^{238}U	1.5 – 400.0	Zöller <i>et al.</i> [49]
	1.4 – 28.3	Duke <i>et al.</i> [50]
^{239}Pu	0.05 – 5.3	Akimov <i>et al.</i> [43]
	2.53×10^{-8} – 3.55	Vorobeva <i>et al.</i> [51]
	0.5 – 50	Meierbachtol <i>et al.</i> [52]

where σ_i and Δ_i are the Gaussian parameters, the index i runs from the low mass side, and the component of $i = 3$ is for the symmetric distribution ($\Delta_3 = 0$). $A_m = A_{\text{CN}}/2$ is the mid-point of the mass distribution, A_{CN} is the mass number of fissioning compound nucleus, and F_i is the fraction of each Gaussian component. The symmetric shape of $Y_P(A)$ ensures implicit relations of $F_1 = F_5$, $F_2 = F_4$, etc.

We assume that the energy sharing between the complementary light and heavy fragments is followed by the anisothermal model [53,54], which is defined by the ratio of effective temperature T_L and T_H in the light and heavy fission fragments,

$$R_T = \frac{T_L}{T_H} = \sqrt{\frac{a_H U_L}{a_L U_H}}, \quad (4)$$

where U is the excitation energy corrected by the pairing energy [55], and a is the level density parameter including the shell correction energy.

There are several estimates of R_T for different fissioning systems. In the case of thermal neutron induced fission on ^{235}U , a constant R_T reasonably reproduces the experimental $\nu(A)$ data [4], and Talou *et al.* [56,57] showed the cases of $^{239}\text{Pu}(\text{n}_{\text{th}}, \text{f})$, and ^{252}Cf spontaneous fission. However, it has been reported that better reproduction of experimental data is achieved by mass-dependent R_T parameters [58–61]. In the present work, we do not explore all possible functional forms of R_T . Instead, a simple energy-dependence is introduced as

$$R_T = \begin{cases} R_{T0} + E_n R_{T1}, & R_{T0} + E_n R_{T1} \geq 1 \\ 1, & \text{otherwise} \end{cases}, \quad (5)$$

where R_{T0} and R_{T1} are model parameters. As we expect R_T decreases as the incident

energy, $R_{T1} < 0$.

In Wahl's Z_p model the even-odd effect in the Z -distribution is given as

$$f = \begin{cases} F_Z F_N & Z_{\text{even}} & N_{\text{even}} \\ F_Z / F_N & Z_{\text{even}} & N_{\text{odd}} \\ F_N / F_Z & Z_{\text{odd}} & N_{\text{even}} \\ 1 / (F_Z F_N) & Z_{\text{odd}} & N_{\text{odd}} \end{cases}, \quad (6)$$

where $F_Z \geq 1$ and $F_N \geq 1$ are parameterized and tabulated by Wahl. This equation gives higher yields when Z and/or N are the even number. We expect such even-odd staggering will be mitigated when a fissioning system has higher excitation energy. We model the reduction in the even-odd effect by

$$F_Z = 1.0 + (F_Z^W - 1.0)f_Z, \quad (7)$$

$$F_N = 1.0 + (F_N^W - 1.0)f_N, \quad (8)$$

where F_Z^W and F_N^W are the parameters in Wahl's systematics, and f_Z and f_N are the scaling factor as inputs. These scaling factors are also linear functions of incident neutron energy, $f_i = f_{i0} + E_n f_{i1}$, $i = Z, N$.

2.2. β -decay calculation

The HF³D model produces the independent fission product yields $Y_I(Z, A)$, as well as the meta-stable state production when the nuclear structure data indicate that the level half-life is long enough (typically more than 1 ms.) Here we add a meta-state index M to specify the isomers explicitly, $Y_I(Z, A, M)$ and $Y_C(Z, A, M)$. The cumulative yields are calculated in a time-independent manner, hence $Y_I(Z, A, M)$ and $Y_C(Z, A, M)$ are simply connected by the decay branching ratios [62]. The decay data included are the half-lives $T_{1/2}$, the decay mode (α -decay, β^- -decay, delayed neutron emission, *etc.*), and the branching ratios to each decay mode. They are taken from ENDF/B-VIII decay data library. We also considered JENDL-4.0 decay data library, however the result is not so different.

When a decay branch includes a neutron emission mode, this nuclide is identified as a β -delayed neutron precursor. The delayed neutron yield from this i -th precursor is calculated as $\nu_d(i) = Y_C(i)b_iN_d$, where b_i is the branching ratio to the neutron-decay mode, and N_d is usually one unless multiple neutron emission is allowed. The total delayed neutron yield $\bar{\nu}_d$ is $\sum_i \nu_d(i)$.

2.3. Adjustment of Hauser-Feshbach model calculation parameters by Bayesian technique

An optimization procedure of the HF³D model parameters is a non-linear multi-dimensional least-squares problem. Albeit such complex problem might be solved by the modern technology, this will be a hefty computation and beyond our scope. Instead, we perform a relatively small-scale adjustment of the model parameters to reproduce some of the fission product yield data by applying the Bayesian technique with the KALMAN code [63]. The model parameters are first estimated by comparing with the most sensitive quantities. They are our prior. Then the prior parameters are adjusted simultaneously by fitting to the experimental data. Although it is always ideal to use raw experimental data, we use the evaluated values that should be representative of available experimental data. However, it should be noted that we are not trying to reproduce the evaluation, but to find a consistent solution among different fission observable.

The model parameters to be included in the KALMAN calculation are the first and second Gaussian parameters (fraction F_i , width σ_i , and mass shift Δ_i for $i = 1$ and 2.) We fix the symmetric Gaussian, because it does not have any sensitivities to the experimental data included in this study, and its fraction is too small anyway. We also include the anisothermal R_T parameter, the spin factor f_J , and the scaling factor in Eqs. (7) and (8). The adjustment is performed at the thermal energy (or at relatively low energy for ^{238}U), and the energy-dependent parts in these model parameters are fixed.

The sensitivity matrix C is defined as

$$c_{ij} = \frac{\partial d_i}{\partial p_j}, \quad 1 \leq i \leq N, \quad 1 \leq j \leq M, \quad (9)$$

where $P = (p_1, p_2, \dots)$ is the model parameter vector, and $D = (d_1, d_2, \dots)$ is the data vector containing the calculated values. The partial derivatives are calculated numerically. The KALMAN code linearizes the model calculation as

$$D = F(P) \simeq F(P_0) + C(P - P_0), \quad (10)$$

where $F(P)$ stands for a model calculation with a given parameter P , and P_0 is the prior parameter vector.

It is not so easy to impose a constraint $2F_1 + 2F_2 + F_3 = 2$ on the Gaussian fractions during the adjustment process, *e.g.*, when the F_1 parameter is perturbed as $F_1 + \delta$, the sum exceeds 2; $2(F_1 + \delta) + 2F_2 + F_3 = 2 + 2\delta$. However, we renormalize the fractions internally

$$F'_j = F_j - \frac{2\delta}{2 + 2\delta} F_j = F_j \left(1 - \frac{\delta}{1 + \delta} \right) \quad (11)$$

to assure the sum to be 2. F'_j is the actual fraction inside the calculations, and F_j is not necessarily normalized but represents a model input.

3. Results

3.1. Adjusted model parameters at thermal energy

3.1.1. Parameter adjustment for $^{235,238}\text{U}$ and ^{239}Pu

The prior Gaussian parameters, R_{T0} , f_J , and TKE for ^{235}U at the thermal energy are taken from our previous study [4]. When we modify TKE, $\text{TKE}(A)$ is automatically shifted to make sure the A -average coincides with the given TKE value. The original Walh's Z_p model is also employed as the prior parameter, which means $f_{Z0} = f_{N0} = 1$.

They are shown in the second column of Table 4. These parameters are adjusted to reproduce the cumulative fission product yields of ^{95}Zr , ^{97}Zr , ^{99}Mo , ^{132}Te , ^{140}Ba , and ^{147}Nd at thermal, as well as $\bar{\nu}_p$ and $\bar{\nu}_d$. Now we have 11 parameters ($M = 11$) and 8 data ($N = 8$.)

With the prior parameters, the calculated $\bar{\nu}_p$ of 2.38 is slightly lower than the evaluated values of 2.41 (ENDF/B-VIII) and 2.42 (JENDL-4.0), while the prior $\bar{\nu}_d$ of 0.0195 is 23% larger than the value found in both libraries, 0.0159. The adjustment reconciles these discrepancies with the better known values, and the posterior parameters yield $\bar{\nu}_p = 2.415$ and $\bar{\nu}_d = 0.0169$. The posterior parameters with their uncertainties and correlation matrix are given in Table 4. Since the actual changes in $Y_P(A)$ are very modest, and the posterior parameters equally reproduce the experimental data of mass distribution, we do not include the comparison plot here. Figure 1 (a) is the mass chain yield with the prior and posterior parameters. The ENDF evaluated values are also compared. This figure also shows some mass-chains that contain major β -delayed neutron emitters. The reduction in $\bar{\nu}_d$ is, in part, caused by the smaller posterior yields of $A = 137$ and 94 , which include ^{137}I and ^{94}Rb . While these masses were not included in the adjustment, the sensitivity of $\bar{\nu}_d$ to these masses implicitly demands the reduction of these mass-chains.

When the prior R_T and f_J parameters are determined, we compare the neutron multiplicity distribution $P(\nu)$ with the experimental data. The posterior parameters modify the calculated $P(\nu)$ but not so significantly. The calculated $P(\nu)$ still agrees fairly well with the data.

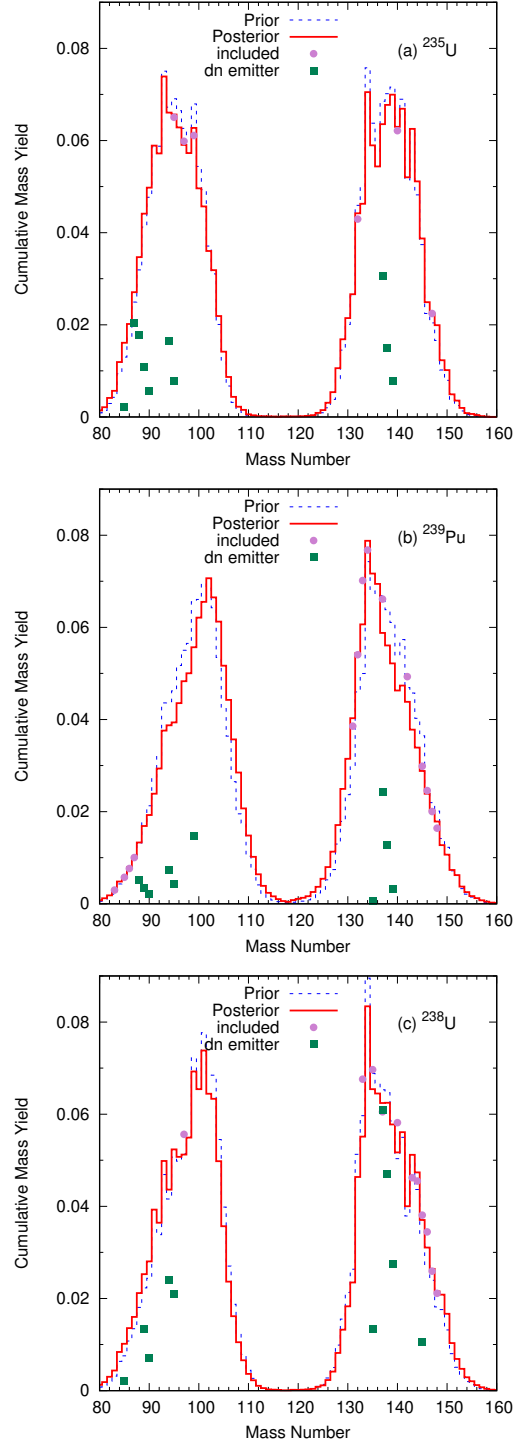


Figure 1. Calculated mass chain yields with the prior and posterior model parameters for ^{235}U , ^{239}Pu , and ^{238}U . They are compared with some selected cumulative fission product yield data by the circles. The squares are the mass chains that include major delayed neutron emitters.

Table 4. Prior and posterior model parameters defined in Eqs. (3), (5), (7), and (8), as well as the spin saling factor f_J . These parameters are dimensionless quantities, except TKE is in MeV.

	pri	post	uncertainty[%] and correlation [%]											
F_1	0.793	0.824	4.3	100										
σ_1	4.83	5.05	1.4	41	100									
Δ_1	23.0	23.1	0.5	-36	-56	100								
F_2	0.205	0.197	4.7	22	-40	36	100							
σ_2	2.73	2.92	3.1	-33	1	28	34	100						
Δ_2	15.6	15.2	0.7	-1	14	39	0	11	100					
f_{Z0}	1.00	1.78	6.6	0	-7	48	0	1	41	100				
f_{N0}	1.00	0.97	20.6	0	2	0	0	3	-1	2	100			
R_{T0}	1.20	1.29	3.8	-3	-10	-64	3	-13	-49	-51	0	100		
f_J	3.00	2.96	4.9	6	-30	16	-6	-9	-23	7	0	0	100	
TKE	170.5	170.1	0.1	-7	25	-27	6	11	1	-5	-1	13	-82	100

3.1.2. Parameter adjustment for ^{239}Pu

The Gaussian parameters obtained by fitting to the experimental $Y_P(A)$ for ^{239}Pu are

$$\Delta_1 = -\Delta_5 = 14.09 + 0.0994E_n , \quad (12)$$

$$\Delta_2 = -\Delta_4 = 20.08 + 0.2940E_n , \quad (13)$$

$$\sigma_1 = \sigma_5 = 3.26 + 0.2000E_n , \quad (14)$$

$$\sigma_2 = \sigma_4 = 6.58 + 0.1969E_n , \quad (15)$$

$$\sigma_3 = 10.0 , \quad (16)$$

where E_n in MeV. The fractions of each Gaussian are given by

$$F_1 = F_5 = 0.2483 + 0.0074E_n , \quad (17)$$

$$F_2 = F_4 = 0.7184 - 0.0075E_n , \quad (18)$$

$$F_3 = 0.003 + 0.003E_n . \quad (19)$$

The adjustment procedure for ^{239}Pu at thermal includes the same parameters as those in the ^{235}U case. These parameters are fitted to $\bar{\nu}_p$, $\bar{\nu}_d$, and cumulative FPY of ^{83}Kr , ^{85}Rb , ^{86}Kr , ^{87}Sr , ^{131}Xe , ^{132}Xe , ^{133}Xe , ^{134}Xe , ^{137}Ba , ^{142}Ce , ^{143}Pr , ^{144}Nd , ^{145}Nd , ^{146}Nd , ^{147}Nd , ^{148}Nd , and ^{150}Nd . They were chosen from the mass chain evaluation by

Table 5. Prior and posterior model parameters for ^{239}Pu . See Table 4 for parameter descriptions.

	pri	post	uncertainty[%] and correlation [%]											
F_1	0.234	0.248	4.1	100										
σ_1	3.51	3.26	5.2	2	100									
Δ_1	14.9	14.1	1.8	2	18	100								
F_2	0.765	0.718	4.6	31	-2	-3	100							
σ_2	6.06	6.58	0.5	-20	21	17	20	100						
Δ_2	20.8	20.1	0.5	40	-20	-9	-41	-66	100					
f_{Z0}	1.00	2.58	4.4	1	0	69	-1	-21	30	100				
f_{N0}	1.00	0.93	21.3	-1	0	-4	1	1	0	2	100			
R_{T0}	1.20	1.30	2.4	-6	22	10	6	29	-69	-31	-2	100		
f_J	2.50	1.58	5.7	-24	10	-24	24	10	-33	14	1	28	100	
TKE	178.2	179.4	0.1	26	-21	1	-26	-7	31	-28	0	-26	-92	100

England and Rider [64], where relatively small uncertainties are assigned. The prior and posterior model parameters are given in Table 5, and the comparison of mass yields are in Fig. 1 (b). Similar to the ^{235}U case, the prior parameter set produces $\bar{\nu}_d = 0.00888$, which is too large compared to the evaluated value of 0.00645. The smaller $\bar{\nu}_d$ is achieved by the adjustment, and the posterior set gives 0.00665.

3.1.3. Parameter adjustment for ^{238}U

Because fission observable data for ^{238}U are only available in the fast energy range and above, the procedure is slightly different from the ^{235}U and ^{239}Pu cases. The adjusted Gaussian parameters were obtained at 1.1 and 1.25 MeV by Goverdovskiy [41] and 1.2 MeV [42] by Vives. The adjusted Gaussian parameters are

$$\Delta_1 = -\Delta_5 = 15.66 - 0.0679E_n , \quad (20)$$

$$\Delta_2 = -\Delta_4 = 23.36 - 0.1929E_n , \quad (21)$$

$$\sigma_1 = \sigma_5 = 3.31 + 0.0159E_n , \quad (22)$$

$$\sigma_2 = \sigma_4 = 5.43 + 0.1267E_n , \quad (23)$$

$$\sigma_3 = 4.50 + 0.267100E_n . \quad (24)$$

Table 6. Prior and posterior model parameters for ^{238}U at ≈ 1 MeV. See Table 4 for parameter descriptions.

	pri	post	uncertainty[%] and correlation [%]											
F_1	0.587	0.625	3.6	100										
σ_1	5.405	5.580	1.4	16	100									
Δ_1	22.879	23.128	0.5	-43	-18	100								
F_2	0.413	0.380	4.4	33	-10	55	100							
σ_2	3.459	3.326	2.6	-40	8	42	31	100						
Δ_2	15.515	15.584	0.7	-11	27	43	30	13	100					
f_{Z0}	1.00	2.386	5.3	0	-16	25	20	-14	48	100				
f_{N0}	1.00	0.736	52.8	0	-8	-2	0	-2	3	13	100			
R_{T0}	1.30	1.327	1.0	-1	-1	-6	2	0	-9	22	0	100		
f_J	3.00	2.956	1.0	3	6	-6	-3	1	-4	-4	0	2	100	
TKE	171.4	170.5	0.1	-5	-29	-9	5	-2	-30	28	0	-4	-18	100

The fractions of each Gaussian are given by

$$F_1 = F_5 = 0.5876 + 0.032E_n, \quad (25)$$

$$F_2 = F_4 = 0.4203 - 0.034E_n, \quad (26)$$

$$F_3 = 0.0006 + 0.001E_n, \quad (27)$$

and the covariance matrix is given in Table 6. These parameters are fitted to $\bar{\nu}_p$, $\bar{\nu}_d$, and cumulative FPY of ^{97}Zr , ^{133}I , ^{135}Xe , ^{137}Cs , ^{140}Ba , ^{143}Ce , ^{14}Ce , ^{145}Pr , ^{147}Nd , and ^{148}Nd .

3.2. Energy dependence of $\bar{\nu}_p$ and $\bar{\nu}_d$

3.2.1. Energy-dependent inputs and pivots

Some of the Gaussian parameters are weakly energy-dependent, and often expressed by a linear function of the incident energy as in Eqs. (17) – (19). The energy-dependent terms are obtained by fitting to the experimental $Y_P(A)$ data, and we do not attempt to tune these parameters. We consider other parameters, R_T , f_Z , f_N , and TKE, to be energy-dependent, and simple linear functions are assumed as in Eqs.(5), (7) and (8). Since the energy-dependence of TKE is rather well known experimentally, we study the energy-dependence of the FPYs, $\bar{\nu}_p$, and $\bar{\nu}_d$ by assuming a simple form for the

mode inputs for ^{235}U and ^{239}Pu first. We exclude ^{238}U for now, as it is a threshold fissioner. The R_{T1} parameter in Eq. (5) is roughly $-(R_T(0) - 1)/6.0 \text{ MeV}^{-1}$ to make $R_T = 1$ at the opening of second chance fission, hence $R_{T1} = -0.0476$ and -0.0507 for ^{235}U and ^{239}Pu . Similarly, f_{Z1} and f_{N1} are estimated to be $f_{Z1} = -0.296 \text{ MeV}^{-1}$ and $f_{N1} = -0.161 \text{ MeV}^{-1}$ for ^{235}U , and $f_{Z1} = -0.430 \text{ MeV}^{-1}$ and $f_{N1} = -0.156 \text{ MeV}^{-1}$ for ^{239}Pu , which ensures that the even-odd effect disappears at $E_n = 6 \text{ MeV}$.

First we consider four cases; (1) both R_T and $f_{Z,N}$ are constant, (2) constant R_T and energy-dependent $f_{Z,N}$, (3) energy-dependent R_T and constant $f_{Z,N}$, and (4) both energy-dependent. By comparing the calculated $\bar{\nu}_p$ and $\bar{\nu}_d$ with experimental data, we found that the energy-dependence of R_T modestly impacts on the results, and probably the modeling uncertainty conceals the importance of R_T . Whereas we also noticed that the energy-dependence of $f_{Z,N}$ is crucial for $\bar{\nu}_d$. Hereafter we assume R_T is constant, while $f_{Z,N}$ is energy-dependent.

When an independent or cumulative FPY is almost energy-independent,

$$\frac{dY_{I,C}(Z, A, E)}{dE} \simeq 0, \quad (28)$$

it is easier to see the mass region where this condition happens by calculating the derivative of mass yields,

$$\frac{dY_{I,C}(A, E)}{dE} = \sum_Z \frac{dY_{I,C}(Z, A, E)}{dE} \simeq 0. \quad (29)$$

We approximate the derivative by coarse numerical derivative $(Y_{I,C}(A, 2[\text{MeV}]) - Y_{I,C}(A, 0[\text{MeV}]))/2$, which is shown in Fig. 2. The general shape of dY_I/dE does not change too much in the energy range below the second chance fission. This implies the cumulative FPYs vary monotonously with the incident neutron energy.

The derivative plot for ^{235}U indicates FPYs near $A = 85, 100$, and 135 vary slowly with the energy, while FPYs near $A = 90, 104, 129$, and 143 should have steeper energy-dependence. These energy-independent regions, or the pivots, appear due to complicated interplay among the energy-dependent model parameters. In the case of ^{239}Pu , the pivots locate near $A = 92, 109, 129$, and 142 , and the FPYs in the

peak regions ($A = 103$ and 133) may show the largest reduction rate.

In Fig. 3 we compare some of our calculated $Y_C(Z, A, E)$ with the experimental data of Gooden et al. [65], measurements at LANL in the critical assemblies [66], as well as other published data. From the derivative plot in Fig. 2, we expect Y_C of ^{235}U decreases in the $A \simeq 90$ region, while Y_C of ^{239}Pu increases. For the both isotopes, the pivot will be seen in $A = 95 - 100$. The comparisons of ^{91}Sr , ^{97}Zr , and ^{99}Mo clearly show these behavior, and ^{103}Ru now shows an opposite tendency as the incident neutron energy.

On the heavier mass side, the slope of $Y_C(Z, A, E)$ changes the sign from positive to negative around $A = 134$ for ^{235}U , with one exception of the $A = 137$ case that has the positive slope. For ^{239}Pu , the sign change happens twice, near $A = 130$ and 145 . This is shown in Fig. 4; ^{132}Te , ^{137}Cs , ^{140}Ba , and ^{147}Nd . Our model calculation also reproduces other isotopes with the similar quality.

Although we didn't include the ^{238}U case in Fig. 2 as FPY at thermal is only given by extrapolation, Figs. 3 and 4 include ^{238}U too.

3.2.2. Energy-dependence of $\bar{\nu}_p$ and $\bar{\nu}_d$

The calculated $\bar{\nu}_p$ and $\bar{\nu}_d$ for $^{235,238}\text{U}$ and ^{239}Pu are compared with experimental data in Figs. 5 and 6. The evaluated $\bar{\nu}_p$ and $\bar{\nu}_d$ in ENDF/B-VIII and JENDL-4.0, which are evaluated by least-squares fitting to the available experimental data, are also compared. In general $\bar{\nu}_p$ increases as the incident neutron energy goes higher, simply because of the energy conservation. However, its slope $d\bar{\nu}_p/dE$ strongly depends on the behavior of TKE. Although the mechanism for the incident-energy dependence of TKE is still unclear, we take the energy dependence of TKE from experimental data, and it enables us to reproduce $\bar{\nu}_p$ by our model. Other parameters, the Gaussian shape and $f_{Z,N}$, also change the slope of $\bar{\nu}_p$, but they have a much more modest impact on the calculated result.

The energy-dependence of $\bar{\nu}_d$ is caused mainly by changing the yields of the delayed neutron precursors. Interestingly the calculated and experimental $\bar{\nu}_d$'s reveal very weak energy-dependency for these isotopes. As we noted large fractions of delayed neutron emission are from the mass regions of $A = 137$ and 94 , and according to Fig. 2,

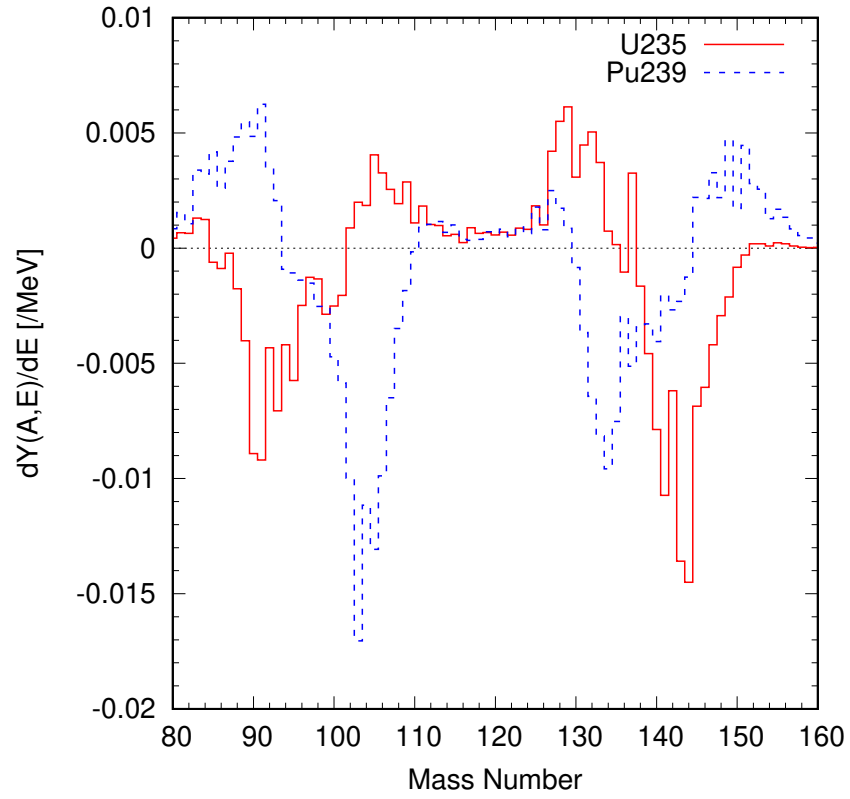


Figure 2. Energy-dependence of the cumulative mass yields, $\partial Y(A, E)/\partial E$, approximated by $(Y(A, 2[\text{MeV}]) - Y(A, 0[\text{MeV}]))/2$.

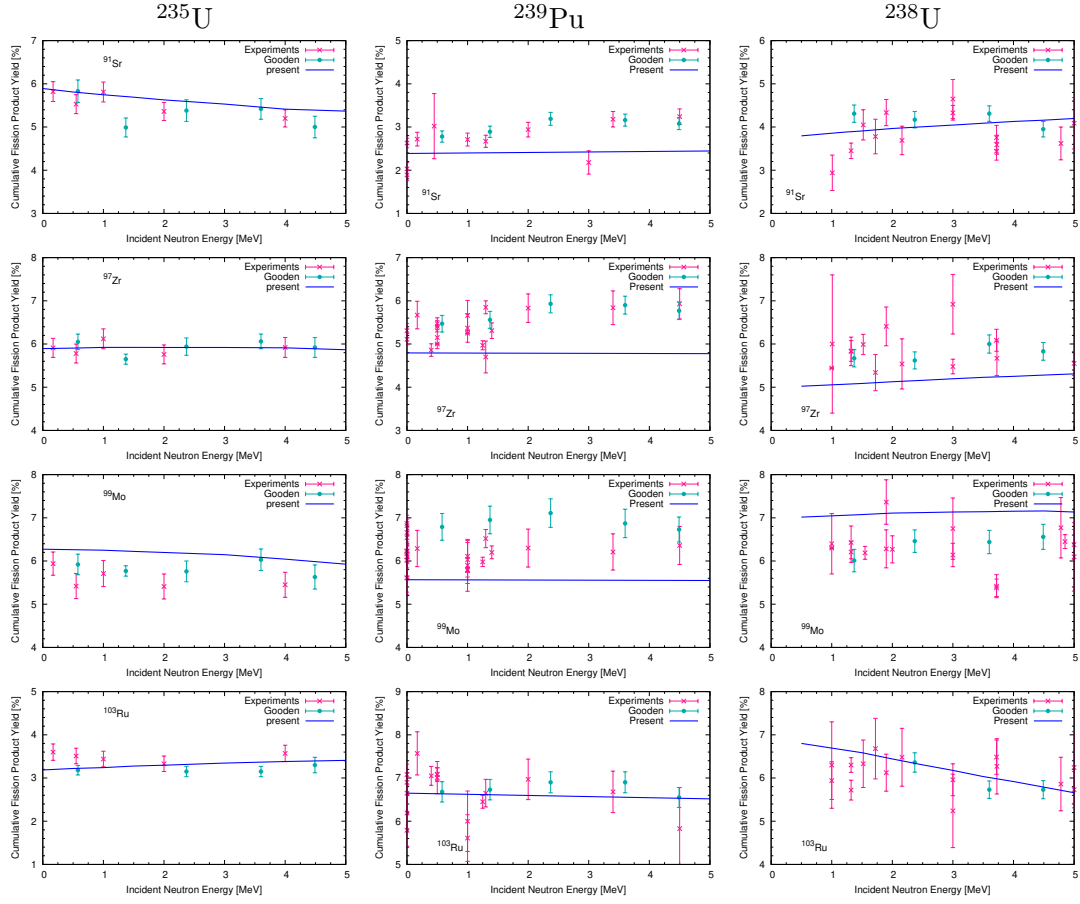


Figure 3. Energy dependence of cumulative fission product yields of ^{91}Sr , ^{97}Zr , ^{99}Mo , and ^{103}Ru for the neutron-induced fission on ^{235}U (left), ^{239}Pu (middle), and ^{238}U (right) of calculated data (solid line) compared with with the experimental data of Gooden et al. [65], as well as other published data.

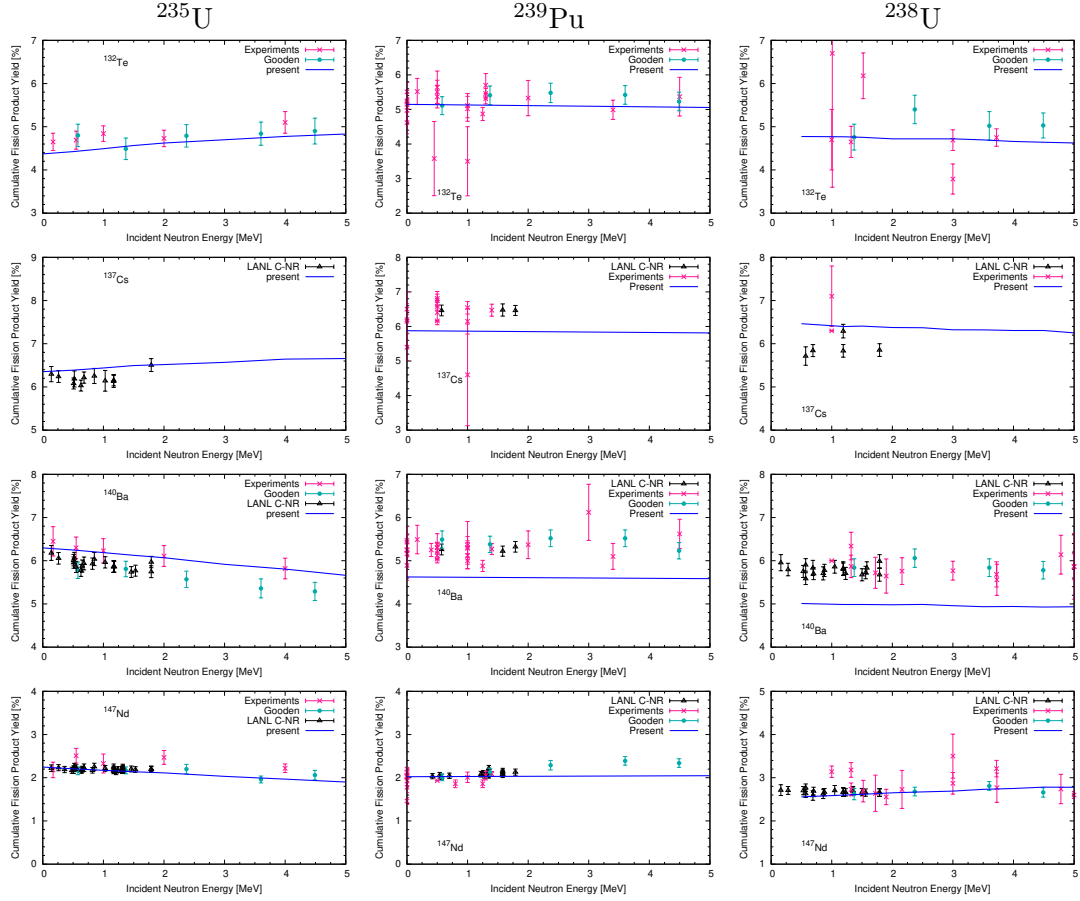


Figure 4. Incident neutron energy dependence of cumulative fission product yields of ^{132}Te , ^{137}Cs , ^{140}Ba , and ^{147}Nd for the neutron-induced fission on ^{235}U (left), ^{239}Pu (middle), and ^{238}U (right) of calculated data (solid line) compared with the experimental data of Gooden et al. [65] and measurements at LANL in the critical assemblies (LANL C-NR) [66], as well as other published data.

we expect $\bar{\nu}_d$ to decrease.

As it is not so convenient to survey the delayed neutron precursors individually, we lump the precursors into the well-known six groups according to their half-lives $T_{1/2}$, and calculate the energy-dependence of the six-group yields. The group structure is usually defined by the isotopes included in each group. This is convenient for the longer $T_{1/2}$ groups, but it is ambiguous for the shorter groups. For the sake of convenience, we define the six-group structure as (1) $T_{1/2} > 40$ s, (2) $8 < T_{1/2} \leq 40$ s, (3) $3 < T_{1/2} \leq 8$ s, (4) $1 < T_{1/2} \leq 3$ s, (5) $0.3 < T_{1/2} \leq 1$ s, and (6) $T_{1/2} \leq 0.3$ s. The fractions of each group are shown in Fig. 7. In the case of ^{235}U , the largest contribution is from the Group 4, which slightly decreases as the incident neutron energy. This is compensated by the increasing Group 2, resulting in the flat behavior of $\bar{\nu}_d$. The energy variation of each group is more visible for the ^{239}Pu case. Obviously the energy-dependence of $\bar{\nu}_d$ does not originate from specific fission products, but a consequence of their competition.

We studied sensitivities of the model parameters to $d\bar{\nu}_d/dE$, and found that the f_{Z1} and f_{N1} terms change the slope. When $f_{Z1} = f_{N1} = 0$, or a constant odd-even effect, $\bar{\nu}_d$ decreases for both ^{235}U and ^{239}Pu cases. We briefly estimated the energy-dependence of the odd-even term so that this effect fades away toward the second chance fission. Nonetheless, this ansatz was not so unrealistic. Better reproduction of the experimental data can be achieved by adjusting the f_{Z1} and f_{N1} parameters, yet the currently available data have rather large uncertainties to estimate these parameters precisely.

3.3. Extrapolating to the second chance fission

The experimental data of $\bar{\nu}_d$ for ^{235}U drop sharply near 5 MeV [8,10], and the evaluated data often include a curious kink to reproduce this behavior. As we demonstrated that $\bar{\nu}_d$ is weakly energy-dependent up to the second chance fission, the kink could be hypothetically the evidence of the second-chance contribution, namely transition of major fissioning system from ^{236}U to ^{235}U . The full-extension of our FPY model by including the multi-chance fission is underway [26], and here we extrapolate our calculations beyond the second-chance fission threshold. We do not intend to perform

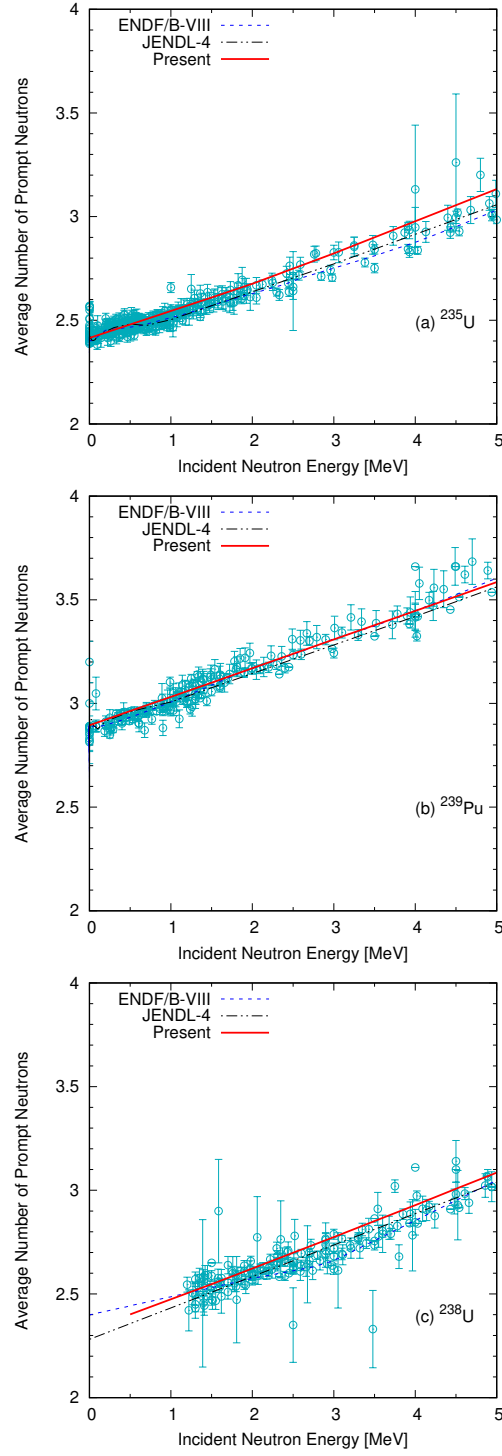


Figure 5. Calculated incident neutron energy dependence of $\bar{\nu}_p$ for ^{235}U , ^{239}Pu , and ^{238}U (solid line) compared with the evaluated $\bar{\nu}_p$ in ENDF/B-VIII (dotted line) and JENDL-4.0 (dot-dashed line) and available experimental data.

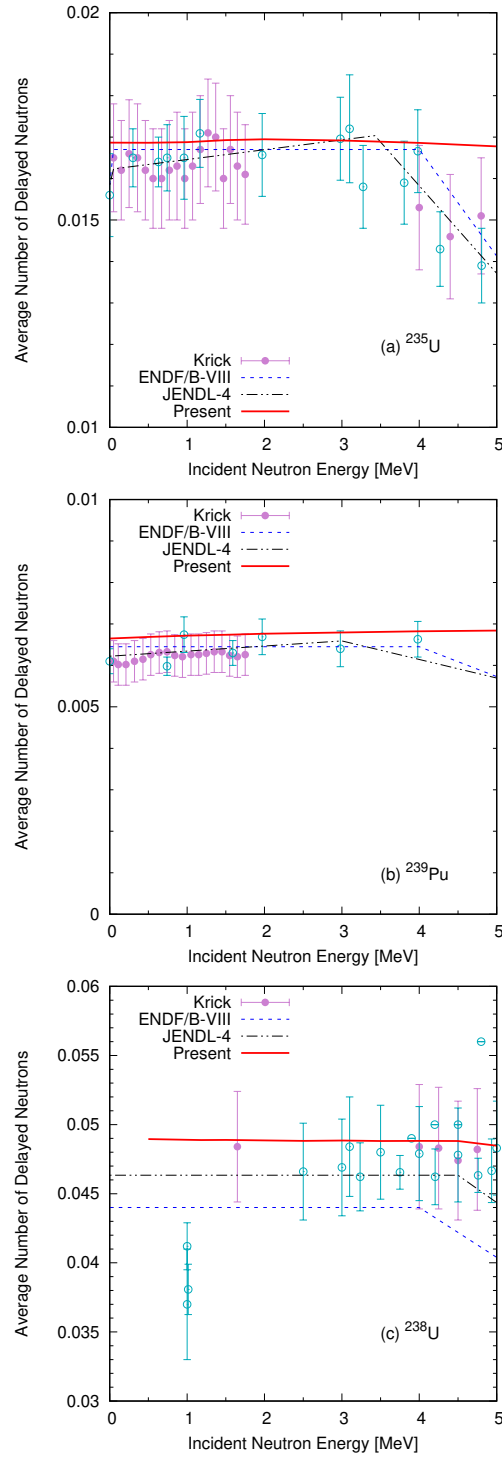


Figure 6. Calculated incident neutron energy dependence of $\bar{\nu}_d$ for ^{235}U , ^{239}Pu , and ^{238}U (solid line) compared with the evaluated $\bar{\nu}_d$ in ENDF/B-VIII (dotted line) and JENDL-4.0 (dot-dashed line), and experimental measurements by Krick *et.al* [8] (filled circle) and the other available experimental data (open circle).

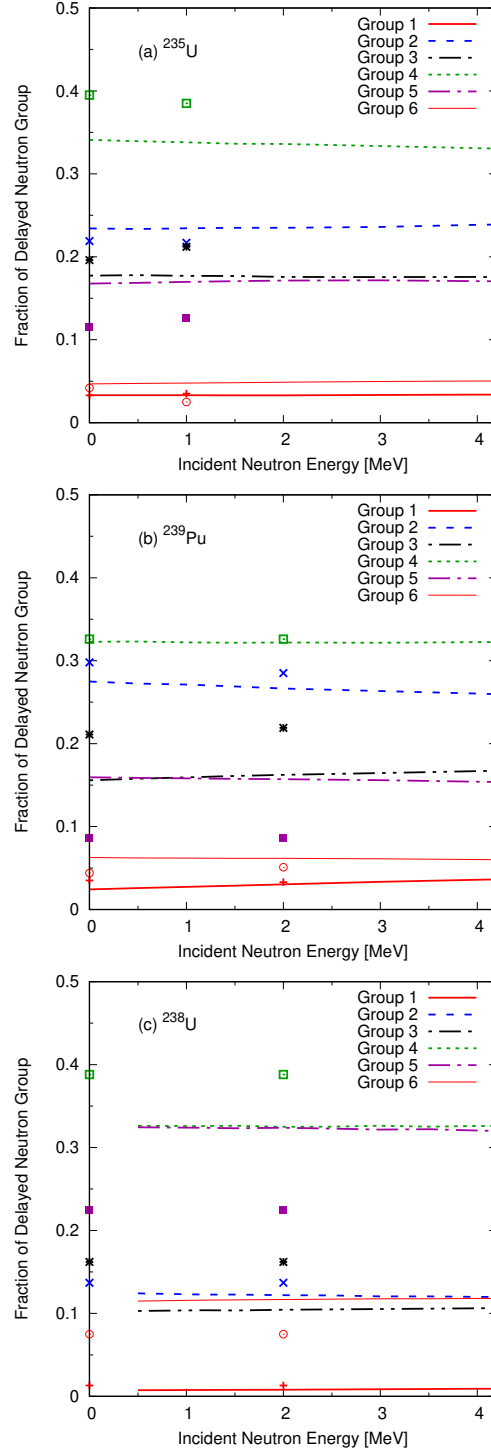


Figure 7. Calculated relative contribution to $\bar{\nu}_d$ from each of the delayed neutron groups for ^{235}U , ^{239}Pu , and ^{238}U . Evaluated $\bar{\nu}_d$ fractions were taken from JENDL-4.0 and are shown by the symbols (+: Group 1, x: Group 2, *: Group 3, □: Group 4, ■: Group 5, and ○: Group 6) at thermal and 1 MeV for ^{235}U , and at thermal and 2 MeV for ^{239}Pu , and ^{238}U .

a detailed model parameter adjustment as done for the first chance case, but similar parameters obtained by the first-chance calculation were plugged into the second-chance fission to see if we will be able to reproduce the kink. This exercise is done for the ^{235}U case only.

The fission probabilities $P_f(E)$ for the first and second chances are calculated with the CoH3 code [67]. The fission parameters, such as the fission barrier, curvature, and level density, are adjusted to reproduce the evaluated fission cross section of ^{235}U . We use the same $Y_P(A)$ for the second chance, but shifted the mid-point by 1/2 mass unit to the lower mass side. f_J , TKE, and $f_{Z,N}$ for both ^{236}U and ^{235}U are the same.

The calculated $\bar{\nu}_d$ is shown in Fig. 8. Albeit the calculated $\bar{\nu}_d$ drops at the energy that is about 1.5 MeV higher than the experimental data, the shape is well reproduced. This supports our hypothesis of the transition of fissioning systems from the first compound nucleus to the second one. At 8 MeV the probability of second chance fission reaches 80%, and a new set of delayed neutron emitters again forms a new plateau above that energy. The step-function-like behavior of $\bar{\nu}_d$ is thus understood.

The calculated transition energy, which is basically the second-chance fission threshold, is higher than the experimental data, and this is still an open question. Despite the fact that our fission barrier parameters could have some uncertainties, the 1.5-MeV change in the fission barriers makes a significant suppression of the fission cross section above 5 MeV. At this moment we don't have a simple solution of matching the kink point in the experimental data and theoretical calculation.

4. Conclusion

The Hauser-Feshbach Fission Fragment Decay (HF³D) model was extended to calculate β -delayed quantities such as the cumulative yield calculation and the delayed neutron yield $\bar{\nu}_d$, where consistency of prompt products retained. The model parameters for ^{235}U , ^{239}Pu , and ^{238}U — the Gaussian functions to characterize the primary fission yields, the anisothermal parameter R_T , the spin parameter f_J , TKE, and the odd-even term of Wahl's Z_p model — were estimated by employing the Bayesian technique with the KALMAN code at the thermal energy for ^{235}U , ^{239}Pu and 1.2 MeV for

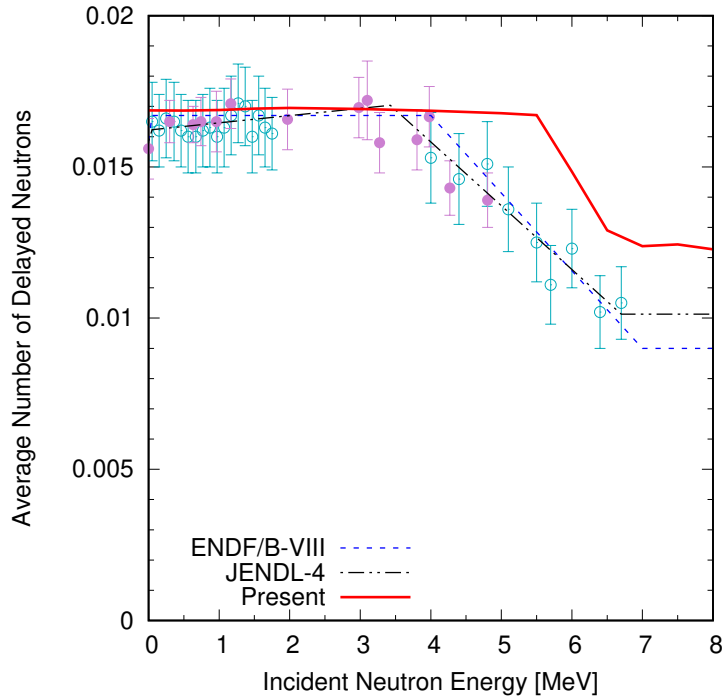


Figure 8. Energy dependence of $\bar{\nu}_d$ for ^{235}U when the second-chance fission is involved.

^{238}U . The result implies that a stronger odd-even effect is required to reproduce the experimental $\bar{\nu}_d$, which is also reported by Minato [23].

Anchoring the statistical decay calculations to experimental data available at the thermal energy for ^{235}U , ^{239}Pu and 1.2 MeV for ^{238}U , we extrapolated the HF³D model to the second chance fission threshold energy, and demonstrated that the calculated cumulative FPYs fairly reproduced the experimental data, as well as $\bar{\nu}_p$ and $\bar{\nu}_d$ simultaneously. The flat behavior of $\bar{\nu}_d$ along the neutron-incident energy seen in the experimental data of ^{235}U and ^{239}Pu was attributed to a coincidental compensation of increasing and decreasing delayed neutron precursors.

To examine the sudden change in $\bar{\nu}_d$ near 5 MeV, we extrapolated our calculations beyond the second-chance fission by assuming the same parameters as the first chance. Indeed this is a crude assumption, nevertheless we were able to reproduce the step-function-like variation of $\bar{\nu}_d$. This is promising, and our HF³D model for the independent and cumulative FPY should be the most advanced tool for evaluating the FPY data, because it produces many fission observable quantities in a consistent manner. Unfortunately our calculation drops at around 5.5 MeV, despite the kink in

the experimental data is seen near 4 MeV. This discrepancy should be explained by further investigation in both the theory and experimental data. Having said that, the HF³D model qualitatively explains that the variation seen in $\bar{\nu}_d$ is a result of different precursors produced by fission at each fission-chance.

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