Model Parameter Adjustment for Independent Fission Yield Mass Distribution Based on Gradient Descent Method

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1. Introduction

Fission product yields are fundamental nuclear data in the field of nuclear reactor physics, which play important role in nuclear reactor and fuel cycle calculations [1]. Independent fission yields correspond to the situation after prompt neutron emission and before the decay of fission products, which reflect the information of fission process from the macro- and microperspectives [2]. Evaluated nuclear data libraries [3-5] serve as the principal source of fission yields in current reactor engineering applications. The fission yield data of these libraries have been carefully evaluated. The evaluation process generally consists of a combination of experimental measurements and theoretical model calculations. Some fission product yields are challenging to be measured directly due to limitations in experimental conditions. In such cases, theoretical models are employed to fill in the gaps with calculated data.

The fission yield systematics [6, 7] proposed by Wahl is a classic and practical theoretical model for evaluation. Assuming that a certain fission product is an isomer I with mass number A and charge number Z, its independent fission yield $IY(A, Z, I)$ is calculated by the following systematic equation:

$$IY(A,Z,I) = MY(A) \times FI(A,Z) \times R(A,Z,I) \quad (1)$$

where $MY(A)$ is the independent fission yield mass distribution, or mass yield, which equals the summation of the independent fission yields of all fission products of mass number A; $FI(A,Z)$ is the fractional independent yield of all isomers with mass number A and charge number Z; $R(A,Z,I)$ is the isomeric yield ratio, which equals the fraction of (A,Z) produced directly as isomer I. In the framework of Wahl's systematics, these three factors are calculated by their respective models. Generally, fission yield systematics have been very successful and widely used in the evaluation process for multiple libraries such as ENDF/B-VI library [8] and its later versions, JEFF-3.1.1 library [9], and JENDL/FPY-2011 fission yield library [10]. However, the latest set of systematic model parameters, introduced by Wahl in 2002 [7], was derived through a least square fitting of experimental data available at that time, accompanied by substantial artificial empirical processing. It is necessary to replace the earlier and empirical parametrization used in the model with recent advanced experimental and theoretical knowledge [11]. Analyzing Eq. (1), it can be seen that the mass yield $MY(A)$ is the basis of systematic calculation process. For the calculation of any fission product yield, its accurate mass yield value must be determined first. Therefore, this paper will focus on adjusting the parameters of the model for mass yields. The model used to calculate mass yields is called a multi-Gaussian model, which describes the fission yield mass distribution through the linear combination of several Gaussian functions [12].

The commonly used nuclear data adjustment methods in the field of reactor physics are generally based on Bayesian theory, including Generalized Least Squares method [13, 14], Bayesian updating method [15], Bayesian Monte Carlo method [16, 17], etc. In addition, the gradient descent method is a classic numerical optimization method and is currently one of the core methods for parameter optimization in machine learning [18]. To adjust the model parameters, the loss function is generally constructed based on the model parameters, and then the parameters are updated by solving the gradient of the loss function. The final minimum value of the loss function corresponds to the optimal model parameters. Similar ideas are seldom applied to the adjustment of nuclear data and nuclear model parameters. Therefore, this paper proposes a model parameter adjustment framework based on the gradient descent method and applies it to the multi-Gaussian model. The multi-Gaussian model parameters for major fissioning systems of $^{235, 238}$U and $^{239, 241}$Pu have been adjusted based on the mass yield data of the evaluated nuclear data library ENDF/B-VIII.0 [3].

This paper is organized as follows. Section 2 introduces the multi-Gaussian model and the model parameter adjustment framework based on gradient descent method in detail. Section 3 presents the implementation of the framework, as well as the results and discussion of multi-Gaussian model parameter adjustment. Conclusions and perspectives are summarized in Section 4.

2. Theoretical Framework

This section first introduces the multi-Gaussian model, then introduces the framework for model parameter adjustment based on the gradient descent method in detail.

2.1 Multi-Gaussian Model

The mass distributions of neutron-induced fission yield generally have some common features. The distribution usually contains a pair of peaks with a valley between the two peaks. The distribution is approximately symmetrical about the center of the valley. And the central mass number of the valley is usually considered to be the average mass of the distribution [1]. Fig. 1 taken from reference [1] (Original data extracted from the JEF-2.2 evaluated nuclear data library [19]) shows the mass yield curve for the thermal neutron-induced fission of ^{235}U, which shows the above features.

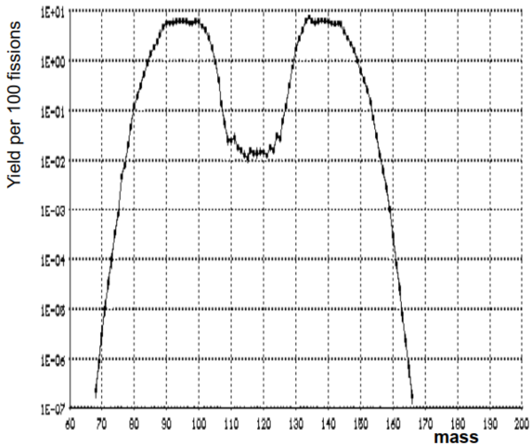

Fig. 1. The fission yield mass distribution of ^{235}U thermal neutron-induced fission.

To quantitatively describe the above features, Musgrove et al. [12] adopted five Gaussian functions (Four asymmetric and one central) to approximate the mass distribution. Later, Mills et al. did the same in the evaluation of the UKFY fission yield libraries [1, 20]. In Wahl's work [7], the model was further extended to seven Gaussian functions (Six asymmetric and one central). In order to achieve a better approximation effect, this paper also uses a seven-Gaussian model.

The multi-Gaussian model assumes that the distribution is symmetrical about an average fission product mass $\overline{A}$. On this basis, the multi-Gaussian model contains three types of unknown parameters. 1) The magnitude *N* of each Gaussian function, which represents the contribution of the Gaussian function to the mass yield. According to the constraints of binary fission, the sum of all magnitudes is 200%. 2) The width σ of each Gaussian function. 3) The offset *D* of each off-center Gaussian function from the average mass $\overline{A}$. Thus, the seven-Gaussian model is expressed as follows:

$$
\begin{aligned}
MY(A) = {} & \frac{N_1}{\sigma_1\sqrt{2\pi}}\left[e^{-\left(\frac{(A-\overline{A}-D_1)^2}{2\sigma_1^2}\right)} + e^{-\left(\frac{(A-\overline{A}+D_1)^2}{2\sigma_1^2}\right)} \right] \\
& + \frac{N_2}{\sigma_2\sqrt{2\pi}}\left[e^{-\left(\frac{(A-\overline{A}-D_2)^2}{2\sigma_2^2}\right)} + e^{-\left(\frac{(A-\overline{A}+D_2)^2}{2\sigma_2^2}\right)} \right] \\
& + \frac{N_3}{\sigma_3\sqrt{2\pi}} e^{-\left(\frac{(A-\overline{A})^2}{2\sigma_3^2}\right)} \\
& + \frac{N_4}{\sigma_4\sqrt{2\pi}}\left[e^{-\left(\frac{(A-\overline{A}-D_4)^2}{2\sigma_4^2}\right)} + e^{-\left(\frac{(A-\overline{A}+D_4)^2}{2\sigma_4^2}\right)} \right]
\end{aligned} \qquad (2)
$$

with $2N_1 + 2N_2 + N_3 + 2N_4 = 200\%$.

Next, Eq. (2) is further explained:

1) The two Gaussian functions with the **parameter subscript 1** describe the **principal peaks** of the mass distribution. This pair of curves is the principal contributor to the mass yields.
2) The two Gaussian functions with the **parameter subscript 2** describe the **inner peaks** of the mass distribution, which represent the sharp change in yields that occurs below the heavy mass number ($A_{\mathrm{H}} \approx 130$), and above the light complement.
3) The Gaussian function with the **parameter subscript 3** describes the **central valley** of the mass distribution. The function is centered on the average mass $\overline{A}$, so it is not offset to the left or right. This part of the curve varies significantly with the incident neutron energy, which will be reflected in the variety of the magnitude parameter N_3 in the model.
4) The two Gaussian functions with the **parameter subscript 4** describe the **two wings** ($A_{\mathrm{H}} > 160$) of the mass distribution. The mass yield contribution of these two curves is also very small.

Although the mass distributions of different fissioning systems have similar shapes, there are some differences in detail. Therefore, for different fissioning systems, the three types of parameters of the multi-Gaussian model need to be fitted separately with experimental data. In addition, there is still an unknown parameter in Eq. (2), which is the average fission product mass $\overline{A}$. Mills et al. previously fitted this quantity with least squares, while Wahl calculated it by the equation:

$$\overline{A} = \left(A_{\mathrm{cn}} - NT\right)/2 \qquad (3)$$

where A_{cn} is the mass number of the fission compound nuclide; *NT* is the total number of prompt neutrons, including the sum before and after fission. Wahl calculated *NT* by some empirical equations. This approach is also adopted in this paper, which will be introduced below.

First, calculate the excitation energy *PE* of fission compound nuclide:

$$PE = BN + E_{\mathrm{N}} \tag{4}$$

where *BN* is the neutron binding energy/MeV; E_{N} is the incident neutron energy/MeV. The binding energy *BN* is calculated with the mass excess data (Converted into energy units):

$$BN = MEX_{\mathrm{N}} + MEX(Z_{\mathrm{cn}}, A_{\mathrm{cn}} - 1) - MEX(Z_{\mathrm{cn}}, A_{\mathrm{cn}}) \tag{5}$$

where MEX_{N} is the mass excess of neutron, a constant of 8.071 MeV; $MEX(Z_{\mathrm{cn}}, A_{\mathrm{cn}}\text{-}1)$ and $MEX(Z_{\mathrm{cn}}, A_{\mathrm{cn}})$ represent the mass excess data of the fission target nuclide and fission compound nuclide, respectively; Z_{cn} is the charge number of the fission compound nuclide. The mass excess data of nuclides are extracted from reference [21].

Next, according to the different excitation energies, different equations are used to calculate the total prompt neutron number *NT*:

1) $PE \le 8\ \mathrm{MeV}$:

$$\begin{aligned} NT = {} & 2.286 + 0.147(Z_{\mathrm{cn}} - 92) + 0.054(A_{\mathrm{cn}} - 236) \\ & + 0.040(2 - F_{\mathrm{Z}} - F_{\mathrm{N}}) \\ & + [0.145 - 0.0043(A_{\mathrm{cn}} - 236)](E_{\mathrm{N}} - TH) \end{aligned} \tag{6}$$

where F_{Z} and F_{N} are coefficients associated with odd-even behavior; *TH* is the threshold energy/MeV. They are calculated by the following empirical equations:

$$F_{\mathrm{Z}} = [-1]^{Z_{\mathrm{cn}}}, F_{\mathrm{N}} = [-1]^{(A_{\mathrm{cn}} - Z_{\mathrm{cn}})} \tag{7}$$

$$\begin{aligned} TH = {} & 11.47 - 0.166(Z_{\mathrm{cn}})^2 / A_{\mathrm{cn}} \\ & + 0.093(2 - F_{\mathrm{Z}} - F_{\mathrm{N}}) - BN \end{aligned} \tag{8}$$

2) $PE \ge 20\ \mathrm{MeV}$:

$$NT = P_1 + (16.66 - P_1)(1.0 - e^{-0.00804 * PE}) \tag{9}$$

where P_1 is a coefficient in the fitting process:

$$P_1 = 1.563 + 0.0918(Z_{\mathrm{cn}} - 92) \tag{10}$$

3) $8\ \mathrm{MeV} < PE < 20\ \mathrm{MeV}$:

When the excitation energy is in this intermediate energy range, the value of *NT* is obtained by linear interpolation in the interval [NT_8, NT_{20}], where NT_8 is calculated with Eq. (6) when *PE* = 8 MeV, and NT_{20} is calculated with Eq. (9) when *PE* = 20 MeV.

After the above process, the calculated total number of prompt neutrons is substituted into Eq. (3) to obtain the average fission product mass $\overline{A}$. The other parameters in Eq. (2) are adjusted from Wahl's work through the model parameter adjustment framework presented later in this paper. Afterwards, the independent fission yield mass distribution of a certain fissioning system can be calculated with Eq. (2).

2.2 Framework of Multi-Gaussian Model Parameter Adjustment Based on Gradient Descent Method

The framework of adjusting the multi-Gaussian model parameters for a certain fissioning system based on gradient descent method is shown in Fig. 2. The flow of the framework can be divided into the following steps:

1) Initialization of parameters.

The initial values for the ten unknown parameters in Eq. (2) should be set carefully, because the gradient descent method updates parameters based on iteration and the initial values have a great influence on the convergence.

2) Construction and calculation of loss function.

Input the model parameters into the multi-Gaussian model to obtain the calculated mass yield vector $\boldsymbol{MY}_{\mathbf{Calc}}$. Then compare $\boldsymbol{MY}_{\mathbf{Calc}}$ with the experimental data $\boldsymbol{MY}_{\mathbf{E}}$ of the certain fissioning system to construct and calculate a loss function. The loss function is constructed to measure the predictive ability of the model.

3) Gradient calculation and parameter update.

The loss function is derived for each model parameter, and the gradient is calculated. The model parameters are updated using the calculated gradient, along with a hyperparameter called the learning rate. This idea is expressed as follows:

$$\boldsymbol{p}^{(n+1)} = \boldsymbol{p}^{(n)} - lr \cdot \nabla Loss\left(\boldsymbol{p}^{(n)}\right) \tag{11}$$

where $\boldsymbol{p}$ is a vector of model parameters; *n* is the number of iterations; *lr* is the learning rate, which controls the pace of parameter update; *Loss* is the loss function. The learning rate can be fixed or can be gradually reduced through certain algorithms during iteration.

4) Iterations of the update process.

Steps 2 and 3 are repeated for multiple iterations to continuously update the parameters. During this process the loss function will keep decreasing.

When the number of iterations is sufficient, the change of the loss function is small, or the loss function oscillates within a certain range, thus it can be judged that the model parameters have converged.

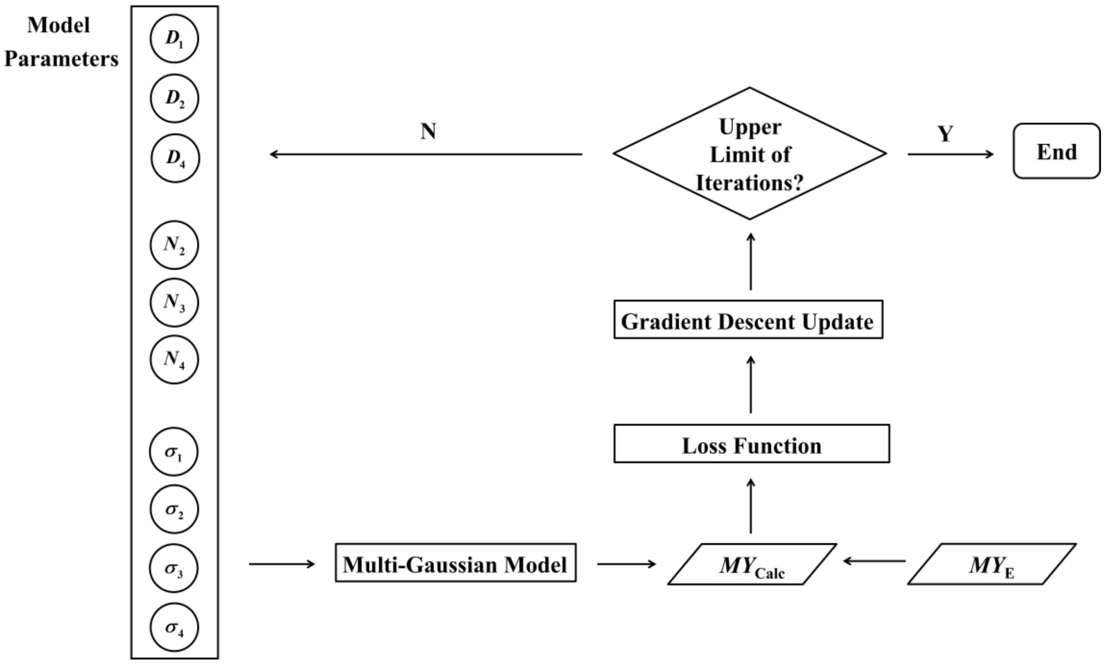

Fig. 2. The framework of multi-Gaussian model parameter adjustment.

3. Parameter Adjustment for Multi-Gaussian Model

This section introduces the implementation of the multi-Gaussian model parameter adjustment framework based on the gradient descent method. Based on the parameter values originally given by Wahl [7], the model parameters of several important fissioning systems of $^{235,\,238}$U and $^{239,\,241}$Pu are adjusted. The numerical results are given and discussed.

3.1 Implementation of the Framework

This paper implements the model parameter adjustment framework based on TensorFlow [22], a machine learning framework developed by Google. A variety of parameter optimization algorithms based on the gradient descent method have been implemented in the TensorFlow framework, thus greatly reducing the workload of code writing. At the same time, the TensorFlow framework has a built-in backpropagation algorithm [23] for efficiently solving the gradient of the loss function.

To implement the framework shown in Fig. 2, the following points need to be explained in detail.

1) The initial values of model parameters.

The initial values of the model parameters are set to be the model parameter values of Wahl, which helps to converge faster in the iterative process.

2) Construction of loss function.

The loss function used in this paper is the mean square error (MSE) of the calculated mass yields compared with the experimental data, which is expressed as follows:

$$Loss = \frac{1}{N_{\mathrm{A}}} \sum_{A=66}^{A=172} \left| MY_{\mathrm{Calc}}(A) - MY_{\mathrm{E}}(A) \right|^2 \qquad (12)$$

where N_{A} is the number of mass points, here it is 107 due to the upper and lower limits of mass point are 172 and 66, respectively, which are the same as the upper and lower limits of most evaluated nuclear data libraries; MY_{Calc} (A) and MY_{E} (A) represent calculated and experimental mass yields of mass number A, respectively.

3) Selection of gradient descent optimizer.

Different model parameters have different magnitudes and trends. To ensure the overall convergence, different learning rates in Eq. (11) should be assigned to different model parameters, and the learning rate should be adaptively adjusted during the iteration process. There are a variety of optimizers built into TensorFlow for this purpose, including Adagrad [24], Adadelta [25], RMSprop [26], Adam [27], etc.

This paper selects from the above four optimizers through the following method. Adjust the multi-Gaussian model parameters of ^{235}U thermal (0.0253 eV) neutron-induced fission based on the proposed framework with the mass yield data of evaluated nuclear data library ENDF/B-VIII.0 [3]. The parameters adjusted by different optimizers are input into the multi-Gaussian model to calculate the mass yields and compared with the results calculated based on the original parameters proposed by Wahl [7]. The metric for comparison is the reduced χ^2 which has been used in reference [1]. This metric measures how well the model fits while considering the uncertainty of the experimental data. The closer the metric value is to one, the better the model fit. The equation of reduced χ^2 is:

$$\text{reduced } \chi^2 = \sqrt{\frac{1}{DOF}\sum_{n=1}^{N_A}\frac{\left(MY_E(n)-MY_{Calc}(n)\right)^2}{\sigma^2(n)}} \quad (13)$$

where *DOF* is the degree of freedom, which is equal to the number of experimental data minus the number of model parameters, that is, 107-10=97 in this paper; $\sigma(n)$ is the absolute uncertainty of the experimental mass yield of the *n*-th mass number, which is taken from the work of England and Rider [8].

The initial learning rates of the four optimizers are all 0.01. This value has been tested. When it turns smaller, it has little effect on the adjustment results, but the convergence speed will be seriously deteriorated. Based on this, the number of iterations is selected as 400, and all four optimizers converge. The results of reduced χ^2 of different optimizers are shown in Table I. The results show that the adjustment effects of Adagrad and Adadelta are poor, while RMSprop and Adam are significantly improved compared with the original parameters. Considering that the distributions of mass yield data of different fissioning systems are similar, this paper selects the RMSprop and Adam optimizers for all the fissioning systems.

Table I: Results of reduced χ^2 of different optimizers

Original Parameters	Adagrad	Adadelta	RMSprop	Adam
33.50	33.98	33.12	14.50	14.06

4) Upper limit of iterations.

In the process of the above-mentioned optimizer selection, 400 iterations are enough to converge, and the change of the loss function with the number of iterations in Fig. 3 and Fig. 4 illustrates this. It has been tested and found that for other fissioning systems, 400 iterations can also make the loss function well converged. Therefore, the upper limit of iterations in this paper is 400. Again, the initial learning rate is 0.01.

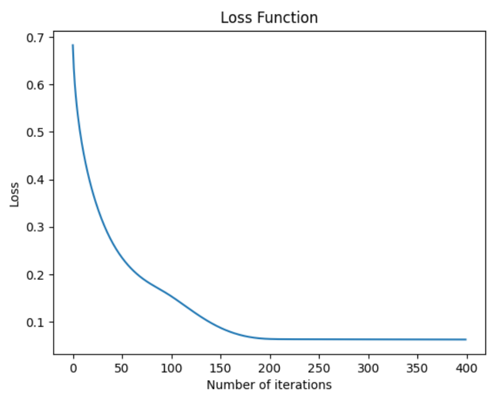

Fig. 3. The trend of the loss function as the number of iterations increases when adjusting for $^{235}U+n_{th}$ (Optimizer: RMSprop).

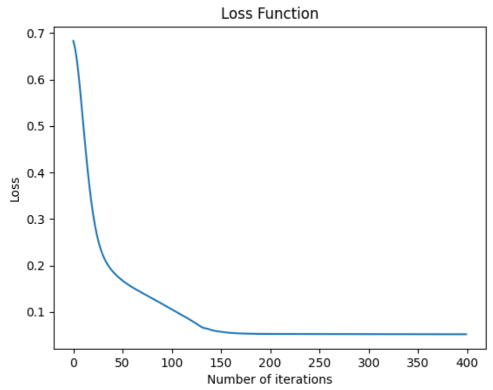

Fig. 4. The trend of the loss function as the number of iterations increases when adjusting for $^{235}U+n_{th}$ (Optimizer: Adam).

3.2 Numerical Results

To exclude the influence of the quality of the experimental data when testing the method, this paper takes the mass yield data of the evaluated nuclear data library ENDF/B-VIII.0 as the simulated experimental data. The multi-Gaussian model parameter adjustment is mainly aimed at thermal (0.0253 eV) neutron, fast (0.5 MeV) neutron, and high-energy (14 MeV) neutron-induced fission. Specifically, the fissioning systems adjusted in this paper include (The subscript 'th' means thermal neutron, 'f' means fast neutron, and 'he' means high-energy neutron): $^{235}U+n_{th}$, $^{235}U+n_f$, $^{235}U+n_{he}$, $^{238}U+n_f$, $^{238}U+n_{he}$, $^{239}Pu+n_{th}$, $^{239}Pu+n_f$, $^{239}Pu+n_{he}$, $^{241}Pu+n_{th}$, $^{241}Pu+n_f$. For comparison, the results based on the original model parameters are calculated with the fission yield calculation code CYFP [7] developed by Wahl in 2002.

3.2.1 Fission Yield Mass Distributions

Mass yields calculated based on adjusted parameters and Wahl's original parameters are compared with data of ENDF/B-VIII.0. The comparison is shown in Fig. 5 to Fig. 14 for ten fissioning systems.

Overall, the degree of agreement with the ENDF/B-VIII.0 data is significantly improved after the model parameter adjustment, especially at the positions of the two principal peaks and the two inner peaks (The sharp change in yields that occurs below the heavy mass number, $A_H \approx 130$). At these positions, the height and width of each Gaussian curve is significantly adjusted. In addition, for high-energy neutron-induced fission, the adjusted central valley of the distribution agrees much better with the evaluated nuclear data.

However, it can still be seen from the figures that the adjusted results do not completely fit the evaluated nuclear data. In the curves of evaluated nuclear data, there are fluctuations especially at the two main peaks and the central valley. These fluctuations are believed to be caused by odd-even and shell effects [1]. The multi-Gaussian model uses smooth Gaussian functions to fit the mass distribution curve, so it is difficult to describe these fluctuations. Furthermore, these fluctuations cause the fission product mass distribution curve to be not symmetrical about the average mass, which conflicts with the symmetry approximated by the multi-Gaussian model.

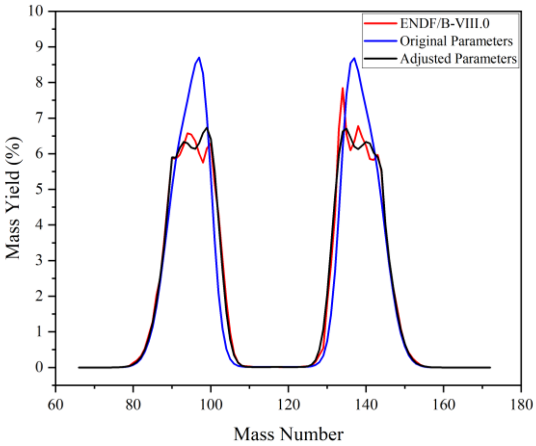

Fig. 5. Mass yield comparison for $^{235}U+n_{th}$.

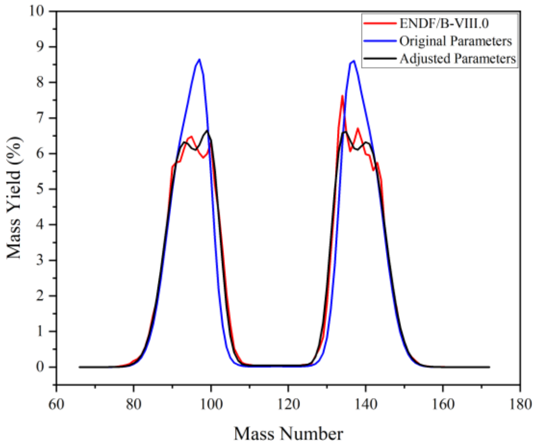

Fig. 6. Mass yield comparison for $^{235}U+n_{f}$.

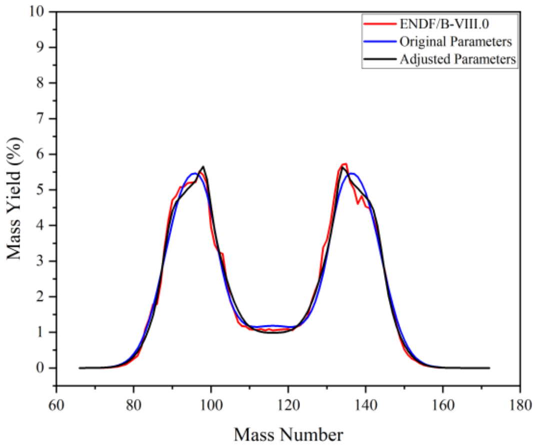

Fig. 7. Mass yield comparison for $^{235}U+n_{he}$.

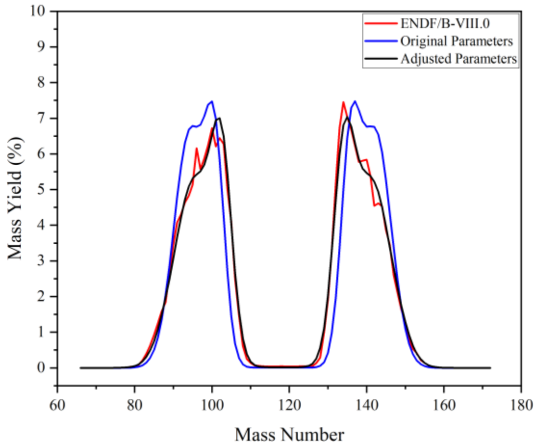

Fig. 8. Mass yield comparison for $^{238}U+n_{f}$.

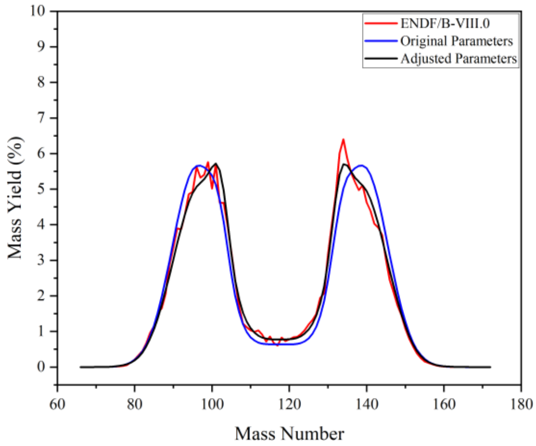

Fig. 9. Mass yield comparison for $^{238}U+n_{he}$.

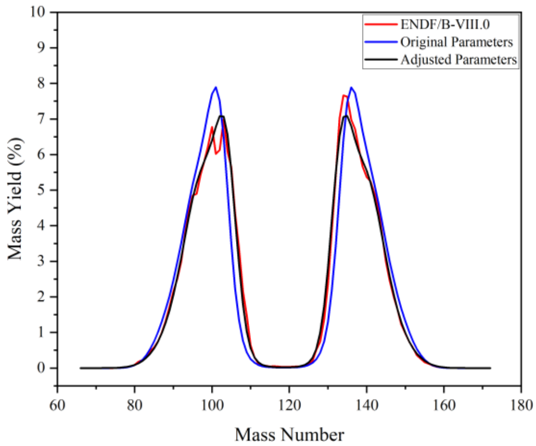

Fig. 10. Mass yield comparison for $^{239}Pu+n_{th}$.

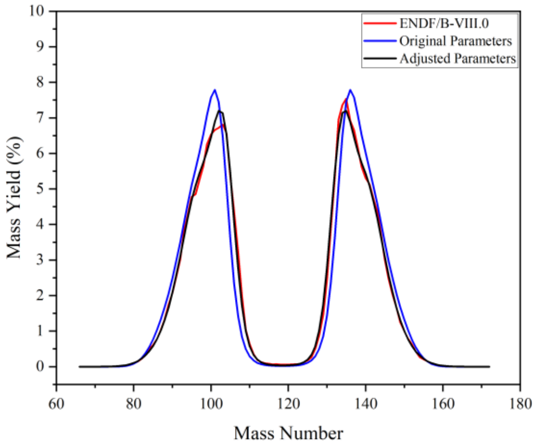

Fig. 11. Mass yield comparison for $^{239}Pu+n_f$.

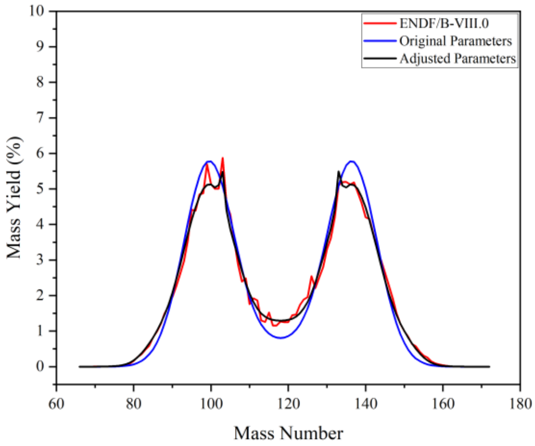

Fig. 12. Mass yield comparison for $^{239}Pu+n_{he}$.

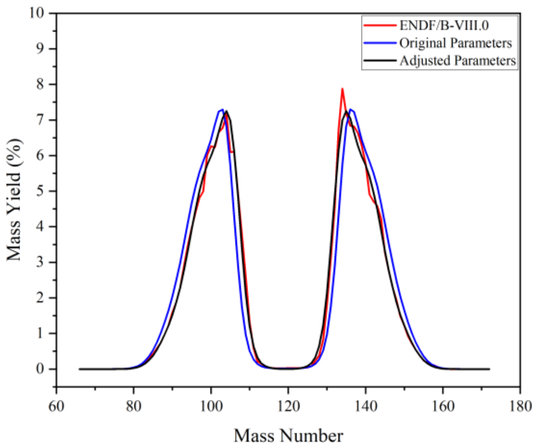

Fig. 13. Mass yield comparison for $^{241}Pu+n_{th}$.

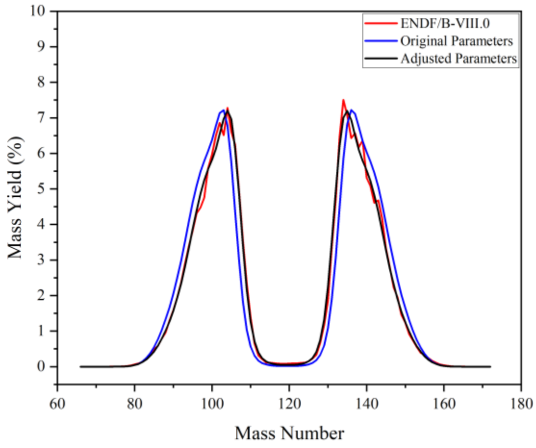

Fig. 14. Mass yield comparison for $^{241}Pu+n_f$.

3.2.2 Reduced χ^2

The reduced χ^2 calculated with Eq. (13) of each fissioning system before and after adjustment is summarized in Table II. This table also summarizes the optimizer used for each fissioning systems. Since the adjustment of the model parameters of each fissioning system is carried out independently, the suitable optimizers are not the same.

Among these results, the thermal neutron-induced fission of ^{241}Pu has a large reduced χ^2 before adjustment, although it is significantly reduced after adjustment, it is still greater than that of other fissioning systems. It is found that the mass yields of mass numbers 162~167 calculated by the multi-Gaussian model are much higher than the data of ENDF/B-VIII.0 for $^{241}Pu+n_{th}$. This phenomenon also exists for $^{239}Pu+n_{th}$. It is believed to be mainly due to the original model parameters in the CYFP code are unreasonable for these two fissioning systems in the two wings of the mass distribution, so this paper is still affected when the parameters are adjusted.

Overall, the reduced χ^2 in Table II are all reduced after adjustment, which illustrates the effectiveness of the model parameter adjustment. Nevertheless, some reduced χ^2 values are still significantly greater than one after adjustment. For this phenomenon, in addition to the influence of the above-mentioned multi-Gaussian model itself, the combination of manually determined TensorFlow hyperparameters adopted in this paper also has an influence. The hyperparameters include the type of loss function, the number of iterations, the optimizer, and the parameters of the optimizer (Such as the initial learning rate). The above results are based on the combination of MSE-type loss function, 400 iterations, RMSprop or Adam optimizer, and an initial learning rate of 0.01 (Other parameters of the optimizer are default values). There is still room for further exploration in the combination. By testing different combinations of hyperparameters, the effect of adjustment has the potential to become better.

Table II: Summary of reduced χ^2 and optimizers

Fissioning System	Reduced χ^2 Based on Original Parameters	Reduced χ^2 Based on Adjusted Parameters	Optimizer
$^{235}U+n_{th}$	33.98	14.06	Adam
$^{235}U+n_{f}$	30.02	8.58	Adam
$^{235}U+n_{he}$	3.92	2.84	Adam
$^{238}U+n_{f}$	26.09	8.55	Adam
$^{238}U+n_{he}$	6.17	3.19	RMSprop
$^{239}Pu+n_{th}$	26.28	18.88	RMSprop
$^{239}Pu+n_{f}$	23.28	8.79	RMSprop
$^{239}Pu+n_{he}$	4.02	1.77	Adam
$^{241}Pu+n_{th}$	274.77	110.42	Adam
$^{241}Pu+n_{f}$	11.03	2.56	RMSprop

4. Conclusions and Perspectives

In this paper, a framework for adjusting model parameters based on the gradient descent method is proposed and applied to the multi-Gaussian model of independent fission yield mass distribution. Based on the mass yield data of ENDF/B-VIII.0, model parameters are adjusted for ten fissioning systems of $^{235,238}U$ and $^{239,241}Pu$. The results calculated based on the adjusted model parameters has better agreement with the evaluated nuclear data than those based on original parameters. It illustrates that this framework will improve the performance of the multi-Gaussian model in the fission yield evaluation process and will also improve the overall evaluation accuracy of fission yield systematics.

In the future, multi-Gaussian model parameters for more fissioning systems can be adjusted based on this framework, and the hyperparameter combinations within the framework can be further tested. Furthermore, neural networks can be added to this framework for learning the physical laws in the mass yields of different fissioning systems to find out correlations between model parameters and fissioning systems.

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