Chapter 5

Neutron Spectrum

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Abstract The subcritical multiplication factor is considered an important index for recognizing, in the core, the number of fission neutrons induced by an external neutron source. In this study, the influences of different external neutron sources on core characteristics are carefully monitored. Here, the high-energy neutrons generated by the neutron yield at the location of the target are attained by the injection of 100 MeV protons onto these targets. In actual ADS cores, liquid Pb–Bi has been selected as a material for the target that generates spallation neutrons and for the coolant in fast neutron spectrum cores. The neutron spectrum information is acquired by the foil activation method in the ^{235}U -fueled and Pb–Bi-zoned fuel region of the core, modeling the Pb–Bi coolant core locally around the central region. The neutron spectrum is considered an important parameter for recognizing information on neutron energy at the target. Also, the neutron spectrum evaluated by reliable methodologies could contribute to the accurate prediction of reactor physics parameters in the core through numerical simulations of desired precision. In the present chapter, experimental analyses of high-energy neutrons over 20 MeV are conducted after adequate preparation of experimental settings.

Keywords Subcritical multiplication factor · Reaction rates · Spectrum index · Spallation neutrons

5.1 Subcritical Multiplication Factor

5.1.1 Theoretical Background

In the Accelerator-Driven System (ADS) study on the subcritical system, neutron multiplication M and subcritical multiplication factor k_s [1] are theoretically expressed with the combined use of total fission reaction rates F in the core (fuel region) and external neutron source rates S at the target, respectively, as follows:

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$$M = \frac{F + S}{S}, \quad (5.1)$$

$$k_s = \frac{F}{F + S}. \quad (5.2)$$

Assuming that the fission reaction rates F do not vary along the wire radius in the thermal neutron field, the total fission reaction rates can be expressed approximately by a the ^{115}In wire length $(n, \gamma)^{116\text{m}}\text{In}$ reaction rates R_{In} as follows:

$$F = \int_V \int_0^\infty \nu \Sigma_f(\underline{r}, E) \phi_s(\underline{r}, E) d\underline{r} dE = C_{\text{Fission}}^{\text{Core}} C_{3D \rightarrow 1D}^{\text{Dimension}} \int_a R_{\text{In}}(x, y_0, z_0) dx, \quad (5.3)$$

where V indicates the whole volume in the system, ν the average number of fission neutrons per fission reaction, Σ_f the fission cross sections, ϕ_s the neutron flux at position r with energy E in the presence of an external source, $C_{\text{Fission}}^{\text{Core}}$ the conversion coefficient of ^{115}In capture reactions to the ^{235}U fission ones, $C_{3D \rightarrow 1D}^{\text{Dimension}}$ the dimension coefficient of x -direction (1D) to x - y - z directions (3D) and a the ^{115}In wire along the fuel regions in the core. The external neutron source rates S can be expressed approximately by source reaction rates R_{source} of $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reactions as follows:

$$S = \int_{r_s} \int_0^\infty s(\underline{r}, E) d\underline{r} dE = C_{\text{Source}}^{\text{Target}} \int_{r_s} R_{\text{source}}(\underline{r}) d\underline{r}, \quad (5.4)$$

where $s(r, E)$ indicates the external neutron source rate at position r with energy E , $C_{\text{Source}}^{\text{Target}}$ the conversion coefficient of $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reactions to neutron generation and r_s the position of the external neutron source. The validity of approximation and the applicability of the methodology mentioned above have been demonstrated previously [2].

Finally, using Eqs. (5.3) and (5.4), M and k_s in Eqs. (5.1) and (5.2), respectively, can be expressed as follows:

$$M = \frac{C_{\text{Fission}}^{\text{Core}} \cdot C_{3D \rightarrow 1D}^{\text{Dimension}} \int_a R_{\text{In}}(x, y_0, z_0) dx}{C_{\text{Source}}^{\text{Target}} \int_{r_s} R_{\text{source}}(\underline{r}) d\underline{r}} + 1, \quad (5.5)$$

$$k_s = \frac{C_{\text{Fission}}^{\text{Core}} \cdot C_{3D \rightarrow 1D}^{\text{Dimension}} \int_a R_{\text{In}}(x, y_0, z_0) dx}{C_{\text{Fission}}^{\text{Core}} \cdot C_{3D \rightarrow 1D}^{\text{Dimension}} \int_a R_{\text{In}}(x, y_0, z_0) dx + C_{\text{Source}}^{\text{Target}} \int_{r_s} R_{\text{source}}(\underline{r}) d\underline{r}}. \quad (5.6)$$

5.1.2 Characteristics of the Target

5.1.2.1 Dimension of Solid Targets

The combined use of the heavy (tungsten, lead and bismuth: W, Pb and Bi) [3] and the light nuclides (beryllium: Be; lithium) was considered useful in accomplishing the study objectives related to the neutron spectrum and the neutron yield. The neutron yield was obtained from the neutrons produced at the surface of the target. The two targets laid with the combined use of heavy and light nuclides are termed “two-layer” targets in this study, and their characteristics were numerically investigated by the MCNPX [4] and SRIM codes [5] with the JENDL/HE-2007 [6] library. The aim of these numerical analyses was to investigate the neutron spectrum in the high-energy region and to determine the thickness of solid targets so that incident protons could be fully stopped inside the target.

From the numerical results by MCNPX, the neutron spectrum was observed in the high-energy region of the solid target used: W, W–Be or Pb–Bi, as shown in Figs. A2.5 and A2.6, when 100 MeV protons were injected onto them. The neutron spectrum was somewhat similar, regardless of the kind of solid target used as shown in Fig. A2.5, although at each target it was comparatively different ranging from 1 to 10 MeV. Of particular interest here is the influence of the difference in the neutron spectrum, caused by the kind of target used, on neutron multiplication in the core. In the numerical simulations of neutron generation of the target, the neutron spectrum of W-Be target (two-layer target) was compared remarkably with other single targets, ranging from 85 to 100 MeV, as shown in Fig. A2.6, and the difference between the two spectra was attributed to the scattering reactions of Be to high-energy protons. The aim in using the two-layer target was to acquire the neutron spectrum in the high-energy region and the neutron yield of high-energy neutrons. Consequently, the proton beams actually penetrated the Be target, and conversely stopped inside the W target. The thickness of the solid targets was correctly determined by the numerical results of the range of high-energy protons with the use of the SRIM code. Lastly, the dimensions of the solid targets were determined as shown in Table A2.6, and since the size of the proton beam spot was 40 mm in diameter, the targets were adequately covered with the proton beams and satisfactorily penetrated and stopped fully in the solid targets.

5.1.2.2 Experimental Settings

The ADS experiments were carried out in the A-core (Fig. A2.1) with the combined use of fuel and polyethylene reflector rods. In the A-core, the fuel assembly shown in Fig. A2.2 is composed of 60 unit cells, and upper and lower polyethylene blocks about 25" and 20" long, respectively, in an aluminum (Al) sheath $2.1 \times 2.1 \times 60''$. In the A-core, the neutron flux information was acquired from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reactions using the indium (In) wire (1 mm diameter and 800 mm length), under

the assumption that, in the thermal neutron region, the cross sections of $^{235}\text{U}(n, f)$ are proportional to those of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ shown in Fig. A2.7. The In wire was set along the vertical direction (14, 13-P, A') at the axial center position shown in Fig. A2.1. An Al ($10 \times 10 \times 1$ mm, (15, H) in Fig. A2.1) and an In ($10 \times 10 \times 1$ mm, (15, H) in Fig. A2.1) foils were attached at the location of the target to monitor information on the generation of protons and spallation neutrons through $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ and $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reactions (threshold energy of 0.3 MeV), respectively. For an easy understanding of experimental analyses, the methodology of normalized reaction rates was introduced with the comparison between the experimental and calculated results at the location of the target and in the core region: the $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ and $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates normalized by $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ and $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ ones, respectively, were estimated in the experiments, and interpreted as actual values of proton yield and neutron yield, respectively, in terms of the influence of the external neutron source. The main characteristics of proton beams were as follows: 100 MeV energy, 0.7 nA intensity, 20 Hz beam repetition, 100 ns beam width and $1.0 \times 10^7 \text{ s}^{-1}$ neutron generation. The irradiation time of all the foils and wire was about 3 h. The measured subcriticality, 2,900 pcm, of the core was obtained by the full insertion of control (C1, C2 and C3) and safety (S4, S5 and S6) rods, as shown in Fig. A2.1. The reactivity worth of all control and safety rods was evaluated by the rod drop method and the positive period method beforehand. The measured reaction rates varied according to the kind of solid target used, as shown in Fig. A2.8, under the subcritical level 2,900 pcm. The reaction rates were high with the combined use of W and Be targets, and the moderation (thermal) peak caused by the high-energy neutrons was observed in the polyethylene region, mostly in the two-layer target (W-Be target). Neutron multiplication has been obtained successfully by the $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates in the core region because the relation between $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ and $^{235}\text{U}(n, f)$ reaction rates in the core region is apparently applicable to the subcritical multiplication analyses through the proportionality of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ and $^{235}\text{U}(n, f)$ cross sections in the thermal region [2].

5.1.2.3 Numerical Simulations

The numerical calculations were performed with the combined use of MCNPX and JENDL/HE-2007 for high-energy protons and high-energy neutrons, JENDL-4.0 [7] for transport, and JENDL/D-99 [8] for reaction rates. With MCNPX, the calculated reaction rates were obtained from the evaluation of volume tallies of activation foils, and since the effects of their reactivity are not negligible, they were included in the simulated geometry and transport calculations. The eigenvalue calculations were conducted for 1,000 active cycles of 100,000 histories. The subcriticalities in the eigenvalue calculations had statistical errors within 0.01 % $\Delta k/k$ (10 pcm), and the reaction rates in the fixed source calculations were within 3% as determined with the use of the total 1×10^8 histories. The precision of numerical subcriticality in the

eigenvalue calculations was attained within the relative difference of 5% between the experimental and the numerical results.

The measured reaction rates of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ (wire: 1 mm diameter and 800 mm long) in the core were normalized by those of $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ (foil: $10 \times 10 \times 1$ mm) at the location of the target. The experimental errors in the activation wire and foil were estimated within 15% and 5%, respectively, including the statistical error of γ -ray counts and the full width at half maximum (FWHM) of the γ -ray spectrum peak. The calculated reaction rates of the ^{115}In wire and foil in the core were included in the simulated geometry and transport calculations, and deduced from tallies taken in the fixed source calculations. Also, the calculated reaction rates of the In foil at the target were obtained by the previously fixed source calculations, modeling the proton injection on the solid target.

The calculation/experiment (C/E) value of the experiments and the calculations of M in Eq. (5.5), as shown in Table 5.1, was good within an error of 7%, and the absolute value of M was large in the W-Be target, compared with that in the other two. Also, the values of neutron multiplication were differently compared with the W and Pb-Bi targets, indicating that neutron multiplication was mostly influenced by the neutron spectrum of the external neutron source, such as the W-Be target. For any target, F and S were numerically estimated with the use of conversion coefficients $C_{\text{Fission}}^{\text{Core}} (\simeq 0.25)$, $C_{3D \rightarrow 1D}^{\text{Dimension}} (\simeq 1.05)$ and $C_{\text{Source}}^{\text{Target}} (\simeq 1.0 \times 10^5)$ as shown in Eqs. (5.3) and (5.4), including the proportionality between ^{235}U fission and ^{115}In capture cross sections in the thermal neutron field, the dimension effect and the source conversion, respectively. These coefficients were applied to the evaluation of M because it is difficult to obtain F and S in Eqs. (5.1) and (5.2) directly by the experiments.

While the accuracy of M was attributable to the experimental validation of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates, the actual influence of the kind of solid target used was considered significant: neutron multiplication increased over 30% in the two-layer target, compared with that in the W target. These experimental results clearly demonstrated the influence of the two-layer target on neutron multiplication, and the high-energy neutrons in the region ranging from 85 to 100 MeV contributed significantly to the neutron characteristics of ADS through the selection of the appropriate target.

The C/E values of the experiments and the calculations of k_s in Eq. (5.6) are shown in Table 5.2: the discrepancy between the experiments and the calculations was within the relative difference of 8%, as in M . The values of the measured and the calculated k_s demonstrated that the source term contributed largely to the estimation of k_s , since the external source was located inside the core (core target location

Table 5.1 Neutron multiplication M in Eq. (5.1) deduced from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates in subcriticality 2,900 pcm (Ref. [3])

Target	Calculation	Experiment	C/E value
W	1.73 ± 0.01	1.85 ± 0.02	0.93 ± 0.01
W-Be	2.29 ± 0.01	2.36 ± 0.03	0.97 ± 0.01
Pb-Bi	1.95 ± 0.01	1.94 ± 0.02	1.01 ± 0.01

Table 5.2 Subcritical multiplication factor k_s in Eq. (5.2) deduced from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates in subcriticality 2,900 pcm (Ref. [3])

Target	Calculation	Experiment	C/E value
W	0.42033 ± 0.00100	0.45874 ± 0.01003	0.92 ± 0.02
W–Be	0.56355 ± 0.00100	0.57662 ± 0.01510	0.98 ± 0.03
Pb–Bi	0.48830 ± 0.00100	0.48488 ± 0.01335	1.01 ± 0.03

(15, H) in Fig. A2.1). Moreover, the results demonstrated that the Pb–Bi target was confirmed fairly well for an upcoming target in actual ADS, as predicted in several experimental and numerical analyses, by its comparison with the W target, although its static core characteristics were almost the same as those of the W target.

5.1.3 Effects of Neutron Spectrum

5.1.3.1 Core Configuration

Starting from the reference core (Fig. A2.11) that reached a criticality, reaction rate experiments [9] (Cases II-1 to II-4 in Fig. 2A.16) in ADS were carried out in the KUCA A-core by varying subcriticality ranging from 2,483 to 11,556 pcm, as shown in Figs. A2.16a through A2.16d. The ADS cores comprise normal fuel assembly “F,” partial fuel assembly “16,” Pb–Bi-zoned fuel assembly “f,” polyethylene moderator “p” and reflector rods. Normal fuel assembly “F” is composed of 36 cells of a highly-enriched uranium (HEU) fuel plate 1/16" thick, polyethylene (p) plates 3/8" thick, and upper and lower polyethylene blocks about 25" and 20" long, respectively, in an aluminum (Al) sheath $2.1 \times 2.1 \times 60$ ", as shown in Fig. A2.12. The numeral “16” corresponds to the number of unit cells in the partial fuel assemblies for reaching criticality in the core shown in Fig. A2.14. The Pb–Bi-zoned fuel assembly “f” is composed of 30 unit cells with two HEU plates 1/8" thick, Pb–Bi plate 1/8" thick, 30 unit cells of two HEU plates 1/8" thick and polyethylene plates 1/8" thick, as shown in Fig. A2.13.

The subcriticality in the pcm units was experimentally obtained with the combined use of control rod worth and its calibration curve measured by the rod drop method and the positive period method, respectively, in Case II-1 (Fig. A2.16a). For the deduction of subcriticality in Case II-1, the effective delayed neutron fraction β_{eff} and the neutron generation time Λ were obtained with 783 pcm and 4.64E-05 s, respectively, with MCNP6.1 and JENDL-4.0. In Cases II-2 to II-4 (Figs. A2.16b–d), the reference of subcriticality in the pcm units was attained with MCNP6.1 eigenvalue calculations and the JENDL-4.0 library, because the reactivity of control and safety rods was made by the substitution of fuel assembly rods for the polyethylene ones.

5.1.3.2 Experimental Settings

In reaction rate experiments, an In foil ($10 \times 10 \times 1$ mm) was attached at the boundary between (15, L) and (15, M) shown in Figs. A2.16a, d, in order to obtain the information on spallation neutrons with the use of $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reactions (threshold energy 0.3 MeV). Six foils, including gold (^{197}Au ; bare and cadmium: Cd covered), iron (^{56}Fe), aluminum (^{27}Al), ^{115}In and nickel (^{58}Ni), were used as activation foils (Table A2.22) to cover a wide range of threshold energy and acquire neutron spectrum information in the region of the Pb–Bi-zoned core affected by spallation neutrons. The foils were attached at the boundary between fuel assemblies (15, M) and (15, O) (Figs. A2.16a–d) with the dimensions and variations as shown in Fig. A2.19. Among the six foils, the ^{197}Au foil (Au-bare) was taken as a normalization factor of reactor power for ^{56}Fe , ^{27}Al , ^{115}In and ^{58}Ni foils. Furthermore, the Cd ratio was experimentally obtained by the combination of two Au foils: Au-bare and Au-Cd sandwiched between two Cd plates (10 mm diameter and 1 mm thick), as a spectrum index. For an easy understanding of neutron irradiation, neutron spectrum by the MCNP calculations was obtained at several significant positions around the Pb–Bi-zoned core (Fig. A2.16a), as shown in Fig. 5.1: at the location of the target, the Pb–Bi-zoned fuel assembly (15, M) and two positions (14-13, P-O) and (14-13, L-K) at the boundary between Pb–Bi-zoned and normal fuel regions.

The information on neutron flux distribution was acquired from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rate distribution with the use of the In wire (1 mm diameter and 680 mm long), which was set at the gap between the Pb–Bi-zoned and the normal fuel regions along (14-13, P-A) at a height of 700 mm from the bottom of the core. The reaction rates of the $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ foil at the Pb–Bi target (the boundary between (15, L) and (15, M)) were taken as the normalization factor of the source generating spallation neutrons.

Spallation neutrons were generated by bombarding 100 MeV proton beams from the FFAG accelerator onto the Pb–Bi target. To compensate for the drawback of

Fig. 5.1 Neutron spectra at target and several locations in Fig. A2.16a during injection of 100 MeV protons onto the Pb–Bi target (Ref. [9])

Table 5.3 Measured and calculated (MCNP6.1) subcriticalities [pcm] in Cases II-1 through II-4 (Ref. [9])

Case	Calculation	Experiment	C/E
II-1	2483 ± 12	2302 ± 6	1.08 ± 0.01
II-2	4812 ± 12	–	–
II-3	9895 ± 12	–	–
II-4	11556 ± 12	–	–

locating the original target outside the core at (15, A'), the Pb–Bi target was moved to (15, L) inside the core on the basis of experimental results obtained in a previous study [10]. Specifications of proton beams were as follows: 100 MeV energy, 1.0 nA intensity, 20 Hz pulsed frequency, 50 ns beam width and 40 mm diameter spot size at the location of the target.

5.1.3.3 Numerical Simulations

To validate the accuracy of calculated subcriticality, eigenvalue calculations were performed with MCNP6.1 [11] and JENDL-4.0 for transport, by comparison with measured subcriticality in Case II-1. Here, since the effect of neutron detectors, control (safety) rods and irradiation materials is not negligible, they were included in simulated geometry and transport calculations with MCNP6.1. The total number of histories used in the eigenvalue calculations was $1\text{E} + 08$ ($1\text{E} + 05$ histories; $1\text{E} + 03$ cycles) with a standard deviation of 8 pcm. A comparison between measured and calculated subcriticalities revealed an agreement with a relative difference of 8%, through the calculation/experiment (C/E) value, as shown in Table 5.3. Also, these results demonstrated that the precision of numerical subcriticality by the eigenvalue calculations was considered carefully to ensure the reliability of the fixed-source calculations with MCNP6.1 in the ADS core with spallation neutrons.

Reaction rate calculations with spallation neutrons were performed by the fixed-source calculations (total number of histories: $1\text{E} + 08$) with the combined use of MCNP6.1 and JENDL/HE-2007 for high-energy protons and spallation process, JENDL-4.0 for transport and JENDL/D-99 for reaction rates. Proton beams (100 MeV) were modeled as a spot size of 40 mm in diameter injected onto the Pb–Bi target at location (15, L). The reaction rates by the fixed-source calculations were numerically obtained by the evaluation of volume tallies of activation foils and the In wire with a statistical error within 5%.

5.1.3.4 Indium Wire Distribution

Thermal neutron flux information was acquired from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates with the use of the In wire, assuming that, in the thermal neutron region, the cross sections of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ are proportional to those of $^{235}\text{U}(n, f)$, as shown in

Fig. A2.7. To investigate the accuracy of reaction rate analyses at different subcritical states, the $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rate distributions along the (14-13, P-A) region in Figs. A2.16a–d were measured with subcriticality ranging from 2,483 to 11,556 pcm. From a comparison between the measured and the calculated $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rate distributions in Cases II-1 and II-3, as shown in Figs. 5.2a, b, respectively, the calculated $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rates reproduced successfully the measured ones at an acceptable accuracy with the MCNP fixed-source calculations in the subcritical states, demonstrating the allowance within the experimental statistical error.

To estimate the effect of spallation neutrons on the neutron multiplication in ADS, the subcritical multiplication factor k_s is acquired as Eq. (5.6), on the basis of a theoretical background [5]. A comparison between experiments and calculations of k_s

Fig. 5.2 Comparison between measured and calculated $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rate distributions along (14-13, P-A) region (Ref. [9])

(a) Case II-1

(b) Case II-3

Table 5.4 Comparison between measured and calculated (MCNP6.1) k_s values in Cases II-1 through II-4 (Ref. [9])

Case	Calculation	Experiment
II-1	0.9989 ± 0.0005	0.9996 ± 0.0008
II-2	0.9997 ± 0.0005	0.9991 ± 0.0009
II-3	0.9990 ± 0.0005	0.9991 ± 0.0009
II-4	0.9985 ± 0.0005	0.9989 ± 0.0005

deduced by Eq. (5.6) showed fairly good agreement within a relative difference of 1%, as shown in Table 5.4, and demonstrated an exact reproduction of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ and $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reaction rates with the MCNP fixed-source calculations.

5.2 Threshold Energy Reactions

5.2.1 Foil Activation Method

The experimental results of reaction rates (RR) [$\text{s}^{-1} \text{ cm}^{-3}$] were obtained by measuring total counts of the peak energy of γ -ray emissions. The value of RR was deduced from that of saturation activity D_∞ [s^{-1}] that is proportional to the reaction rates by using the following equations:

$$D_\infty = \frac{\lambda T_c C}{\varepsilon_D \varepsilon_E (1 - e^{-\lambda T_i}) e^{-\lambda T_w} (1 - e^{-\lambda T_c})}, \quad (5.7)$$

$$RR = D_\infty \frac{\rho}{M} = \frac{\lambda T_c C \rho}{\varepsilon_D \varepsilon_E (1 - e^{-\lambda T_i}) e^{-\lambda T_w} (1 - e^{-\lambda T_c}) M}, \quad (5.8)$$

where λ [s^{-1}] indicates the decay constant, T_c —[s] the measurement counting time, C [1/s] the counting rate, ε_D [%] the detection efficiency, ε_E [%] the emission rate, T_i [s] the irradiation time, T_w [s] the waiting time until the measurement starting after the irradiation, ρ [g cm^{-3}] the density and M [g] the mass of the foil. The experimental errors in the activation wire and foils were estimated within 15% and 5%, respectively, including the statistical errors of γ -ray counts and full width at half maximum of the γ -ray spectrum peak.

5.2.2 Activation Foils

The measured reaction rates of ^{197}Au (bare), ^{197}Au (Cd), ^{56}Fe , ^{27}Al , ^{115}In and ^{58}Ni foils irradiated in the subcritical states are shown in Table 5.5. A comparison between measured and calculated reaction rates is presented in Table 5.6 and Fig. 5.3. As shown in Table 5.6, in Case II-1 (2483 pcm), non-threshold and low-threshold energy

Table 5.5 Measured reaction rates of activation foils in Cases II-1 through II-4 (Ref. [9])

Reaction	Measured reaction rate ($\text{s}^{-1} \text{ cm}^{-3}$)			
	Case II-1	Case II-2	Case II-3	Case II-4
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ (target)	$(1.87 \pm 0.24) \times 10^5$	$(1.12 \pm 0.04) \times 10^5$	$(1.62 \pm 0.03) \times 10^5$	$(1.02 \pm 0.04) \times 10^5$
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ (bare)	$(8.88 \pm 0.02) \times 10^6$	$(4.88 \pm 0.04) \times 10^6$	$(3.51 \pm 0.08) \times 10^6$	$(2.53 \pm 0.04) \times 10^6$
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ (Cd)	$(7.84 \pm 0.09) \times 10^6$	$(4.46 \pm 0.04) \times 10^6$	$(3.11 \pm 0.04) \times 10^6$	$(2.30 \pm 0.03) \times 10^6$
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ (Core)	$(8.60 \pm 0.13) \times 10^4$	$(4.27 \pm 0.22) \times 10^4$	$(4.27 \pm 0.03) \times 10^4$	$(2.86 \pm 0.06) \times 10^4$
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	$(4.90 \pm 0.08) \times 10^4$	$(3.18 \pm 0.03) \times 10^4$	$(3.23 \pm 0.16) \times 10^4$	$(2.00 \pm 0.10) \times 10^4$
$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	$(1.82 \pm 0.07) \times 10^3$	$(1.26 \pm 0.02) \times 10^3$	$(1.55 \pm 0.03) \times 10^3$	$(1.39 \pm 0.02) \times 10^3$
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	$(1.54 \pm 0.05) \times 10^3$	$(1.11 \pm 0.02) \times 10^3$	$(1.62 \pm 0.03) \times 10^3$	$(1.10 \pm 0.01) \times 10^3$

Table 5.6 C/E values between measured and calculated reaction rates in Cases II-1 through II-4 (Ref. [9])

Reaction	Case II-1	Case II-2	Case II-3	Case II-4
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ (bare)	1.14 ± 0.09	0.96 ± 0.06	0.90 ± 0.06	0.75 ± 0.07
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ (Cd)	1.10 ± 0.09	0.81 ± 0.05	0.80 ± 0.06	0.71 ± 0.07
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ (Core)	0.86 ± 0.02	0.69 ± 0.04	0.52 ± 0.01	0.47 ± 0.02
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	0.99 ± 0.04	0.74 ± 0.03	0.59 ± 0.03	0.67 ± 0.04
$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	0.60 ± 0.03	0.54 ± 0.03	0.47 ± 0.02	0.40 ± 0.02
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	0.47 ± 0.01	0.26 ± 0.02	0.19 ± 0.01	0.22 ± 0.02

Fig. 5.3 C/E value between measured and calculated reaction rates by varying subcriticality in Cases II-1 through II-4 (Ref. [9])

reaction rates showed good agreement between experimental and numerical reaction rates, within a relative difference of around 10%. Conversely, the numerical calculations for high-threshold reaction rates of ^{27}Al and ^{56}Fe foils revealed an underestimation of about 50% at most. Besides, by varying subcriticality, the deeper its level, the smaller the C/E value, as shown in Fig. 5.3. Notably, in ADS with spallation neutrons, the dependence of reaction rates on subcriticality was revealed in the accuracy of C/E, under subcriticality ranging from 2,483 to 11,556 pcm.

Compared with previous analyses of ADS with 14 MeV neutrons [12], a discrepancy between measured and calculated reaction rates was, by contrast, larger in ADS with spallation neutrons, and considered attributable mainly to the uncertainty of reaction rates in the high-energy thresholds and the difficulty in the exact simulation of defocused proton beams. In the reaction rate experiments at KUCA, the proton beams were transported through an un-vacuumed air space from the location of the original target to that of the Pb–Bi target, although the proton beams were in a vacuum until they reached the location of the original target. As a result, the proton beam spot was easily defocused at the location of the Pb–Bi target by the scattering reactions of high-energy protons in the air space (15, A-L; Figs. A2.16a–d).

5.3 Spectrum Index

5.3.1 Cd Ratio

In reactor physics experiments, the Cd ratio is generally interpreted as the neutron spectrum index, where the relation between absorption cross sections of Au and Cd is used in foil activation analyses. For thermal and resonance regions, 0.18 eV corresponds to the boundary energy (cut-off energy) between thermal and epithermal neutron regions. By defining the Cd ratio as that of activation reaction rates $R_{\text{Au-bare}}$ and $R_{\text{Au-Cd}}$ of Au foils (Au-bare and Au-Cd-sandwiched plates, respectively), the Cd ratio can be expressed as follows:

$$\text{Cd ratio} = \frac{R_{\text{Au-bare}}}{R_{\text{Au-Cd}}}. \tag{5.9}$$

On the basis of the definition in Eq. (5.9), the Cd ratio was evaluated by two Au foils (bare and Cd-sandwiched plates) in subcritical core configurations, as shown in Table 5.7. The C/E values of the Cd ratio were found in good agreement in terms of the relative difference error, demonstrating the accuracy of Au absorption reaction rates with MCNP simulations in both thermal and epithermal neutron regions.

5.3.2 In Ratio

As discussed in Sect. 5.3.1, the ratio of thermal neutrons to epithermal ones obtained by the Cd ratio is readily understood as a simple interpretation of the spectrum index being about 10%, by subtracting the unity value from the experimental results of the Cd ratio shown in Table 5.7. Moreover, among several spectrum indexes of fast reactors, the value of F8/F5, which is the ratio of fast fission and total fission reaction rates of ^{238}U and ^{235}U , respectively, is especially introduced as a quantitative evaluation of the neutron spectrum. From the point of view of the spectrum index, a new assumption suggested here is supportable by introducing the same concept as the Cd ratio and the value of F8/F5, regarding thermal and fast neutrons.

This section is devoted to a worthwhile discussion of a new index of the neutron spectrum in ADS through a new definition of ^{235}U fission ratio of thermal and fission

Table 5.7 Comparison between measured and calculated Cd ratios in Cases II-1 through II-4 (Ref. [9])

Case	Calculation	Experiment	C/E
II-1	1.15 ± 0.09	1.11 ± 0.06	1.04 ± 0.10
II-2	1.24 ± 0.09	1.09 ± 0.02	1.13 ± 0.09
II-3	1.27 ± 0.09	1.12 ± 0.04	1.13 ± 0.09
II-4	1.16 ± 0.09	1.10 ± 0.04	1.05 ± 0.09

neutrons in the specified Pb–Bi-zoned fuel region from (14-13, L; y_1) to (14-13, O; y_2) shown in Figs. A2.16a–d, with an interpretation similar to the Cd ratio mentioned in Sect. 5.3.1 as follows:

$$^{235}\text{U fission ratio} = \frac{\int_{y_1}^{y_2} R_{U-\text{fission}}^{\text{thermal}}(x_0, y, z_0)dy}{\int_{y_1}^{y_2} R_{U-\text{fission}}^{\text{fast}}(x_0, y, z_0)dy}, \quad (5.10)$$

where $R_{U-\text{fission}}^{\text{thermal}}$ and $R_{U-\text{fission}}^{\text{fast}}$ indicate $^{235}\text{U}(n, f)$ reaction rates in the thermal (less than 0.1 eV) and fast (more than 0.1 MeV) neutron regions, respectively.

The numerator in Eq. (5.10) is expressed as follows, with the assumption as discussed in Sect. 5.1.1:

$$\int_{y_1}^{y_2} R R_{U-\text{fission}}^{\text{thermal}}(x_0, y, z_0)dy = C_{\text{fission}}^{\text{thermal}} \int_{y_1}^{y_2} R_{\text{In}(n, \gamma)}^{\text{thermal}}(x_0, y, z_0)dy, \quad (5.11)$$

where $C_{\text{fission}}^{\text{thermal}}$ indicates the proportionality coefficient of cross sections between $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ and $^{235}\text{U}(n, f)$ reactions in the thermal neutron region. Similarly, the denominator in Eq. (5.10), by a new assumption of the proportionality of the In inelastic scattering (threshold energy 0.4 MeV) and ^{235}U fast fission reactions, is written as follows:

$$\int_{y_1}^{y_2} R_{U-\text{fission}}^{\text{fast}}(x_0, y, z_0)dy = C_{\text{fission}}^{\text{fast}} \int_{y_1}^{y_2} R_{\text{In}(n, n')}^{\text{fast}}(x_0, y, z_0)dy, \quad (5.12)$$

where $C_{\text{fission}}^{\text{fast}}$ indicates the proportionality coefficient of cross sections between $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ and $^{235}\text{U}(n, f)$ reactions in the fast neutron region, although the assumption of proportionality in the fast neutron region is somewhat complicated from the viewpoint of the characteristics of cross sections. On the basis of Eqs. (5.11) and (5.12), the results of $C_{\text{fission}}^{\text{thermal}}$ and $C_{\text{fission}}^{\text{fast}}$ were found to be nearly constant around 1.11 and 6.25, respectively, with the MCNP fixed-source calculations.

Generally, in the experiments, while it is apparently difficult to measure $^{235}\text{U}(n, f)$ reaction rates directly, a convenient alternative is to introduce the proportionality of cross sections discussed in Sect. 5.1.2. Subsequently, special attention was directed to the property of In wire reaction rates to experimentally obtain neutron flux information on both thermal and fast energy regions simultaneously. Assuming that the ^{235}U fission ratio in Eq. (5.9) corresponds approximately to the ratio of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ and $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reaction rate distributions, a new spectrum index of the In ratio is, by introducing the coefficients of $C_{\text{fission}}^{\text{thermal}}$ and $C_{\text{fission}}^{\text{fast}}$, defined as follows:

$$\text{In ratio} = \frac{C_{\text{fission}}^{\text{thermal}} \int_{y_1}^{y_2} R_{\text{In}(n, \gamma)}^{\text{thermal}}(x_0, y, z_0)dy}{C_{\text{fission}}^{\text{fast}} \int_{y_1}^{y_2} R_{\text{In}(n, n')}^{\text{fast}}(x_0, y, z_0)dy}. \quad (5.13)$$

Compared with the numerical results of ^{235}U fission ratio and In ratio, the assumption in Eqs. (5.11) and (5.12) is considered mostly valid with a relative difference of around 10%, as shown in Table 5.8. A comparison between measured and calculated In ratios revealed fairly good agreement within a relative difference of 5% in Cases II-1 through II-4 the ratio, as shown in Fig. 5.4, and was found constant especially in the Pb–Bi-zoned fuel regions, except at the boundaries between the fuel regions. Furthermore, the results in Fig. 5.4 demonstrated that fast neutrons were dominant over the Pb–Bi-zoned fuel region through the ratio of thermal and fast neutrons shown in Eq. (5.13). From the results in Table 5.8 and Fig. 5.4, the consistency between measured and calculated In ratios supported an interesting interpretation that the In ratio is useful in conveniently determining the ratio of thermal and fast neutrons as another neutron spectrum index of ADS, in addition to the Cd ratio, through two different In reaction rate distributions obtained by the In wire.

Table 5.8 Comparison between measured and calculated In ratios in Cases II-1 through II-4 (Ref. [9])

Case	^{235}U fission ratio in Eq. (5.10)	In ratio in Eq. (5.13)		
	Calculation	Calculation	Experiment	C/E
II-1	$(5.16 \pm 0.08) \times 10^{-3}$	$(4.62 \pm 0.08) \times 10^{-3}$	$(4.52 \pm 0.10) \times 10^{-3}$	1.02 ± 0.03
II-2	$(2.21 \pm 0.10) \times 10^{-3}$	$(2.15 \pm 0.10) \times 10^{-3}$	$(2.17 \pm 0.10) \times 10^{-3}$	0.99 ± 0.07
II-3	$(1.79 \pm 0.07) \times 10^{-3}$	$(1.60 \pm 0.07) \times 10^{-3}$	$(1.57 \pm 0.11) \times 10^{-3}$	1.02 ± 0.08
II-4	$(1.70 \pm 0.09) \times 10^{-3}$	$(1.54 \pm 0.09) \times 10^{-3}$	$(1.48 \pm 0.10) \times 10^{-3}$	1.04 ± 0.10

Fig. 5.4 Comparison between measured and calculated In ratios (Eq. (5.12)) in Case II-1 (Ref. [9])

5.4 Spallation Neutrons

5.4.1 Neutron Spectrum Analyses

5.4.1.1 Experimental Settings

High-energy protons were generated by the FFAG accelerator under the following parameters: 100 MeV energy, 30 pA intensity, 30 Hz repetition rate and 200 ns beam width. On the downstream of the FFAG beam line, the W was set at the location (15, A'; Fig. A2.1) of the original target (80 mm diameter and 10 mm thick); the thickness was determined on the basis of previous experimental and numerical analyses [13] for the injection of high-energy proton beams onto the W target. For the proton beam configuration modeled by numerical simulations, the size of the proton beam spot was requisite experimentally and precisely, when 100 MeV protons were injected onto the tungsten target where the spallation neutrons are generated. The Gafchromic film [14], which is very sensitive to the charged particles, was then used to evaluate the size of the proton beam spot injected onto the W target, since a graphic image on the film is acquired quickly after the irradiation of charged particles for a short time.

The reaction rates for threshold energy of high-energy neutrons and the continuous energy distribution of the spallation neutrons at the target were acquired by the foil activation method and the organic liquid scintillator, respectively [15]. The high-energy neutrons (spallation neutrons) of threshold reactions $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ ($n = 3, \text{ to } 12$) over 15 MeV have been generated by the injection of high-energy protons over 100 MeV. Here, to obtain the reaction rates by high-energy neutrons at the target, ^{209}Bi was selected as an activation foil (Table 5.9) to cover threshold energies over 15 MeV, and ^{115}In was selected as a normalization factor for monitoring the spallation neutrons at the target to cover threshold reactions $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ over 0.3 MeV. Foil dimensions at the target were as follows: ^{209}Bi , 50 mm in diameter and 3 mm thick, ^{115}In , $10 \times 10 \times 1$ mm, and two foils were set around the target region as shown in Fig. 5.5. Additionally, nine other ^{115}In foils ($10 \times 10 \times 3$ mm) were placed in a circle (100 mm radius) around the target at 30° intervals on an acrylic plate, to investigate the angular distribution of spallation neutrons as shown in Fig. 5.6. The irradiation time of ^{209}Bi and ^{115}In foils was four hours for measuring the neutron yield of spallation neutrons, and their reaction rates were measured by the high-purity germanium detector (ORTEC, GEM60P). Besides the previous study [12, 13, 16], the detection efficiency of the germanium detector was determined by the fitting line obtained from the energy calibration with the use of several γ -ray standard sources.

The continuous energy distribution of spallation neutrons was determined by the organic liquid scintillator (Nuclear Enterprises Ltd., NE213 Scintillator; 5" in diameter and 5" long) set directly facing the W target without any reactor components as shown in Fig. 5.7. The measurement circulation of the organic liquid scintillator was as indicated in Fig. 5.8. The main advantage of the measurement system is that the two signals (rise time and light output of γ -ray and neutron) acquired coincidentally

Table 5.9 Threshold energy, half-life and γ -ray energy of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reactions ($x = 3$ to 12) (Ref. [15])

x	Threshold energy (MeV)	Half-life ($T_{1/2}$)	Emission γ -ray energy (keV)
3	14.42	38.3 y	569.7 (97.8) 1063.6 (74.9)
4	22.52	6.24 d	803.1 (98.9) 881.1 (66.2) 1718.7 (31.8)
5	29.62	15.31 d	703.4 (31.5) 1763.4 (32.5)
6	38.13	11.22 h	374.8 (81.8) 899.2 (98.5) 983.9 (59.1)
7	45.37	11.76 h	820.2 (29.6) 825.2 (14.6) 1847.3 (11.4)
8	54.24	1.67 h	422.1 (83.7) 658.5 (60.6) 961.7 (99.3)
9	61.69	1.77 h	786.4 (9.5) 935.7 (11.3)
10	70.89	36.45 m	419.7 (91.3) 462.3 (98.3) 1026.4 (100.0)
11	78.47	27.12 m	425.3 (22.0) 560.1 (22.0)
12	87.94	11.85 m	562.4 (79.0) 1063.4 (100.0)

(): Emission rate [%]

Fig. 5.5 Experimental setting of activation foils (Bi and In) for measuring the reaction rates at the target position (Ref. [15])

Fig. 5.6 Experimental setting of tungsten target for angular distribution of spallation neutrons (Ref. [15])

Fig. 5.7 Experimental setting of the organic liquid scintillator (NE213) (Ref. [15])

with the use of specific equipment (Laboratory Equipment Corp., Dual MCA) readily provide a two-dimensional graphic image (rise time and light output). This system allows the rise time information on two signals to accomplish easy discrimination between the γ -ray and the neutron, and the signals can be converted into fluorescence signals of charged particles (recoil protons generated in reaction with neutrons).

Fig. 5.8 Measurement circulation of the organic liquid scintillator (NE213) (Ref. [15])

5.4.1.2 Experimental Analyses

The 10 mm-thick W target was determined on the basis of experimental and numerical analyses [13] from the viewpoint of the full stop of proton beams within the W target. On the other hand, the proton beam irradiation experiments were carried out to monitor the size of the proton beam spot with the use of a Gafchromic film, which is highly sensitive to charged particles. The Gafchromic film was irradiated for two minutes and set on the surface (downstream beam) outside the stainless steel flange (Fig. 5.5) without the W target, although the influence of the stainless steel flange was slightly found in the proton beam profile. It demonstrated that the proton strength was distributed by the downstream beams (Fig. 5.9), with a relative distribution within an approximately 40 mm diameter spot (Fig. 5.10). The experimental result of the Gafchromic film was very useful for modeling the size of the proton beam

Fig. 5.9 Measured result (Gafchromic film) of proton strength distribution of 100 MeV protons at downstream beam without the tungsten target (Ref. [15])

Fig. 5.10 Scanning relative result of proton strength distribution in Fig. 5.9 of 100 MeV protons at the downstream beam without the tungsten target (Ref. [15])

spot in numerical simulations, therefore, the irradiation experiments were important for evaluating the size of the proton beam spot for which the W target (80 mm in diameter) was considered sufficient to cover.

No value for the 100 MeV proton irradiation was observed in $3n$, $9n$ through $12n$ reactions of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ ($x = 3\text{--}12$), since little activation was caused by insufficient irradiation with the low proton beam intensity of 30 pA and the long half-life (38.3 y) of $3n$ reactions. The MCNPX calculations with JENDL/HE-2007 for nuclear data and ENDF/B-VI for cross sections of Bi were executed by a total number of 2×10^8 histories within a statistical error of 1% to obtain the reaction rates of the irradiated ^{209}Bi foil. The spot size 40 mm diameter of proton beams was modeled in the MCNPX calculations on the basis of the experimental results in the Gafchromic film. For the irradiation experiments of the ^{209}Bi foil, a comparison (Table 5.10) between the experimental and the numerical values showed agreement around a relative difference of 10% in the calculation/experiment (C/E) values, excluding the $^{209}\text{Bi}(n, 8n)^{202}\text{Bi}$ reaction. Here, from the results of the ^{209}Bi foil irradiation, the high-energy neutrons up to 50 MeV generated by $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reactions were confirmed to have been bombarded by the injection of 100 MeV protons.

Table 5.10 C/E values between measured and calculated reaction rates of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reactions ($x = 4\text{--}8$) for 100 MeV proton beams (Ref. [15])

Reaction	C/E value
$^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$	1.00
$^{209}\text{Bi}(n, 5n)^{205}\text{Bi}$	0.94 ± 0.01
$^{209}\text{Bi}(n, 6n)^{204}\text{Bi}$	0.88 ± 0.01
$^{209}\text{Bi}(n, 7n)^{203}\text{Bi}$	0.95 ± 0.02
$^{209}\text{Bi}(n, 8n)^{202}\text{Bi}$	1.65 ± 0.03

The comparison between measured and calculated results of the reaction rates at 90 degrees is listed in Table 5.10. The highest reaction rate was observed at the center of the tungsten target as shown in Table 5.11, and the angular distribution (Fig. 5.11 and Table 5.11) of reaction rates appeared slightly polarized in the upper direction, when all reaction rates were normalized by that at position 210 degrees, which was the largest along the angular. The reaction rate at 210 degrees was larger than at other positions, whereas the effect of the acryl plate was considered insufficient in the measured reaction rates along the angular. Next, to investigate the effect of the acryl plate on measured reaction rates, numerical simulations were executed with the use of MCNPX and JENDL/HE-2007 with (w/) and without (w/o) the acryl plates. A comparison of the results (Fig. 5.12) in the presence or absence of acryl plates showed an apparent effect on the reaction rates: the high-energy neutron flux was attenuated

Fig. 5.11 Measured results of the angular distribution of indium reaction rates (Ref. [15])

Fig. 5.12 Comparison between calculated results of angular distribution of indium reaction rates with and without the acryl plate (Ref. [15])

ahead at the target and influenced by the acryl plate. Thus, the spallation neutrons were considered significantly spherical in the angular distribution through the results in numerical simulations, although their angular distribution was observed actually reversed at the target. Subsequently, the neutron yield at the target was evaluated at $(9.73 \pm 0.12) \times 10^4 \text{ s}^{-1}$ over 20 MeV and $(1.03 \pm 0.04) \times 10^7 \text{ s}^{-1}$ over 0.3 MeV from the measured reaction rates of Bi and In foils, respectively.

The main characteristics of the measurements by the organic liquid scintillator are to acquire two signals of prompt (electrons) and delayed (protons, deuterons, or α -ray, etc.) fluorescence components and to discriminate γ -ray and neutron events caused by the two signals, respectively. Thus, the discrimination between the γ -ray and the neutron was possibly caused by the difference between their fluorescence intensities in a time-dependent manner.

A comparison between the combined (γ -ray and neutron) and the γ -ray events showed apparent discrimination between the γ -ray and the neutron in the experimental results (Fig. 5.13) of fluorescence distributions. The γ -ray events were found to be considerably large in low-fluorescence distribution and difficult to discriminate the two events of the γ -ray and neutron. The amount of fluorescence by high-energy neutrons was found to be small in high-fluorescence distribution because of very small counting rates in the high-channel region. Moreover, the spallation neutrons generated from the FFAG accelerator were considered to be a group of continuous energy neutrons with ambiguity in maximum energy, since an edge of the recoil proton corresponding to the neutron energy was not found in the measurements by the organic liquid scintillator. Thus, the neutron energy calibration [17, 18] of fluorescence to the light unit was conducted with the use of the results of ^{22}Na standard source (γ -ray energy; 1.274 MeV).

The neutron spectrum (Fig. 5.14) was obtained experimentally with the use of the SCINFUL-QMD code [19] for the matrix of response functions and with the UMG code [20] for the unfolding of experimental results (Fig. 5.13) together with the matrix by SCINFUL-QMD. As a reference of the neutron spectrum, the MCNPX

Fig. 5.13 Comparison between measurement results of the light output from the organic liquid scintillator before and after the discrimination of γ -ray and neutron (Ref. [15])

Fig. 5.14 Comparison between measured (Unfolding) and calculated (MCNPX) results of neutron spectrum (Ref. [15])

calculations were executed with the use of ENDF/B-VI.6 for cross section data and of LAHET150 [21] for the high-energy neutron and proton libraries. A comparison between the experiment (UMG) and the calculation (MCNPX) revealed an approximate reconstruction of the neutron spectrum in the experiment, ranging from 5 to 45 MeV neutrons, although the discrepancy was observed in some energy regions. In the measurement system, the amount of fluorescence was insufficiently over 50 MeV neutron, because of detector sensitivity in relating the intensity and the energy of protons. Finally, the spallation neutrons up to 45 MeV were considered successfully detected by the organic liquid scintillator, since the discrimination between the γ -ray and the neutron was satisfactorily conducted.

5.4.2 Reaction Rates

5.4.2.1 Experimental Settings

At KUCA, the proton beam transport facility for injecting 100 MeV protons onto a heavy metal target was used for experiments on ADS [22] equipped with a subcritical core. The main specifications of proton beams were 100 MeV energy, 1 nA intensity, 30 Hz beam repetition, 100 ns beam width and $1 \times 10^7 \text{ s}^{-1}$ neutron yield, as shown in Fig. 5.15. The heavy metal (Pb–Bi) target was set in the downstream of a stainless steel flange, as shown in Fig. 5.16. The Pb–Bi target was 50 mm in diameter for covering the proton beam shape and 18 mm thick for attaining the full stopping of proton beams [3, 13] inside the Pb–Bi target. To monitor the size of the proton beam spot, the Gafchromic film, which is highly sensitive to charged particles, was attached to the surface of the stainless steel flange before setting the Pb–Bi target. Among the main characteristics of the protons, the proton beam shape [23] was considered essential for determining neutron multiplication [3, 9] in the subcritical core, and for demonstrating adequate numerical precision of Monte Carlo calculations.

Fig. 5.15 Numerical results (PHITS3.0 [24]) of spectra of injected proton beams and spallation neutrons generated by injection of protons onto Pb–Bi target (Ref. [22])

Fig. 5.16 Foil settings at stainless steel flange (Ref. [22])

Table 5.11 Measured results of indium reaction rates for the angular (Ref. [15])

Position (°)	Measured reaction rate ($\text{s}^{-1} \text{ cm}^{-3}$)
Center	11216 ± 96
0	162 ± 5
30	229 ± 6
60	254 ± 8
90	297 ± 9
120	311 ± 10
150	343 ± 11
180	269 ± 8
210	448 ± 14
330	331 ± 10

The $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reaction rates of high-energy neutrons generated at the location of the Pb–Bi target were used for the monitoring of the source neutrons, showing the threshold energy as 0.3 MeV (Table 5.12). Also, the $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ reaction rates were selected for monitoring high-energy protons (100 MeV). The number of neutrons per proton ($^{115}\text{In}(n, n')^{115\text{m}}\text{In}/^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$) was then experimentally evaluated with the use of the two reaction rates.

High-energy neutrons over 20 MeV, generated by the interaction between the Pb–Bi target and 100 MeV protons, were evaluated with the use of the neutron threshold energies of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ (Table 5.9). The ^{209}Bi foil was considered a very effective detector for easily acquiring the high-energy neutron spectrum information from the systematic threshold energies of high-energy neutrons ranging from 20 to 50 MeV (Table 5.9). Also, in the reaction rate analyses of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reactions, the $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ reaction rates were taken as a normalization factor of the neutron spectrum for the injection of 100 MeV protons onto the Pb–Bi target.

5.4.2.2 Experimental Analyses

Monte Carlo calculations have been considered useful in accurately obtaining reaction rates in the ADS experimental analyses, with the combined use of major nuclear data libraries. In the present study, a series of numerical simulations was conducted with the PHITS3.0 [24] and the MCNP6.1 codes together with the JENDL libraries, the INCL model [25] and LAHET150, for the transport of neutrons and protons, as shown in Table 5.13. The $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reaction rates of 0.3 MeV neutron threshold energy were obtained by JENDL/D-99 for cross sections under 20 MeV. For over 20 MeV, the $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reaction rates were attained by ENDF/B-VI.8 [21, 26]. Finally, the $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ reaction rates were acquired by the point-wise data of cross sections in JENDL/HE-2007. Reaction rate calculations performed by the fixed-source calculations (total number of histories: $1\text{E} + 08$) were

Table 5.12 Threshold energy, half-life and emission γ -ray energy of activation foils (Ref. [22])

Reactions	Threshold energy (MeV)	Half-life ($T_{1/2}$) (h)	Emission γ -ray energy [keV] (Branching ratio (%))
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$	0.3	4.486	336.2 (37.0)
$^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$	–	15.00	1368.6 (99.9)

numerically obtained by the evaluation of volume tallies of activation foils (In, Bi and Al foils) within a statistical error of 5%.

5.4.2.3 Number of Neutrons Per Proton

To determine the size of the proton beam spot, the Gafchromic film was attached to the stainless steel flange downstream of the proton beam path, because a modeling of the proton beam spot has a significant effect [3, 23] of high-energy neutron generation injected by 100 MeV protons onto the Pb–Bi target. By injecting 100 MeV protons in a few seconds, the configuration of the proton beam spot was found to be a triangle-like configuration, as shown in Fig. 5.17, and the actual size was decided by scanning the result of irradiation shown in Fig. 5.18.

On the basis of the experimental result of the proton beam spot (Fig. 5.17), reaction rates of neutrons (0.3 MeV threshold energy) and protons were numerically acquired by the PHITS3.0 code with JENDL-4.0 and the INCL model for neutron

Fig. 5.17 Measured result (Gafchromic film) of proton strength distribution of 100 MeV protons of 1 nA intensity at upstream beam of the Pb–Bi target (Ref. [22])

Fig. 5.18 Scanning relative result of proton strength distribution of 100 MeV protons in Fig. 5.17 (Ref. [22])

and proton transport, respectively, and JENDL/D-99 and JENDL/HE-2007 for reaction rates of neutrons and protons, respectively. The ratio of neutrons and protons (number of neutrons per proton) by PHITS3.0 was compared with the use of experimental (Table 5.14) and numerical results shown in Table 5.15, demonstrating that the calculation/experiment (C/E) value of the number of neutrons per proton showed agreement with a relative difference about 10%. Meanwhile, the MCNP code yielded a discrepancy between the experiments and the calculations about 30%, with the combined use of nuclear data libraries and data sets shown in Table 5.13.

From the results in Table 5.15, the PHITS3.0 code was successfully validated to obtain the reaction rate ratio of neutrons (under 20 MeV) and protons (100 MeV), together with the neutron spectrum by JENDL-4.0 and the $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ cross

Table 5.13 Combination of spectra and cross sections by PHITS3.0 and MCNP6.1 with nuclear data libraries (Ref. [22])

<i>Spectrum (Transport)</i>		
	PHITS3.0	MCNP6.1
Under 20 MeV (Neutrons)	JENDL-4.0	JENDL/HE-2007
Over 20 MeV (Neutrons)	INCL model	JENDL/HE-2007
100 MeV (Protons)	INCL model	LAHET150
<i>Cross sections (Reaction rates)</i>		
	PHITS3.0	MCNP6.1
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$	JENDL/D-99	
$^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$	ENDF/B-VI.8	
$^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$	JENDL/HE-2007	

Table 5.14 Measured results of $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ and $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ reaction rates (Ref. [22])

Reactions	Measured reaction rates ($\text{s}^{-1} \text{ cm}^{-3}$)
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$	$(2.89 \pm 0.03) \times 10^5$
$^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$	$(1.58 \pm 0.02) \times 10^6$

Fig. 5.19 Cross sections of $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ (JENDL/HE-2007), $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ (JENDL/D-99) and $^{209}\text{Bi}(n, xn)^{201-x}\text{Bi}$ (ENDF/B-VI.8) reactions (Ref. [22])

sections (Fig. 5.19) by JENDL/D-99, the proton spectrum by the INCL model of PHITS3.0 and the $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ cross sections (Fig. 5.19) by JENDL/HE-2007.

5.4.2.4 Neutron Spectrum Over 20 MeV

Interestingly, another attempt with PHITS3.0 was applied to the reaction rate analyses of over 20 MeV neutrons generated by the injection of 100 MeV protons onto the Pb–Bi target. As shown in Fig. 5.16, the Al foil was attached to the downstream side of the stainless steel flange for injecting the proton beam, and the Pb–Bi target was set between the Al and Bi foils, which was attached to the backside of the Pb–Bi target. After the injection of proton beams, the γ -ray spectrum of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ was experimentally obtained for numbers ranging from 4 to 8, as shown in Fig. 5.20. Experimental reaction rates were attained by Eq. (5.8), with the use of total counts of γ -rays in threshold energies, as shown in Table 5.9.

From the results in Table 5.16, the ratios of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ and $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ reaction rates by PHITS3.0 were found to be fairly good with a relative difference of about 10%, except for $x = 7$. Additionally, a comparison of the C/E value demonstrated significant validation of the INCL model of PHITS3.0 and verification of cross sections over 20 MeV threshold energy (ENDF/B-VI.8; Fig. 5.19) and 100 MeV protons (JENDL/HE-2007; Fig. 5.19). Also, the MCNP code showed good agreement with the experimental results shown in Table 5.16, demonstrating the accuracy of a relative difference of about 10%, except for $x = 7$.

Fig. 5.20 Experimental results of the γ -ray spectrum of $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ ($x = 4\text{--}8$) (Ref. [22])

Table 5.15 Comparison between calculations and experiments of reaction rate ratio $^{115}\text{In}(n, n')^{115\text{m}}\text{In}/^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ (Ref. [22])

Ratio (number of neutrons per proton)	Experiment	C/E*	
		PHITS3.0	MCNP6.1
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}/^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$	0.185 ± 0.004	1.14 ± 0.03	1.34 ± 0.03

C/E*: Calculation/Experiment

Table 5.16 Comparison between calculations and experiments of reaction rate ratio $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ ($x = 4$ to 8)/ $^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$ (Ref. [22])

Reactions	Measured reaction rates ($\text{s}^{-1} \text{ cm}^{-3}$)	C/E*	
		PHITS3.0	MCNP6.1
$^{27}\text{Al}(p, n + 3p)^{24}\text{Na}$	$(1.64 \pm 0.01) \times 10^6$	—	—
$^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$	$(8.22 \pm 0.09) \times 10^4$	1.01 ± 0.03	0.83 ± 0.03
$^{209}\text{Bi}(n, 5n)^{205}\text{Bi}$	$(4.86 \pm 0.25) \times 10^4$	1.06 ± 0.06	1.00 ± 0.06
$^{209}\text{Bi}(n, 6n)^{204}\text{Bi}$	$(2.23 \pm 0.02) \times 10^4$	0.83 ± 0.02	0.92 ± 0.04
$^{209}\text{Bi}(n, 7n)^{203}\text{Bi}$	$(1.42 \pm 0.04) \times 10^4$	0.69 ± 0.03	0.81 ± 0.04
$^{209}\text{Bi}(n, 8n)^{202}\text{Bi}$	$(0.35 \pm 0.01) \times 10^4$	0.99 ± 0.06	1.08 ± 0.07

C/E*: Calculation/Experiment

For experimental analyses of over 20 MeV neutrons and 100 MeV protons, a choice of the INCL model of PHITS3.0 or MCNP6.1 with LAHET150 still remained for examining the validity of $^{209}\text{Bi}(n, 7n)^{203}\text{Bi}$ reaction rate analyses. Meanwhile, the PHITS3.0 and the MCNP6.1 codes were significantly similar with respect to the reconstruction of reaction rates over 20 MeV neutrons and 100 MeV protons, through a comparative study on a suitable combination (Table 5.13) of the Monte Carlo codes and nuclear data libraries.

5.5 Conclusion

The ADS experiments with 100 MeV protons were carried out at KUCA to evaluate the accuracy of experiments and calculations in subcritical states, when the kind of solid target (W, W-Be and Pb-Bi) used was varied at the location of the target. The analyses of neutron multiplication and subcritical multiplication factor demonstrated a fairly good comparison between the experiments and the calculations.

Reaction rate experiments were carried out on ADS with spallation neutrons to examine the accuracy of reaction rates by the foil activation method at various subcriticalities. A comparison between measured and calculated $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction rate distributions in the subcritical cores proved the reliability of the MCNP simulations through analyses of k_s . The reaction rates by the activation foils for threshold energy were compared significantly with the discrepancy between experiments and calculations, and the dependence of reaction rates on subcriticality. Furthermore, the In ratio was proposed to examine neutron spectrum information on ADS by two different In reaction rate distributions, as another neutron spectrum index, and the validation of the In ratio was well supported by the comparison between experiments and calculations.

Neutron spectrum experiments on spallation neutrons were conducted in the ADS facility at KUCA to investigate the neutronic characteristics of spallation neutrons generated at the target. The reaction rates and the continuous energy distribution of spallation neutrons were measured by the foil activation method and by the organic liquid scintillator, respectively. For the reaction rate experiments of ^{209}Bi foil, the C/E values between the experiments and the calculations (MCNPX and ENDF/B-VI) were found well within the relative difference of 10% ($^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$; $x = 4-7$), except for some reactions ($x = 8$). For continuous energy distribution experiments, the spallation neutrons were observed up to 45 MeV with the use of the organic liquid scintillator and the numerical simulations (MCNPX with JENDL/HE-2007 and ENDF/B-VI.6). Moreover, a suitable combination of Monte Carlo codes and the nuclear data libraries was investigated for reaction rate analyses of high-energy neutrons and protons. The ratio of high-energy neutrons under 20 and 100 MeV protons was successfully reconstructed by combining JENDL-4.0 and the INCL model of the PHITS3.0 code, showing a relative difference of about 10% between experiments and calculations. For high-energy neutrons over 20 MeV, the Monte Carlo codes together with JENDL/HE-2007 for high-energy neutrons and LAHET150 for protons demonstrated significant agreement through $^{209}\text{Bi}(n, xn)^{210-x}\text{Bi}$ reaction rate analyses.

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