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# Measurement of High-energy Neutron Fluxes and Spectra around the J-PARC Mercury Spallation Neutron Target using Multi-foil Activation Method

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Measurement of High-energy Neutron Fluxes and Spectra around the J-PARC Mercury Spallation  
Neutron Target using Multi-foil Activation Method

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The high-energy neutron fluxes and spectra around the mercury spallation neutron source at the Materials and Life Science Experimental Facility (MLF) of J-PARC were measured by the multi-foil activation method. The threshold energies of neutron reactions utilized in this experiment covered from 0.1 to 50 MeV. The foil irradiation was carried out on the first beam-run of MLF from May 30th to 31th, 2008. After the irradiation, the induced radioactivity of each foil was measured using an HPGe detector, and the neutron-induced reaction-rate distribution around the mercury target was determined. Using these data, the high-energy neutron fluxes and spectra were deduced with unfolding method in which the neutron spectra calculated with PHITS code were used as the initial-guess spectra. By comparison between the initial and the unfolded spectra, it was shown that most of the calculation results, which had been the basis of the neutronics design of the MLF target assembly, were consistent with the experimental data within  $\pm 30\%$ .

Keywords: J-PARC, MLF, JSNS, Neutron Source, Mercury, Spallation Reactions, Threshold Reaction Rate, High-energy Neutron Spectra, Multi-foil Activation Method, PHITS, Unfolding

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J-PARC 水銀核破碎中性子源まわりにおける多数箔放射化法を用いた  
高エネルギー中性子束とスペクトルの測定

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J-PARC、物質・生命科学実験施設 (MLF) の水銀を使った核破碎中性子源周りの高エネルギー中性子束及びスペクトルを、多数箔放射化法で測定した。この実験で使った中性子反応のしきい値は 0.1 から 50 MeV であった。実験における箔の照射は、2008 年 5 月 30 日から 31 日にかけて実施された MLF における初めてのビーム運転の際に行われたものである。照射後、各金属箔の放射能を HPGe 検出器で測定し、中性子誘導反応のターゲット周りにおける反応率分布のデータを得た。これらのデータを使い、各測定位置における高エネルギー中性子束及びスペクトルをアンフォールディング法で導出した。その際、初期スペクトルとして PHITS による計算値を用いた。初期スペクトルとアンフォールディングスペクトルを比較したところ、計算結果（これは MLF のターゲット集合体の中性子工学設計の基礎となったものであるが）は、実験値に $\pm 30\%$ で一致することがわかった。

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## 1. Introduction

Materials and Life Science Experimental Facility (MLF) in J-PARC<sup>1)</sup> is a multipurpose experimental facility equipped with two kinds of secondary-particle sources: a muon source and a neutron source. The secondary neutrons are produced by bombarding a mercury target with the pulsed proton beam accelerated to 3 GeV by the linac and the rapid cycling synchrotron (RCS). The final goal of the incident proton beam intensity is 1 MW. The high-energy neutrons generated from the mercury target are moderated in the ultra-low-temperature-hydrogen moderators and are supplied to neutron beam lines to be used for various kinds of material analysis.

The first-beam run of MLF was carried out on May 30 and 31 in 2008. At this chance, the realistic evaluation of the neutronics performance was important mission for the purpose of validation of the neutron source design based on Monte Carlo simulation. Therefore, on the first beam run, we conducted on the following measurements:

- (1) High-energy neutron spectra around the mercury target vessel,
- (2) Cold neutron flux and spectra at a neutron beam line.

The former one, which is called "source neutron spectra", is important as the start point of neutron transportation because the source-neutron properties make a great impacts on the final goal of the transportation such as the cold-neutron character at the neutron beam line and the shielding performance of the neutron target station as a steel-and-concrete complex. The measurement was carried out with multi-foil activation method. The multi-foil stacks were pasted at the various positions on the surface of the mercury target vessel before the first beam operation. The proton beam operation dedicated to this experiment was done on May 31. After the beam operation, the foils were removed from the vessel, and the induced activities were measured using an HPGe detector.

The measurement on item (2) was carried out using the beam line No.10, called NOBORU<sup>2)</sup>, as the first neutron detection of MLF on May 30, and a neutron beam pulse was successfully detected.<sup>3)</sup>

In this report, we focus on the measurement of the source neutron spectra and report the procedure, results and analysis. In the former part of Chapter 2, the experimental procedure on the activation-foil setup, neutron irradiation, and activity measurement are shown. In the latter part, the process of the activation data is shown. In Chapter 3, the reaction-rate data obtained by processing the activation data are shown, and the high-energy neutron spectra are deduced using unfolding method<sup>4)</sup>. The comparisons between experimental data and the calculation results are also mentioned in this chapter. Finally the summary of this report is given in Chapter 4.

## 2. Experiment

### 2.1 Activation Foil Setup

The mercury target vessel consists of the mercury vessel and the safety hull with cooling-water channel; the mercury vessel is covered by the safety hull as shown in Fig. 1.

Before the first beam run, stacks of indium, aluminum, niobium and bismuth foils were pasted at various positions on the surface of the safety hull. The thulium foils were also installed to the stacks pasted at the former positions. The size and weight of each kind of foil are tabulated in Table 1, and the positions of the sample stacks are shown in Fig. 2. The stacks were pasted on every side, "Up", "Down", "East" and "West", at the distance 75, 160, 255, 370, 455 and 750 mm from the incident-beam surface. An aluminum foil was pasted on the beam-incident surface in order to measure the incident-beam profile by the activation. The photographs of the sample set-up are presented in Fig. 3.

### 2.2 Irradiation

The foil irradiation was carried out through the first-beam run on May 30th and 31th, 2008. The history of the beam intensity is shown in Fig. 4. On May 30th, the first-beam event and the conditioning operation were performed with one-pulse operation. On May 31th, the beam operation dedicated for the foil irradiation was implemented. On the dedicated run, 2000 pulses corresponding to  $8.4 \times 10^{14}$  protons were bombarded with the neutron target. Consequently the total number of the incident protons through the whole beam run was  $1.0 \times 10^{15}$  protons.<sup>i</sup>

### 2.3 Activity Measurement

The activities of the radioactive products were measured using an HPGe detector. The full-energy-peak efficiency was calibrated at the distance of 5 cm from the detector surface; we called it "the standard position". In order to measure a lot of foils efficiently and get good statistics of counting, the irradiated foils were positioned at distance of 0.5 cm from the detector surface; we called it "the close positions". In general, it is difficult to determine the gamma-ray detection efficiencies precisely at positions close to detector surface because it is too sensitive for setting position of samples and the sum effect of cascade-coincidence. Therefore, we determined the

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<sup>i</sup> The total number of the incident protons was adjusted in order to keep accessibility of persons around the target vessel after the beam operation because we had to remove the foils by hand after the beam operation.



detection efficiencies at the close position by measuring the counting ratios between the close positions and the standard position using the samples whose activities were relatively higher.<sup>ii</sup>

An imaging plate (IP) was also used for measurement of  $^{24}\text{Na}$  induced in the aluminum foils. The detail of the measurement is shown in Appendix A.

## 2.4 Deduction of the Reaction Rates

We determined the reaction rates in Table 2 from the activation data. The excitation functions of the reactions<sup>5)</sup> are shown in Fig. 5. The reaction rates  $RR$  were deduced using the following equation:

$$RR = \frac{C\lambda}{NF_{irr}e^{-\lambda t_c}(1 - e^{-\lambda t_m})I\epsilon} \quad , \quad (1)$$

where

- $C$ : Gamma-ray peak counts,
- $\lambda$ : Decay constant of a product [ $\text{s}^{-1}$ ],
- $N$ : Number of target nucleus,
- $t_c$ : Cooling time [s],
- $t_m$ : Measurement time [s],
- $I$ : Gamma-ray branching ratio,
- $\epsilon$ : Full-energy-peak efficiency of a gamma-ray.

The decay constants and the gamma-ray branching ratios<sup>6)</sup> used in this work are shown in Table 2. In the equation (1),  $F_{irr}$  is an irradiation factor defined as follows:

$$F_{irr} = \sum_{n=0}^{N-1} I_p(t_n)(1 - e^{-\lambda\Delta t})e^{-\lambda\{t_f - (t_n + \Delta t)\}} \quad , \quad (2)$$

where

- $\Delta t$ : Sampling time,
- $t_i$ : Irradiation start time,
- $t_f$ : Irradiation termination time,
- $t_n$ :  $= n\Delta t$
- $I_p(t_n)$ : Number of incident protons per second averaged between  $t_n$  and  $t_n + \Delta t$ .

In this equation,  $N = (t_f - t_i)/\Delta t$ , and we set  $\Delta t = 300$  s. The factor  $I_p(t_n)(1 - e^{-\lambda\Delta t})$  corresponds to the generation of products between  $t_n$  and  $t_n + \Delta t$ , and  $e^{-\lambda\{t_f - (t_n + \Delta t)\}}$  corresponds to

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<sup>ii</sup> The close-to-standard efficiency ratios are largely depends on the following factors: (i) shapes, size and thickness of samples, (ii) property of gamma-ray cascade decay. Therefore the measurement needs to be done once at least for each kind of sample.

the decay of the elements from  $t_n + \Delta t$  to the irradiation termination  $t_f$ . Supposing that  $I_p(t_n) = I_p$  is constant, the factor  $F_{irr}$  reduced to  $I_p(1 - e^{-\lambda(t_f - t_i)})$ .<sup>iii</sup>

## 2.5 Error Estimation

Systematic and statistical errors were considered for deduction of the reaction-rate data. The systematic errors included errors of the detection-efficiency calibrations ( $\pm 5-9\%$ ) and the nuclear data of gamma-ray branching ratios (5% at a maximum). The statistical errors were 25% at a maximum while the most of them are less than 5%. The total relative-errors were deduced as

$$\delta_t^2 = \sum_i \delta_i^2 \quad . \quad (3)$$

The value of  $\delta_i$  is the relative error for  $i$ 'th item.

The data deduced from the activation measurements of the irradiated foils are shown in Table 3 for all reactions and positions with the total errors.

Note that the total errors shown in Table 3 do not include the error coming from the number of incident protons because that does not contribute to the comparison among the reaction-rate data. The number of incident protons was measured using the current transformer located at the closest position from the proton beam window, and the relative error  $\delta_p$  was assumed to  $\pm 10\%$ . If you use them as absolute reaction-rate values, you should consider that the total errors ( $\delta_t'$ ) become to the following<sup>iv</sup> :

$$\delta_t' = \sqrt{\delta_t^2 + \delta_p^2} \quad . \quad (4)$$

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<sup>iii</sup> Deduction of the equation is shown in the Appendix B.

<sup>iv</sup> The values of  $\delta_i$  can be calculated as (Error) $\div$ (Reaction Rate) in Table 3.

### 3. Results

#### 3.1 Reaction Rate Distribution

The reaction-rate distributions for  $^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$  are shown in Fig. 6. The proton-beam profile was obtained using the activation of the aluminum foil pasted at the beam incident surface. The radioactivity distribution of the aluminum foil was transferred to an IP after the irradiation. The IP image of the aluminum foil is shown in Fig. 7. The effective center of the profile slightly shifted to "down" and "east", so the reaction rates at the "down" positions were larger than those of the "up" positions. Similarly the reaction rates at the "East" positions were larger than those of the "West" positions.

#### 3.2 Deduction of Neutron Spectra

The high-energy neutron spectrum at each position was deduced using the unfolding method with the SAND-II code<sup>4)</sup>. The initial guess spectra were calculated using PHITS code<sup>7)</sup> with 3-dimensional geometry model including the target vessel and other components around the target such as moderators, reflectors and shielding. The calculation model and the calculation parameters were mentioned in Ref. 8) in detail. The same proton beam condition shown in Fig.7 was considered. The neutron spectra for the initial guess were calculated by track length tallies. The statistical errors at each energy bin (5% of 1 decade in logarithm scale) were within 5 % below 100 MeV and around 20% above 100 MeV. In Fig. 8, the initial guess and adjusted spectra are shown with the C/E-values, the ratios between the calculated and experimental reaction-rate, where the calculated reaction-rate are the integration of the product of the neutron spectrum and the cross section at individual energy bin over whole energy range.<sup>v</sup> It is shown that most of the C/E-values are ranged from 0.7 to 1.3, indicating that the initial-guess spectra calculated with the PHITS code are quite consistent with the experimental data within  $\pm 30\%$ .

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<sup>v</sup> Actually, the reaction rates were calculated using following:  $RR = \sum \phi(E)\sigma(E)\Delta E$ , where  $RR$ =(Reaction Rate),  $E$ =(Neutron Energy),  $\Delta E$ =(Neutron-Energy Bin),  $\phi(E)$ =(Neutron Flux from  $E$  to  $E+\Delta E$ ), and  $\sigma(E)$ =(Cross Section).

#### 4. Summary

The high-energy neutron fluxes and spectra around the mercury target of the pulsed spallation neutron source of J-PARC were measured by the multi-foil activation method, where the threshold energies of neutron reactions ranged from 0.1 to 50 MeV. The foil irradiation was carried out for the first beam-run of MLF from May 30 to 31, 2008. After the irradiation, the induced radioactivity of each foil was measured using an HPGe detector to determine the neutron-induced reaction-rate distribution around the mercury target. Moreover, the high-energy neutron fluxes and spectra were deduced with the unfolding method using the SAND-II code, in which the initial guess spectra were calculated with the PHITS code. As a result of the comparison between the initial and the unfolded spectra, it was concluded that the calculated results were in good agreement with the experimental one within  $\pm 30\%$ .

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Table 1 Nominal weight, shape, size and thickness of foils

Material	Weight	Shape	Size	Thickness
In	1.6 g	Square	1.5 cm, 1.5 cm	1 mm
	3.0 g	Square	2 cm, 2 cm	1 mm
Al	0.21 g	Disk	1 cm in diameter	1 mm
Nb	0.66 g	Disk	1 cm in diameter	1 mm
Bi	2.5 g	Disk	1.5 cm in diameter	1.5 mm
Tm	0.9 g	Square	1 cm, 1 cm	1 mm

Table 2 Reactions with high-energy neutrons and the decay data of the products

	Reaction	Q-value [MeV]	$E_{th}$ [MeV] <sup>a)</sup>	Half-life	Energy [keV]	Intensity	
<sup>115</sup> In	(n, n')	<sup>115m</sup> In	-0.34	0.34	4.486 h	336.2 <sup>b)</sup>	45.9%
<sup>27</sup> Al	(n, $\alpha$ )	<sup>24</sup> Na	-3.13	3.25	14.959 h	1368.6	100%
<sup>93</sup> Nb	(n, 2n)	<sup>92m</sup> Nb	-8.97	9.06	10.15 d	934.5	99.07%
<sup>209</sup> Bi	(n, 4n)	<sup>206</sup> Bi	-22.45	22.56	6.243 d	803.1	98.9%
	(n, 5n)	<sup>205</sup> Bi	-29.48	29.63	15.31 d	703.4	31.1%
	(n, 6n)	<sup>204</sup> Bi	-37.90	38.08	11.22 h	374	81.8%
	(n, 7n)	<sup>203</sup> Bi	-45.12	45.34	11.76 h	820.3	29.6%
<sup>169</sup> Tm	(n, 2n)	<sup>168</sup> Tm	-8.03	8.08	93.1 d	816.0	48.99%
	(n, 3n)	<sup>167</sup> Tm	-14.87	14.96	9.25 d	207.8	41%
	(n, 4n)	<sup>166</sup> Tm	-23.60	23.74	7.20 h	778.8	18.9%
	(n, 5n)	<sup>165</sup> Tm	-30.63	30.82	1.253 d	242.9	35.5%

a)  $E_{th}$ : Threshold energy.

b) The contribution of <sup>115m</sup>Cd ( $T_{1/2} = 44.6$  d) produced via <sup>115</sup>In(n, p)<sup>115m</sup>Cd reaction to 336 keV gamma-ray counts were taken into consideration in the analysis.

Table 3(a) Reaction rates on the "Up" side of the target vessel

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]		
		<sup>115</sup> In(n, n') <sup>115m</sup> In	<sup>27</sup> Al(n, α) <sup>24</sup> Na	<sup>93</sup> Nb(n, 2n) <sup>92m</sup> Nb
Up	75	8.9(7)×10 <sup>-27</sup>	4.81(25)×10 <sup>-28</sup>	1.30(7)×10 <sup>-27</sup>
Up-East	160	3.4(4)×10 <sup>-27</sup>	1.31(7)×10 <sup>-28</sup>	4.2(21)×10 <sup>-28</sup>
Up-West	160	3.4(4)×10 <sup>-27</sup>	1.41(8)×10 <sup>-27</sup>	3.5(18)×10 <sup>-28</sup>
Up	255	6.2(5)×10 <sup>-27</sup>	2.80(15)×10 <sup>-28</sup>	7.7(4)×10 <sup>-28</sup>
Up	370	3.4(4)×10 <sup>-27</sup>	1.56(9)×10 <sup>-28</sup>	4.22(22)×10 <sup>-28</sup>
Up	455	2.22(24)×10 <sup>-27</sup>	9.7(5)×10 <sup>-29</sup>	2.63(14)×10 <sup>-28</sup>
Up	750	3.3(4) ×10 <sup>-28</sup>	1.57(9)×10 <sup>-29</sup>	4.52(24)×10 <sup>-29</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>209</sup> Bi(n, 4n) <sup>206</sup> Bi	<sup>209</sup> Bi(n, 5n) <sup>205</sup> Bi	<sup>209</sup> Bi(n, 6n) <sup>204</sup> Bi	<sup>209</sup> Bi(n, 7n) <sup>203</sup> Bi
Up	75	1.48(7)×10 <sup>-27</sup>	1.13(7)×10 <sup>-27</sup>	7.0(5)×10 <sup>-28</sup>	3.51(27)×10 <sup>-28</sup>
Up-East	160	7.1(4)×10 <sup>-28</sup>	5.1(3)×10 <sup>-28</sup>	3.60(28)×10 <sup>-28</sup>	1.83(19)×10 <sup>-28</sup>
Up-West	160	5.43(27)×10 <sup>-28</sup>	3.86(24)×10 <sup>-28</sup>	2.77(22)×10 <sup>-28</sup>	1.41(14)×10 <sup>-28</sup>
Up	255	1.13(6)×10 <sup>-27</sup>	8.9(5)×10 <sup>-28</sup>	5.9(4)×10 <sup>-28</sup>	3.20(25)×10 <sup>-28</sup>
Up	370	6.7(3)×10 <sup>-28</sup>	5.2(3)×10 <sup>-28</sup>	3.75(29)×10 <sup>-28</sup>	2.10(22)×10 <sup>-28</sup>
Up	455	4.28(22)×10 <sup>-28</sup>	3.43(22)×10 <sup>-28</sup>	2.27(18)×10 <sup>-28</sup>	1.53(15)×10 <sup>-28</sup>
Up	750	7.6(4)×10 <sup>-29</sup>	6.2(4)×10 <sup>-29</sup>	4.1(4)×10 <sup>-29</sup>	2.1(3)×10 <sup>-29</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>169</sup> Tm(n, 2n) <sup>168</sup> Tm	<sup>169</sup> Tm(n, 3n) <sup>167</sup> Tm	<sup>169</sup> Tm(n, 4n) <sup>166</sup> Tm	<sup>169</sup> Tm(n, 5n) <sup>165</sup> Tm
Up	75	5.42(28)×10 <sup>-27</sup>	3.3(5)×10 <sup>-27</sup>	1.90(12)×10 <sup>-27</sup>	1.11(6)×10 <sup>-27</sup>
Up-East	160	2.07(11)×10 <sup>-27</sup>	1.30(20)×10 <sup>-27</sup>	8.4(6)×10 <sup>-28</sup>	4.84(26)×10 <sup>-28</sup>
Up-West	160	1.33(8)×10 <sup>-27</sup>	8.8(14)×10 <sup>-28</sup>	4.7(4)×10 <sup>-28</sup>	3.18(17)×10 <sup>-28</sup>
Up	255	3.22(17)×10 <sup>-27</sup>	2.1(3)×10 <sup>-27</sup>	1.66(11)×10 <sup>-27</sup>	7.8(4)×10 <sup>-28</sup>

\* The errors of last digits are given in parentheses.

\* The errors shown in these tables do not include the uncertainty of the incident-proton number; it is estimated to be ±10%.



Table 3(b) Reaction rates on the "Down" side of the target vessel

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]		
		<sup>115</sup> In(n, n') <sup>115m</sup> In	<sup>27</sup> Al(n, α) <sup>24</sup> Na	<sup>93</sup> Nb(n, 2n) <sup>92m</sup> Nb
Down	75	1.08(11)×10 <sup>-26</sup>	5.18(28)×10 <sup>-28</sup>	1.50(8)×10 <sup>-27</sup>
Down	160	9.9(11)×10 <sup>-27</sup>	5.46(29)×10 <sup>-28</sup>	1.48(8)×10 <sup>-27</sup>
Down	255	6.8(7)×10 <sup>-27</sup>	3.77(20)×10 <sup>-28</sup>	9.9(5)×10 <sup>-28</sup>
Down	370	3.9(4)×10 <sup>-27</sup>	1.82(10)×10 <sup>-28</sup>	5.29(28)×10 <sup>-28</sup>
Down	455	1.94(22)×10 <sup>-27</sup>	9.7(5)×10 <sup>-29</sup>	2.72(14)×10 <sup>-28</sup>
Down	750	3.2(4)×10 <sup>-28</sup>	1.48(9)×10 <sup>-29</sup>	4.59(25)×10 <sup>-29</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>209</sup> Bi(n, 4n) <sup>206</sup> Bi	<sup>209</sup> Bi(n, 5n) <sup>205</sup> Bi	<sup>209</sup> Bi(n, 6n) <sup>204</sup> Bi	<sup>209</sup> Bi(n, 7n) <sup>203</sup> Bi
Down	75	1.79(9)×10 <sup>-27</sup>	1.37(8)×10 <sup>-27</sup>	8.4(7)×10 <sup>-28</sup>	4.1(4)×10 <sup>-28</sup>
Down	160	1.77(9)×10 <sup>-27</sup>	1.36(8)×10 <sup>-27</sup>	9.2(7)×10 <sup>-28</sup>	4.8(5)×10 <sup>-28</sup>
Down	255	1.40(7)×10 <sup>-27</sup>	1.10(7)×10 <sup>-27</sup>	7.4(6)×10 <sup>-28</sup>	4.3(5)×10 <sup>-28</sup>
Down	370	8.4(4)×10 <sup>-28</sup>	6.5(4)×10 <sup>-28</sup>	4.4(4)×10 <sup>-28</sup>	2.64(27)×10 <sup>-28</sup>
Down	455	4.67(24)×10 <sup>-28</sup>	3.62(23)×10 <sup>-28</sup>	2.61(21)×10 <sup>-28</sup>	1.50(16)×10 <sup>-28</sup>
Down	750	8.3(4)×10 <sup>-29</sup>	6.8(5)×10 <sup>-29</sup>	4.7(4)×10 <sup>-29</sup>	3.2(4)×10 <sup>-29</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>169</sup> Tm(n, 2n) <sup>168</sup> Tm	<sup>169</sup> Tm(n, 3n) <sup>167</sup> Tm	<sup>169</sup> Tm(n, 4n) <sup>166</sup> Tm	<sup>169</sup> Tm(n, 5n) <sup>165</sup> Tm
Down	75	6.1(3)×10 <sup>-27</sup>	3.6(6)×10 <sup>-27</sup>	2.17(16)×10 <sup>-27</sup>	1.20(6)×10 <sup>-27</sup>
Down	160	6.0(3)×10 <sup>-27</sup>	3.9(6)×10 <sup>-27</sup>	2.55(19)×10 <sup>-27</sup>	1.39(8)×10 <sup>-27</sup>

\* The errors of last digits are given in parentheses.

\* The errors shown in these tables do not include the uncertainty of the incident-proton number; it is estimated to be ±10%.

Table 3(c) Reaction rates on the "East" side of the target vessel

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]		
		<sup>115</sup> In(n, n') <sup>115m</sup> In	<sup>27</sup> Al(n, α) <sup>24</sup> Na	<sup>93</sup> Nb(n, 2n) <sup>92m</sup> Nb
East	75	1.28(14)×10 <sup>-27</sup>	4.53(25)×10 <sup>-29</sup>	1.40(7)×10 <sup>-28</sup>
East	160	1.04(11)×10 <sup>-27</sup>	3.95(22)×10 <sup>-29</sup>	1.24(7)×10 <sup>-28</sup>
East	255	7.8(8)×10 <sup>-28</sup>	2.67(16)×10 <sup>-29</sup>	8.4(4)×10 <sup>-29</sup>
East	370	4.9(5)×10 <sup>-28</sup>	1.73(10)×10 <sup>-29</sup>	4.90(26)×10 <sup>-29</sup>
East	455	3.7(4)×10 <sup>-28</sup>	1.08(7)×10 <sup>-29</sup>	3.19(17)×10 <sup>-29</sup>
East	750	1.20(15)×10 <sup>-28</sup>	3.98(27)×10 <sup>-30</sup>	9.2(5)×10 <sup>-30</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>209</sup> Bi(n, 4n) <sup>206</sup> Bi	<sup>209</sup> Bi(n, 5n) <sup>205</sup> Bi	<sup>209</sup> Bi(n, 6n) <sup>204</sup> Bi	<sup>209</sup> Bi(n, 7n) <sup>203</sup> Bi
East	75	1.96(10)×10 <sup>-28</sup>	1.36(9)×10 <sup>-28</sup>	8.3(7)×10 <sup>-29</sup>	3.3(5)×10 <sup>-29</sup>
East	160	1.89(10)×10 <sup>-28</sup>	1.33(9)×10 <sup>-28</sup>	8.7(7)×10 <sup>-29</sup>	4.5(5)×10 <sup>-29</sup>
East	255	1.35(7)×10 <sup>-28</sup>	9.7(6)×10 <sup>-29</sup>	6.7(6)×10 <sup>-29</sup>	4.4(5)×10 <sup>-29</sup>
East	370	8.3(4)×10 <sup>-29</sup>	6.1(4)×10 <sup>-29</sup>	4.7(4)×10 <sup>-29</sup>	2.3(3)×10 <sup>-29</sup>
East	455	5.59(29)×10 <sup>-29</sup>	4.05(28)×10 <sup>-29</sup>	2.94(27)×10 <sup>-29</sup>	1.55(25)×10 <sup>-29</sup>
East	750	1.80(9)×10 <sup>-29</sup>	1.38(10)×10 <sup>-29</sup>	1.05(10)×10 <sup>-29</sup>	3.51(99)×10 <sup>-30</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>169</sup> Tm(n, 2n) <sup>168</sup> Tm	<sup>169</sup> Tm(n, 3n) <sup>167</sup> Tm	<sup>169</sup> Tm(n, 4n) <sup>166</sup> Tm	<sup>169</sup> Tm(n, 5n) <sup>165</sup> Tm
East	75	6.0(3)×10 <sup>-28</sup>	3.8(6)×10 <sup>-28</sup>	2.10(19)×10 <sup>-28</sup>	1.24(7)×10 <sup>-28</sup>
East	160	5.1(3)×10 <sup>-28</sup>	3.4(5)×10 <sup>-28</sup>	2.01(19)×10 <sup>-28</sup>	1.25(7)×10 <sup>-28</sup>

\* The errors of last digits are given in parentheses.

\* The errors shown in these tables do not include the uncertainty of the incident-proton number; it is estimated to be ±10%.

Table 3(d) Reaction rates on the "West" side of the target vessel

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>115</sup> In(n, n') <sup>115m</sup> In	<sup>27</sup> Al(n, α) <sup>24</sup> Na	<sup>93</sup> Nb(n, 2n) <sup>92m</sup> Nb	
West	75	9.5(10)×10 <sup>-28</sup>	3.39(19)×10 <sup>-29</sup>	1.01(5)×10 <sup>-28</sup>	
West	160	9.10(10)×10 <sup>-28</sup>	3.03(17)×10 <sup>-29</sup>	8.9(5)×10 <sup>-29</sup>	
West	255	6.1(7)×10 <sup>-28</sup>	2.26(13)×10 <sup>-29</sup>	6.3(3)×10 <sup>-29</sup>	
West	370	3.4(4)×10 <sup>-28</sup>	1.38(8)×10 <sup>-29</sup>	3.65(20)×10 <sup>-29</sup>	
West	455	2.04(25)×10 <sup>-28</sup>	9.4(6)×10 <sup>-30</sup>	2.43(13)×10 <sup>-29</sup>	
West	750	8.5(11)×10 <sup>-29</sup>	4.08(28)×10 <sup>-30</sup>	7.9(5)×10 <sup>-30</sup>	

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>209</sup> Bi(n, 4n) <sup>206</sup> Bi	<sup>209</sup> Bi(n, 5n) <sup>205</sup> Bi	<sup>209</sup> Bi(n, 6n) <sup>204</sup> Bi	<sup>209</sup> Bi(n, 7n) <sup>203</sup> Bi
West	75	1.56(8)×10 <sup>-28</sup>	9.6(6)×10 <sup>-29</sup>	7.4(6)×10 <sup>-29</sup>	3.2(4)×10 <sup>-29</sup>
West	160	1.38(7)×10 <sup>-28</sup>	9.4(6)×10 <sup>-29</sup>	6.5(5)×10 <sup>-29</sup>	3.4(4)×10 <sup>-29</sup>
West	255	1.05(5)×10 <sup>-28</sup>	7.7(5)×10 <sup>-29</sup>	5.8(5)×10 <sup>-29</sup>	3.1(4)×10 <sup>-29</sup>
West	370	6.7(3)×10 <sup>-29</sup>	5.0(3)×10 <sup>-29</sup>	3.5(3)×10 <sup>-29</sup>	1.66(27)×10 <sup>-29</sup>
West	455	4.45(23)×10 <sup>-29</sup>	3.37(22)×10 <sup>-29</sup>	2.22(18)×10 <sup>-29</sup>	1.66(19)×10 <sup>-29</sup>
West	750	1.64(8)×10 <sup>-29</sup>	1.24(8)×10 <sup>-29</sup>	8.1(6)×10 <sup>-30</sup>	4.7(5)×10 <sup>-30</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>169</sup> Tm(n, 2n) <sup>168</sup> Tm	<sup>169</sup> Tm(n, 3n) <sup>167</sup> Tm	<sup>169</sup> Tm(n, 4n) <sup>166</sup> Tm	<sup>169</sup> Tm(n, 5n) <sup>165</sup> Tm
West	75	4.29(25)×10 <sup>-28</sup>	2.9(4)×10 <sup>-28</sup>	1.45(14)×10 <sup>-28</sup>	9.4(5)×10 <sup>-29</sup>
West	160	3.91(24)×10 <sup>-28</sup>	2.6(4)×10 <sup>-28</sup>	1.99(18)×10 <sup>-28</sup>	9.3(5)×10 <sup>-29</sup>

\* The errors of last digits are given in parentheses.

\* The errors shown in these tables do not include the uncertainty of the incident-proton number; it is estimated to be ±10%.

Table 3(e) Reaction rates on the "Front" side of the target vessel

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]		
		<sup>115</sup> In(n, n') <sup>115m</sup> In	<sup>27</sup> Al(n, α) <sup>24</sup> Na	<sup>93</sup> Nb(n, 2n) <sup>92m</sup> Nb
Front	0	1.28(14)×10 <sup>-27</sup>	4.35(24)×10 <sup>-29</sup>	1.15(6)×10 <sup>-28</sup>

Position Surface	Dist.	Reaction Rates [Nucleus <sup>-1</sup> Proton <sup>-1</sup> ]			
		<sup>209</sup> Bi(n, 4n) <sup>206</sup> Bi	<sup>209</sup> Bi(n, 5n) <sup>205</sup> Bi	<sup>209</sup> Bi(n, 6n) <sup>204</sup> Bi	<sup>209</sup> Bi(n, 7n) <sup>203</sup> Bi
Front	0	1.53(8)×10 <sup>-28</sup>	1.06(7)×10 <sup>-28</sup>	7.1(6)×10 <sup>-29</sup>	3.1(5)×10 <sup>-29</sup>

\* The errors of last digits are given in parentheses.

\* The errors shown in these tables do not include the uncertainty of the incident-proton number; it is estimated to be ±10%.

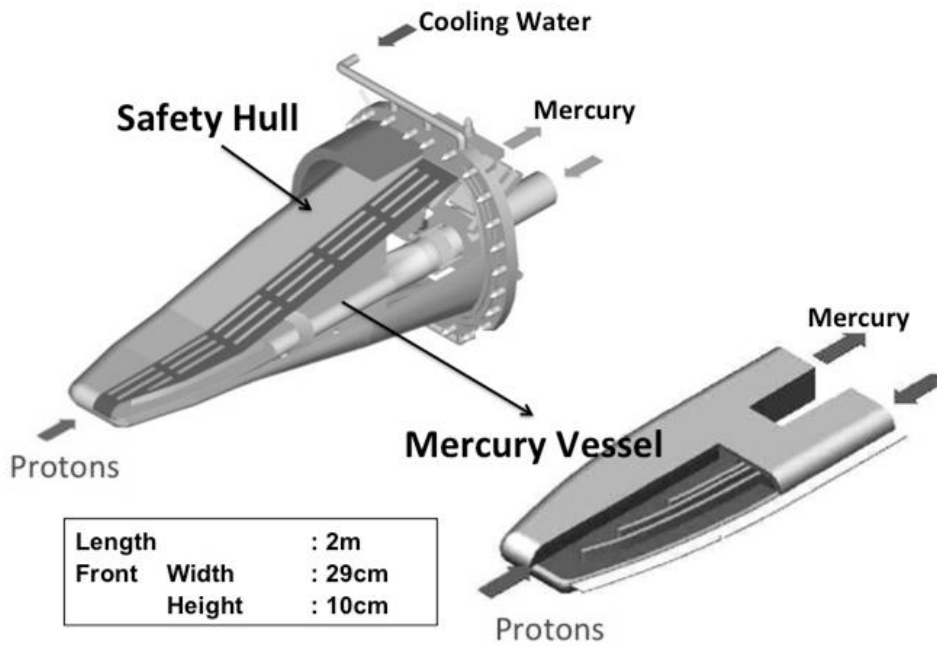


Fig. 1 Configuration of the mercury target. The mercury vessel is covered by the safety hull with a cooling water channel. Helium gas is contained in the intermediate layer between the mercury target and the safety hull. The main constructional material is SUS316L.

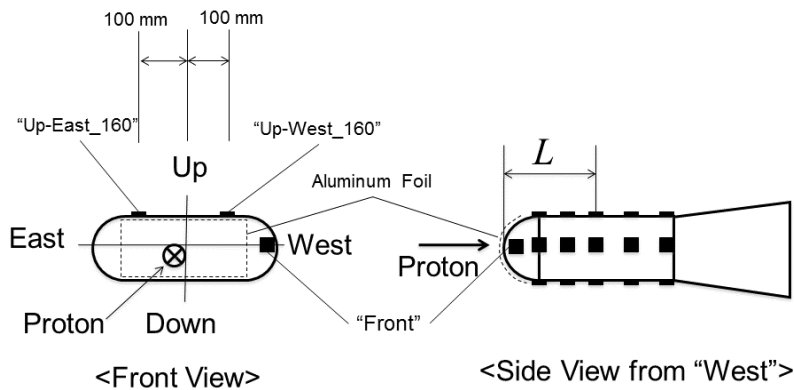


Fig. 2 Activation foils were set on the surface of the mercury target vessel at the positions labeled as "Surface-Length": the "Surfaces" were labeled as "Up", "Down", "East" and "West", and the "Length" means the distances between the vessel top and the sample positions in millimeters. For example, the label of "East-160" means the position at 160-mm distance from the vessel top on the surface of the east side. The foils labeled as "Up-East\_160" and "Up-West\_160" were pasted on the off-center positions because the position corresponding to "Up\_160" was just on the reflecting mirror of laser beam for monitoring the vibration induced on the vessel surface by the pulsed proton beam. The "Front" foils were set on the beam-incident surface. The beam-incident front was covered by the aluminum foil for measurement of the incident proton beam profile.

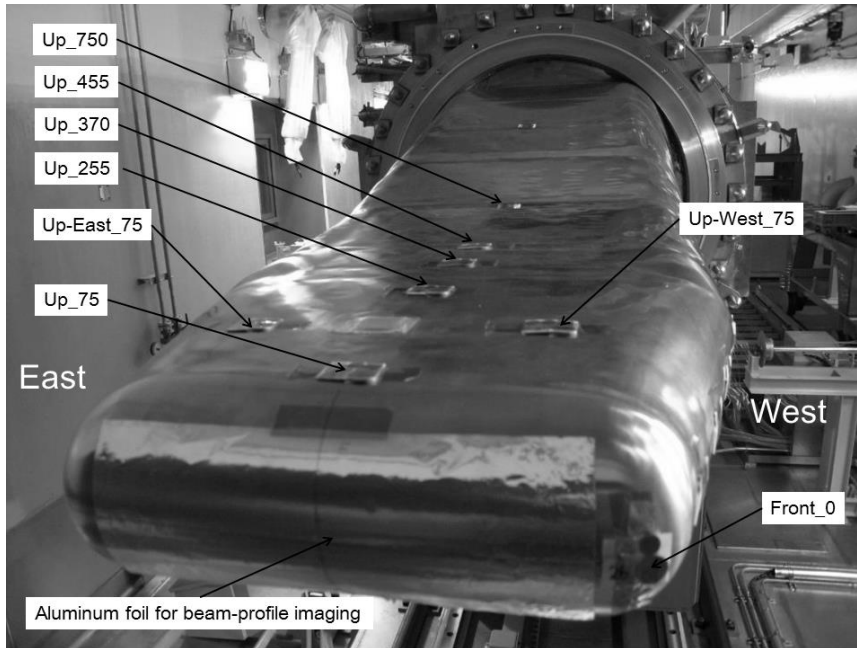


Fig. 3 Photos of sample setting. The labels such as "Up\_75" show the sample positions: the "Up\_75" means the samples on the "Up"-side at 75-mm distance from the vessel top.

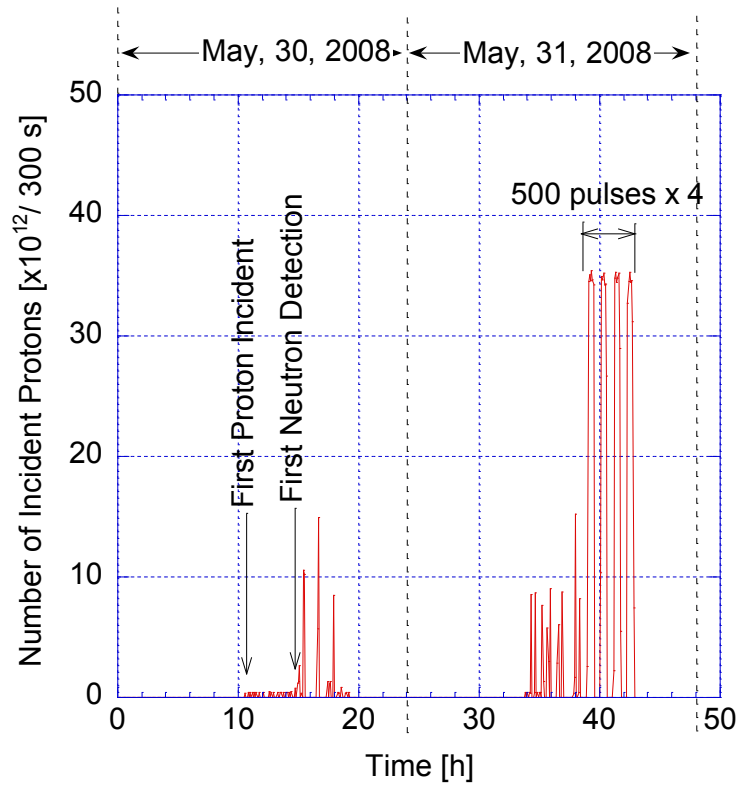


Fig. 4 History of the incident proton-beam intensity during the first beam run on May 30 and 31.

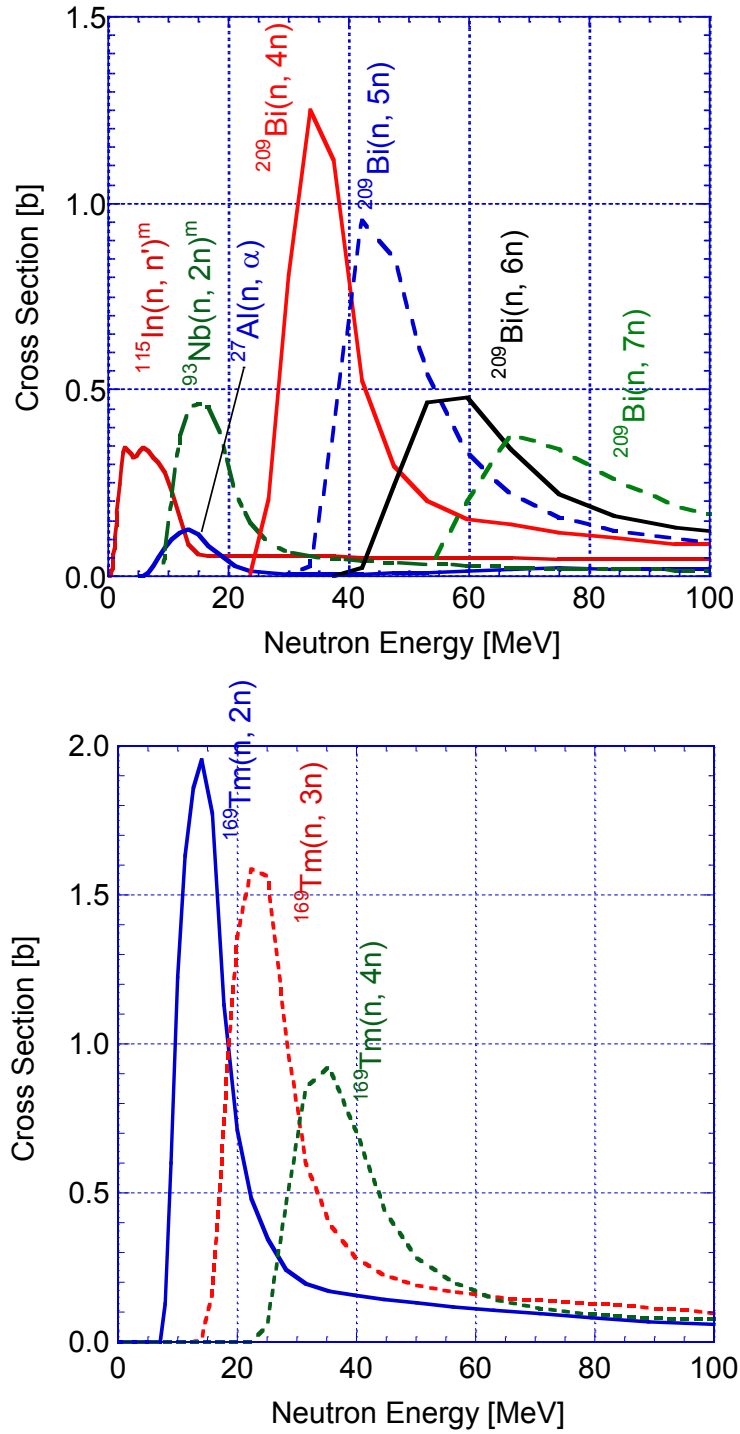


Fig. 5 Cross-section curves of the neutron induced reactions used in this experiment.

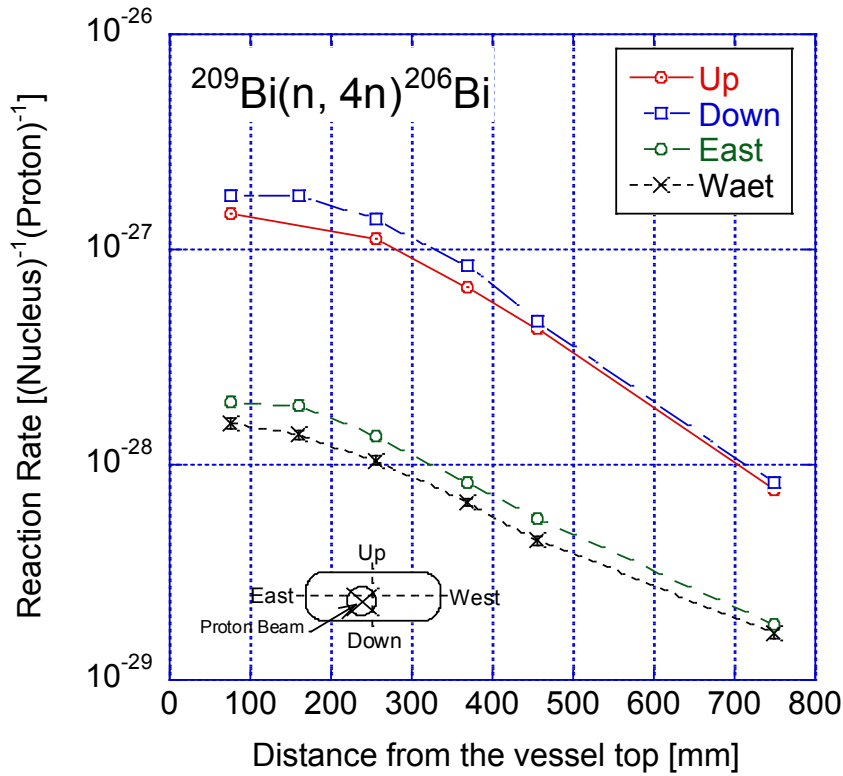


Fig. 6 Reaction rate distributions of  $^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$  on every sides of the mercury vessel. The reaction rates of "Down" are larger than the those of "Up", and "East" is larger than "West"; these results show that the center of the proton beam profile was shifted to "Down"- "East" for the target center as shown in the lower part of the figure. The shift of the proton beam center corresponds to the result of the activation of the aluminum foil pasted on the beam incident surface of the mercury target vessel shown in Fig. 7.

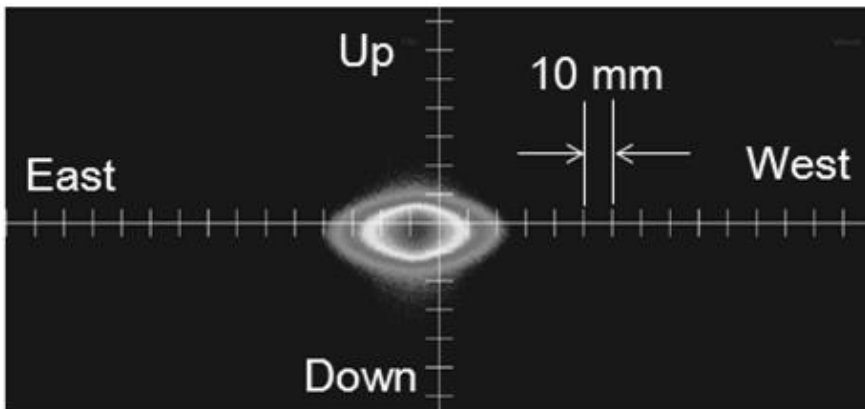


Fig. 7 Image of the activation distribution of the aluminum foil pasted on the beam-incident front of the mercury target vessel. The image corresponded to the proton beam profile accumulated during the first beam-run. The profile center was shifted to 3 mm-down and 7 mm east from the target center.

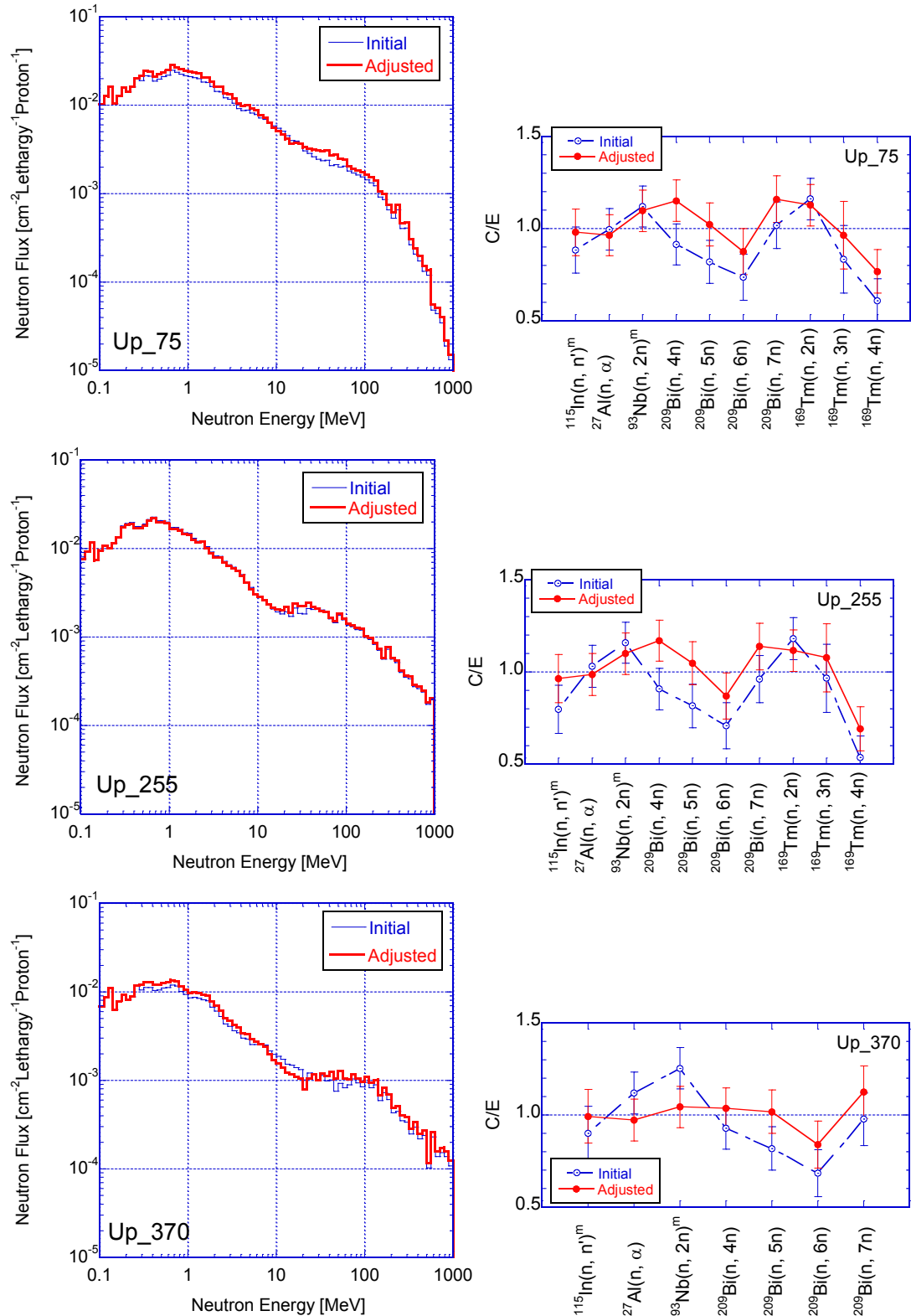


Fig. 8(a) Neutron spectra and the C/E-values of the reaction rates on the "Up" positions. The "unfolded" neutron spectra were deduced with unfolding method from the "initial" spectra estimated by the calculation using PHITS code. The values of C/E mean the ratios of the reaction rates deduced from the neutron spectrum ("initial" and "unfolded") to the experimental reaction-rate data.



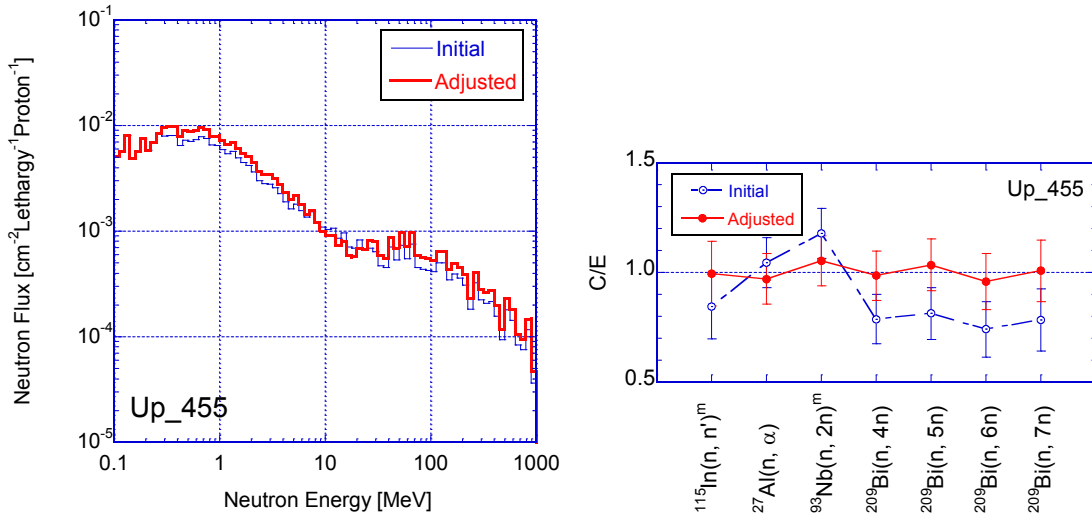


Fig. 8(a) (Continued)

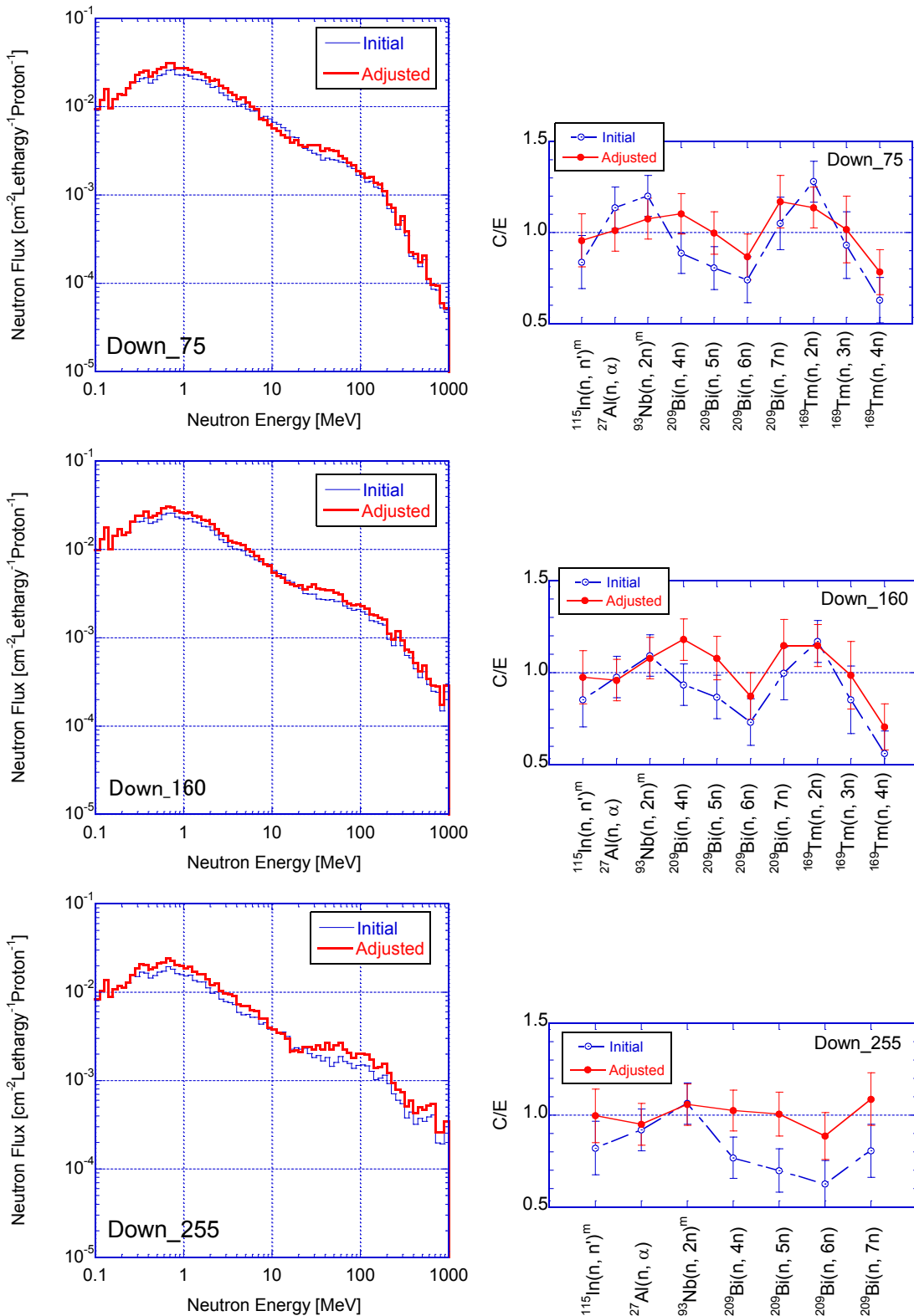


Fig. 8(b) Neutron spectra and the C/E-values of the reaction rates on the "Down" positions.

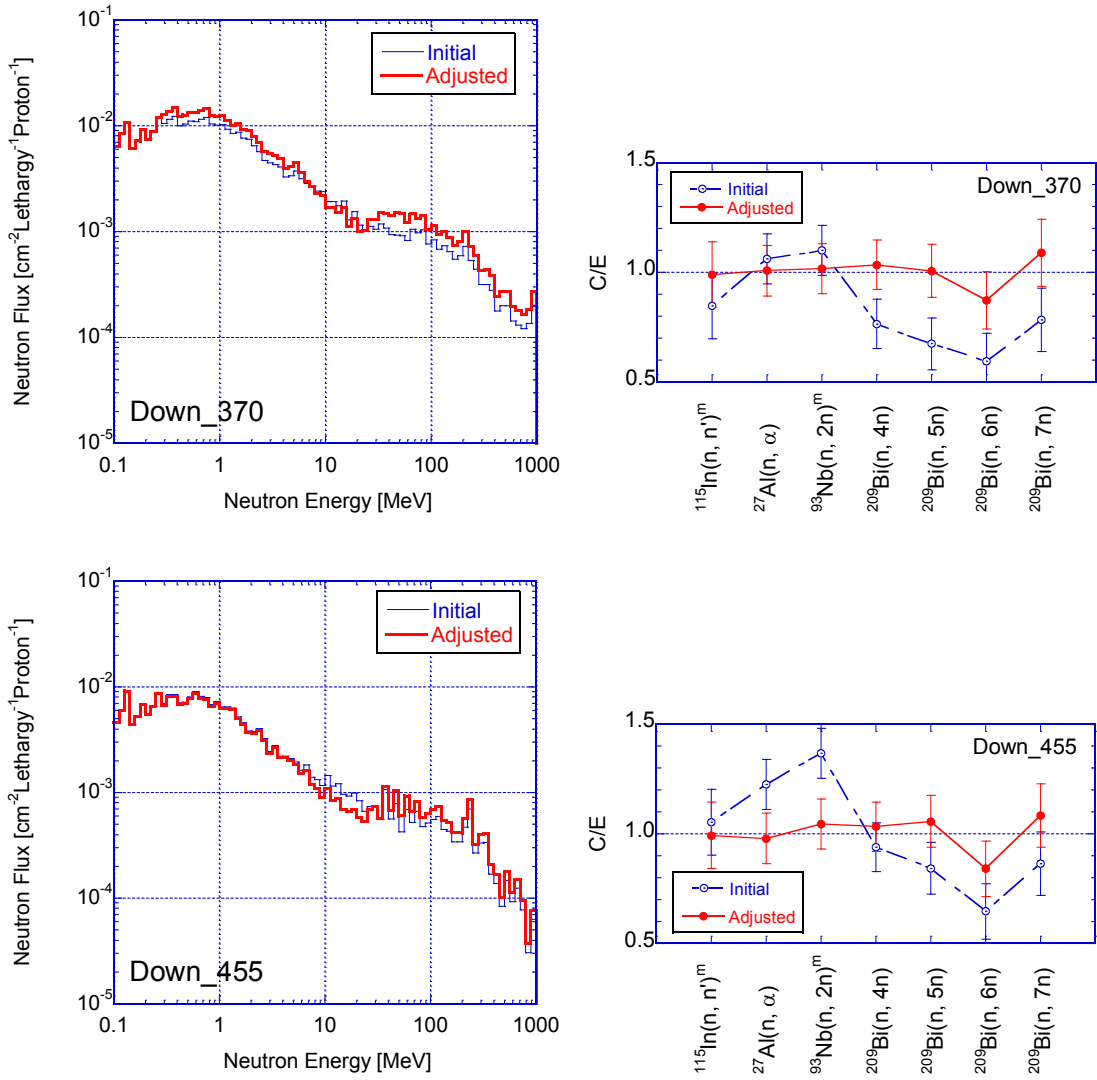


Fig. 8(b) Neutron spectra and the C/E-values of the reaction rates on the "Down" positions. (Continued)

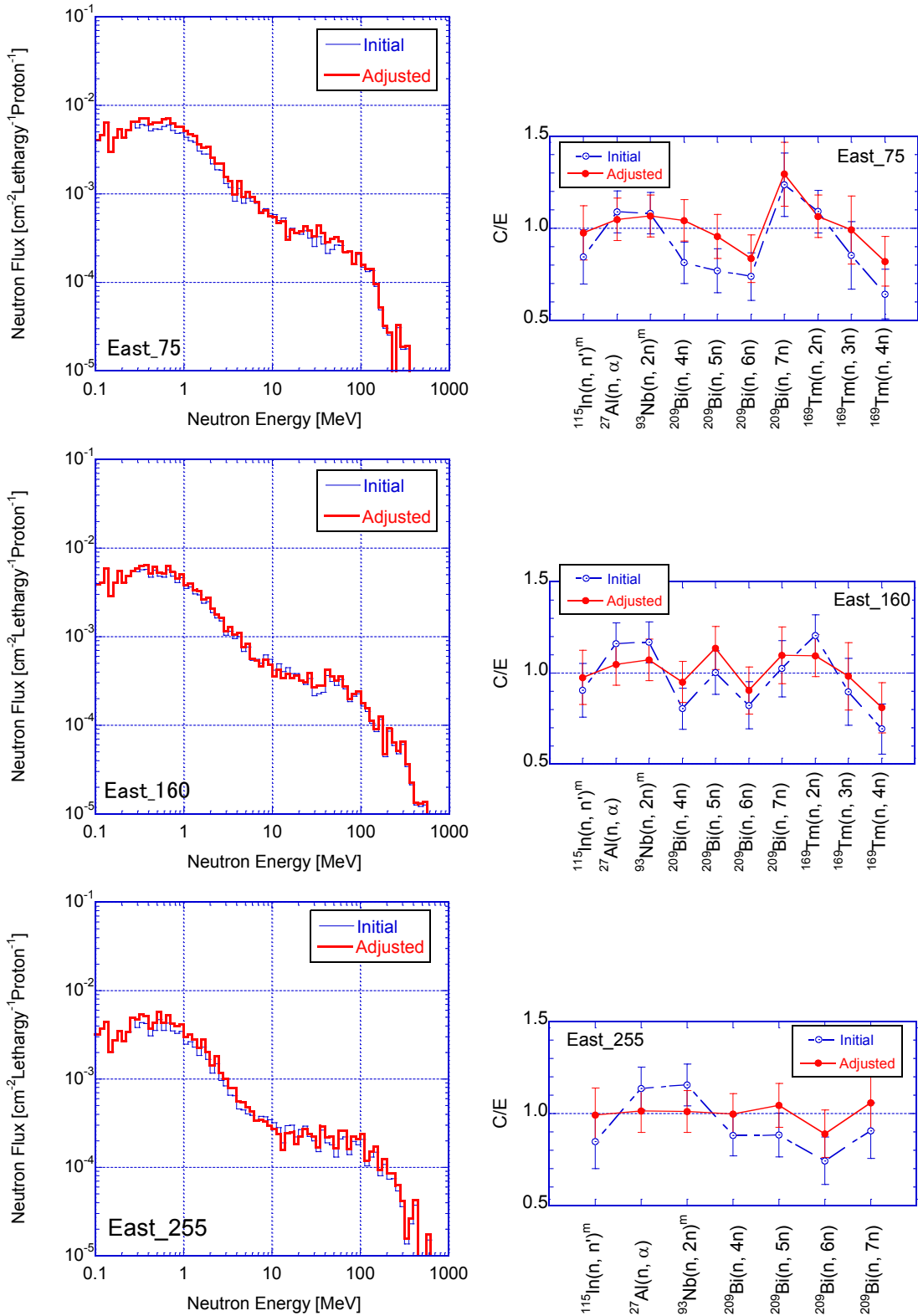


Fig. 8(c) Neutron spectra and the C/E-values of the reaction rates on the "East" positions.

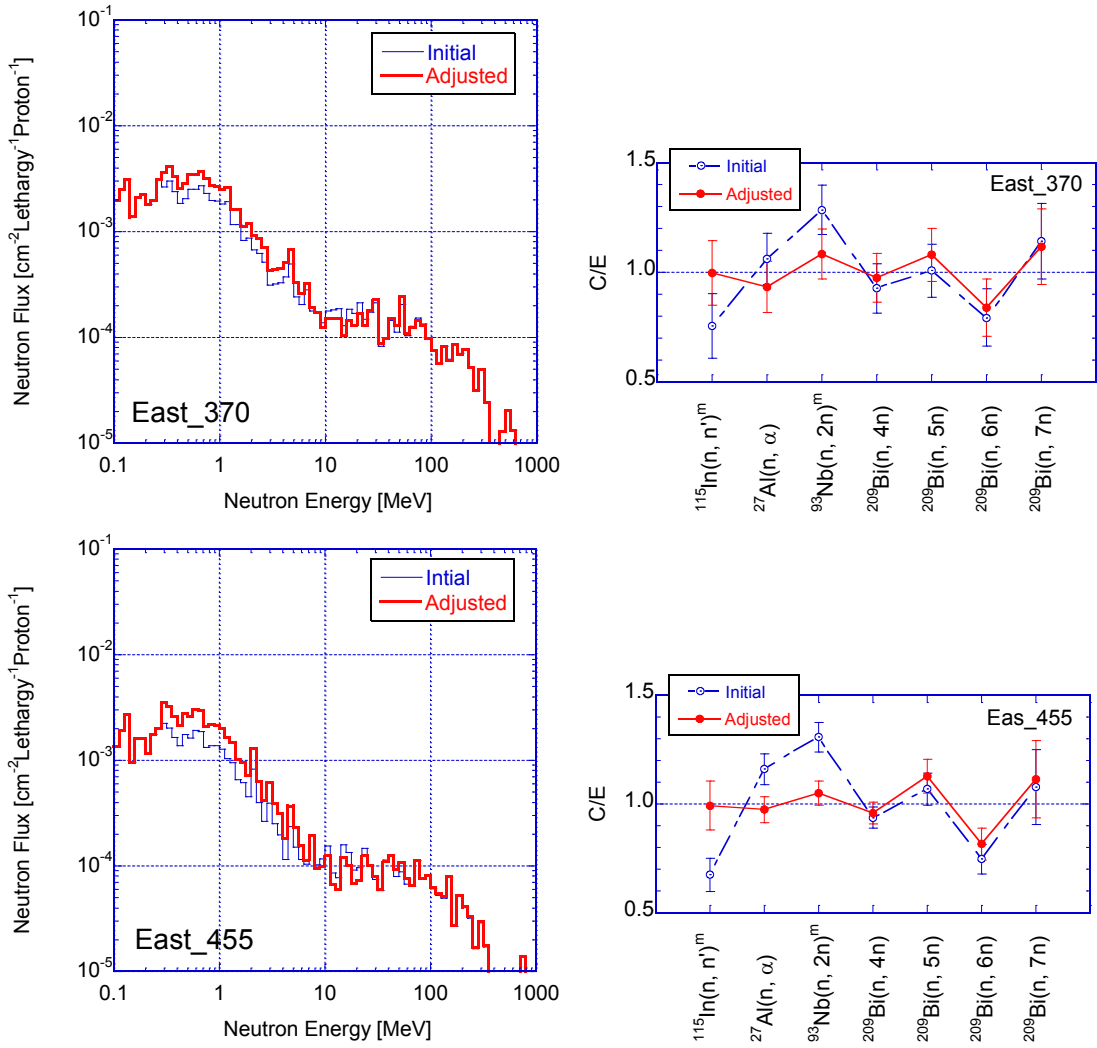


Fig. 8(c) Neutron spectra and the C/E-values of the reaction rates on the "East" positions.  
(Continued)

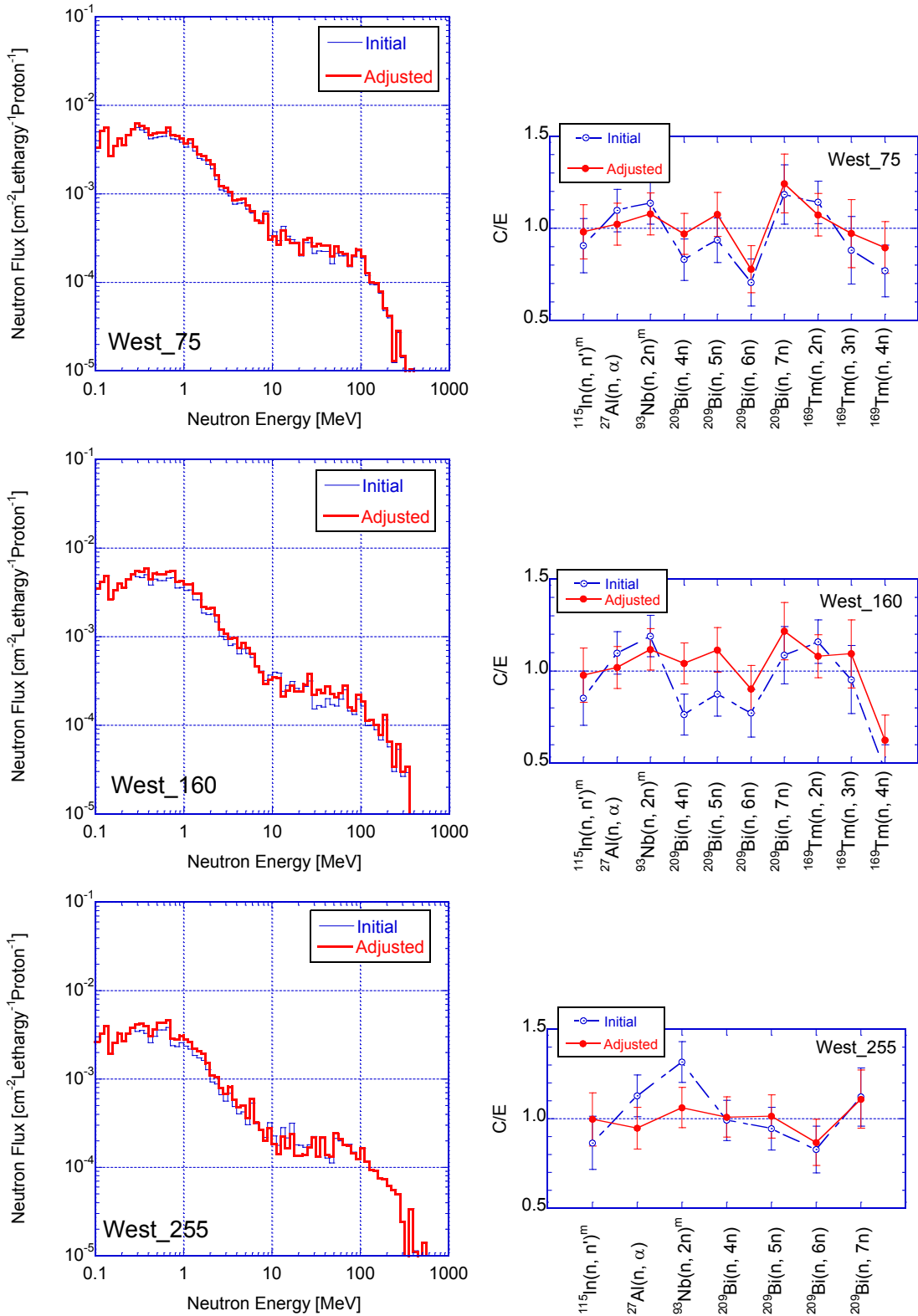


Fig. 8(d) Neutron spectra and the C/E-values of the reaction rates on the "West" positions.

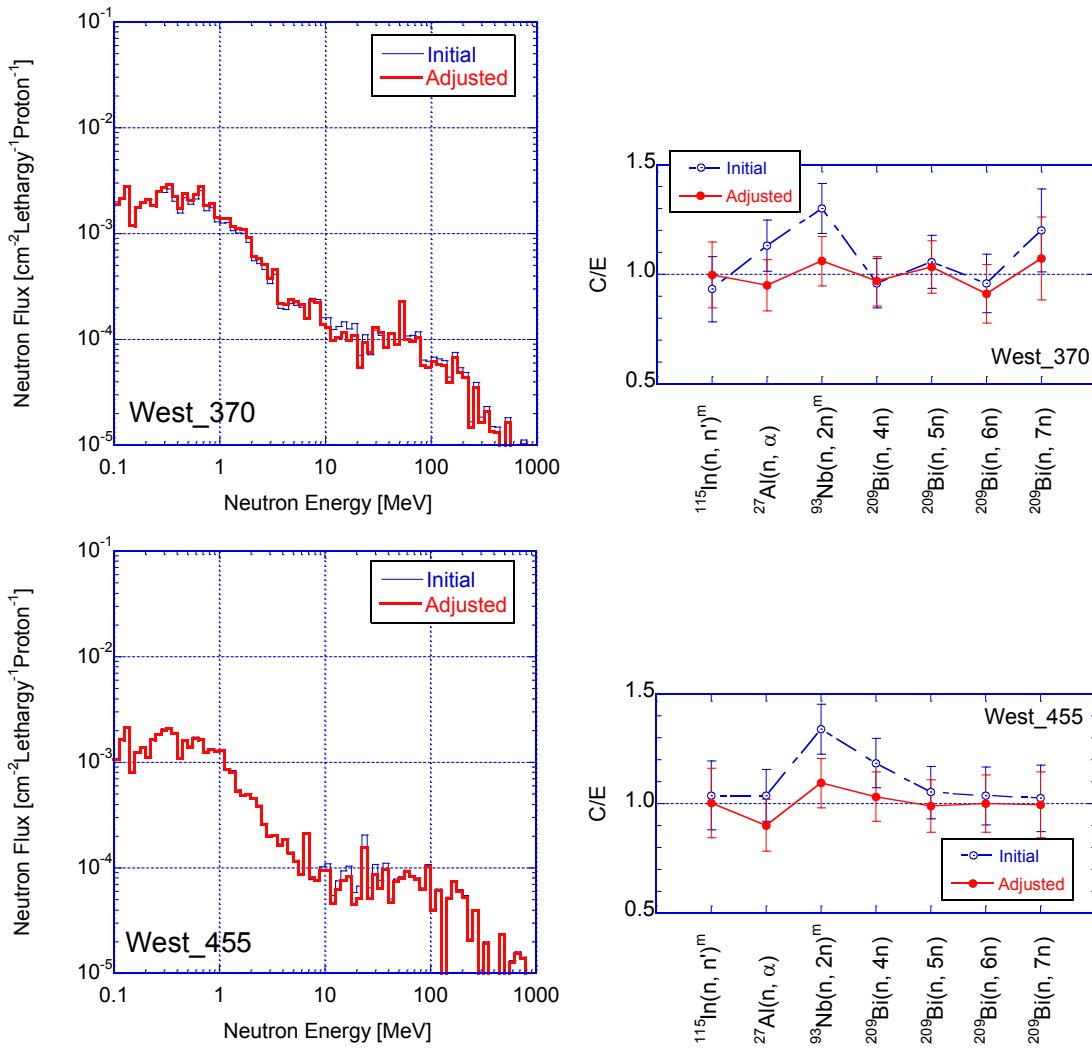


Fig. 8(d) Neutron spectra and the C/E-values of the reaction rates on the "West" positions.  
(Continued)

## Appendix A Measurement of $^{24}\text{Na}$ Induced in Aluminum Foils using an Imaging Plate

After the proton-beam operation, we need to measure more than 100 activation foils using only one HPGe detector, and the half-lives of some radioactive products to be measured is short: that of  $^{115\text{m}}\text{In}$  (4.5 h) is the shortest. In order to measure all the activation foils efficiently without sacrificing the statistics of gamma-ray counting, we also utilize an imaging plate (IP).

In order to use the IP for radioactivity determination, it is required that only a single radioactive nuclide remains as residual in an activation foils. The main products of aluminum foils for high-energy neutron irradiation are  $^{24}\text{Na}$  ( $T_{1/2}=15$  h) produced via  $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$  reaction and  $^{27}\text{Mg}$  ( $T_{1/2}=9.6$  m) via  $^{27}\text{Al}(n, p)^{27}\text{Mg}$  reaction. After considerable cooling time, the  $^{24}\text{Na}$  radioactivity only remains as residual.

In the measurement, all of the aluminum foils was placed on an IP. Exposing and analyzing the IP, the Imaging Plate Luminescence (PSL), corresponding to the relative intensities of  $^{24}\text{Na}$ , were obtained for each foil at once. For getting the absolute activities of  $^{24}\text{Na}$ , some of the aluminum foils was measured with the HPGe detector. The relation between  $^{24}\text{Na}$  activity and PSL was drawn in Fig. A1, which shows good linearity between them. We estimated that the radioactivity determination with IP method contains  $\pm 6\%$  uncertainty.

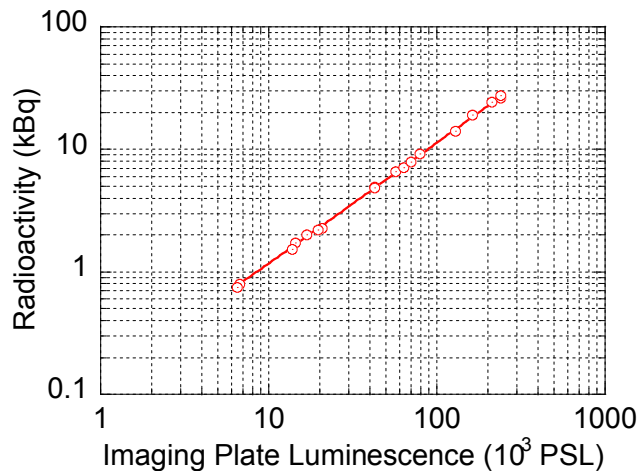


Fig. A1 Relation between the imaging plate luminescence (PSL) and activities of  $^{24}\text{Na}$  induced in the aluminum foils.



## Appendix B Deduction of the Irradiation Factor $F_{irr}$ when the Proton Beam Intensity is Constant in Time

The irradiation factor in this work was defined as follows:

$$F_{irr} = \sum_{n=0}^{N-1} I_p(t_n)(1 - e^{-\lambda\Delta t})e^{-\lambda\{t_f - (t_n + \Delta t)\}} \quad , \quad (B1)$$

where

$\Delta t$ : Sampling time,

$t_i$ : Irradiation start time,

$t_f$ : Irradiation termination time,

$I_p(t_n)$ : Number of incident protons per second averaged between  $t_n$  and  $t_n + \Delta t$ .

In addition, we set  $N = (t_f - t_i)/\Delta t$  and  $t_n = t_i + n\Delta t$ . The mathematical model of this equation is shown in Fig. B1. We rewrite the equation using  $T = t_f - t_i$  as

$$F_{irr} = \sum_{n=0}^{N-1} I_p(t_n)(1 - e^{-\lambda\Delta t})e^{-\lambda T} e^{\lambda\Delta t(n+1)} \quad . \quad (B2)$$

Suppose that  $I_p(t_n) = I_p$  is constant, the equation (B2) can be written as

$$F_{irr}^c = I_p(1 - e^{-\lambda\Delta t})e^{-\lambda T} \sum_{n=0}^{N-1} e^{\lambda\Delta t(n+1)} \quad . \quad (B3)$$

Here we change the notation  $F_{irr}$  to  $F_{irr}^c$ . Then,<sup>vi</sup>

$$\begin{aligned} \sum_{n=0}^{N-1} e^{\lambda\Delta t(n+1)} &= e^{\lambda\Delta T} + e^{\lambda \cdot 2\Delta T} + \dots + e^{\lambda \cdot N\Delta t} \\ &= \frac{e^{\lambda\Delta t} - e^{\lambda(N+1)\Delta t}}{1 - e^{\lambda\Delta t}} = \frac{1 - e^{\lambda N\Delta t}}{e^{-\lambda\Delta t} - 1} \\ &= \frac{e^{\lambda T} - 1}{1 - e^{-\lambda\Delta t}} \quad . \end{aligned} \quad (B4)$$

Therefore, we get

$$F_{irr}^c = I_p(1 - e^{-\lambda\Delta t})e^{-\lambda T} \frac{e^{\lambda T} - 1}{1 - e^{-\lambda\Delta t}} = I_p(1 - e^{-\lambda T}) \quad . \quad (B5)$$

In Fig. B2, the ratios of  $F_{irr}^c$  to  $F_i$  are shown as a function of the half-lives of radioactive products. In the calculation of  $F_{irr}^c$ , the values of  $I_p$  and  $T$  were set to be  $8.4 \times 10^{14}$  and 3.8 hours corresponding to the factor for the dedicated run on May 31. The graph shows that the contribution of the conditioning run on May 30 to the radioactivity productions was larger as the half-lives of the products were getting longer.

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<sup>vi</sup>  $S = a + a^2 + \dots + a^N = (a - a^{N+1})/(1 - a)$

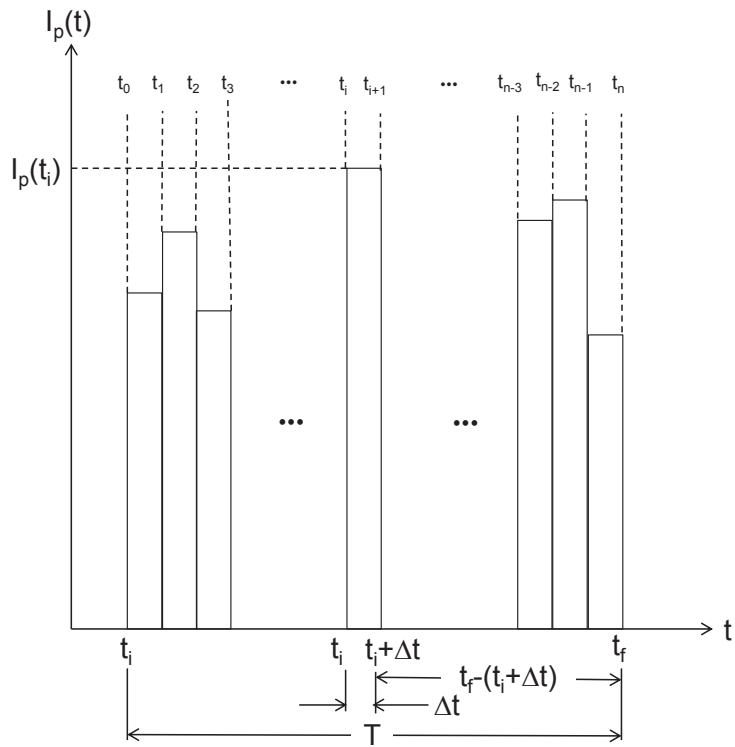


Fig. B1 Mathematical model for deduction of the equation (A1).

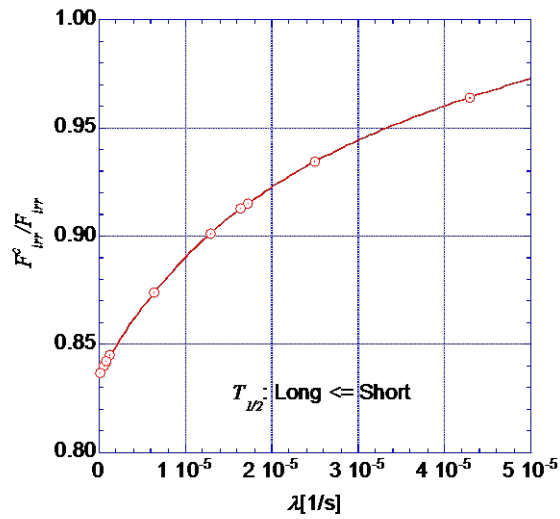


Fig. B2 The values of  $F_{irr}^c / F_{irr}$  as a function of half-lives of the products



