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The dependence of cumulative ^{238}U(n,f) fission yield on incident-neutron energy

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Abstract This work is aim at studying the dependence of fission yields on incident neutron energy, so as to produce evaluated yield sets of the energy dependence. Experimental data at different neutron energies for gas fission products ^{85m,87,88}Kr and ^{138}Xe resulting from the ^{238}U(n, f) reaction are processed using codes AVERAGE for weighed average and ZOTT for simultaneous evaluation. Energy dependence of the cumulative fission product yields on the incident neutron is presented. The evaluated curve of product yield is compared with the results calculated by the TALYS-0.64 code. The present evaluation is consistent with other main libraries in error permission. The fit curve of ^{87,88}Kr can be recommended to predict the unmeasured fission yields. Comparisons of the evaluated energy dependence curves with theoretical calculated results show that the predictions using purely theoretical model for the fission process are not sufficiently accurate and reliable for the calculations of the cumulative fission yields for the ^{238}U(n,f).

Key words Cumulative yield, Evaluation, ^{238}U, Gas nuclides

1 Introduction

For developing new reactors and recycling nuclear wastes, fission product yields of actinides should be known as a function of incident-particle energy. The energy dependence of fission yields, however, is a quite complex problem, due to the complicated fission mechanism. Studies, experimental or theoretical, are needed to seek answers to many open questions in this field of research[1]. In the theoretical studies, several models have been developed, such as combining the nuclear reaction code ALICE-91[2] with a temperature-dependent version of the BROSA model[3], combining the BROSA model with TALYS-0.64[4] (a newly developed nuclear reaction code), and using the five-Gaussian[5]. The new methods can predict mass yield of the fission fragments and products, but most of them are suitable for intermediate energy range.

Data evaluation is a useful way for having a better understanding of the fission yield, especially for fission reactions induced by lower energy neutrons. Rider[6] has produced successive versions of his libraries over the years. Wahl[7] has spent many years in evaluating especially fraction- independent fission yields. Mills[8] has evaluated independent and cumulative fission yields systemically. However, only the cumulative yields in fission spectrum and around 14 MeV can be found in major international nuclear data libraries of ENDF/B-VII[9], JEF-2.2[10], JENDL-3.2[11], and CENDL-2[12] and other databases such as UCRL-51458[13]. With the growing nuclear data form new experiments, it is necessary to update the evaluated data.

Because of large yield, long half life and convenience in their measurement, some inert gas nuclides can be used to study the energy dependence of yields for neutron-induced actinide fission reactions,

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such as ^{85m}Kr, ^{87}Kr, ^{88}Kr and ^{138}Xe from the ^{238}U(n, f) reaction. This work is to evaluate new experimental data of these nuclides to produce a recommended set of values, for dependence of cumulative yields on incident-neutron energy below 15 MeV. The recommended energy dependence curves of yields are compared with the results calculated by the code TALYS-0.64.

2 Evaluation method

2.1 Data collection and selection

The experimental data were obtained in recent years by measurements, absolute or relative, of radiochemistry and mass-spectrometry, γ-ray spectrometry with chemical separation of fission products, fission product recoil separators and isotope-separator on-line systems[14]. Experimental data from the IAEA nuclear data services, the experimental nuclear reaction database EXFOR[15] and the neutron reaction bibliography CINDA[16] are used in this study.

The experimental data are evaluated by the fission yield data evaluation system FYDES[17]. The EXFOR bibliographic information sections and papers concerned are studied carefully and analyzed in physics. A decision to take or drop the data is made according to the date and method of measurement, and the data discrepancy.

2.2 Experimental data correction

The yields are corrected for gamma intensity and fission cross section with the recent standards recommend by IAEA nuclear data services[9,18,19], and the measurement-provided relative values are updated using modern reference values[20].

2.2.1 Cumulative yield and chain yield

In the present evaluation, the cumulative yield (CUM) data are used to describe the relationship between the fission yields and the incident-neutron energy. The relevant chain yield (CHN) data are used if the CUM does not differ greatly from CHN. The factor α is defined to describe the difference, namely

$$\alpha=(Y_{\mathrm{CUM}}/Y_{\mathrm{CUM}})\times 100\%. \quad (1)$$

The differences between of CUM and CHN are less than 1% for ^{85m,87,88}Kr at fission spectrum and 14MeV, for ^{138}Xe at fission spectrum, but about 9% for ^{138}Xe at 14 MeV. Thus, the CHN data of ^{85m,87,88}Kr are used to substitute CUM directly. The CHN data of ^{138}Xe are used after modifying by Eq.(1). The factor α is calculated from JAERI-M[21].

2.2.2 Fission spectrum energy

The average energy of the reactor-neutron spectrum, E_{ave}, is calculated by Eq.(2)

$$E_{\mathrm{ave}}=\frac{\int E\Phi(E)\mathrm{d}E}{\int \Phi(E)\mathrm{d}E}, \quad (2)$$

where E is the spectrum energy. If $\Phi(E)\propto E^{1/2}\mathrm{e}^{-E/T_{\mathrm{M}}}$, the average energy becomes

$$E_{\mathrm{ave}}=2T_{\mathrm{M}}/3, \quad (3)$$

where T_{M} is the core-temperature of neutrons from reactor.

Then, effective average energy of the spectrum neutron induced fission of ^{238}U is defined as

$$E_{\mathrm{eff}}=\frac{\int E\sigma_f(E)\Phi(E)\mathrm{d}E}{\int \sigma_f(E)\Phi(E)\mathrm{d}E}, \quad (4)$$

where $\sigma_f(E)$ is the fission cross section taken form ENDF/B-VII[9]. The values of E_{ave} and E_{eff} are given in Table 1.

Table 1 E_{eff} values of different reactor average energy of ^{238}U fission

E_{ave} /MeV	0.4	0.5	1	1.3	1.5	2
E_{eff} /MeV	1.53	1.67	2.17	2.44	2.62	3.07

2.3 Error processing

The errors given in the EXFOR data are different and complicated, but the total error should be of the same level for the same method. According to error propagation, the errors are processed simultaneously with the data correction. Generally, there is an error region for different methods and periods[20]. An error may differ from each other in a region for the same method and period, depending on the value of the yield, measured energy point, data and library.

2.4 Data processing

The collected and corrected data with their processed errors are processed, including average with weight and simultaneous evaluation.

2.4.1 Data average

The data, measured at the same energy points for a nuclide, are averaged using the code AVERAGE[17]. The mean value with weight $\overline{Y}$ and its external error Σ_E are calculated and recommended. For n measurements the weighted mean is calculated by $Y_i \pm \sigma_i$, $1 \le i \le n$,

$$\overline{Y} = \sum_{i=1}^{n} \frac{Y_i / \sigma_i^2}{\sum_{i=1}^{n} 1/\sigma_i^2}, \tag{5}$$

with external error,

$$\Sigma_E = \sqrt{\frac{\sum_{i=1}^{n} (Y_i - \overline{Y})^2 / \sigma_i^2}{\sum_{i=1}^{n} (n-1)/\sigma_i^2}}. \tag{6}$$

2.4.2 Simultaneous evaluation

The data, for which not only absolute yield $Y_{ab,i} \pm e_{ab,i} (i=1,\ldots,k)$ but also their ratios $Y_{ra,j} \pm e_{ra,j} (j=1,\ldots, m)$ are measured, are simultaneously evaluated by employing the ZOTT[22] code. That is, the measurement vector $\boldsymbol{Y}$, the measurements errors $\boldsymbol{e}$ and the associated covariance matrix $\boldsymbol{D}(Y)$ are defined as

$$\boldsymbol{Y} = \begin{bmatrix} Y_{ab,i} \\ Y_{ra,j} \end{bmatrix}, \qquad \boldsymbol{e} = \begin{bmatrix} e_{ab,i} \\ e_{ra,j} \end{bmatrix},$$

and

$$\boldsymbol{D}(Y) = \boldsymbol{E}(\boldsymbol{ee}^*) = \begin{bmatrix} D(Y_{ab,i}) & Cov(Y_{ab,i}, Y_{ra,j}) \\ Cov(Y_{ra,j}, Y_{ab,i}) & D(Y_{ra,j}) \end{bmatrix}. \tag{7}$$

where $\boldsymbol{e}^*$ is the transpose of matrix $\boldsymbol{e}$, the symbol E ( ) indicates the expectation values of a scalar.

The partitioned matrix is given by

$$\boldsymbol{C} = \left[-R_{m,k} \middle| I_{m,m} \right], \tag{8}$$

where the submatrix dimensions are indicated by subscripts, $\boldsymbol{R}$ is the sensitivity matrix between Y_{ab} and Y_{ra}, and $\boldsymbol{I}$ is the identity matrix. Thus, the discrepancy vector can be written as

$$\boldsymbol{P} = Y_{ra} - RY_{ab} = CY, \tag{9}$$

and the covariance matrix is defined as

$$Cov(Y, P) = \begin{bmatrix} Cov(Y_{ab,} P) \\ Cov(Y_{ra}, P) \end{bmatrix}. \tag{10}$$

Finally, the adjusted yields can be written as

$$\boldsymbol{Y}' = \begin{bmatrix} Y'_{ab} \\ Y'_{ra} \end{bmatrix} = Y - Cov(Y, P) D^{-1}(P) P. \tag{11}$$

2.5 Data fitting

In order to study the relationship between the cumulative yields and incident-neutron energy, the SPCC[23] code, which is a general spline fit program of multiple sets of correlated data with knot optimization, is used, and the correlation among the experimental data is also considered.

In fission yield measurement, the main system error comes from the gamma intensity ΔP_γ, the calibration of the detector $\Delta\varepsilon_D$ and the measurement of fission number ΔN_f. For different methods, different nuclides and different energy points, the influences of these errors are different. In general the influence of ΔP_γ is long-correlation for the same nuclide at different energy, the influence of $\Delta\varepsilon_D$ is mid-correlation for different nuclides at different energy but long-correlation for the same nuclide at different energy, and the influence of ΔN_f is always long-correlation for the different nuclides at same energy, but no-correlation for the different energy. All of these correlations have been considered in both ZOTT and SPCC processing. The main γ-ray intensity used in the evaluated yield data for ^{85m,87,88}Kr and ^{138}Xe are listed in Table 2.

Table 2 Values of the main gamma intensity of ^{85m,87,88}Kr and ^{138}Xe used in fission yield measurement

Nuclides	E_γ/keV	P_γ/%	ΔP/%	$(\Delta P/P)$ /%
^{85m}Kr	151.1	74.9	0.4	0.53
	304.9	14.2	0.2	2.1
^{87}Kr	402.4	50.5	1.5	3
^{88}Kr	196.1	26.9	1	3.7
	834.9	13.4	0.5	3.7
^{138}Xe	258.3	31.5	1.1	3.5

3 Results and discussion

The evaluated cumulative fission yields of ^{85m,87,88}Kr and ^{138}Xe resulting from ^{238}U(n, f) at fission spectrum and 14 MeV are processed by using the AVERAGE and ZOTT codes. The comparisons of present evaluation with major international nuclear data libraries are shown in Fig.1. From this figure we can see that the present evaluation is consistent with other main libraries in error permission, but the errors of this work are a little large. The error of ENDF/B-VII is very small, while the error of the JEF-2.2 is very large; the data presented by this work are based on abundant experiment data, and evaluated by rigorous process in physics and statistics, so they are recommended.

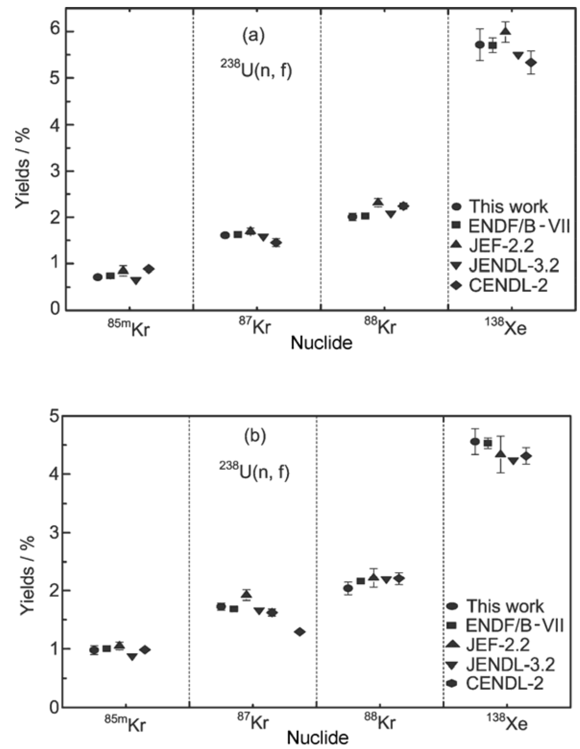

Fig.1 Comparisons of present evaluation with other evaluations for ^{238}U (a) fission spectrum, (b) at 14 MeV.

The evaluated yield data for ^{85m,87,88}Kr and ^{138}Xe are fitted by using the SPCC code, respectively. The reduced χ^2 values for ^{88}Kr and ^{138}Xe are 0.954 and 0.306, while for ^{85m}Kr and ^{87}Kr the values are 1.333 and 1.077, respectively. The comparisons of evaluated experimental data for ^{85m,87,88}Kr and ^{138}Xe are shown in Fig. 2. The experimental data are marked by their subentry numbers in the EXFOR library. From the Fig.2, the curvilinear fit data of ^{87}Kr, ^{88}Kr and ^{138}Xe represent distribution trends of the experiment data. The fit curve of ^{85m}Kr shown in Fig.2(a) can be recommended as reference to predict all the unmeasured fission yields because of lack of experiment data in this neutron energy range. More experiment data will be needed to confirm and update the neutron energy dependence of the cumulative fission yield for other product nuclides.

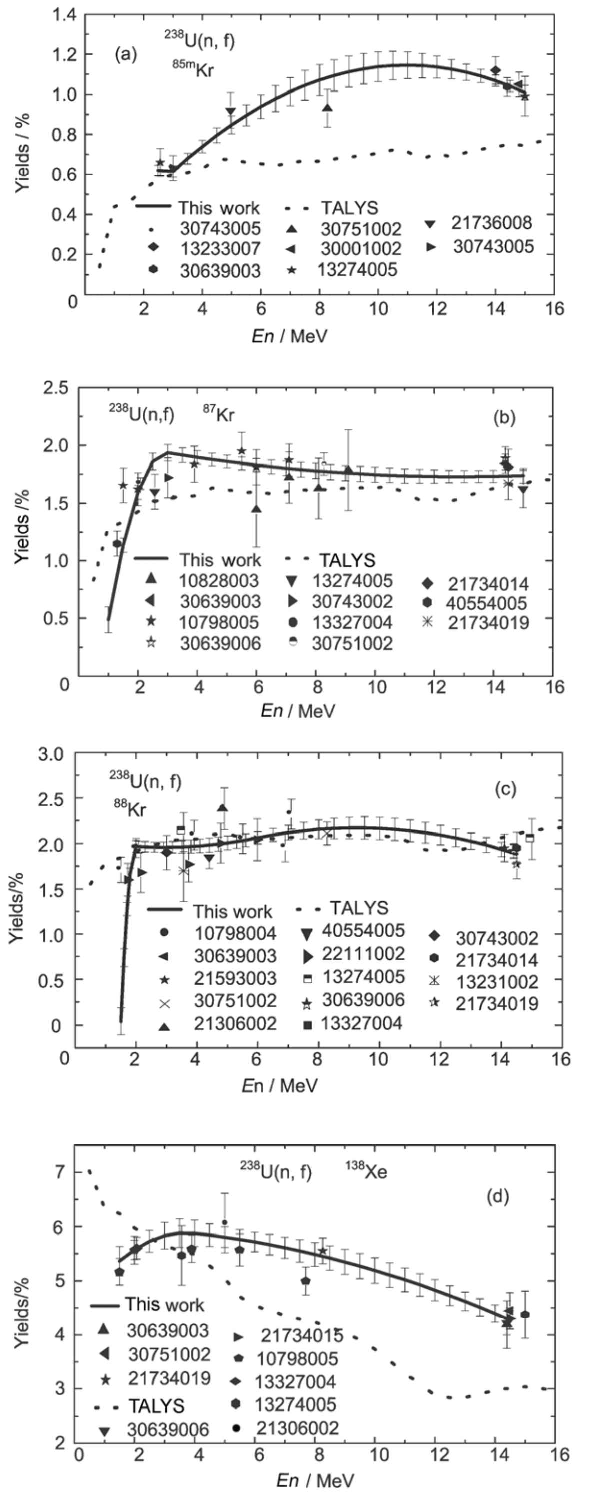

Fig.2 Comparisons of evaluated yield data of (a) ^{85m}Kr, (b) ^{87}Kr, (c) ^{88}Kr and (d) ^{138}Xe for ^{238}U(n, f) with the curve fit data and the theoretical calculated data.

The TALYS-0.64 code is used to calculate the cumulative fission yield dependence on neutron energy for ^{238}U. The comparisons of the theoretical calculated results with the evaluated curve are also shown in Fig.2. Figs.2(b) and 2(c) show that the energy dependence curves of ^{87}Kr and ^{88}Kr are similar. But examining Figs.2(a) and 2(d) for ^{85m}Kr and ^{138}Xe between 4 and 15 MeV shows that the theoretical calculated data are systematically lower than the evaluated curve, even they give similar trends in the energy dependence. It may be concluded that the predictions using purely theoretical model for the fission process are not sufficiently accurate and reliable for applied purposes.

4 Conclusion

Based on available experimental data, this work judged whether the data are available according to the methods, laboratories and other information; used the new standard and γ-ray intensity to correct the data and adjust the error; and processed the data by using codes AVERAGE, ZOTT, and SPCC. The cumulative fission yield data of ^{85m,87,88}Kr and ^{138}Xe at fission spectrum and around 14 MeV are evaluated. The dependence of the cumulative fission yields of ^{85m}Kr, ^{87}Kr, ^{88}Kr and ^{138}Xe on the incident neutron energy from ^{238}U in the energy range of 0–15 MeV is presented. Comparisons of the evaluated energy dependence curves with theoretical results show that the predictions using purely theoretical model for the fission process are not sufficiently accurate and reliable for calculating the cumulative fission yields of ^{238}U(n, f).

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