Measurement and Evaluation of Leakage Neutron Spectrum at TRACY

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The leakage neutron spectrum of the Transient Experiment Critical Facility (TRACY) at the Japan Atomic Energy Research Institute was measured. The experimental core is composed of 10wt% enriched uranyl nitrate solution in a 50cm inner diameter cylindrical tank. As measuring methods, we used the recoil proton proportional counter method and the multi-foil activation method. In order to evaluate a spectrum, the former unfolding code is SPEC-4[1)] and the latter is NEUPAC-JLOG. We had improved the NEUPAC-JLOG[2)] in order to use for the thermal reactor. In the case of measurement by the recoil proton proportional counter, it was performed under the stationary state operating condition. The evaluated neutron spectrum was in good agreement with the tendency of calculation using MCNP4B[3)] with JENDL3.2[4)] library. For the multi-foil activation method, they were performed under the stationary state and the transient state operating conditions. The neutron spectrum as a guess spectrum was calculated using MCNP4B with JENDL3.2. The result of neutron spectrum analyzed by MCNP is reliable for the steady state. In case of the transient state, the unfolding spectrum of the fast neutron region was almost the same result as the initial spectrum. In the thermal neutron region, the unfolding spectrum was somewhat greater than the initial spectrum. This is considered to be caused by generation of void by the transient.

KEYWORDS: Recoil Proton Proportional Counter, NEUPAC, Neutron Spectrum, Unfolding, Covariance, TRACY, MCNP

1. Introduction

The information of the neutron spectrum is fundamental characteristics for all reactor and neutron irradiation field. Generally, it can be obtained by experiment and calculation. At the moment, the neutron spectrum with remarkable accuracy and precision is calculated by using Monte Carlo code.

In this report, the evaluated method of the leakage neutron spectrum at TRACY is described. Until now, the evaluation of the leakage neutron spectrum was not performed for TRACY. The experiment was conducted by two measuring methods. We used recoil proton proportional counter method and multi-foil activation method. The measurement of the recoil proton proportional counter was performed under the stationary state operating condition that the temperature of the solution did not change. For the multi-foil activation method, it was performed under the stationary state and the transient state operating conditions. The temperature of solution was increased. The neutron spectrum was used unfolding code. The initial neutron spectrum was calculated using MCNP4B with JENDL3.2.

2. Description of TRACY

2.1 Core Tank

TRACY is designed to obtain the data on super-critical phenomena using 10wt% enriched uranyl nitrate solution. The concentration of uranium in the solution can be varied up to 500 gU/L. Figure 1 show a photograph of TRACY core tank.

TRACY is operated at first by feeding the uranyl nitrate solution with a pump into a cylindrical shaped core tank made of stainless steel. The diameter and height of the core tank are 50cm and 200cm, respectively. Delayed criticality is attained by the feeding the solution. Then, transient operation is initiated by either withdrawal of a unique control rod inside the tank, named ‘transient rod’, or continuous feed (max. 65L/min.) of the solution exceeding criticality. The transient rod can be driven by two sorts of mechanisms, i.e. (1) rapid withdrawal by compressed air (within 0.2 sec.), (2) ramped withdrawal with an electric motor (max. 900cm.min.). The reactivity insertion is licensed up to 3 dollars for the transient operation. Peak power and integrated power are also limited up to 5,000MW and 32MJ (equal to 10^{18} fissions), respectively. Table 1 shows major specifications of TRACY.

2.2 Experimental System

The multi-foil activation method was performed with ramped withdrawal. Addition reactivity was 2.98$, and withdrawal speed was 875cm/min, and

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initial power was 1mW. Moreover, in the stationary state, the reactor power was kept at 10kW, which produced 5MJ of integral power. On the other hand, for the measurement of recoil proton proportional counter, the reactor power was kept at 30mW, which produced 408J of integral power. Details of the experimental conditions are shown in Table 2. A reactor room arrangement is shown in Figure 2.

3. Measuring Method

3.1 Recoil Proton Proportional Counter Method

The recoil proton spectrometer[5)] consists of four spherical hydrogenous proportional counters. Each proportional counter covers the following energy range.

SP2-1 : 2-inch inner diameter, hydrogen gas 1 atmospheric pressure, (50keV～200keV)
SP2-4 : 2-inch inner diameter, hydrogen gas 4 atmospheric pressure, (100keV～500keV)
SP2-10 : 2-inch inner diameter, hydrogen gas 10 atmospheric pressure, (400keV～1.5MeV)
SP6: 6-inch inner diameter, Ar- methane gas 5 atmospheric pressure, (1.0MeV～4.5MeV)

Figure 3 show a photograph of the recoil proton proportional counter.

The recoil proton proportional counter does not depend on the direction of the incidence of neutron, because proportional counters have the sphere shape. These detectors are on a rotating platform. One rotation time is about 20 seconds. The measurement energy range for the recoil proton proportional counter is from the thermal energy to 4.5MeV. However, the energy range for unfolding is from 50keV to 4.5MeV.

From the result, the amount of differentiation is calculated using unfolding code SPEC4. The maximum dose of these detectors is 1mSv/h. Therefore, accurate measurement of a neutron spectrum demands long measurement time.

3.2 Multi-Foil Activation Method

The transient operation of TRACY is usually performed with several 100 MW power. Foil activation/unfolding method is a general method in such the neutron and gamma ray mixed field in high dose rate.

This method adjust from initial neutron and several reaction rate to best estimate spectrum. The C/E's of all reaction rates set equal to 1.0; this compares C/E of calculated reaction rate $\phi i\sigma$ (C) with measurement reaction rate (E). In this case, $\phi i(E)$ and $\sigma(E)$ are important and especially the former influences a result. As most important one for the unfolding method, we have to adopt $\phi i(E)$ that is best estimation using radiation transport code. Now, continuous energy Monte Carlo code MCNP is widely used in the world.

We used NEUPAC-JLOG unfolding code that is improved for thermal reactor. The code is processing the covariance data of $\phi i(E)$, $\sigma(E)$, and reaction rate. As a result to the adjusted spectrum, covariance data is also given. The microscopic cross sections for NEUPAC-JLOG are edited with MGCL137 groups from JENDL Dosimetry File[6)]. The microscopic cross section for (n,γ) reaction was treated in consideration of the resonance self-shielding effect correction and the self-shielding correction for the foil thickness.

4. Measurement and Evaluation

4.1 Measurement and Evaluation with the Recoil Proton Proportional Counter

The recoil proton spectrometer was installed 2 m away from TRACY reactor core tank surface. Measurement was performed at the 30mW critical state for 4 hours. The dose rate at the measurement location was 1mSv/h. The error of adjustment for recoil proton proportional counter in the range of 50keV to 4.5MeV, when based on the result calibrated using the Cf-252 standard source calibrated by NIST in Bubble Technology Industries, was about 7% (1 σ)[7)] of the integrated fluence value. For fluence, dose, and neutron spectrum, errors are not shown about the results after adjustment. For this reason, the error guaranteed by the maker is estimated to standard value.

Unfolding results are shown in Figure 4 and Table 3. For the neutron spectrum, a tendency which resembled well the result calculated by MCNP is shown. There is a region where the ratio of MCNP/ recoil proton proportional counter is greater than 20% at below 500keV. However, this is due to the influence by the end of the energy measurement range of each globular form detector. For the neutron fluence, the adjustment is less than 1% in the range of 50keV to 4.5MeV.

4.2 Measurement and Evaluation with Multi-Foil Activation Method

Foils used multi-foil activation method are Au-197, Au-Cd, Cu-63 for the thermal neutron region and In-115, and Ti-48 for the fast neutron region. Twelve kind reactions were prepared for the multi-foil activation method. Less than half of them were actually irradiated. This number is a limit when the irradiation environment of the present TRACY is considered. However, five kinds of foil, which can be measured, cover the neutron energy range of the thermal and fast energy region. Details of each activation foil are shown in Table 4. The gamma ray emitted from activation foil measured with the high purity germanium detector (HPGe).

In regards to the covariance for NEUPAC-JLOG, only the diagonal element of each activation foil and an initial spectrum were taken into consideration. All of the off-diagonal elements were made into zero.

4.2.1 Stationary Sate

The value by the indicator with which the integral power was proofread by FP analysis is 5MJ. The temperature of solution was about 35 °C. For such operating conditions, unfolding was performed using five kinds of activation foils, and the neutron spectrum was evaluated. MCNP4B with JENDL3.2 were used for calculation of an initial neutron spectrum. As calculation conditions, eigenvalue calculation of the usual steady state was performed. The 3σ of the statistical error (FSD) of each energy bin in MCNP calculation was used on the covariance in the diagonal element from the initial spectrum. The unfolding result is shown in Figure 5. The adjusted neutron spectrum is overestimated about 1% in the thermal neutron region, although it becomes the almost same shape to an initial neutron spectrum. Moreover, in near 10MeV, the adjusted spectrum is large about 5%. The C/E (see Table 5) of the reaction rate of integration value is adjusted so that 1.0 may be approached; the amount of change of Au, In, and Ti after adjustment is 0.5%, 0.3%, and 1.4%, respectively. However, this result means that Au-Cd and Cu do not necessarily approach 1.0 from overall adjustment of five reaction rate values. The difference is about 0.4%, which has been disregarded. The C/E's of reaction rates were hardly changed after adjustment. It means that the calculation of initial neutron spectrum is best estimation.

4.2.2 Transient State

The transient operation mode is ramped withdrawal. The power change of the operation is shown in Figure 6. From the figure, the 2nd and 3rd peak are seen after the 1st peak. The integral power is 19.2MJ. The solution temperature was about 65 °C. During this time, the radiolysis gas was generated, and change of a solution level was created by void. Although attached in the same radial as the foil for thermal neutrons, large variation is conspicuous. In this time, the standard deviation that was calculated by the reaction rate of three Au foils was about ±7%. The covariance of the diagonal element of the initial spectrum was estimated about 20% for the thermal and the fast region, and others were estimated as 3σ of FSD in the Monte Carlo calculation. The unfolding result is shown in Figure 7. As Monte Carlo calculation conditions, the eigenvalue calculation for the usual steady state was performed. The adjusted neutron spectrum is high about 3% in the thermal neutron region, although the shape of the spectrum is same as initial neutron spectrum. The C/E (see Table 6) of the reaction rate is adjusted to 1.0; the change of Au-Cd, Cu, and Ti after adjustment is 1.9%, 1.9%, and 0.2%, respectively. The difference is about 0.3%, which has been disregarded. Here, the $\phi i(E)$ is incalculable with sufficient accuracy because of complicated conditions of the transient state.

5. Summary

The information of the neutron spectrum is fundamental characteristics for all reactor and neutron irradiation field. Therefore, the leakage neutron spectrum was evaluated using the Transient Experiment Critical Facility (TRACY) at the Japan Atomic Energy Research Institute. As measuring methods, we used the recoil proton proportional counter method and the multi-foil activation method.

[Recoil Proton Proportional Counter Method]

When the measurement was performed by using the recoil proton spectrometer, the reactor power was kept at 30mW, which produced 408J of integral power. The measurement was performed for 4 hours. For the neutron spectrum, a tendency which resembled well the result calculated by MCNP is shown. For the neutron fluence, the value of the C/E is in agreement by less than 1% in the range of 50keV to 4.5MeV.

[Multi-Foil Activation Method - Stationary Sate]

The integral power is 5MJ, and the temperature of solution was about 35 °C. Under such operating conditions, reaction rate measurement was performed using five kinds of the activation foil, and the neutron spectrum was adjusted. MCNP4B with JENDL3.2 were used for calculating of an initial neutron spectrum. As calculation conditions, eigenvalue calculation of the usual steady state was performed. The adjusted neutron spectrum is high about 1% in the thermal neutron region, although it becomes the almost same shape to an initial neutron spectrum. The C/E's of reaction rates were hardly changed after adjustment. It means that the calculation of initial neutron spectrum is best estimation.

[Multi-Foil Activation Method - Transient State]

The transient operation mode is ramped withdrawal. The integral power is 19.2MJ, and the solution temperature was about 65 °C. During this time, radiolysis gas was generated, and change of a solution level was created by void. From the above phenomenon, the standard deviation of thermal neutron region was about ±7% that was calculated by the reaction rates of three Au foils. As calculation conditions, eigenvalue calculation of the usual steady state was performed. The adjusted neutron spectrum is high about 3% in the thermal neutron region, although it becomes the almost same shape to an initial neutron spectrum. Here, the reliability of $\phi i(E)$ is incalculable with sufficient accuracy because of the complicated conditions of the transient state.

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Table 1 Specification of TRACY

Core tank	Diameter	Approx. 50cm
	Height	Approx. 200cm
Solution fuel: $UO_2(NO_3)_2$ aq.	Uraniu-235 enrichment	Approx. 10%
	Uranium concentration	up to 500gU/L, currently 400gU/L
Critical height		40 to 100cm, currently 50cm
Excess reactivity for transient operation		up to 3$
Power	Peak	up to 5000MW
	Integrated	up to 32MJ (=10^{18} fission)
Initial temperature		Room temperature, Typically 26 °C up to 40 °C

Table 2 Experiment condition of TRACY

Run No.	Operation mode	Power (W)	Integral power(J)	Measuring Method
R148	Stationary state	30m	408	Recoil proton
R175	Transient State (Ramped withdrawal)	-	19M	Multi-foil activation
R183	Stationary state	10k	5M	Multi-foil activation

Table 3 Comparison of neutron fluence

Energy range (MeV)	Proton recoil (E)	MCNP (C)	C/E
0.05 – 4.5	1.18E+07	1.17E+07	0.992

Units : (n/cm^2)

Table 4 The details of each activation foil in NEUPAC-JLOG

Nuclear reaction	Half life	Energy range		Cross section (barn)	Resonance energy	γ ray (keV)
		min.	max.			
^{197}Au(n,γ)	2.694d	2.44E-02	6.39E+00	98.65	4.906eV	412
^{197}Au(n,γ)-Cd	2.694d	6.90E-01	6.00E+02	98.65	4.906eV	412
^{63}Cu(n,γ)	12.70h	2.03E-02	9.61E+03	4.51	580eV	1346
^{115}In(n,n')	4.486h	1.07E+06	5.62E+06	170m	1.2MeV	336
^{48}Ti(n,p)	43.68h	5.76E+06	1.23E+07	30.3m	7.6MeV	1312

*The energy range is decided by TRACY spectrum.
In and Ti is threshold value

Table 5 C/E of the reaction rate in the analysis result of NEUPAC-JLOG (Stationary sate)

Nuclear reaction	Before unfolding	After unfolding
	C/E	C/E
^{197}Au(n,γ)	0.984	0.989
^{197}Au(n,γ)-Cd	1.013	1.017
^{63}Cu(n,γ)	1.000	1.004
^{115}In(n,n')	1.037	1.034
^{48}Ti(n,p)	0.971	0.984

Table 6 C/E of the reaction rate in the analysis result of NEUPAC-JLOG (Transient state)

Nuclear reaction	Before unfolding	After unfolding
	C/E	C/E
^{197}Au(n,γ)	0.997	0.994
^{197}Au(n,γ)-Cd	1.028	1.009
^{63}Cu(n,γ)	0.971	0.989
^{115}In(n,n')	0.999	1.001
^{48}Ti(n,p)	0.877	0.919

Fig. 1 Overall view of TRACY

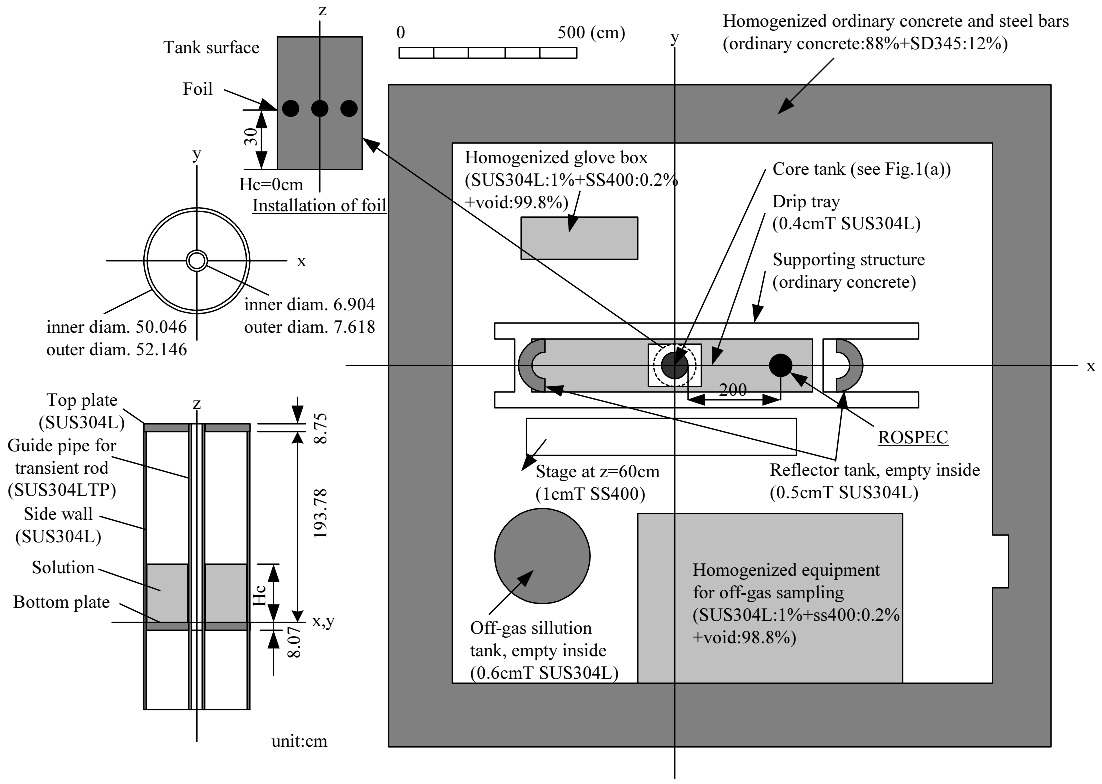

Fig. 2 Installation requirements in TRACY reactor room
* A brand name of recoil proton proportional counter is ROSPEC.

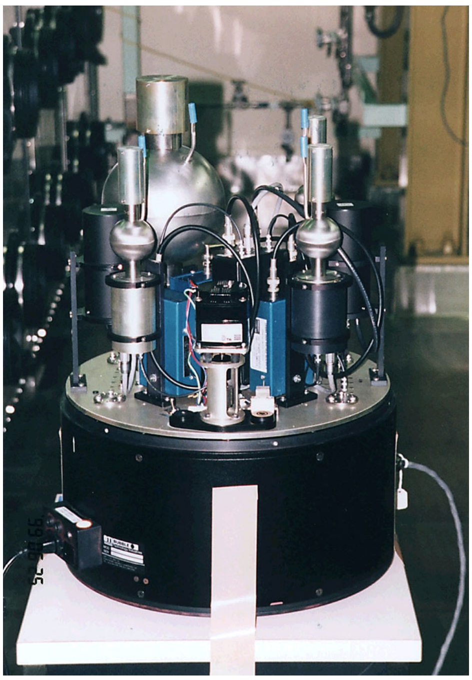

Fig. 3 Recoil proton proportional counter overall view

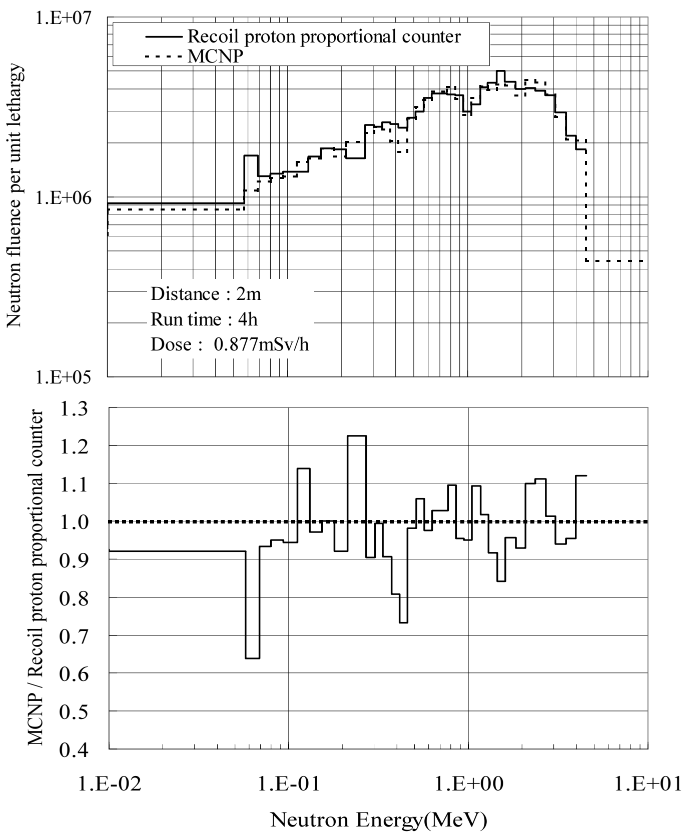

Fig. 4 Comparison with recoil proton proportional counter and MCNP

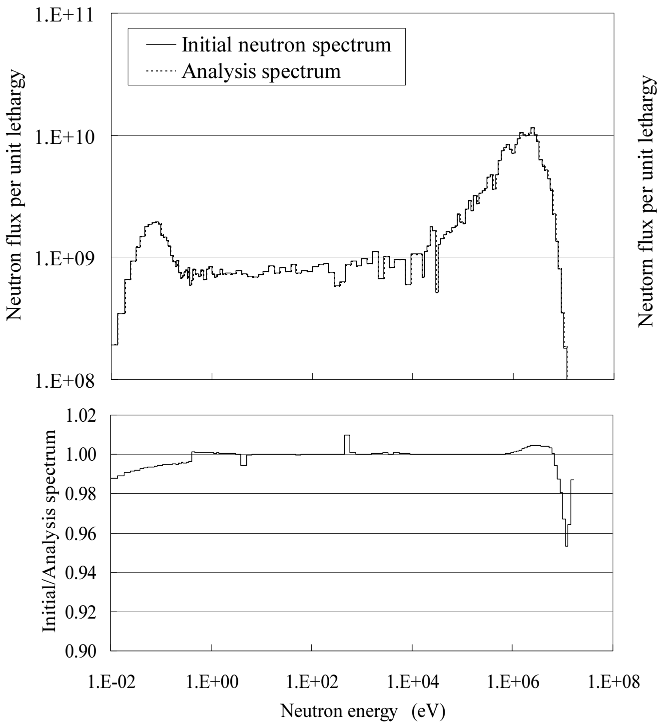

Fig. 5 Unfolding result of NEUPAC-JLOG in stationary state

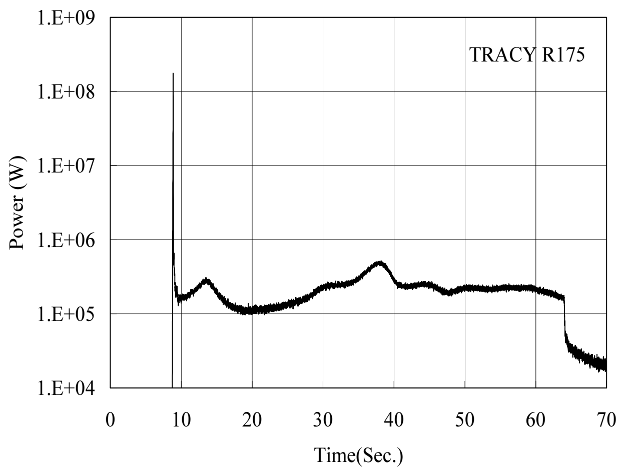

Fig. 6 Power change of transient state operation on TRACY

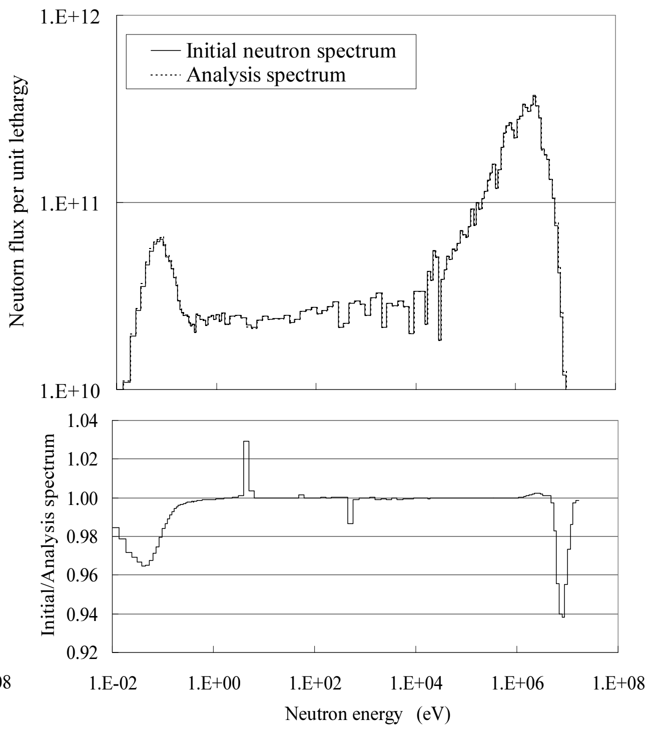

Fig. 7 Unfolding result of NEUPAC-JLOG in transient state