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 \(^{235}\text{U} \), 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 1,000 fission fragments, and 2–3 prompt neutrons per fission are emitted. A small fraction (~1%) of the total neutron yield is produced during the \( \beta \)-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 the delayed neutron by Roberts et al. [5] shortly after the discovery of nuclear fission in 1939, despite its tiny fraction, the delayed neutron has attracted people in various scientific communities, due to its quite important role in keeping the thermal reactors critical, in reactor systems containing high burn-up fuel, and in the 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 the 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 \(^{235}\text{U} \) and \(^{238}\text{U} \). 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.

As 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 $\tau_d$ based on the fission yield using Katakur’a’s systematics [24]. Although an explicit statistical decay calculation was not performed in Minato’s model – hence the calculated $\tau_p$ and $\tau_d$ are independent of one another – it also supports Alexander’s observation: the odd-even effect in 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 (HF$^3$D) model [4] to the $\beta$-decay process, consistency among the independent and cumulative fission yields $Y_f(Z,A)$ and $Y_c(Z,A)$, and neutron multiplicities $\tau_p$ and $\tau_d$ 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 calculation for the excited fission fragments to calculate the independent fission yields $Y_f(Z,A)$ and $\tau_p$. A successive $\beta$-decay calculation gives the cumulative fission yields $Y_c(Z,A)$ and $\tau_d$. 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 $\tau_d$.

2. Methods

2.1. Hauser-feshbach statistical decay and $\beta$-decay calculations

2.1.1. Sources of energy dependency

The energy dependence of the independent and cumulative yields arises from the properties of some model parameters in our modeling for the fission process. The primary fission fragment distribution $Y_f(Z,A)$, often approximated by a few Gaussian forms, gradually changes the shape as the incident neutron energy increases.

The energy dependence of total kinetic energy (TKE) is also one of the related physical observables of predicting energy-dependent independent and cumulative yield. We often see that the experimental data of TKE decrease monotonously for some major fissioning nuclides such as $^{235,238}$U and $^{239}$Pu [27–29], except at very low energies [30,31].

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 for 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 [32,33] might decrease at higher excitation energies, where a particular nuclear structure effect no longer persists. Since the original Whal 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 (HF$^3$D) model. The model produces various fission observables simultaneously, e.g., the prompt neutron multiplicity $\tau_p$, the independent yields $Y_f(Z,A)$, 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 $Y_f(Z,A)$ is constructed by five (or seven if needed) Gaussians fitted to experimental primary fission fragment mass distributions of neutron-induced reaction on $^{235}$U, $^{238}$U, and $^{239}$Pu. A charge distribution for a given mass number is generated by the $Z_p$ model [33] of Wahl’s systematics [32] implemented in the HF$^3$D 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 this with the distributions of excitation energy $E_{ex}$, spin $J$, and parity $\Pi$ described in the previous work [4], an initial configuration of fission fragment compound nucleus $Y_f(Z,A,E_{ex},J,\Pi)$ is fully characterized. The Hauser-Feshbach theory is applied to the statistical decay of generated $Y_f(Z,A,E_{ex},J,\Pi)$. 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 $^{235}$U are the same as before. Those
Table 1. Experimental data of the fission fragment mass distributions included in the parameter fitting of \( Y_p(A,E) \).

| Nuclide | Energy (MeV) | Author & Reference |
|---------|--------------|---------------------|
| \( ^{238}\text{U} \) | \( 2.53 \times 10^{-8} \) | \([58]\) |
| \( 2.53 \times 10^{-8} \) | Hambisch \([59]\) |
| \( 2.53 \times 10^{-8} \) | Pleasonton et al. \([60]\) |
| \( 2.53 \times 10^{-8} \) | Simon et al. \([61]\) |
| \( 2.53 \times 10^{-8} \) | Straede et al. \([62]\) |
| \( 2.53 \times 10^{-8} \) | Zeynalov et al. \([63]\) |
| \( 2.53 \times 10^{-8} \) | D'yachenko et al. \([64]\) |
| \( 1.11, 1.25 \) | Goverdovskiy et al. \([51]\) |
| \( 1.2-5.8 \) | Vives et al. \([52]\) |

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

| Nuclide | Energy (MeV) | Author & Reference |
|---------|--------------|---------------------|
| \( ^{235}\text{U} \) | \( 2.53 \times 10^{-8} \) | \([58]\) |
| \( 2.53 \times 10^{-8} \) | Hambisch \([59]\) |
| \( 2.53 \times 10^{-8} \) | Simon et al. \([61]\) |
| \( 2.53 \times 10^{-8} \) | Zeynalov et al. \([63]\) |
| \( 2.53 \times 10^{-8} \) | D'yachenko et al. \([64]\) |
| \( 1.2 \) | Vives et al. \([52]\) |
| \( ^{239}\text{Pu} \) | \( 2.53 \times 10^{-8} \) | \([58]\) |
| \( 2.53 \times 10^{-8} \) | Surin et al. \([66]\) |
| \( 2.53 \times 10^{-8} \) | Wagemans et al. \([67]\) |
| \( 2.53 \times 10^{-8} \) | Schillebeeckx et al. \([68]\) |
| \( 2.53 \times 10^{-8} \) | Nishio et al. \([69]\) |
| \( 2.53 \times 10^{-8} \) | Tsuchiya et al. \([70]\) |

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

| Nuclide | Energy (MeV) | Author & Reference |
|---------|--------------|---------------------|
| \( ^{235}\text{U} \) | \( 0.18-8.83 \) | Meadows and Budtz-Jørgensen \([27]\) |
| \( 2.53 \times 10^{-8} \) | \( 35.5 \) | Duke \([30]\) |
| \( 1.5-400.0 \) | Zäler et al. \([29]\) |
| \( 1.4-28.3 \) | Duke et al. \([71]\) |
| \( 0.05-5.3 \) | Akimov et al. \([65]\) |
| \( 2.53 \times 10^{-8} \) | \( 3.55 \) | Vorobeva et al. \([72]\) |
| \( 0.5-50 \) | Meierbach et al. \([73]\) |

for \( ^{239}\text{Pu} \) were taken from the CGMF code \([34]\). Because there is no primary fission fragment data for \( ^{238}\text{U} \) 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

\[
\text{TKE}(A) = (348.371 - 1.274A) \left( 1 - 0.1800 \exp \left( -\frac{(A - A_m)^2}{59.199} \right) \right) \text{ MeV},
\]

and TKE(\(E\)) is

\[
\text{TKE}(E) = 171.11 - 0.320E_n \text{ MeV},
\]

where the incident energy \( E_n \) is in MeV.

2.1.3. Model parameters

The Gaussian terms for \( Y_p(A) \) are parameterized as

\[
Y_p(A) = \sum_{i=1}^{5} \frac{F_i}{\sqrt{2\pi} \sigma_i} \exp \left\{ -\frac{(A - A_m + \Delta_i)^2}{2\sigma_i^2} \right\},
\]

where \( \sigma_i \) and \( \Delta_i \) are the Gaussian parameters, the index \( i \) runs from the low mass side, and the component of \( i = 3 \) is for the symmetric distribution (\( \Delta_i = 0 \)). \( A_m = A_{CN}/2 \) 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 \([35,36]\), which is defined by the ratio of effective temperature \( T_L \) and \( T_H \) in the light and heavy fission fragments,

\[
\frac{R_T}{T_H} = \frac{T_L}{T_H} = \frac{\alpha E}{\alpha L U_L},
\]

where \( U \) is the excitation energy corrected by the pairing energy \([37]\), and \( \alpha \) 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 \( ^{235}\text{U} \), a constant \( R_T \) reasonably reproduces the experimental \( \nu(A) \) data \([4,38]\), and \( \text{Talou et al.} [38,39] \) showed the cases of \( ^{239}\text{Pu}(n_{th},f) \), and \( ^{252}\text{Cf} \) spontaneous fission. However, it has been reported that better reproduction of experimental data is achieved by mass-dependent \( R_T \) parameters \([40-43]\). In the present work, we do not explore all possible functional forms of \( R_T \). Instead, simple energy dependence is introduced as

\[
R_T = \begin{cases} 
R_{T0} + E_n R_{T1}, & R_{T0} + E_n R_{T1} \geq 1 \\
1, & \text{otherwise}
\end{cases}
\]

where \( R_{T0} \) and \( R_{T1} \) are model parameters. As some experimental data imply \([44]\), \( R_T \) decreases as the incident energy increases, \( R_{T1} < 0 \).

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

\[
f = \begin{cases} 
F_Z F_N \text{ Even,} & \text{Neven} \\
F_Z/F_N \text{ Even,} & \text{Nodd} \\
F_N/F_Z \text{ Zodd,} & \text{Neven} \\
1/(F_Z F_N) \text{ Zodd,} & \text{Nodd}
\end{cases}
\]

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 even. 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

\[
F_Z = 1.0 + (F_Z^W - 1.0) f_Z,
\]

\[
F_N = 1.0 + (F_N^W - 1.0) f_N,
\]

where \( F_Z^W \) and \( F_N^W \) are the parameters in Wahl's systematics, and \( f_Z \) and \( f_N \) are the scaling factors as inputs. These scaling factors are also linear functions of incident neutron energy, \( f_i = f_0 + E_{dfi}, \) \( i = Z, N.\)
2.2. β-decay calculation

The HF3D model produces the independent yields $Y_j(Z, A)$, 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, $Y_j(Z, A, M)$ and $Y_C(Z, A, M)$. The cumulative fission yields are calculated in a time-independent manner, hence $Y_j(Z, A, M)$ and $Y_C(Z, A, M)$ are simply connected by the decay branching ratios [45]. 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. For example, in the $^{235}$U(n^5,f) case, the differences in the calculated cumulative yields caused by the ENDF and JENDL decay data libraries are at most 0.02%, except for $^{77}$Ge, $^{83}$Se, $^{96}$Rb, and $^{102}$Nb, for which these libraries give very different branching ratios to the ground and metastable states. We confirmed that the sum of the cumulative yields to these states is still consistent.

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 $v_d(i) = Y_C(i) b_i N_d$, 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 $v_d$ is $\sum_i v_d(i)$.

2.3. Adjustment of calculation parameters by bayesian technique

Although the model parameters have been already tuned to some experimental data, these parameters often do not reproduce other fission observables simultaneously. For example, the Gaussian parameters reported by Katakura [46] do not necessarily reproduce the experimental cumulative yields, since they depend on the statistical decay calculation.

An optimization procedure of the HF3D model parameters is a non-linear multi-dimensional least-squares problem. Although such a complex problem might be solved by 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 cumulative yield data by applying the Bayesian technique with the KALMAN code [47]. The model parameters are first estimated by comparing with the most sensitive quantities. They are our prior parameters. 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 observable.

The model parameters to be included in the KALMAN calculation are the first and second Gaussian parameters (fraction $F_i$, width $\sigma_i$, and mass shift $\Delta_i$ for $i = 1 \text{ and } 2$). 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 $f_i$, 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

$$\left( \frac{\partial c_i}{\partial p_j} \right)_{1 \leq i \leq N, \quad 1 \leq j \leq M},$$

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

$$D = F(P) \cong F(P_0) + C(P - P_0),$$

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

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

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 of the HF3D calculation

3.1.1. Parameter adjustment for $^{235}$U

The prior Gaussian parameters, $R_{TH}, f_i$, and TKE for $^{235}$U at the thermal energy are taken from our previous study [4]. As aforementioned, the adjustment is performed only for the energy-independent terms of the model parameters. Energy-dependent parameters are not adjusted. The same procedure of parameterization is introduced. When we modify TKE, TKE(A) is automatically shifted to make sure the A-average coincides with the given TKE value.
The original Wahl’s $Z_p$ model is also employed as the prior parameter, which means $f_{Z0} = f_{N0} = 1$ for all energy ranges. They are shown in the second column of Table 4. These parameters are adjusted to reproduce the cumulative yields of $^{95}$Zr, $^{97}$Zr, $^{99}$Mo, $^{132}$Te, $^{140}$Ba, and $^{147}$Nd at thermal, as well as $\sigma_p$ and $\sigma_d$.

Now, we have 11 parameters ($M = 11$) and 8 data ($N = 8$). With the prior parameters, the calculated $\sigma_p$ 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 $\sigma_d$ 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 $\sigma_p = 2.415$ and $\sigma_d = 0.0169$. The reduction in $\sigma_d$ is primarily due to the largely increased $f_{Z0}$ in Wahl’s $Z_p$ model. The odd-even staggering in the charge number of the primary fission fragments not so sensitive to the chain yields, which are the cumulative yields of the stable or long-lived fission product at a specific isobaric mass chain [48], but it directly modifies the yields of $\beta$-delayed neutron precursors. 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) shows the chain yields calculated with the prior and posterior parameters. This figure also shows some cumulative yields of major $\beta$-delayed neutron emitters from the ENDF/B-VIII evaluated values for comparison. The reduction in $\sigma_d$ 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 $\sigma_d$ to these masses implicitly demands the reduction of these mass-chains.

When the prior $R_f$ and $f_p$ 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. We also calculated the prompt fission neutron spectra (PFNS) with the prior and posterior parameters and compared them with available experimental data. We confirmed that the posterior parameters better fit the data than the prior, albeit the PFNS data were not included in the data fitting. This result will be further investigated in a separate paper [49].

### 3.1.2. Parameter adjustment for $^{239}$Pu

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

$$\Delta_1 = -\Delta_5 = 20.80 + 0.2940E_n \, ,$$

$$\Delta_2 = -\Delta_4 = 14.90 + 0.0994E_n \, ,$$

$$\sigma_1 = \sigma_5 = 6.06 + 0.1969E_n \, ,$$

$$\sigma_2 = \sigma_4 = 3.51 + 0.2000E_n \, ,$$

$$\sigma_3 = 10.0 \, ,$$

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

$$F_1 = F_5 = 0.765 - 0.0075E_n \, ,$$

$$F_2 = F_4 = 0.234 + 0.0074E_n \, ,$$

$$F_3 = 0.003 + 0.003E_n \, .$$

Equation (17) shows that the asymmetric distribution increases as the incident energy increases, which is against our intuition. This is because the increase of $F_{1,5}$ is really modest, and this is also compensated by the increasing width as in Eq. (12).

The adjustment procedure for $^{239}$Pu at the thermal incident energy includes the same parameters as those in the $^{235}$U case. These parameters are fitted to $\sigma_p$, $\sigma_d$, and cumulative yield of $^{85}$Kr, $^{85}$Rb, $^{86}$Kr, $^{87}$Sr, $^{131}$Xe, $^{132}$Xe, $^{133}$Xe, $^{134}$Xe, $^{135}$Ba, $^{142}$Ce, $^{143}$Pr, $^{144}$Nd, $^{146}$Nd, $^{147}$Nd, $^{148}$Nd, $^{150}$Nd. They were chosen from the chain yields evaluation by England and Rider [50], where relatively small uncertainties are assigned. The prior and posterior model parameters are given in Table 5, and the comparison of chain yields is in Figure 1 (b). Similar to the $^{235}$U case, the prior parameter set produces $\sigma_d = 0.00848$, which is too large compared to the evaluated value of 0.00645.

### Table 4. Prior and posterior model parameters for $^{235}$U defined in Eqs. (3), (5), (7), and (8) and its covariance matrix, as well as the spin salting factor $f_s$. These parameters are dimensionless quantities, except TKE is in MeV.

| pri. | post. | Uncertainty[$\%$] and correlation [ ] |
|------|-------|---------------------------------------|
| $f_1$ | 0.793 | 0.824 | 4.3 | 100 |
| $\sigma_1$ | 4.83 | 5.05 | 1.4 | 41 | 100 |
| $\Delta_1$ | 23.00 | 23.1 | 0.5 | 36 | 56 | 100 |
| $F_2$ | 0.205 | 0.197 | 4.7 | 22 | 40 | 36 | 100 |
| $\sigma_2$ | 2.73 | 2.92 | 3.1 | 33 | 1 | 28 | 34 | 100 |
| $\Delta_2$ | 15.63 | 15.2 | 0.7 | 14 | 0 | 39 | 11 | 100 |
| $f_3$ | 1.00 | 1.78 | 6.6 | 0 | 7 | 48 | 0 | 1 | 41 | 100 |
| $f_0$ | 1.00 | 0.97 | 20.6 | 0 | 2 | 0 | 0 | 0 | 3 | 1 | 2 | 100 |
| $R_f$ | 1.20 | 1.29 | 3.8 | 3 | 10 | 64 | 3 | 13 | 49 | 51 | 0 | 100 |
| $f_s$ | 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 |
3.1.3. Parameter adjustment for $^{238}\text{U}$

Because fission observable data for $^{238}\text{U}$ are only available in the fast energy range and above, the procedure is slightly different from the $^{235}\text{U}$ and $^{239}\text{Pu}$ cases. The adjusted Gaussian parameters were obtained at 1.1 and 1.25 MeV by Goverdovskiy [51] and 1.2 MeV [52] by Vives. The adjusted Gaussian parameters are

$$\Delta_1 = -\Delta_5 = 22.879 - 0.1929E_n,$$

$$\Delta_2 = -\Delta_4 = 15.515 - 0.0679E_n,$$

$$\sigma_1 = \sigma_5 = 5.405 - 0.1267E_n,$$

$$\sigma_2 = \sigma_4 = 3.459 + 0.0159E_n,$$

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

The fractions of each Gaussian are given by

$$F_1 = F_5 = 0.587 + 0.032E_n, \quad (25)$$

$$F_2 = F_4 = 0.413 - 0.034E_n, \quad (26)$$

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

and the covariance matrix is given in Table 6. These parameters are fitted to $\bar{\nu}_p, \bar{\nu}_d$, and cumulative yield of $^{92}\text{Zr}, ^{133}\text{I}, ^{135}\text{Xe}, ^{137}\text{Cs}, ^{140}\text{Ba}, ^{143}\text{Ce}, ^{144}\text{Ce}, ^{145}\text{Pr}, ^{147}\text{Nd}$, and $^{148}\text{Nd}$.

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, $R_T, f_{2}, f_{0},$ 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 independent and cumulative yields, $\bar{\nu}_{d}$ and $\bar{\nu}_{p}$ by assuming a simple form for the model inputs for $^{235}\text{U}$ and $^{239}\text{Pu}$ first. We exclude $^{238}\text{U}$ for now, as it is a threshold fissioner. The $R_{T1}$ parameter in Eq. (5) is roughly $-(R_{T}(0) - 1)/6.0$ MeV$^{-1}$ to make $R_{T} = 1$ at the opening of second chance fission, hence $R_{T1} = -0.0476$ and $-0.0507$ for $^{235}\text{U}$ and $^{238}\text{Pu}$. Similarly, $f_{21}$ and $f_{01}$ are estimated to be $f_{21} = -0.296$ MeV$^{-1}$ and $f_{01} = -0.161$ MeV$^{-1}$ for $^{235}\text{U}$, and $f_{21} = -0.430$ MeV$^{-1}$ and $f_{01} = -0.156$ MeV$^{-1}$ for $^{239}\text{Pu}$, which ensures that the even-odd effect disappears at $E_n = 6$ MeV. This is a very rough estimate of even-odd effect damping, which should be better quantified by theories in the future [26]. Here, we selected 6 MeV by just an anzatz, and roughly determined by the energy dependence of $\bar{\nu}_{d}$ as shown later.
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 $\overline{\nu_p}$ and $\overline{\nu_d}$ 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 $\overline{\nu_d}$. Hereafter, we assume $R_T$ is constant, while $f_{Z,N}$ is energy-dependent.

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

$$\frac{dY_{1,C}(Z, A, E)}{dE} \approx 0\,.$$

(28)

It is easier to see the mass region where this condition happens by calculating the derivative of the independent or cumulative yield at a particular mass number,

$$\frac{dY_{1,C}(A, E)}{dE} = \sum_{Z} \frac{dY_{1,C}(Z, A, E)}{dE} \approx 0\,.$$

(29)

We approximate the derivative by coarse numerical derivative \((Y_{1,C}(A, 2\,[\text{MeV}]) - Y_{1,C}(A, 0\,[\text{MeV}]))/2\), which is shown in Figure 2. We took this large energy interval to avoid numerical errors due to local fluctuations. The general shape of $dY_1/dE$ does not change too much in the energy range below the second chance fission. This implies the cumulative yields vary monotonously with the incident neutron energy.

The derivative plot for $^{235}$U indicates the chain yield near $A = 85, 100$, and $135$ vary slowly with the energy, while 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 chain yield in the peak regions ($A = 103$ and $133$) may show the largest reduction rate.

In Figure 3, we compare some of our calculated $Y_{1,C}(Z, A, E)$ with the experimental data of Gooden et al. [53], measurements at LANL in the critical assemblies [54], as well as other published data. From the derivative plot in Figure 2, we expect $Y_{1,C}$ of $^{235}$U

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

| $f_1$ | 0.765 | 0.718 | 4.1 | 100 |
| $\sigma_1$ | 6.06 | 6.58 | 5.2 | 20 | 100 |
| $\Delta_1$ | 20.80 | 20.1 | 1.8 | 41 | 66 | 100 |
| $F_2$ | 0.234 | 0.248 | 4.6 | 31 | 2 | 3 | 100 |
| $\sigma_2$ | 3.51 | 3.26 | 0.5 | 20 | 21 | 17 | 2 | 100 |
| $\Delta_2$ | 14.90 | 14.1 | 0.5 | 40 | 20 | 9 | 2 | 18 | 100 |
| $t_{F_0}$ | 1.00 | 2.58 | 4.4 | 1 | 21 | 42 | 1 | 0 | 69 | 100 |
| $r_{t_0}$ | 1.00 | 0.93 | 3.0 | 1 | 1 | 0 | 1 | 0 | 2 | 4 | 100 |
| $R_T$ | 1.00 | 2.00 | 2.4 | 1 | 6 | 29 | 6 | 22 | 10 | 9 | 31 | 2 | 100 |
| $t_{F_0}$ | 2.50 | 1.58 | 5.7 | 31 | 10 | 24 | 31 | 10 | 24 | 14 | 1 | 31 | 100 |

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

| $f_1$ | 0.587 | 0.625 | 3.6 | 100 |
| $\sigma_1$ | 5.405 | 5.580 | 1.4 | 16 | 100 |
| $\Delta_1$ | 22.879 | 23.128 | 0.5 | 43 | 18 | 100 |
| $F_2$ | 0.413 | 0.380 | 4.4 | 33 | 10 | 55 | 100 |
| $\sigma_2$ | 3.459 | 3.326 | 2.6 | 40 | 8 | 42 | 100 |
| $\Delta_2$ | 15.515 | 15.584 | 0.7 | 11 | 27 | 43 | 30 | 13 | 100 |
| $t_{F_0}$ | 1.00 | 2.396 | 5.3 | 0 | 16 | 25 | 20 | 14 | 48 | 100 |
| $r_{t_0}$ | 1.00 | 0.736 | 52.8 | 0 | 8 | 2 | 0 | 2 | 3 | 13 | 100 |
| $R_T$ | 1.30 | 1.327 | 1.0 | 1 | 1 | 6 | 2 | 0 | 9 | 22 | 0 | 100 |
| $t_{F_0}$ | 3.00 | 8.06 | 1.0 | 6 | 6 | 3 | 1 | 0 | 1 | 4 | 100 |
| TKE | 171.4 | 170.5 | 0.1 | 5 | 29 | 9 | 28 | 0 | 100 | 100 |

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

Figure 3. Meaning of the term ‘energy-independent’, $Y_{1,C}(A, E)$ for $^{235}$U and $^{239}$Pu is compared with experimental data from Gooden et al. [53], critical assembly measurements [54], and LANL data [55].
3.2.2. Energy dependence of $\nu_p$ and $\nu_d$

The calculated $\nu_p$ and $\nu_d$ for $^{235}$U, $^{238}$U, and $^{239}$Pu are compared with experimental data in Figures 5 and 6. The evaluated $\nu_p$ and $\nu_d$ 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, $\nu_p$ increases as the incident neutron energy goes higher, simply because of the energy conservation. However, its slope $d\nu_p/dE$ 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 $\nu_p$ by our model. Other parameters, the Gaussian shape and $f_{EA}$, also change the slope of $\nu_p$, but they have a much more modest impact on the calculated result.

The energy dependence of $\nu_d$ is caused mainly by changing the yields of the delayed neutron precursors. Interestingly the calculated and experimental $\nu_d$'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 Figure 2, we expect $\nu_d$ to decrease.

Figure 3. Energy dependence of cumulative 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 the experimental data of Gooden et al. [53], as well as other published data obtained from the EXFOR database [56,57].

decreases in the $A \approx 90$ 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 behaviors, 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 a positive slope. For $^{239}$Pu, the sign change happens twice, near $A = 130$ and 145. This is shown in Figure 4: $^{132}$Te, $^{137}$Cs, $^{140}$Ba, and $^{147}$Nd. Our model calculation also reproduces other isotopes with a similar quality.

Although we didn’t include the $^{238}$U case in Figure 2 as the cumulative yields at thermal are only given by extrapolation, Figures 3 and 4 include $^{238}$U too.
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) \( T_{1/2} = 40 \), (2) \( 8 < T_{1/2} \leq 40 \), (3) \( 3 < T_{1/2} \leq 8 \), (4) \( 1 < T_{1/2} \leq 3 \), (5) \( 0.5 < T_{1/2} \leq 1 \), and (6) \( T_{1/2} \leq 0.3 \). The fractions of each group are shown in Figure 7. In the case of \(^{235}\text{U}\), the largest contribution is from Group 4, which slightly decreases as the incident neutron energy. This is compensated for the increasing Group 2, resulting in the flat behavior of \( \nu_d \). The energy variation of each group is more visible for the \(^{239}\text{Pu}\) case. Obviously, the energy dependence of \( \nu_d \) does not originate from specific fission products but is a consequence of their competition.

As noted before, the overestimation of calculated \( \nu_d \) with the prior parameters is resolved by the adjustment procedure. However, the changes in each group are not so uniform. In the case of \(^{235}\text{U}\), the changes are; +2.8% (Group 1), −9.1(3), −15% (4), −16% (5), and 19% (6). Because Group 4 has the largest fraction to the total \( \nu_d \), this group is responsible for the reduction in \( \nu_d \).

We studied sensitivities of the model parameters to \( d\nu_d/dE \), and found that the \( f_{22} \) and \( f_{23} \) terms change the slope. When \( f_{22} = f_{23} = 0 \), or a constant odd-even effect, \( \nu_d \) decreases for both \(^{235}\text{U}\) and \(^{239}\text{Pu}\) 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 \( f_{22} \) and \( f_{23} \) 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 $\nu_d$ for $^{235}\text{U}$ drop sharply near 5 MeV [8,10], and the evaluated data often include a curious kink to reproduce this behavior. As we demonstrated that $\nu_d$ 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 $^{236}\text{U}$ to $^{235}\text{U}$. The full-extension of our fission yield calculation model by including the multi-chance
This exercise is done for the \(^{235}\text{U}\) second-chance fission calculation case only. The fission probabilities \(P_f(E)\) for the first and second chances are calculated with the CoH3 code [55]. The fission parameters, such as the fission barrier, curvature, and level density, are adjusted to reproduce the evaluated fission cross section of \(^{235}\text{U}\). We use the same \(Y_f(A)\) for the second chance, but shifted the midpoint by 1/2 mass unit to the lower mass side. \(f_f, TKE, \) and \(f_{Z,N}\) for both \(^{236}\text{U}\) and \(^{235}\text{U}\) are the same.

The calculated \(\bar{\nu}_d\) is shown in Figure 8. Although 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 \(\beta\)-delayed quantities.
such as the cumulative yields and the delayed neutron yield $\nu_d$, where consistency of prompt products is retained. The model parameters for $^{235}\text{U}$, $^{239}\text{Pu}$, and $^{238}\text{U}$ – the Gaussian functions to characterize the primary fission fragment yields, the anisothermal parameter $R_T$, the spin parameter $f_s$, 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 $^{235}\text{U}$, $^{239}\text{Pu}$ and 1.2 MeV for $^{238}\text{U}$. The result implies that a stronger odd-even effect is required to reproduce the experimental $\nu_d$ at thermal and low incident energies, which is also important for the energy-dependent calculation that was reported by Minato [23].

Anchoring the statistical decay calculations to experimental data available at the thermal energy for $^{235}\text{U}$, $^{239}\text{Pu}$, and 1.2 MeV for $^{238}\text{U}$, we extrapolated the $\text{HF}^3\text{D}$ model to the second chance fission threshold energy, and demonstrated that the calculated cumulative yields fairly reproduced the experimental data, as well as $\nu_p$ and $\nu_d$ simultaneously. The flat behavior of $\nu_d$ along the neutron-incident energy seen in the experimental data of $^{235}\text{U}$ and $^{239}\text{Pu}$ was attributed to a coincidentional compensation of increasing and decreasing delayed neutron precursors.

To examine the sudden change in $\nu_d$ 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 could reproduce the step-function-like variation of $\nu_d$. This is promising, and our $\text{HF}^3\text{D}$ model calculation for the independent and cumulative fission yields should be the most advanced tool for evaluating the fission yield 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 $\text{HF}^3\text{D}$ model qualitatively explains that the variation seen in $\nu_d$ is a result of different precursors produced by fission at each fission-chance.

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