Article ID: 1007-4627(2009)04-0360-07

Evaluation of Cumulative Yields for Short-lived Fission Products from $^{238}U(n, f)$*

ZHENG Na[1], CHEN Zhong-jing[1], SUN Ji-ning[1], LIU Ting-jin[2],
CHEN Jin-xiang[1], FAN Tie-shuan[1,#]

(1 *State Key Laboratory of Nuclear Physics and Nuclear Technology and School of Physics, Peking University, Beijing* 100871, *China*;
2 *China Nuclear Data Center, Beijing* 102413, *China*)

Abstract: For reliable and consistent nuclear calculation, the cumulative yields for short-lived fission products are evaluated based on the available experimental data for ^{238}U fission induced by fission spectrum neutrons and ~14 MeV neutrons. The data are processed using codes AVERAGE for weighed average and ZOOT for simultaneous evaluation. The evaluated data are compared with those in the major international nuclear data libraries, including ENDF/B-VII, JEF-2.2, JENDL-3.2 and CENDL-2. The present evaluation will be used to improve and update the CENDL-2 library.

Key words: $^{238}U(n, f)$ reaction; cumulative yield; evaluation for fission product

CLC numbers: O571.4 **Document Code**: A

1 Introduction

For reliable and consistent nuclear calculations in the advanced nuclear reactors and other nuclear engineering designs, it is desirable to have an accurate set of nuclear data. The cumulative fission yield data of short life products from $^{238}U(n, f)$ are of special importance in some cases, such as decay heat estimation in the nuclear industry, burn-up credit study etc.[1], but those data have been paid less attention either on measurements or on evaluation, especially in the China Evaluation Nuclear Data Library (CENDL) the cumulative fission yields of short life products have not been updated since 1990s. Recently, the measurent precision of the fission yields are improved greatly[2-5], so that it is necessary to re-evaluate and improve the fission yields for some important actinide nuclei like ^{235}U, ^{238}U and ^{239}Pu[6].

In this paper, the codes AVERAGE and ZOOT are used to evaluate the cumulative yields for some short-lived fission products from the ^{238}U fission induced by the fission spectrum neutrons and ~14 MeV neutrons. The evaluation methods are presented in Sec. 2. The following section shows the recommended cumulative fission yields of 44 products, including ^{72}Ga, ^{76}As, ^{83}Se, ^{84}Br, $^{87,88}Kr$, ^{88}Rb, ^{89}Rb, $^{91,92,93}Sr$, $^{93,94,95}Y$, ^{97}Zr, ^{101}Mo, ^{104}Tc, ^{106}Ru, $^{112,115}Ag$, ^{117}Cd, ^{116m}In,

* **Received date**: 22 Jan. 2009; **Revesed date**: 20 Sep. 2009

* **Foundation item**: Major State Basic Research Development Programs for Basic Research of China(2007CB209903, 2008CB717803); Research Fund for Doctoral Program of Higher Education of China(200610001023); National Found for Fostering Talents of Basic Science of China(J0630311)

Biography: Zheng Na(1981-), female(Han Nationality), Donge, Shandong, Postgraduate, working on neutron physics and fission physics; E-mail: nnazheng@gmail.com

Corresponding author: Fan Tie-shuan, E-mail: tsfan@pku.edu.cn

^{127m}Sn, ^{129,130,131}Sb, ^{131,133}Tf, $^{132-135}$I, ^{136,138,139}Cs, ^{141}Ba, ^{140,142,143}La, ^{144}Ce, ^{142,146}Pr, ^{151}Nd, ^{156,158}Eu and the comparisons with those in other evaluated libraries[7], such as ENDF/B-VII[5], JEF-2.2[8], JENDL-3.2[9] and CENDL-2[7]. Conclusions of the present work are summarized in Sec. 4.

2 Evaluation Methods

2.1 Experimental data collection and selection

The experimental data used for this evaluation are retrived from the IAEA nuclear data services, the new version of the experimental nuclear database EXFOR[10] and the neutron reaction bibliography CINDA[11]. The experimental data are evaluated by the fission yield data evaluation system FYDES[12]. The experimental data are obtained by different methods, including radiochemical methods, classical mass spectrometry, direct γ-ray spectroscopy, and measurements of unstopped fission fragments, and are classified as absolute yields, relative yields and *R*-values. The EXFOR bibliographic information section and papers concerned are studied carefully and analyzed in this work. The original yield data are decided to be taken or abandoned according to both the concerned measurement methods and the data deviation with other measurement results. Each experimental method has its own advantages and faults[13], which have been taken into account for the evaluation of data.

2.2 Experimental data correction

The standard, gamma intensity and fission cross section used in this evaluation are presented as follows.

2.2.1 Standards

The original data from relative measurements are corrected by using modern reference values[2]. It is simple that

$$Y = Y_0 \frac{1}{N}\sum_{i=1}^{N}\frac{Y_{si}}{Y_{s0i}},\ N=1,\ 2,\ 3,\ \ldots \qquad (1)$$

where Y_0 is the original experiment fission yield, Y is the yield to be evaluated (the same definition in the following), Y_s is the new standard, Y_{s0} is the standard originally used, and N is the number of the standards used in experimental measurement.

Generally speaking, there is only one monitor nuclide used in experiment, for example, D. C. Harris et al. used ^{140}Ba in their experiment[14], while in some cases several nuclide are used as monitors, for example ^{88}Kr, ^{88}Rb, ^{138}Xe and ^{138}Cs were employed in N. E. Ballou's experiment[15].

2.2.2 Gamma intensity

If the gamma intensity used is given by the experimentalists for the yields obtained from radiochemical method or γ-ray spectroscopy, the fission yields are corrected by recent standards[3,4] which is recommend by IAEA nuclear data services. The evaluated yield is given by

$$Y = Y_0 \frac{1}{M}\sum_{i=1}^{M}\frac{I_{0i}}{I_i},\ M=1,\ 2,\ 3,\ \ldots \qquad (2)$$

where I is the new gamma intensity, I_0 is the originally used one and M is the number of the gamma lines used in experimental measurement.

For most fission products, one γ-ray is detected to determine the cumulative yield, while for some products, several gamma rays are employed. For example, in the experiment[16] of S. Nagy, one γ-ray was employed to measure the yields of ^{87}Kr, ^{88}Kr and ^{92}Sr, while three rays with different energies were employed to measure the yields of ^{91}Sr, and six rays were employed in the case of ^{135}I.

2.2.3 Cross section

In some fission yield measurements, cross sections are employed to be monitor, for example the cross section of ^{65}Cu(n, 2n) was used in Gevaert's experiment[17]. Thus, the original yields Y_0 are corrected by using the formula

$$Y = Y_0 \frac{\sigma_0}{\sigma}, \qquad (3)$$

where σ is the new evaluated fission cross section and σ_0 is the originally used one. The new values of fission cross section are taken from ENDF/B-VII[5].

2.2.4 Cumulate yield and chain yield

For some fission products, such as ^{83}Se, ^{141}Ba and ^{151}Nd, there are not enough experimental data of cumulative yield Y_{CUM}. The relevant chain yields Y_{CHN} can be used to the present evaluation after corrected by the factor α, which is defined to describe the difference between Y_{CUM} and Y_{CHN}.

$$\alpha = \frac{Y_{\mathrm{CUM}}}{Y_{\mathrm{CHN}}} \times 100\% \ . \tag{4}$$

The factor α is calculated with Y_{CUM} and Y_{CHN} served by JAERI-M[18].

2.3 Error processing

The errors given in the EXFOR library are different and complicated, but the total error should be kept at same level for the same method. Generally, there are error regions for different methods, detectors and periods. The errors are listed in Table 1[2].

Table 1 The relative errors (%) of measured fission yield data with different methods, detectors and periods

Methods		Fission yields Before 1965	Fission yields After 1965	R-value
Radiochemical	GeLi	7—15	4—8	3—4
	NaI	8—16	6—9	3—5
	Geiger	15—25		5—6
γ-spectrometry		6—10	3—6	2—3
Mass spectrometry		2—3	1—2	1—2

According to error propagation, the errors are simultaneously processed with the data correction. With the data processing by correcting the standard, gamma intensity and cross section, the error must have been processed as following.

For the standard fission yield and fission cross section correction, the relative error of yield Y, $\Delta Y' = \Delta Y / Y$, is given by

$$\Delta Y' = (Y_0'^2 + \Delta Y_s'^2 + \Delta Y_{s0}'^2)^{1/2} \ , \tag{5}$$

$$\Delta Y' = (\Delta Y_0'^2 + \Delta\sigma_s'^2 + \Delta\sigma_{s0}'^2)^{1/2} \ , \tag{6}$$

Where $\Delta Y_{s0}'$ and $\Delta\sigma_{s0}'$ are the original relative error of the standard and the cross section, respectively; $\Delta Y_s'$ and $\Delta\sigma_{s0}'^2$ are the relative error of the standard and the cross section used in the present evaluation, respectively.

The error propagation induced by gamma intensity correction is not considered in this work.

The relative error of yield Y, which is calculated from the measured R-value R_v, is given by

$$\Delta Y' = (\Delta R_0'^2 + \Delta Y_s'^2)^{1/2} \ , \tag{7}$$

$$\Delta R'_0 = (\Delta R_v'^2 + \Delta Y_{\mathrm{mxw}}'^2 + \Delta Y_{\mathrm{smxw}}'^2)^{1/2} \ , \tag{8}$$

where $\Delta Y_{\mathrm{mxw}}'$ and $\Delta Y_{\mathrm{smxw}}'$ are the relative error of the nuclide to be measured and used as standard at thermal energy point, respectively. $\Delta R_v'$ is the relative error of the R-value.

2.4 Data processing

The corrected fission yield data and errors are processed by weighted average and simultaneous evaluation methods.

2.4.1 Data average

The data, measured at same energy for a nuclide, are averaged using the code AVERAGE[8]. The mean value with weight $\bar{Y}$ and its external error σ_E are calculated and recommended. For n measurements $Y_i \pm \sigma_i$, $1 \leqslant i \leqslant n$, the mean value is given by

$$\bar{Y} = \frac{\sum_{i=1}^{n} Y_i/\sigma_i^2}{\sum_{i=1}^{n} 1/\sigma_i^2}\ , \tag{9}$$

With external error

$$\sigma_E = \sqrt{\frac{\sum_{i=1}^{n}(Y_i - \bar{Y})^2/\sigma_i^2}{(n-1)\sum_{i=1}^{n} 1/\sigma_i^2}}\ . \tag{10}$$

2.4.2 Simultaneous evaluation

The fission product yields, of which there are not only absolute yield values ($Y_{ab,i} \pm e_{ab,i}$, $i=1$, k) but also their ratios ($Y_{ra,j} \pm e_{ra,j}$, $j=1$, m) are measured, are simultaneously evaluated by employing the ZOTT[19] code. The measurement vector Y, the measurement error vector e and the associated covariance matrix $D(Y)$ are defined respectively as

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

and

$$D(Y) = E(e, e^*) = \begin{bmatrix} D(Y_{ab,i}) & Cov(Y_{ab,i}, Y_{ra,j}) \\ Cov(Y_{ra,j}, Y_{ab,i}) & D(Y_{ra,j}) \end{bmatrix} \tag{11}$$

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

The partitioned matrix is given by

$$C = [-R_{m,k} \mid I_{m,m}]\ , \tag{12}$$

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

$$P = Y_{ra} - RY_{ab} = CY, \tag{13}$$

and the covariance matrix is defined as

$$Cov(Y, P) = \begin{bmatrix} Cov(Y_{ab}, P) \\ Cov(Y_{ra}, P) \end{bmatrix}. \tag{14}$$

Then the adjusted yields can be written as

$$Y' = \begin{bmatrix} Y'_{ab} \\ Y'_{ra} \end{bmatrix} = Y - Cov(Y, P)D^{-1}(P)P\ . \tag{15}$$

3 Results and Recommendation

In comparison of the cumulative fission yield in present evaluation with the experimental data[20] are shown in Fig. 1. The experimental data are marked by their subentry numbers in the EXFOR library. In general, if the data are obtained based on the measured data more than three sets, they are reliable and recommended.

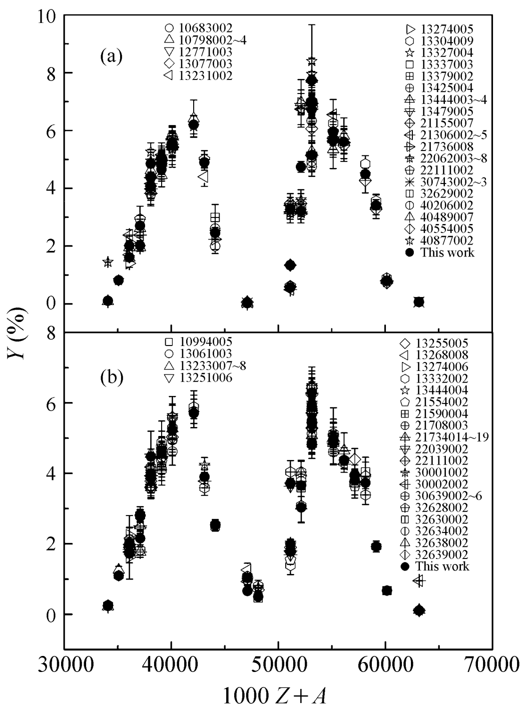

Fig. 1 Comparison of cumulative fission yields in present evaluation with experimental data of ^{238}U (n, f) induced by (a) fission spectrum neutrons and (b) the ~14 MeV neutrons, where Z and A are the charge number and the mass number of the fission product nuclides, respectively. In order to differentiate varieties of fission nuclides, the figure ($1000 \times Z + A$) is served as the abscissa.

In the present evaluation, the errors are 1%—6% for most fission product yields. The errors of 1%—2% for the evaluated data come from the fact that the data measured by mass spectrometry and multiple sets of measurements also make error reduced. The errors of 3%—6% result from the later measurements by using γ-spectrometry and radiochemistry methods, and the errors of larger than 6% are from the earlier measurements. In addition, if the deviations among several data sets for the same fission product are large, the errors will be larger.

The comparisons of the evaluated data with ENDF/B-VII, JEF-2.2, JENDL-3.2 and CENDL-2 are given in Fig. 2, where the differences mean the ratios of the divergences between the present evaluated data and the library recommended ones

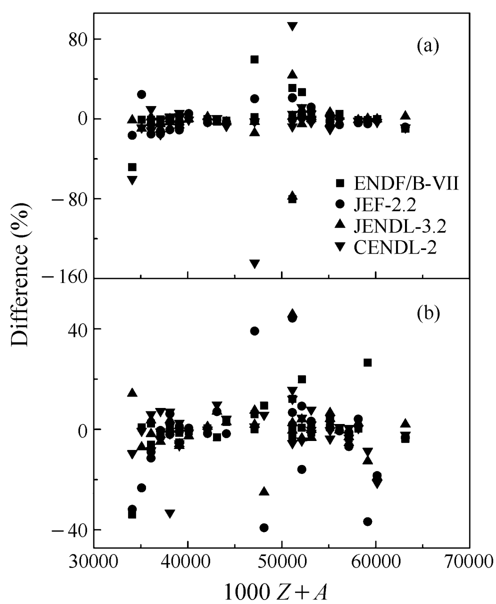

Fig. 2 Comparisons of present evaluation with other evaluated libraries of ^{238}U (n, f) induced by (a) the fission spectrum neutrons and (b) the ~14 MeV neutrons.

to the present evaluated data. From these figures we can see that for most cases the differences of present evaluated data with others are not large. In general, ~16% of the present evaluated fission products which have a difference at least of 10% in comparision with those of other libraries. For fission spectrum neutron induced fission, ~10%, ~22%, ~11% and ~16% of the present evaluated fission products have the differences of at least 10%, compared with ENDF/B-VII, JEF-2.2, JENDL-3.2 and CENDL-2 respectively, while the similar comparisons for the 14 MeV neutron fission lead to the corresponding figures of 10%, 22%, 11% and 10%. It can be seen that the differences of present evaluated values with ENDF/B-VII are smaller than those with JEF-2.2 and CENDL-2.

In addition, the present values, ^{112}Ag (F), ^{117}Cd (H), ^{130}Sb (H) and ^{83}Se (F, H), are in agreement with those given by one or two of the four libraries, where the symbols (F), (H) and (F, H) stand for the data at fission spectrum neutron and 14 MeV neutrons as well as for both, respectively, but the differences with others are large (20%—60%). Because there are more than one set of experimental data, those evaluated data are recommended as references.

The error level analyses about the present evaluation and the other libraries are shown in Fig. 3. It can be seen from Fig. 3 that the errors of

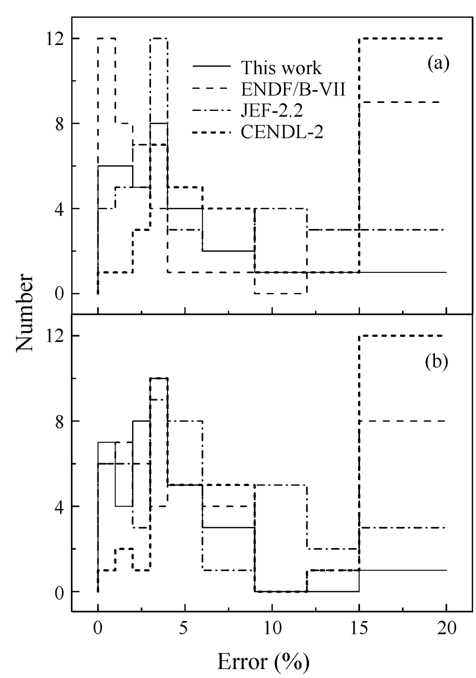

Fig. 3 The error comparisons of present evaluated yields with other evaluated libraries of ^{238}U (n, f) induced by (a) the fission spectrum neutrons and (b) the ~14 MeV neutrons. To express the error level, the errors of 1%, 2%, 3%, 4%, 6%, 9%, 12%, 15%, 20% are chosen to be the nodes, and the ordinate means the number of the product nuclei, whose error values are in the interval between two nodes.

present evaluated data are at the same level with the ENDF/B-VII, but smaller than those given in JEF-2.2 and CENDL-2. The error values of the most the present evaluation are in agreement with ENDF/B-Ⅶ within 1%—6% for fission spectrum and around 14 MeV neutron induced fission. There are also quite a lot of nuclides, for which the error is larger than 20%. This is because the cumulative yields of some nuclides are too small to be accurately measured.

4 Conclusion

Based on available experimental data the fission yield data of some short-lived products are evaluated for fission spectrum neutron and 14 MeV neutron induced fission of ^{238}U. The codes AVERAG and ZOTT are used for averaging with weight and for simultaneous evaluation, respectively. 90 cumulative fission yield data of 45 product nuclides are evaluated, among which 58 are recommended, 3 are not recommended, 6 are recommended only as reference and need to be improved, and 23 have no experimental data. Most of the present recommended data are in agreement with the recommended data in the ENDF/B-VII, JEF-2.2, JENDL-3.2 and CENDL-2 libraries. For about 16% of the present evaluated nuclides, the differences of the recommended data with those of other four libraries are larger than 10%. The error of the present evaluation is at the same level as ENDF/B-VII and effectively reduced compared with JEF-2.2 and CENDL-2. The present evaluation is approved by the China Nuclear Data Center, and will be used to update the CENDL-2 library.

References:

[1] Denschlag H O, England T R, Goverdovski A A, *et al*. Compilation and Evaluation of Fission Yield Nuclear Data. IAEA, Vienna, 2000, Report IAEA-TECDOC-1168: 9－22.

[2] Liu Tingjin, Liang Qichang. Commutation of Nuclear Data Progress, 2002, **22**: 78.

[3] Nichols A L. IAEA Co-ordinated Research Project: update of X-ray and Gamma-ray Decay Data Standards for Detector Calibration and other Applications, 2004, 29－34.

[4] Firestone R B, Chu S Y F, Baglin C M. 8th Edition of the Table of Isotopes. New York: John Wiley and Sons, 1998.

[5] Chadwick M B, Oblozinsky P, Herman M, *et al*. Nuclear Data Sheets, 2006, **107**(12): 2931.

[6] Maslov V M, Porodzinskij Yu V, Baba M, *et al*. Physical Review, 2004, **C69**: 034607.

[7] Liu Tingjin. Nuclear Physics Review, 2001, **18**(3): 192 (in Chinese).
(刘廷进. 原子核物理评论, 2001, **18**(3): 192.)

[8] Koning A, Forrest R, Kellett M, *et al*. JEFF Report 21, the JEFF-3.1 Nuclear Data Library, 2006, Report ISBN 92-64-02314-3: 1—130.

[9] Nakagawa T, Shibata K, Chiba S, *et al*. Journal of Nuclear Science and Technology, 1995, **32**(12): 1259.

[10] EXFOR, Database Version of November 27, 2008, http://www-nds.iaea.org/exfor/exfor.htm.

[11] CINDA, database Version of November 27, 2008, http://www-nds.iaea.org/exfor/cinda.htm.

[12] Liu Tingjin. Commutation of Nuclear Data Progress, 1998, **19**: 103.

[13] Wahl A C. Atomic Data and Nuclear Data Tables, 1988, **39**(1): 1.

[14] Harris D C, Beck J N, Raines W L, *et al*. Nuclear Science and Engineering, 1977, **63**: 504.

[15] Ballou N E, Kaye J H, Reeder P L, *et al*. Journal of the American Chemical Society, 1974, **16**: 156.

[16] Nagy S, Flynn K F, Gindler J E, *et al*. Phys Rev, 1978, **C17**:163.

[17] Gevaert L H, Jervis R E, Subbarao S C, *et al*. Can J Chem, 1970, **48**: 652.

[18] Ihara H. Tables and Figures from JNDC Nuclear Data Library for Fission Products, Version 2, 1989, JAERI-M 89-204: 85-138.

[19] Muir D W. Nuclear Science and Engineering, 1989, **101**: 88.

[20] EXFOR, 2008, Experimental Nuclear Reaction Data, nucleus ZA92238, quantity "FY".

^{238}U(n，f)短寿命产物核累积产额的评价*

郑 娜[1]，陈忠靖[1]，孙吉宁[1]，刘廷进[2]，陈金象[1]，樊铁栓[1,#]
（1 北京大学物理学院核物理与核技术国家重点实验室，北京 100871；
2 中国核数据中心，北京 102413）

摘　要：为满足堆内或核爆中链式反应过程的燃耗值的计算需要，对裂变谱中子和 14 MeV 中子诱发^{238}U 裂变的短寿命产物核的累积产额进行了评价。评价利用了所有可利用的实验数据，经物理分析后，使用 AVERAGE 程序加权平均，用 ZOTT 程序进行同时评价，给出了所要求能点的唯一最佳推荐数据，并将评价结果与 ENDF/B-VII，JEF-2.2，JENDL-3.2 和 CENDL-2 的推荐数据进行了比较。评价结果将用于 CENDL-2 的更新与升级。

关 键 词：^{238}U(n，f)反应；累积产额；裂变产物的评价

* **收稿日期**：2009－01－22；**修改日期**：2009－09－20

* **基金项目**：国家重点基础研究发展计划项目(2007CB209903，2008CB717803)；教育部博士点基金资助项目(20061000l023)；国家基础科学人才培养基金资助项目(J0630311)

通讯联系人：樊铁栓，E-mail：tsfan@pku.edu.cn