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ARTICLE

Development of neutron spectrum unfolding method for advanced nuclear emulsion

Shigetaka Maeda[a*], Chikara Ito[a], Kohei Ishihara[b], Keisuke Takagi[b], Haruna Minato[b],
Yousuke Sakai[b], Jun Kawarabayashi[b], Hideki Tomita[b] and Tetsuo Iguchi[b]

[a]*Japan Atomic Energy Agency, 4002 Narita-cho, Oarai-machi, Ibaraki, 311-1393, Japan;*
[b]*Nagoya University, Furo-cho, Chikusa-ku, Nagoya, Aichi, 464-8603, Japan*

In order to realize neutron spectroscopy in high-intensity gamma ray fields such as those present in spent nuclear fuel storage facilities using an advanced nuclear emulsion based on a non-sensitized OPERA film with AgBr grain sizes of 60 nm, a new neutron spectrum unfolding method has been developed. The response functions were evaluated in the neutron energy range between 0.1 and 4.5 MeV by Monte Carlo calculations. To realize a highly reliable unfolding method that does not require an initial guess spectrum, an algorithm was formulized based on the maximum entropy principle and the maximum likelihood method, and the new unfolding code MEALU was developed. The code performance was checked by analyses of mock-up data. As an example, the neutron spectrum from fresh fuel of the experimental fast reactor Joyo was calculated and track lengths were simulated using the calculated response function and the estimated spectrum. Comparison between the original neutron spectrum and the unfolded one confirmed the effectiveness of the proposed method.

Keywords: nuclear emulsion; neutron spectrum; radiation measurement; unfolding

1. Introduction

A nuclear emulsion [1] is a promising detector, in which neutrons can be identified as recoil proton tracks with a high position resolution, typically within 1 μm [2]. The darkness of a track, which represents the linear density of developed silver clusters, is proportional to the linear energy transfer of the incident charged particle. Furthermore, the sensitivity of the nuclear emulsion can be controlled by pre-varying parameters such as gold sensitization or AgBr grain size. Consequently, nuclear emulsions with well controlled sensitivity are promising candidates as neutron detectors for use in high-intensity gamma ray fields such as those present in spent nuclear fuel storage facilities.

In this study, neutron spectrum unfolding method for advanced nuclear emulsion based on a non-sensitized OPERA film with AgBr grain sizes of 60 nm [3] has been developed. The response functions for the nuclear emulsion have been evaluated in the neutron energy range between 0.1 and 4.5 MeV by Monte Carlo calculations. In addition, an unfolding code suitable for high-resolution neutron spectroscopy without *a priori* information has been developed on the basis of the maximum likelihood principle combined with the maximum entropy principle.

The effectiveness of the proposed neutron spectrum measurement method was demonstrated through analysis of mock-up data.

2. Neutron spectrum measurement using a nuclear emulsion

A nuclear emulsion based on a non-sensitized OPERA film containing AgBr nano-grains, whose sensitivity to gamma rays could be well controlled by the grain size, was used in the present study. Its efficiency was calculated to be 0.7×10^{-4} for neutrons with energies from 0.3 to 2 MeV. The tracks in a nuclear

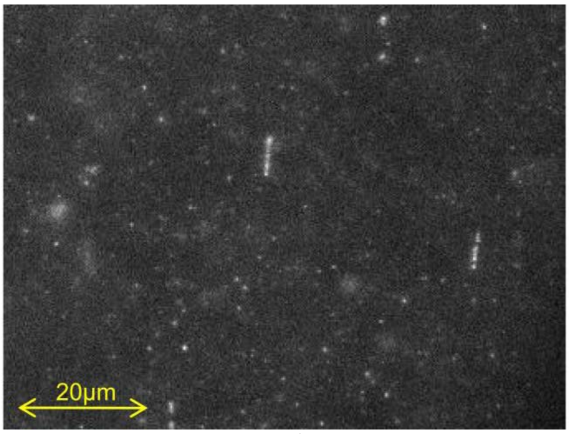

Figure 1. Typical micrograph of recoil proton tracks produced by 565keV neutron.

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*Corresponding author. Email: maeda.shigetaka@jaea.go.jp emulsion are formed by proton recoil following collision with neutrons, and their lengths depend on the incident neutron energy. A typical micrograph of recoil proton tracks produced by 565keV neutrons in the emulsion after image development is shown in **Figure 1**. Information concerning each track was accumulated using an automatic scanning system, which is currently under development. A detailed description of this scanning system is outside of the scope of the present paper. However, the accumulated data included the length and number of proton tracks, which is then converted to a frequency distribution of track lengths. The neutron energy spectrum in the field can be unfolded based on this measured data.

3. Evaluation of response function

The response function was evaluated based on the detailed information on the properties and behavior of recoil protons in a nuclear emulsion. First, the position, energy, and direction of protons were calculated as a function of neutron energy using the particle and heavy ion transport code system PHITS[4]. The calculation model used to determine the response function is shown in **Figure 2**, and the composition of a nuclear emulsion is listed in Table 1. Proton track lengths were calculated using the transport of ions in matter code SRIM[5] based on the PHITS results. The resulting relation between the energy of a recoil proton and its track length is shown in **Figure 3**.

Figure 4 shows the response function of the nuclear emulsion for protons, which is the frequency distribution of recoil proton track lengths. **Figure 5** shows the corresponding response function for neutrons, which is the frequency distribution of neutron energies. The neutron energy range from 0.1 to 4.5 MeV is divided into 45 bins with an interval of 0.1 MeV, and the track length is divided into 23 bins with an interval of 3 μm.

Table 1. Composition of nuclear emulsion.

Nuclide	Ratio (wt.%)
Ag	38.34
Br	27.86
I	0.81
C	13.00
O	12.43
N	4.81
H	2.40
S	0.09

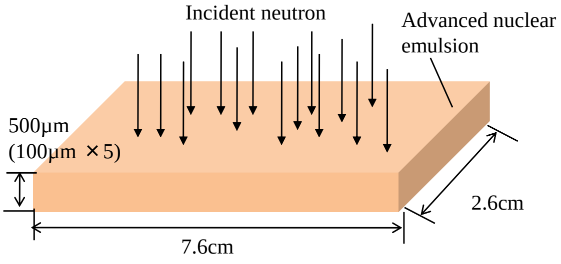

Figure 2. Calculation model used to determine response function in PHITS.

The response function of the nuclear emulsion for neutrons is similar to that of a typical recoil proton spectrometer. However, the direction of incident neutron to nuclear emulsion affects response efficiency. It is necessary to prepare several response functions for each incident neutron condition for applying to a measured data.

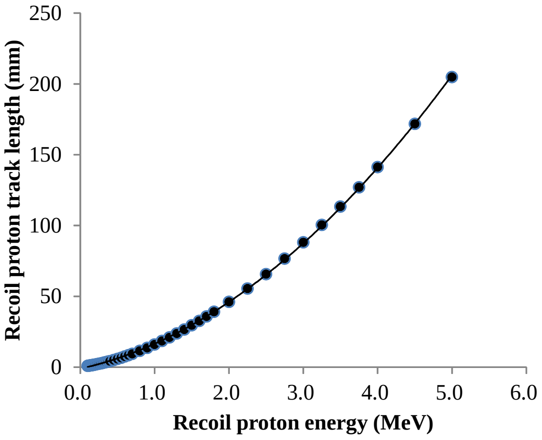

Figure 3. Relation between recoil proton energy and track length.

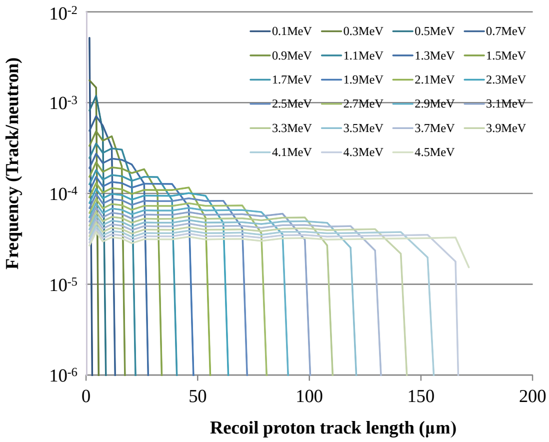

Figure 4. Response function for protons (frequency vs. track length).

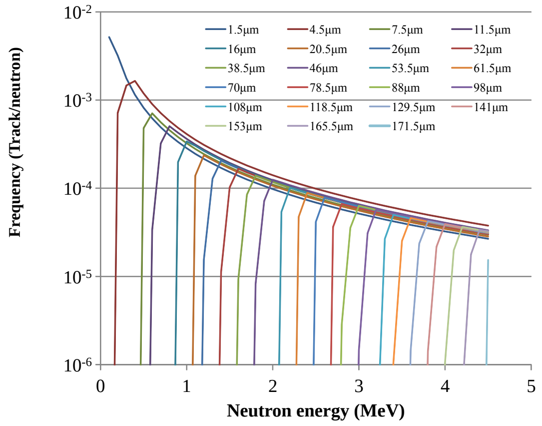

Figure 5. Response function for neutrons (frequency vs. energy).

4. Spectrum unfolding procedure

The *M* kinds of detector responses c_i are related to the irradiating neutron spectrum $\phi(E)$ by a Fredholm integral equation of the first kind,

$$c_i \approx \int_0^\infty R_i(E)\phi(E)dE, \quad i = 1,2,\cdots,M \tag{1}$$

where $R_i(E)$ is the response function of the *i*-th detector. The problem involved in neutron spectrum unfolding is to estimate $\phi(E)$ that satisfies Eq.(1) based on the measured data c_i and well evaluated data for $R_i(E)$ within a reasonable uncertainty. The usual approach is to divide the energy *E* into *N* discrete groups and rewrite Eq. (1) in sum (or matrix) notation as

$$c_i \approx \sum_{k=1}^{N} R_{ik}\phi_k, \quad i = 1,2,\cdots,M \tag{2}$$

where R_{ik} is the average value of $R_i(E)$ and ϕ_k is the flux in the *k*-th energy group. In general, since the number *N* of energy groups is larger than the number *M* of measured detector responses in the case of the proposed method, the foil activation method and multisphere neutron spectrometer (Bonner Ball), the set of simultaneous linear equations represented by Eq. (2) represents a mathematically insoluble problem.

In the present study, to realize a highly reliable unfolding method that does not require an initial guess spectrum, the algorithm based on the maximum entropy principle and the maximum likelihood method was formulized, and the new unfolding code MEALU was developed.

Taking the vector *p* as a probability, the quantity

$$H = -\sum_{k=1}^{N} p_k \ln p_k \tag{3}$$

where p_k is the normalized value of ϕ_k, is known as Shannon's information entropy. Based on the maximum entropy principle, an energy distribution *p* that maximizes Eq. (3) is chosen.

On the other hand, the likelihood relevant to the Poisson statistics of neutron detection [6] is given by the following:

$$L = \sum_{i=1}^{M} \left(c_i \ln < c_i > - \ln(c_i!) - < c_i > \right) \tag{4}$$

In this study, *p* is selected to maximize the linear combination of the entropy and the likelihood. This study adopts other formulations to improve the reproducibility of the unfolded neutron spectrum compared with conventional algorithms that use the maximum entropy principle, such as that of Itoh and Tsunoda [1].

The combination of entropy and likelihood, *Q*, is given by

$$\begin{aligned} Q &= H + L \\ &= -\sum_{k=1}^{N} p_k \ln p_k \\ &\quad + \sum_{i=1}^{M} \left(c_i \ln < c_i > - \ln(c_i!) - < c_i > \right) \end{aligned} \tag{5}$$

A solution ϕ is determined that maximizes the objective function *Q* using a modified Newton method based on the Broyden–Fletcher–Goldfarb–Shanno (BFGS) formula, one of the most efficient numerical algorithms for nonlinear optimization programming.

In this code, the need for the arbitrary initial guess spectrum required when applying conventional unfolding codes to underdetermined problems is eliminated.

An analytical estimation method is adopted for error propagation. To analyze the propagation of uncertainties in the MEALU solutions and their sensitivity to such uncertainties, the effect of variations δc_i in the measurements, δR_{ik} in the response function and $\delta\alpha$ in total number of neutrons incident on the detector are considered. If a maximum entropy and maximum likelihood solution exists, it can be shown that explicit solutions can be found for the matrices $\delta\phi_k/\delta c_i$, $\delta\phi_k/\delta R_{ik}$ and $\delta\phi_k/\delta\alpha$. An $N \times N$ total uncertainty matrix *U* can then be introduced, where

$$\begin{aligned} U_{kl} &= \sum_{i,j} \frac{\delta\phi_k}{\delta c_i} K_{ij} \frac{\delta\phi_l}{\delta c_j} \\ &\quad + \sum_{i} \sum_{k,l} \frac{\delta\phi_k}{\delta R_k} K'_{ikl} \frac{\delta\phi_l}{\delta R_l} \\ &\quad + \sum_{kl} \frac{\delta\phi_k}{\delta\alpha} K'' \frac{\delta\phi_l}{\delta\alpha} \end{aligned} \tag{6}$$

provided that the covariance matrices *K*, *K'* and *K''* are available. The usual assumption is made that *K* is an $M \times M$ diagonal matrix with elements δc_i^2 on the diagonal. For the $N \times N$ covariance matrix *K'*, it is useful to consider the full covariance matrix in each detector response. *K''* is the uncertainty of α. The matrix *U* can be used to assign an uncertainty to any integral quantity *H* of the form $H=\Sigma_i h_k\phi_k$ where h_k is the response function. The uncertainty associated with *H* is given by

$$\Delta H = \sqrt{\sum_{kl} h_k U_{kl} h_l} \tag{7}$$

5. Performance test and results

The performance of the proposed method was checked by the analysis of simulated data. As an example, the neutron spectrum obtained from fresh MOX fuel of the experimental fast reactor Joyo was estimated using ORIGEN-2 and MCNP[7]. As shown in **Figure 6**, the calculated neutron spectrum is a typical spontaneous fission spectrum. The neutron emission rate per driver fuel subassembly is 1.3 ×10^6 neutrons/s. The

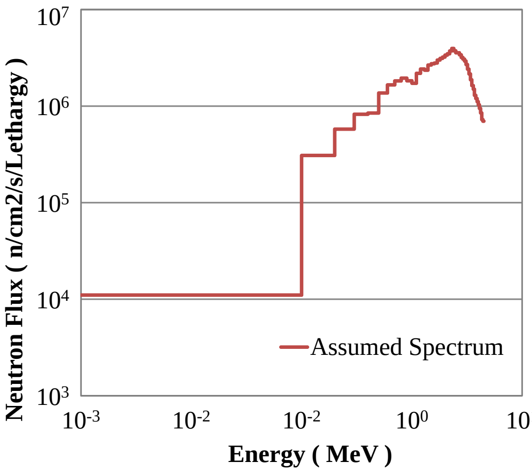

Figure 6. Calculated neutron spectrum close to nuclear fuel.

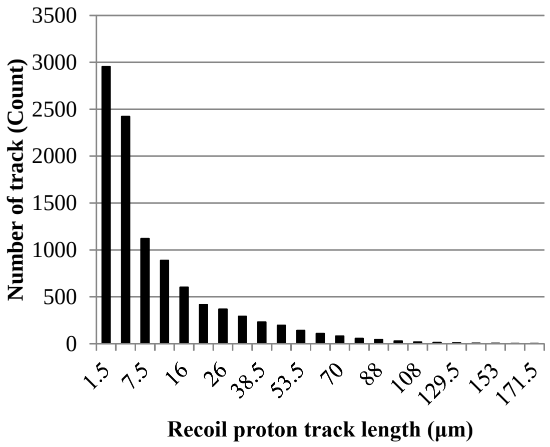

Figure 7. Mock-up response data without measurement error.

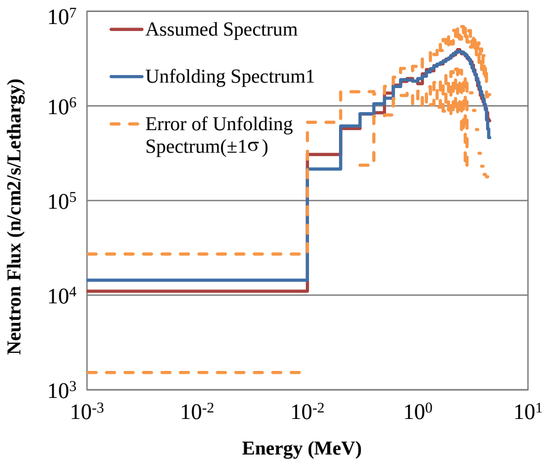

Figure 8. Unfolded neutron spectrum using an ideal response.

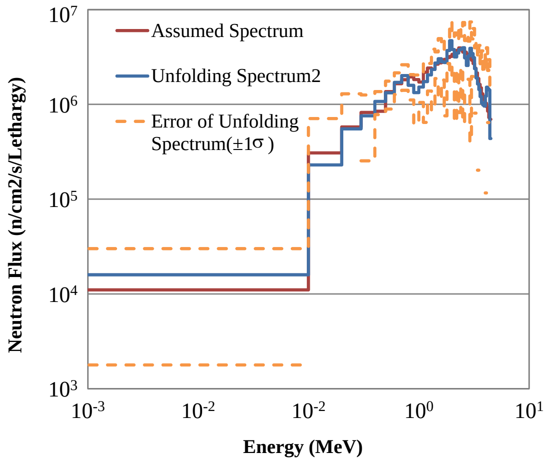

Figure 9. Unfolded neutron spectrum using a response with an artificial error.

neutron flux assumed to be irradiating the nuclear emulsion on the driver fuel side is 1.5×10^{3} neutrons/cm^2/s.

Track length simulations were then carried out using the calculated response function and the calculated spectrum. Distortions of the response function due to inaccuracies in the track recognition process were not considered, i.e., ideal response function was assumed. Two different cases were considered. In the first, no errors were introduced in order to carry out a theoretical verification of the neutron spectrum measurement procedure using a nuclear emulsion. In the second, an artificial measurement error associated with proton track scanning was introduced in order to evaluate the effect of error propagation on the unfolded neutron spectrum. The mock up data for the case without any error is shown in **Figure 7**.

The resulting unfolded spectrum is shown in **Figure 8**, and it can be seen that it is in good agreement with the original spectrum. Thus, the proposed method can reproduce the original spectrum under ideal conditions. The unfolded spectrum for the case where an error was introduced is shown in **Figure 9**. It can be seen that it agrees with the original spectrum within the estimated error. This indicated that even if the measurement data contains errors, the proposed method can be used for measuring and unfolding a neutron spectrum. However, it was found that measurement errors gave rise to unphysical fluctuations in the spectral shape. To avoid this, the energy and track length resolution should be optimized to reduce the influence of measurement errors.

6. Conclusion

In order to realize neutron spectroscopy in high-intensity gamma ray fields such as those present in spent nuclear fuel storage facilities using an advanced nuclear emulsion based on a non-sensitized OPERA film with AgBr grain sizes of 60 nm, a new neutron spectrum unfolding method has been developed.

The response function of the emulsion for recoil protons was evaluated and a spectral unfolding code suitable for this measurement procedure was developed. This unfolding code is based on a combination of the maximum likelihood principle and the maximum entropy principle.

This measurement method was verified and its performance was evaluated using mock-up data. Comparison between the original neutron spectrum and the unfolded one confirmed the effectiveness of the proposed method. In this study, the recoil proton track lengths were divided into 3 μm interval data. The results indicated this was adequate scanning resolution of track length for the recoil proton tracks.

In future work, a track recognition program will be created in order to achieve the desired resolution. Moreover, the validity of the response function will be confirmed by irradiation with known doses. In addition, the method will be applied to actual measurement data.

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