PAPER

The investigation on fragments mass distributions for photofission of $^{232}\text{Th}$, $^{238}\text{U}$, $^{237}\text{Np}$ and $^{240}\text{Pu}$ isotopes by systematic methods

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Abstract

Three neutron induced models have been investigated for photofission of $^{232}\text{Th}$, $^{238}\text{U}$, $^{237}\text{Np}$ and $^{240}\text{Pu}$ isotopes. The fission fragment mass distribution yields were calculated and compared with photofission experimental data on a logarithmic scale. The peak to valley ratio, average light mass ($\langle A_L \rangle$) and heavy mass ($\langle A_H \rangle$) for photofission of $^{232}\text{Th}$ and $^{238}\text{U}$ were calculated at different excitation energies. Also, the yields of symmetric and asymmetric fission products were calculated. The Gorodisskiy model were modified to fit the photofission experimental data on a logarithmic scale. The comparison between calculated results and the experimental data shows that the mentioned neutron models should not be used for the description of photofission reactions.

1. Introduction

Although photofission was discovered more than fifty years ago [1], but its theoretical modeling was produced with many challenges. After the Wilkins presented the fragment mass distribution by Gaussian framework in scission point model [2–4], many researcher developed the multi-Gaussian model up to now [5–10]. Some models have the complicate forms [11] or complicate method [12] and some others have many parameters [13]. In the other hand, abundance models have presented for neutron and proton fission, but the rare works have done in photofission phenomenon [14]. Because of this point, the yields of photofission products are needed to be studied in systematic way. In other hand, the photofission phenomenon is similar to neutron induced fission, then we investigated the photofission fragments mass yields base on systematic method of neutron induced fission.

At first, three important models in neutron induced fission have been discussed. The first systematic model in neutron/proton fission is the Gorodisskiy model [15] in which the variances and average mass are calculated by mass and charge number of compound nuclei. Although the original model has not proper results in photofission process, but [16] modified this model for photofission phenomena to reach the experimental results on a normal scale. The second model is the Robert Mills model [17], which is the easiest model to calculate fission fragment mass distribution yields (section 3). As an old model, the last model is the Wang Fu-Cheng model [18] (section 4).

In section 5, the fission fragment mass distribution of those models are compared with photofission experimental data of $^{232}\text{Th}$, $^{238}\text{U}$, $^{237}\text{Np}$ and $^{240}\text{Pu}$ isotopes on a logarithmic scale. The average light mass number and heavy mass number, the peak to valley ratio ($P/V$) and symmetric and asymmetric fission products are calculated for photofission of $^{232}\text{Th}$ and $^{238}\text{U}$, subsequently. They were compared to experimental data to improve the models.

For rare actinides, such as $^{232}\text{Th}$ [19–23] and $^{235,238}\text{U}$ [24–26], the yields of bremsstrahlung-fission products are available in a wide energy range. For other actinides, such as $^{237}\text{Np}$ [27] and $^{240}\text{Pu}$ [28], the yields of bremsstrahlung-fission products are available within the limited bremsstrahlung energy range. Since the experimental data on this process are not find abundantly, two first actinides are chosen for calculation of their characters such as heavy and light fragment mass average and the peak to valley ratio.
2. The modified Gorodisskiy model

The new model [16], which has developed the Gorodisskiy model in photofission phenomena, is fixed the structure of original model and reclaimed its parameters for photofission phenomena. Here’s a brief overview of the relations of this model to plot the photofission fragment mass distribution:

The heavy mass fragment yields in Gaussian form with little deviation term of Charlier’s distribution is presented as

\[ Y_{H} = \frac{1}{\sqrt{2\pi}} \left( \frac{Ye^{-u_{\text{sym}}^2/\sigma _{\text{sym}}^2}}{\sigma _{\text{sym}}} + \frac{Y_{a}e^{-u_{\text{asym}}^2/\sigma _{\text{asym}}^2}}{\sigma _{\text{asym}}} \right), \]

where \( f(u) \) is the Charlier’s distribution which defined as [15, 29, 30]

\[ f(u) = 1 - \gamma _2(u/2 - u^3/6) + \gamma _2(u^4/24 - u^2/4 + 1/8), \]

and we have \( u_{\text{sym}} = \frac{A - A_{\text{sym}}}{\sigma _{\text{sym}}}, u_{\text{asym}} = \frac{A - A_{\text{asym}}}{\sigma _{\text{asym}}}, A_{\text{sym}} = \frac{A_{\text{sym}}}{2} \) and

\[ A_{\text{asym}} = \begin{cases} 54 \frac{A_{\text{sym}}}{Z_{\text{cn}}} & \text{for } Z_{\text{cn}} = 90 \\ 28.6 \frac{A_{\text{sym}}}{Z_{\text{cn}}} + 0.708Z_{\text{cn}} & \text{for } Z_{\text{cn}} \geq 92. \end{cases} \]

The \( \sigma _{\text{sym}} \) and \( \sigma _{\text{asym}} \) are the mass variance for asymmetric and symmetric fission, respectively. They were parameterised as

\[ \sigma _{\text{sym}} = \frac{0.031 A_{\text{sym}} \sqrt{E}}{\sqrt{90.54 - 1.9 \frac{Z_{\text{cn}}^3}{A_{\text{sym}}}}} + 9.64 \]

and

\[ \sigma _{\text{asym}} = \frac{A_{\text{sym}}(Z_{\text{cn}} - 73)(0.074 + 0.029 \sqrt{E})}{Z_{\text{cn}}}, \]

where \( A_{\text{sym}} \) and \( Z_{\text{cn}} \) are the mass and charge for component nucleus. \( E \) is the end-point energy in MeV. But the \( E \) in \( \sqrt{E} \) is used without dimension. The \( Y_{a} \) and \( Y_{s} \) are the relative contributions of the symmetric and asymmetric modes of fission, respectively. These parameters were evaluated by new ratio as

\[ \frac{Y_{a}}{Y_{s}} = 1.244 \left( 1 - e^{-0.0027 (E - 5.71)^{1/3}} \left( \frac{\sqrt{E}}{100} \frac{Z_{\text{cn}}}{A_{\text{sym}}} - 0.4 \right) \right), \]

which led us to calculation of values of \( Y_{a} \) and \( Y_{s} \) with the following two relations:

\[ Y_{a} = 200 \left( \left| \frac{Y_{a}}{Y_{s}} \right| + 2 \right)^{-1} \]

and

\[ Y_{s} = 200 - 2Y_{a}. \]

After all, the light fragment mass yield must be calculated as the heavy fragment relation (equation (1)) by

\[ Y_{L(A)} = Y_{H(A_{\text{asym}} - A)}. \]

Finally, the photofission fragments mass yields are achieved by

\[ Y = Y_{H} + Y_{L}. \]

3. The Robert William Mills model

In 1995, Robert William Mills [17] examined the mass yields of neutron fission for many nuclei at low energy and found that the fluctuations of calculation results can be neglected by changing energy. Therefore, the independent energy model was born by following relations:

\[ Y = \frac{n_{1}}{\sigma _{1}} e^{\frac{(M-h)^2}{2\sigma _{1}^2}} + e^{\frac{(M-h)^2}{2\sigma _{2}^2}} + \frac{n_{2}}{\sigma _{2}} e^{\frac{(M-h)^2}{2\sigma _{2}^2}} + e^{\frac{(M+h)^2}{2\sigma _{2}^2}} + \frac{n_{3}}{12} e^{\frac{M^2}{3\sigma _{2}^2}}, \]

where \( M = A - A_{\text{sym}}/2, \)

\[ \sigma _{1} = 0.2017 A_{\text{sym}} - 42.906, \]

\[ \sigma _{2} = 0.1125 A_{\text{sym}} - 24.375, \]

and

\[ n_{1} = 0.0027 A_{\text{sym}} - 24.375. \]
\[ d_1 = 183.113 \times 60 - 0.678 \times 32 A_{in} + 0.013 \times 664(A_{in} - 230)^2, \]
\[ d_2 = 156.750 - 0.595 A_{CN} + 0.001 \times 250(A_{in} - 230)^2, \]
\[ n_1 = 0.000 \times 384 A_{in} + 0.621 \times 5, \]
\[ n_2 = 0.286, \]
\[ n_3 = 2 - 2 n_1 - 2 n_2 \]

and \( A_{CN} \) is mass numbers of compound nucleus. We need to multiply the equation (10) in \( \frac{100}{22} \) compare with the experimental data. This model has linear coefficients and five independent parameters.

### 4. The Wang Fu-Cheng model

In Wang Fu-Cheng model [18], the fission mass distribution is presented by a superposition of three modes of fission (S1, S2 and S3). A residual function \( \phi \) is presented to equation for fitting the calculated results with experimental data. However, it could be neglected at low energy range (MeV). The three modes of fission are presented as [31]:

\[ Y_S = \frac{C_3}{\sqrt{2\pi} \sigma_2} e^{-\frac{(A-A_3)^2}{2\sigma_2^2}}, \]
\[ Y_S = \frac{C_3}{\sqrt{2\pi} \sigma_2} e^{-\frac{(A-A_3)^2}{2\sigma_2^2}}, \]
\[ Y_S = \frac{C_3}{\sqrt{2\pi} \sigma_2} e^{-\frac{(A-A_3)^2}{2\sigma_2^2}}, \]

where

\[ A_1 = 141 - 0.053 E, \]
\[ A_2 = 82.3 + 0.293 N_{CN} + 0.1 Z_{CN} - 0.03 E, \]
\[ \sigma_1 = 30.276 + 0.24 N_{CN} + 0.12 E, \]
\[ \sigma_2 = (1.884 - 0.0094 4 N_{CN} + 0.267 e^{- (N_{CN}-142.5)^2} + 0.114 e^{- (N_{CN}-146.81)^2}) \sigma_3, \]
\[ \sigma_3 = 1.4 \sigma_1, \]
\[ C_1 = 451.934 - 2.66 N_{CN} + 0.19 (A_{CN} - 232.2) E, \]
\[ C_2 = 59.3 - 0.263 N_{CN} - 0.017 (A_{CN} - 235.7) E, \]

and \( C_3 = 0.01 e^{0.49 E} \). The \( Z_{CN}, N_{CN} \) and \( A_{CN} \) are proton numbers, neutron number and mass numbers of compound nucleus, respectively. \( E \) is the bremsstrahlung induced energy in MeV for photofission phenomena. The calculated mass distribution is normalized by the \( C \) coefficient as

\[ C = 100(C_3/2 + C_2 + C_1)^{-1}. \]

The fragments mass distribution yield in percentages are calculated by:

\[ Y = C(Y_{S1} + Y_{S2} + Y_{S3}). \]

### 5. Results and discussion

#### 5.1. The fragments mass distribution and photofission experimental data

The fragments mass distributions for photofission of \(^{232}Th \), \(^{238}U \), \(^{237}Np \) and \(^{240}Pu \) isotopes are presented in figures 1–4 on a logarithmic scale. The calculated results of three neutron-fission models accompany with photofission experimental data show that the calculated results are not in agreement to experimental data exactly. In [16], the photofission fragments mass distributions in normal scale were studied only for one model (the Modified Gorodisskiy model). So here, we look more accurately using the logarithmic scale. The figures 1–4 show that the results of all models are almost near photofission experimental data, but they can not calculate the exact values.

For photofission of \(^{232}Th \), the calculated results by using the Modified Gorodisskiy model are better than other models, but it can not calculate the third peak (figure 1). The third hump of photofission of \(^{232}Th \) is not achieved from any models even [14]. In the final section, we will discuss it. For photofission of \(^{238}U \), the calculated results of Robert Mills model are closer to experimental data than other models (figure 2). For photofission of \(^{240}Pu \), the results of the Wang Fu-Cheng model are closer to experimental data than others (figure 4).

Over the 20 MeV bremsstrahlung energy, the Wang Fu-Cheng model can not be used in photofission phenomena. It is because the symmetry mode of neutron fission model increase with increasing energy. It is
clear that symmetry coefficient of this model depends on excitation energy as exponential function. We rewritten that coefficient such as the R Mills model \( (i.e. C_L = 100 - C_1 - C_2) \) but the calculated results did not fit, again.

5.2. The peak to valley ratio

The peak to valley ratio of two models are presented in tables 1 and 2. Although the end-point energy applied to calculate the mass distribution, but the (average) excitation energy must be used to calculate the P/V ratio. The calculated results of these models, similar the other literatures \([23, 28, 32, 33]\), decrease with increasing excitation energy. The decreasing rate is not exactly same as other works, but the trend of changing is the same.

Although the calculated values by the Modified Gorodisskiiy model decrease by increasing the excitation energy, but there is a substantial difference between the calculated values of the Modified Gorodisskiiy model and experimental values. It is seen that the results of the Wang Fu-cheng model is closer to the experimental data than results of the M Gorodisskiiy model. In the last line of table 1, it is seen a significant error in calculation results over 20 MeV endpoint energy in the Wang Fu-cheng model.
Also, Table 2 shows that the differences between the calculated values and experimental data for photofission of $^{232}\text{Th}$ are more considerable than those for photofission of $^{238}\text{U}$. It can be due to the difference in the type of potential barrier of $^{232}\text{Th}$ and $^{238}\text{U}$ nuclei.

![Figure 3](image-url)  
*Figure 3.* Comparison of the calculated photofission fragments mass distribution with experimental data [27] in logarithmic scale for $^{237}\text{Np}$ at 9.5 MeV end-point bremsstrahlung energy.

![Figure 4](image-url)  
*Figure 4.* Comparison of the calculated photofission fragments mass distribution with experimental data [28] in logarithmic scale for $^{239}\text{Pu}$ at 10 MeV end-point bremsstrahlung energy.

### Table 1

|       | $^{232}\text{Th}$ | $^{238}\text{U}$ |
|-------|-------------------|------------------|
| Energy| calculated results | Naik [28]         | Energy| calculated results | Naik [28]         |
| 8 Mev | 219.7             | 696              | 9 MeV | 86                | 310.4            |
| 10 Mev| 83                | 26.6             | 20 MeV| 21                | 24.3             |
| 15 Mev| 8.07              | 9.7              | 25 MeV| 93                | 16–19            |

Also, Table 2 shows that the differences between the calculated values and experimental data for photofission of $^{232}\text{Th}$ are more considerable than those for photofission of $^{238}\text{U}$. It can be due to the difference in the type of potential barrier of $^{232}\text{Th}$ and $^{238}\text{U}$ nuclei.
5.3. The average heavy and light mass fragments

The average light mass ($\langle A_L \rangle$) and heavy mass ($\langle A_H \rangle$) are calculated within the mass ranges of 80–105 and 125–150 by using the following relations [23, 34]:

$$\langle A_L \rangle = \frac{\sum(A \times Y_L)}{\sum Y_L}, \quad \langle A_H \rangle = \frac{\sum(A \times Y_H)}{\sum Y_H}.$$  (30)

The calculated values of $\langle A_L \rangle$ for photofission of $^{232}$Th and $^{238}$U are presented in figure 5. According the Naik work ([26]), the $\langle A_L \rangle$ values decrease by increasing excitation energy in photofission of $^{238}$U. While it is only happen with less slope for the calculated results of Wang Fu-cheng model in figure 5. The $\langle A_L \rangle$ values, according to the [23], increase sharply for photofission of $^{232}$Th but it is happened only for results of the the Wang Fu-cheng model. It expresses that this model calculates the fission characteristics better than other models, if the end-point bremsstrahlung energy does not exceed 20 MeV. In spite of other literatures, the calculated values of $\langle A_L \rangle$ by M Gorodissky model remain constant approximately for these two nuclei.

The $\langle A_H \rangle$ values for photofission of $^{238}$U (left) and $^{232}$Th (right) are presented in figure 6. According to the [23], the $\langle A_H \rangle$ values decrease as the increase of excitation energy in photofission of $^{232}$Th. The results of the Wang Fu-cheng model have a similar trend whereas the results of M Gorodissky model remain constant about 139.1 ± 0.02 (right part of figure 6). From the experimental data of [26], it can be seen that the $\langle A_H \rangle$ values increase slowly with the increase of end-point energy for photofission of $^{238}$U. The results of the M Gorodissky model have the same increase but the results of the Wang Fu-Cheng model has a huge error over 20 Mev endpoint energy (left part of figure 6).

In addition, the differences between calculated results and experimental data for photofission of $^{232}$Th are very deeply while they are close together in photofission of $^{238}$U. It is because of this fact that the mass yield distribution for the photofission of $^{232}$Th is triple-humped whereas that the photofission of $^{238}$U component is double-humped or it is due to the different type of potential barrier of these two nuclei. Even the fissility values ($Z^2/A$) of this nucleus is very special which cause the results of calculating get out of normal values. It is

| Energy (MeV) | $^{232}$Th calculated results Naik [28] | $^{238}$U calculated results Naik [28] |
|-------------|----------------------------------------|----------------------------------------|
| 8           | 8027                                   | 9 MeV                                  |
| 10          | 2037                                   | 20 MeV                                 |
| 15          | 1299                                   | 25 MeV                                 |
| 25          | 87                                     | 40 MeV                                 |
| 35          | 36                                     | 70 MeV                                 |

Table 2. The calculated peak to valley ratio using the modified Gorodissky model for $^{232}$Th and $^{238}$U nuclei along with experimental data. The first columns are end-point bremsstrahlung energy.
recommended that the formula of the thorium be separated from other actinide, such as Modified Gorodisskiy model (section 2).

The Robert Mills model is independent from energy, so it does not change with increasing excitation energy. Therefore, it could not be compared with experimental works. The slight increasing and decreasing of calculated values from these models, despite of experimental results, show that the results of these models are not accurate for photofission phenomena.

5.4. The yields of symmetric and asymmetric fission products

The yields of symmetric (for \( A = 117 \)) and asymmetric (for \( A = 134 \)) photofission products of \(^{238}\text{U}\) for the Wang Fu-Cheng and the Modified Gorodisskiy models are presented in figure 7. According to the other literatures [26, 28, 33], the yields of asymmetric products are decreased, whereas the symmetric products are increased sharply up to 20 MeV excitation energy. It is clearly seen that the results of Wang Fu-Cheng model are better than the results of M Gorodisskiy model.
Otherwise, the calculated yields of symmetric and asymmetric fission products by Wang Fu-Cheng model are not correct over 20 MeV endpoint energy. Because according the experimental data \[26, 28\], the value of symmetric yields at 19.9 MeV excitation energy equal 0.7 but it is about 4 by using the Wang Fu-Cheng model.

The results of M Gorodisskiy model are constant for asymmetric fission products (left part of figure 7) and the symmetric fission products values are much less than the experimental data. This is another remarkable difference, indicating that this model must be modified for photo-fission phenomena. Also, the right part of figure 7 shows that the yields of symmetric products calculated by the Modified Gorodisskiy models differ from the experimental data. To correct this problem, we introduced the $\beta$ parameter in equation (6) as follows:

$$
\frac{Y_d}{Y_s} = 1.244 (\beta e^{-0.0027 (E - 5.7)^{1/2}}) \left( \sqrt{E} + 100 \left( \frac{Z_{on}}{A_{on}} - 0.4 \right) \right).
$$

By fitting the calculated results to the experimental data, we have $\beta = 1.17$. For $\beta = 1$ and 1.17, the photofission fragments mass distributions of $^{232}\text{Th}$ and $^{238}\text{U}$ are plotted and compared with experimental data in figure 8. It is seem that the third hump of photofission of $^{232}\text{Th}$ could be calculated by changing the value of $\beta$.

6. Conclusion

The Wang Fu-Cheng model could not be used over 20 MeV endpoint energy, but at low energy, the calculated results of this model are better than others for photo-fission of $^{240}\text{Pu}$. For photofission of $^{232}\text{Th}$, the results of Modified Gorodisskiy model is better than others. For photofission of $^{238}\text{U}$, the Robert Mills model has better results. Although the photofission fragment mass distributions of some models are closer to the experimental data, but they cannot predict the experimental data with considerable accuracy. The Modified Gorodisskiy model could calculate the third hump of photofission of $^{232}\text{Th}$ by adding the $\beta$ value.

The increase of the yield of symmetric-product and decrease of the P/V ratio is achieved by Modified Gorodisskiy model for photofission of $^{238}\text{U}$, but they did not reach the exact values of experimental works. Despite of the $^{238}\text{U}$ fissioning results, the $^{232}\text{Th}$ fissioning results have huge differences with experimental data for all models, which could be called the $^{232}\text{Th}$ anomaly. It is better to separate the formulas of this nucleus from other nuclei.

The yields of asymmetric products are constant by using the M Gorodisskiy model, while they are slightly decreased in other literatures. The Wang Fu-Cheng model calculates the yields of symmetric fission products and $\langle A_d \rangle$ and $\langle A_s \rangle$ near the experimental values up to 20 MeV endpoint energy for photofission of $^{238}\text{U}$. It is an strong point of this model. Over 20 MeV endpoint energy, the results of this model has not a good agreement to the experimental works. We chose the symmetric mode coefficient ($S_1$) of this model as the Robert Mills model coefficient, but still the results were not correct.

The differences of the $\langle A_{ij} \rangle$ and $\langle A_i \rangle$ values with experimental works could be related to the number of the pre- and post-neutrons which has not included in all described models. The Robert Mills model is independent...
from energy, therefore, its characters (i.e. P/V, yields of symmetric and asymmetric fission products, \(A_H\) and \(A_L\)) can not be calculated by increasing the endpoint energy.

It is seen that only peaks of mass distributions are reproduced by these models satisfactorily. All other calculated results differ substantially from the experimental values. So it shows that the above neutron models should not be used for the description of photo-fission reactions.

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