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Unfolding the measured neutron spectra in the irradiation chamber of the UZrH reactor using iterative method

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Abstract In the procedure of neutron fluence measurement in the whole energy range (10^{-4} eV~18 MeV), in the irradiation chamber of a UZrH reactor, the neutron energy spectra are unfolded using the method of minimizing directed divergence and SAND-II, which are used broadly at home and abroad. These methods belong to the iterative methods. In this article, the procedure of the spectra unfolding using the two methods is described in detail. The neutron spectrum distribution unfolded by the two methods agree well with each other. In the end, the major differences of the two iterative methods are compared with each other, and the main factors affecting the accuracy of the spectra unfolding with the iterative method are discussed.

Keywords Pulsed reactor, Neutron spectra, Iterative method
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1 Introduction

The neutron spectra measurement is often made by means of foils. The foil is placed in the position of interest, irradiated for a known time, and then the quantity of some radioactive nuclides produced is assessed by counting β- or γ- radiation. Thus, an estimate of the reaction rate for the production of the nuclides is obtained.

By using materials with a cross section, which vary with energy in a distinctive manner, an estimate of the neutron spectra can sometimes be obtained. In the pulsed case, the activated foil detectors satisfy the following type of activation integral equations:

$$A_i = \lambda_i \int_0^\infty \sigma_i(E)\phi(E)\mathrm{d}E, \quad i=1,2,\cdots,I \qquad (1)$$

where I is the number of the foil detectors used in the experiment, A_i is the experimentally determined activity per target nucleus of the i^{th} foil detector at the end of the neutron pulse emission, $\sigma_i(E)$ is the energy-dependent activation cross section, λ_i is the decay constant of the i^{th} foil detector, $\phi(E)$ is the neutron fluence per unit energy interval.

The procedure of determining unknown neutron fluence spectra by solving Eq. (1) is called unfolding spectra. As the relations among $\sigma_i(E)$, $\phi(E)$, and E are very complicated, it is very difficult to solve Eq.(1) directly, and the neutron fluence can be approximately solved only under some hypothetic conditions. Hence, different assumed conditions form different methods for unfolding spectra. The development of solving spectra can be divided into two stages: [1, 2] the stage before 1967 can be summed up as the solving "accurate" spectra stage. There are many different methods, such as, the subsection approximation, the zigzag line approximation, various orthogonal polynomial expan-

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sions and so on[3, 4]. The activation equation (1) is expanded in different forms by the methods mentioned above, and later the solution for the spectra is resolved by solving Eq. (1). The solution of neutron fluence is unique, because the number of the unknowns is equal to that of the equation, however, sometimes there may exist some inappropriate perturbations on the shape of the differential fluence, or some values of the solution may be negative, in which case these solutions do not have any physical meaning.

After 1967, the iterative method was introduced to overcome the shortcomings of the methods mentioned above. As the number of equations in this case is much less than that of the unknowns, the freedom of the system is rather high, and the iterative method can only come up with an appropriate solution according to the physical conditions. The iterative method is better in many aspects than the above-mentioned methods, and it is used extensively. The typical method is SAND-II [3]. SAND-II is used most extensively, and the method itself has been improved. Besides, the method of minimizing directed divergence is considered to be one of the better spectra unfolding methods.

2 Basic principle of the iterative method

The iterative procedure of solving neutron fluence using the iterative method consists of the following steps: (1) the discretion of Eq.(1) at the first step; dividing the whole energy region into J intervals , with $J >> I$. Under this condition Eq.(1) can be written as:

$$A_i = \lambda_i \sum_{j=1}^{J} \sigma_{ij} \phi_j \Delta E_j \tag{2}$$

$$\Delta E_j = E_{j+1} - E_j$$
$$i = 1,2, \cdots, I$$
$$j = 1,2, \cdots, J$$

where J is the total number of energy intervals, ϕ_j is the differential fluence of the j^{th} energy interval, ΔE_j is the width of the j^{th} energy interval, and σ_{ij} is the reaction cross section of the i^{th} foil in the j^{th} energy interval; (2) selecting an initial approximate input spectrum ϕ_j^0 (j = 1,2,⋯,J) based on all the physical information available for the given case; (3) calculating the activities A_i^0 (i = 1,2,⋯,I) of the foil detectors by inputting ϕ_j^0 into the linear equation (2); (4) comparing the calculated A_i^0 with the measured activities A_i^{m} (I = 1,2,⋯,I), and then, after the first iteration, the iterative spectra ϕ_j^1 can be obtained by modifying the initial spectra ϕ_j^0 with the difference between the measured and calculated activities; ϕ_j^1 (j = 1,2, ⋯,J), as the new input spectra, are used to calculate the foils' activities again. Repeat steps (3) and (4) until the iterative spectra satisfy a certain criterion. The last iterative spectra are the solution.

SAND-II and the method of minimizing directed divergence, all belong to the iterative method, and the principles of unfolding spectra by the two methods are introduced briefly as follows.

2.1 SAND-II

This method was studied successfully by Berg and McElyoy.[3] The whole energy range of the spectra unfolding is of 10^{-4} eV～18MeV, which is divided into 620 energy intervals. The basic algorithm of the present method is described as follows:

After k^{th} iteration, the calculated activity of the i^{th} foil in the j^{th} energy interval is given as

$$A_{ij}^k = \lambda_i \sigma_{ij} \phi_j^k \left(E_{j+1} - E_j \right) \tag{3}$$

The total activity of the i^{th} foil after k^{th} iteration is written as

$$A_i^k = \sum_{j=1}^{J} A_{ij}^k \tag{4}$$

The ratio of the measured activity to the calculated activity of the i^{th} foil after the k^{th} iteration is defined as

$$M_i^k = A_i^m / A_i^k \tag{5}$$

To minimize the distortion of the solution's shape, the smoothing weight functions are introduced to smooth the spectra before each iteration. The smoothing weight functions are defined as

$$W_{ij}^k=\begin{cases}(5A_{i1}^k+2A_{i2}^k-A_{i3}^k)/6A_i^k & j=1\\ \sum_{l=l_1}^{l_2}A_{il}^k/(l_2-l_1+1)A_i^k & 1<j<J\\ (5A_{iJ}^k+2A_{iJ-1}^k-A_{iJ-2}^k)/6A_i^k & j=J\end{cases} \quad (6)$$

where l_1 and l_2 are defined as

$$l_1=\begin{cases}1 & j=2,3,\cdots,(N\text{-}1)/2\\ j-(N-1)/2 & j=(N\text{+}1)/2,\cdots,J\text{-}(N\text{-}1)/2\\ 2j-J & j=J\text{-}(N\text{-}3)/2,\cdots,J\text{-}1\end{cases} \quad (7)$$

$$l_2=\begin{cases}2j-1 & j=2,3,\cdots,(N\text{-}1)/2\\ j+(N+1)/2 & j=(N\text{+}1)/2,\cdots,J\text{-}(N\text{-}1)/2\\ J & j=J\text{-}(N\text{-}3)/2,\cdots,J\text{-}1\end{cases} \quad (8)$$

where the odd number N is the maximum number of points used in the smoothing weight functions, the minimum value of N is equal to 5. Using Eqs.(5) and (6), an activity-weighted term C_j^k can be calculated using

$$C_j^k=\frac{\sum_{j=1}^{J}W_{ij}^k\ln M_i^k}{\sum_{j=1}^{J}W_{ij}^k} \qquad j=1,2,\cdots,J \quad (9)$$

The iteration is then performed by

$$\phi_j^{k+1}=\phi_j^k\exp(C_j^k) \quad (10)$$

2.2 The method of minimizing directed divergence

The present method is provided by Doroshenko et al. [4]. The energy range of spectra unfolding is of 0.4 eV~10 MeV. The iterative algorithm can be summarized as

$$\phi_j^{k+1}(E)=\frac{\phi_j^k(E)}{\sum_{i=1}^{I}\frac{\sigma_{ij}(E)}{A_i^m}}\sum_{i=1}^{I}\left[(\frac{\sigma_{ij}(E)}{A_i^m})(\frac{A_i^m}{A_i^c})\right] \quad (11)$$

where A_i^m is the measured activity of the i^{th} foil, A_i^c is the calculated activity of the i^{th} foil after k^{th} iteration.

2.3 The criterion of stopping iteration

There are three independent conditions for stopping the iteration procedure: (1) if the value of Q^K is smaller than the one value defined as input, where Q^K is the standard deviation of the ratios of measured activity to calculated activity, which is expressed in percents:

$$Q^k=\left[\frac{1}{I-1}\sum_{i=1}^{I}(\frac{A_i^k-A_i^m}{A_i^k})^2\right]^{1/2}\times 100 \quad (12)$$

(2) if the standard deviation becomes stable within less than 1% in two successive iterations; and (3) if the maximum number of iterations specified by the input are achieved.

2.4 Neutron self-shielding correction

The neutron field will be disturbed when foil detectors (especially the cadmium-covered foils) are placed in it. The disturbed neutron field is generally corrected by a self-shielding factor. If the neutron energy is considered to be the same in every single energy interval, the neutron self-shielding factors $G_{i,j}$ of every energy interval can be calculated. Then the reaction cross section of every energy interval can be expressed as

$$\begin{aligned}&\sigma_{\text{eff},i,j}=G_{i,j}\sigma_{i,j}\\ &i=1,2,\cdots,I\\ &j=1,2,\cdots,J\end{aligned} \quad (13)$$

The algorithm of $G_{i,j}$ can be found in Ref. [5].

3 Solution

To measure neutron fluence spectra of the whole energy range in the irradiation chamber of UZrH reactor, 21 types of foil detectors (listed in Table 1), which are sensitive to different energy regions, are used. The cross-section data are obtained from the cross section library ENDF/B-6. All the foils are placed at the center of the irradiation chamber, with the distance between the experiment point and the front chamber being 50 cm. The foils are activated even as the pulsed reactor emits a pulse. Then the experimental (or measured) activities of the foils are measured with an HpGe spectrometer. The values of both the measured activities and the cross sections of the foils are put into the solving spectra program (NFLUX), which was made by the first author of this article. As no information has been obtained regarding the distribution of the neutron fluence in the whole energy region before the spectra unfolding, an arbitrary initial spectrum has to be se-

lected, to iterate initially, and then smooth the shape of the iterative fluence by hand. The smoothed spectrum, as a new initial spectrum, is put into the program to iterate until an appropriate solution is obtained. The uncertainty of the solution fluence is calculated using the Monte Carlo method [6, 7].

The shapes of the solution neutron fluence obtained using the two methods are plotted in Figs.1 and 2, respectively. The uncertainties of the results in every energy group are from 10% to 30% [6]. The comparison of the measured with the calculated activities is listed in Table 1.

Table 1 Comparison of the measured with the calculated activities

Reactions	Measured activity A_i/Bq per target nucleus	Uncertainty of measured activity/%	Calculated activity(1) A_i/Bq per target nucleus	Deviation(1) /%	Calculated Activity(2) A_i /Bq per target nucleus	Deviation(2) /%
$Na^{23}(n,\gamma)\ Na^{24}$	5.56×10^{-16}	4.69	5.71×10^{-16}	2.63	5.47×10^{-16}	1.68
$Al^{27}(n,\alpha)\ Na^{24}$	3.78×10^{-19}	4.26	3.93×10^{-19}	3.88	4.06×10^{-19}	7.45
$Al^{27}(n,p)\ Mg^{27}$	1.82×10^{-16}	3.98	2.01×10^{-16}	9.76	2.09×10^{-16}	14.3
$In^{115}(n,\gamma)\ In^{116m}$	5.22×10^{-12}	4.42	5.20×10^{-12}	0.34	4.25×10^{-12}	18.7
$Cl^{37}(n,\gamma)\ Cl^{38}$	1.09×10^{-14}	3.78	1.11×10^{-14}	2.06	1.06×10^{-14}	2.44
$Sc^{45}(n,\gamma)\ Sc^{46}$	2.08×10^{-16}	3.07	2.19×10^{-16}	5.34	2.09×10^{-16}	0.61
$Ti^{46}(n,p)\ Sc^{46}$	4.19×10^{-20}	4.19	4.08×10^{-20}	2.71	4.20×10^{-20}	0.26
$Ti^{47}(n,p)\ Sc^{47}$	1.65×10^{-18}	4.32	1.89×10^{-18}	15.2	2.16×10^{-18}	30.1
$Ti^{48}(n,p)\ Sc^{48}$	5.18×10^{-20}	5.07	4.82×10^{-20}	6.89	4.81×10^{-20}	7.04
$Zn^{64}(n,p)\ Cu^{64}$	2.14×10^{-17}	4.16	2.23×10^{-17}	6.92	2.51×10^{-17}	20.3
$Cu^{63}(n,\gamma)\ Cu^{64}$	5.54×10^{-15}	3.28	5.75×10^{-15}	3.90	5.58×10^{-15}	0.73
$Co^{59}(n,\gamma)\ Co^{60}$	1.31×10^{-17}	3.01	1.32×10^{-17}	0.85	1.30×10^{-17}	0.029
$Ni^{58}(n,p)\ Co^{58}$	4.72×10^{-19}	5.12	4.10×10^{-19}	13.1	4.60×10^{-19}	2.53
$Mg^{24}(n,p)\ Na^{24}$	8.90×10^{-19}	4.34	8.95×10^{-19}	0.63	9.23×10^{-19}	3.71
$Mn^{55}(n,\gamma)\ Mn^{56}$	8.61×10^{-14}	3.55	8.51×10^{-14}	1.06	8.16×10^{-14}	5.19
$Fe^{54}(n,p)Mn^{54}$	7.89×10^{-20}	5.01	7.90×10^{-20}	10.1	7.90×10^{-20}	0.15
$Fe^{56}(n,p)Mn^{56}$	3.33×10^{-18}	3.25	3.33×10^{-18}	0.07	3.32×10^{-18}	0.34
$Au^{197}(n,\gamma)\ Au^{198}$	4.63×10^{-14}	3.08	4.63×10^{-14}	0.15	4.59×10^{-14}	0.86
$Mo^{98}(n,\gamma)\ Mo^{99}$	1.09×10^{-16}	3.12	1.09×10^{-16}	0.13	1.09×10^{-16}	0.054
$Lu^{176}(n,\gamma)\ Lu^{177}$	3.96×10^{-13}	3.01	3.91×10^{-13}	1.07	3.36×10^{-13}	15.0
$Dy^{164}(n,\gamma)Dy^{165}$	1.85×10^{-11}	3.17	1.63×10^{-11}	11.0	1.57×10^{-11}	14.5

(1) Results calculated with SAND-II; (2) Results calculated with the method for minimizing directed divergence.

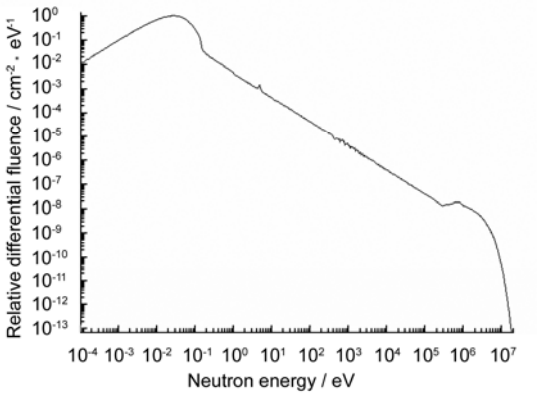

Fig.1 Distribution of $\phi(E)\sim E$ solved by SAND-II.

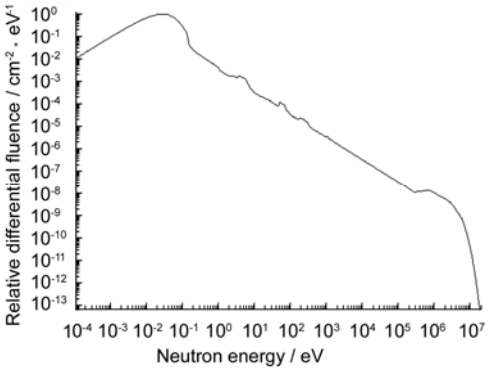

Fig.2 Distribution of $\phi(E)\sim E$ solved by the method of minimizing directed divergence.

4 Conclusions

The results obtained using the two methods mentioned above agree well with each other, but their spectra in the intermediate energy region have a little difference, because the iterative neutron spectrum using the method of minimizing directed divergence is not smoothed before each iteration step. The values of

the calculated activities using the two methods agree well with the measured activities. In the whole procedure of spectra unfolding with the help of the iterative method, the main characteristics of the method can be summarized as follows: (1) the selection of the initial input spectrum has some effect on the accuracy of the solution spectrum. The iterative spectrum is especially dependent on the initial spectrum in the energy region where there are no sensitive detectors; (2) the uncertainty of the measured activity has more effect on the accuracy of the iterative solution than that of the cross section. If the value of the measured activity is bigger or that of the cross section is smaller than the real facts, peaks can form in the iterative spectrum, in the foil's sensitive energy region; on the contrary, dips are formed in the shape. This influence can be a little weak in the foil's insensitive energy region; (3) the distribution of the obtained differential neutron fluence uncertainties is not well proportioned, the values of the solution's uncertainties are smaller in the energy region, which is covered by sensitive foils, otherwise they are larger. The difficulties, rather inherent in the problems of foil coverage and activity measurement, are the non-uniqueness of the solution, and incomplete or inaccurate evaluations of the reaction cross-section data. Physically unreasonable structures in the solution spectrum can therefore be minimized by careful experimental planning and execution, to assure sufficient foil energy coverage and accurate activity measurements. This can be done by utilizing all physical information available, to select an appropriate initial input spectrum, and by acquiring sufficiently accurate cross-section data.

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