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Measurement of Spectrum for Thermal Neutrons Produced from H_2O Moderator coupled with Mercury target

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Abstract

In order to obtain fundamental data for the design of pulsed spallation neutron source, the slowing-down and thermalized neutrons from an $\rm H_2O$ moderator coupled with the mercury target were measured using GeV proton beams at AGS (Alternative Gradient Synchrotron) in BNL under the ASTE collaboration. The mercury target (ϕ 20 cm \times L 130 cm) was surrounded by a lead reflector ($1x1x1m^3$) was irradiated by 1.94-, 12- and 24-GeV protons. The spectral intensities of thermal neutrons from the moderator was measured by the current-mode time-of-flight technique using enriched ⁶Li and ⁷Li glass scintillators. By this technique, only several incident pulses were needed to obtain sufficient statistics for each incident energy. The results have shown that the neutron spectral intensity per proton integrated over the Maxwellian region was almost proportional to the proton energy. By moving the target along the beam direction within 15 cm, the dependence of the relative moderator position to the target on the neutron flux was also measured. With this position change, the difference with flux was found within 10 %.

1 Introduction

Incorporating the development of high intensity and high energy proton accelerator, projects of high power pulse spallation neutron sources using a mercury target are extensively in progress in several countries. On the performance of the spallation neutron source, some experiments have been carried out using activation foil[1, 2], time-of-flight technique[3, 4], and Mn-bath technique[5]. Under the ASTE (AGS-Spallation Target Experiment) collaboration, we have carried out a series of experiments[2] to characterize the neutronics performance of the spallation neutron source using mercury target. To estimate directly overall performance of a spallation neutron source, it is required to measure neutrons produced from the moderator coupled with the target and a reflector. However,

the measurements of the absolute neutron spectrum of slow-neutron from the moderator under the circumstance mentioned above were scarce for proton energies above 0.8 GeV. On the other hand, the planed spallation neutron sources such as JSNS, SNS and ESS will be employing protons with incident energies ranging from 1.0 to 3.0 GeV. Thus, the experimental neutronics validation with a simulated configuration play a key role for the facility design.

As for the measurement of the neutrons produced from the moderator, the configuration of the mercury target system used for the ASTE collaboration was modified to couple with a lead reflector and a light water moderator. With increase in intensity of neutrons per each proton pulse, however, it becomes difficult to apply a conventional pulse counting technique because of the severe pile up of output pulse signals. In the AGS experiment, it is rather difficult to reduce proton intensity at much lower level acceptable for the conventional pulse counting technique. Furthermore, available number of pulses is smaller than 20 for each incident energy. To overcome the problem and to meet the requirement, we have developed a new method by utilizing a Current mode Time of Flight (CTOF) technique[6], which can make it possible, in principle, to measure the neutron spectrum of interest with a single shot pulse. This paper deals with the brief description of CTOF and experimental result in applying this technique to an intense spallation neutron source measurements. and lead reflector curried out at AGS under the ASTE collaboration.

2 Current mode Time of Flight technique

Since details of Current mode Time of Flight (CTOF) technique has already been described in elsewhere [6], a briefly outline is given in this paper.

2.1 Principle of the CTOF technique

The principle of the CTOF technique is to measure an output current of the detector as a function of time-of-light. In case of a low counting rate, the signal of the detector can be resolved for each pulse. We chose enriched ⁶Li and ⁷Li glass scintillators as the detector, by which neutrons are observed via the ⁶Li(n, α) reaction (Q=+4.8 MeV). As increasing of the counting rate, the signal does not revert to the ground level due to the pile up and exhibits the time-of-flight spectrum. The relationship between the neutron spectrum and the time-of-flight spectrum by the pulse mode and the CTOF technique is given by the Eq. (1),

$$dn/dE_n = dY/dt \cdot dt/dE_n \cdot 1/\epsilon = dQ/dt \cdot dt/dE_n \cdot 1/\bar{Q}\epsilon \tag{1}$$

where, dn/dE_n is the energy spectrum of neutrons, Y the count number obtained by the pulse mode technique, dQ/dt the current of the detector, \bar{Q} the average charge for each detection event, ϵ the detection efficiency for neutrons, and E_n and t the energy and time-of-flight of neutrons, respectively. If the mean charge of the pulse for each detection event is constant even in high counting rate, the instantaneous current will be proportional to the number of neutrons striking the detector. Therefore, the neutron energy spectrum can be obtained by observing the current instead of the pulse.

By the CTOF technique, the linearity of the charge for each detection event should be kept constant even in the high counting rate. The linearity will be thought to become break down due to space charge effect in a PMT (Photo Multiplier Tube). Therefore, the characteristics of the current amplification of PMT was examined as described later.

2.2 Neutron detector used in the CTOF technique

As a detector, the enriched ⁶Li glass scintillator, (GS20: Koch-Light Laboratories) of 50.8 mm in diameter and 6.4 mm in thickness was employed. The specification of the GS20 is summarized in Table 1. Scintillation light produced by ⁴He and ³H nuclei via the ⁶Li(n,α)³H reaction is detected in the PMT. As a PMT, we used R1221 (Hamamatsu) consisted of multi-alkali(Na-K-Sb-Cs) photocathode, which has a lower resistance than an ordinary bialkali(Na-K-Cs) photocathode, in order prevent potential between cathode and dynode from significant dropping due to high emission current from photocathode. PMT was coated by the insulator of HA20 and covered by a magnetic shield.

The anode signal of PMT was observed by the Digital Storage Oscilloscope (DSO) of HP2545 (Hewlett Packard) which had the resolution of 8 bit. The average signal for \sim 50 pulses was obtained by the average acquisition mode of DSO. The current of the anode signal was converted to the voltage via DSO with a termination resistance (1k or 10k Ω).

In order to subtract the contribution of the γ rays, an enriched ⁷Li glass scintillator (GS30) was also used. In Table 1, the specification of the GS30 is also summarized. Pulse height distributions of GS20 and GS30 are compared in Fig. 2 for the γ rays of ¹³⁷Cs and the neutrons of Am-Be source. The net signal for neutrons can be obtained by subtracting the GS30 result from the GS20 one. By this technique, common noises due to the spike of the kicker magnet and dark current of PMT can be eliminated.

2.3 Examine experiment for the CTOF technique

In order to develop the CTOF technique, an examine experiment was carried out using a neutron beam at the H9 beam line of the pulsed spallation neutron source, KENS[7], at High Energy Accelerator Research Organization (KEK). A schematic drawing of the experimental setup is shown in Fig. 1. Slow neutrons were produced in a water (H₂O) moderator of with $100 \times 100 \times 50$ mm³ located near the Ta target, which was irradiated by the 500-MeV proton beam. Typical intensity of proton beam was 10^{12} per pulse. The intensity of protons for each pulse was measured by the current transformer(CT)[8] placed in front of the target along the proton beam line.

Neutron detectors were put at 7.3 m from the center of the moderator. For defining the active area of the neutron beam, a B₄C collimator of 50 mm thick with the hole of 10×10 mm², was put in front of the detector. At the detector position, an intensity of the thermal neutron was expected to be $1.0\times10^7/\text{cm}^2/\text{s}$ per each pulse.

For the validation of the result by CTOF technique, the spectrum was also measured by the pulse counting mode using a boron loaded plastic scintillator (BC454). The BC454 is loaded 5% weight of natural boron and has fast decay constant of 2.2 ns for scintillation light. In order to avoid the pile up of pulses, a small scintillator (7 mm in diameter and 1 mm in thickness) was employed. For subtraction of contribution of γ rays, a plastic scintillator without boron, BC408, was utilized. Net counts of neutrons were obtained by subtracting of the BC408 result from BC454 one.

In order to obtain the absolute thermal neutron flux, the activation technique was also employed using a gold foil ($13\times12~\text{mm}^2$ and $50~\mu\text{m}$ in thickness). The Au foil covered with the Cd (0.5~mm in thickness) was also employed to subtract the contribution of the epithermal neutrons above Cd cut off energy. Those foils were put at the detector position of Li-glass and were irradiated by the neutrons for about 9 h. After the irradiation, the γ -rays of ^{198}Au (412~keV) were measured by a Ge detector to derive the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction rate.

2.4 Result of examine experiment and calibration

The comparison of energy spectra obtained by the CTOF and the pulse mode techniques is shown in Fig. 3. The energy spectrum was obtained by the Eq (1). The absolute intensities of the spectrum by the CTOF and pulse counting techniques are normalized so that the integration of Maxwellian fit to the result of the activation of Au foils, which is 1.42×10^{-8} (n/cm²/proton). It is found that both results are in a good agreement each other. Therefore, it is assured that the present CTOF technique can be applied to the absolute spectrum measurement. By the normalization above, the average charge produced from the detector was determined to be 11.9 pC for each neutron detection event. Using this result, the absolute neutron spectrum can be obtained.

3 Experiment at AGS

The experiment was carried out at the U-line of AGS at BNL. A schematic drawing of the experimental setup is shown in Figs. 4 and 5. The mercury target, which had been used for measurements of the activation reaction-rates[2] and pressure wave[9], was employed. Mercury was filled in the cylindrical target container (ϕ 20 cm × L 130 cm), made of the stainless steel (German standard DIN 1.4571) with thickness of 2.5 mm. For the safety reason, the target container was confined in the secondary container which had a rectangular parallelepiped shape. The secondary container was put in a lead reflector (approximately 1m³ cube). The secondary container was put on a movable plat-home and driven by a stepping motor. By moving the container along the beam direction within 150 mm, it was observed that the dependence of the neutron intensity on the relative moderator position to the target. The moderator of water (H₂O) with $100 \times 100 \times 50$ mm³ dimension was placed at 3 cm below of the target container. A slow neutrons were extracted from the moderator surface having a cross section of 100×100 mm toward the detector. An iron collimator was placed at the inside of the U-line tunnel. Neutrons were transported through an evacuated duct having mylar foil window (0.25 mm) at the entrance and the exit.

In the TOF house outside the U-line tunnel, the neutron detectors of $^6\mathrm{Li}$ and $^7\mathrm{Li}$ glass scintillators were placed at a distance of 18 m from the moderator. The same detectors, $\mathrm{B_4C}$ collimator and DSO were employed as used in the examine experiment. In order to confirm the conservation of the average charge for each detection event (\bar{Q}) , the measurement of the pulse height distributions was performed for neutrons from the Am-Be source. The \bar{Q} value agreed well within $2{\sim}3$ % with the data obtained at KENS. After the AGS experiment, an additional calibration run was performed at KENS. It was recognized that \bar{Q} value agreed with the data obtained in examine experiment.

1.94-, 12- and 24-GeV protons with intensities about 10^{12} per pulse were injected in the Hg target. The time width of each pulse was ~ 100 ns. Proton beams of 1.94 and 12-GeV were deliberated by a pulse on demand mode. By this mode, protons were able to be delivered to the target at any time when all data acquisition system were ready. For 1.94 GeV, the proton pulses consisting of 8 bunches were deliberated with a repetition time of several seconds in order to measure efficiently the neutron. In this mode, the average storage mode of DSO was employed to obtain the average current of PMT.

Proton beam diagnostics devices are shown in Fig. 4. For the measurement of individual incident proton-beam-intensity, an integrating current transformer(ICT) of ICT-210-20:1 (Bergoz) was employed. The ICT consisting of a toroidal coil which is covered with a copper shell produces output signals which are exactly 1/40 of the beam pulse

charge. This ICT can be operated for a pulse width in a range from sub ps to several μ s. Also the activation method with Cu foil was employed to measure the total amount of protons during irradiation. A Cu foil was placed in front of the target vessel, After the irradiation, the γ -rays of ²⁴Na (1369 keV) produced via the Cu(p, ²⁴Na) reaction were measured by a Ge detector.

As the proton beam profile monitor, a segmented parallel-plate ion chamber (CHI-DORI)[10] was used. CHIDORI was composed of parallel plates of anode and cathode with active area 170 x 170 mm². Anode and cathode electrodes were consist of Cu-coated polyimide foil, which had thickness of 98.5 and 36 μm , respectively. As the ionization gas, the helium was employed because the mobility of the electron is fast enough to avoid the recombination of the electron/ion pair. In the chamber, helium gas flowed with the pressure less than 108 kPa. Potential of -900 V was supplied to cathode for giving the electric field in the chamber.

As an alternative monitoring of beam profile, an imaging plate (IP) technique with Al activation foils were also employed. The profiles for individual pulses can not be measured by this technique because the Al-foil were all the time exposed to the proton beams during irradiation. On the other hand, the two dimensional profiles was obtained. An aluminum foil in thickness 25 μ m was placed at 6 cm in front of the target and had been irradiated by the proton beams during measurement. After irradiation, the Al foil was attached to an IP. IP was exposed to the radiations emitted from the activitated Al. The image of IP represented to the intensity distribution of the radio activities, which corresponded to the incident proton intensity profile. The relative exposure doses in each pixels with size of 200 μ m² can be measured by an imaging plate reader (Fuji Film).

It is ideal to duplicate the monitors to avoid unexpected failure and malfunctions by the irradiation damage. Other beam monitors were settled by the AGS team. For the measurement of profiles and intensity of the beam, a segmented wire ionization chamber(SWIC) and a secondary emission chamber(SEC) were settled in the beam line.

4 Results and discussion

4.1 Proton beams

The whole number of protons used in the series of experiment are shown in Table 2. The results of the proton intensity obtained by the activation technique are compared with the results of ICT. For 1.94 GeV, the intensity obtained by the activation technique is 50 % smaller than the result of the ICT. However, it is found that the intensity obtained by the ICT are in good agreements with those by the activation technique except for 1.94-GeV protons; for 24 GeV, the intensities obtained by activation technique shows remarkably agreement with the results with the ICT within 1.4 %.

The beam intensity obtained by the SEC is compared in Table 2. Furthermore, the results for TOF measurement obtained by the ICT and SEC are compared in Table 3. It is found that the results by the SEC are in a good agreement with data by the ICT. The difference was 18 % at the worst case. The number of protons pulses used in the CTOF experiment is shown in Table 3. It should be noted that only several pulses are sufficient to obtain good statistics for each incident energy.

The beam profile distribution obtained by the IP technique is shown in Fig 6 The left side of the figure corresponds to the TOF detector side and incident protons penetrate from the front to back surface. It is found that the shape of the beam for 12 and 24 GeV

are well focused at the center and there is no local peak in the distribution. On the other hand, beam for 1.94 GeV is broad and almost uniform distributed around center.

The results with CHIDORI are compared with those by IP in Fig 7. Although a small background exists, the positions and the widths of peak obtained by IP show remarkably agreements with the result of CHIDORI except for 1.94 GeV. The peak by SWIC is in good agreement with the result of CHIDORI. The width obtained by SWIC are, however, slightly larger than the results of CHIDORI and IP. This is probably due to the difference in the number of secondary particles from the target, because the SWIC was located closer to the target than the CHIDORI.

4.2 Neutron spectrum

The time-of-flight spectrum obtained by the CTOF experiment is shown in Fig 8. After subtraction of signals ⁶Li from ⁷Li glass scintillators, the net TOF spectrum of neutrons was obtained. By using Eq. (1), the time-of-flight spectrum was converted to the energy spectrum. The detection efficiency of the scintillator used in Eq. (1) was calculated with the MCNP-4A[11] code using a cross section library FSXLIB-J3R2[12] processed from the nuclear data file JENDL-3.2[13]. By this calculation, it was found that the detection efficiency of the enriched ⁶Li detector was 100 % for the thermal neutrons.

The neutron spectrum obtained by CTOF technique is shown in Fig. 9. In this figure, the Maxwellian peak is clearly seen. It is recognized that the neutron intensity per proton is almost proportional to the incident proton energies.

4.3 Intensity of Maxwellian peak (J)

By the integration of the neutron spectrum in the Maxwellian peak region, the integration value of J was obtained for the comparisons of the dependence of the incident energies and positions. The integrated intensity over Maxwellian peak (J) per incident power of the protons is shown in Fig. 10 for each incident energy and different moderator position to the target position. With this position change, the difference with flux was found within 10 %.

It is recognized that the intensity per power decreases as increasing the proton energy. In the qualitative discussion, the present results show same tendency with the result of the neutron production experiment using lead target by Mn-bath technique[5]. The intensity is approximately proportional to the energy of projectile of protons.

5 Summary

In order to obtain fundamental data for the design of pulsed spallation neutron source, the slowing-down and thermalized neutrons from an H_2O moderator coupled with the mercury target were measured at AGS in BNL under the ASTE collaboration. To measure the neutron spectrum, we have developed current-mode time-of-flight (CTOF) technique using enriched 6 Li and 7 Li glass scintillators. It is confirmed that the result by CTOF technique is reliable comparing with the result by ordinary pulse counting technique. The spectrum for thermal neutrons from moderator was measured by the CTOF technique. Only several incident pulses are sufficient to obtain a good statistics for each incident energy. The results have shown that the spectral intensity per protons integrated over Maxwellian region is almost proportional to the proton energy in the range $12\sim 24$ GeV. By moving the target along beam direction within 15 cm, the dependence of relative

moderator position to the target was measured and resulted in the difference within 10 %. Using present experimental data, the confirmation and development of neutron transport code such as NMTC/JAM[14, 15] can be performed[16].

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JAERI-Conf 2001-002

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JAERI-Conf 2001-002

Table 1: Specification of Li glass scintillator used in the present experiment

	GS20	GS30
Li content by weight (%)	7.2	8.0
Enrichment of ⁶ Li (%)	96	0.1
Density (g/cm^3)	2.5	
Wavelength(nm)	395	
Decay time for γ rays (ns)	100	

Table 2: Comparison of results for proton intensity obtained by ICT, activation technique and SEC.

Proton energy(GeV)		1.9	12	24
Number of pulses		864	56	18
Proton	ICT	6.42×10^{14}	4.52×10^{13}	2.07×10^{13}
Intensity	Activation technique	4.22×10^{14}	$4.37{\times}10^{13}$	$2.05{\times}10^{13}$
	SEC	5.55×10^{14}	$4.09{\times}10^{13}$	$2.05{\times}10^{13}$
Ratio of	ICT/Activation	1.522	1.035	1.009
the results	ICT/SEC	1.157	1.105	1.014

Table 3: Results of proton intensity obtained by ICT and SEC for the time-of-flight measurements.

Proton energy(GeV)		1.9	12	24
Used number of pulses		40	11	14
Proton	ICT	6.00×10^{13}	1.07×10^{13}	1.54×10^{13}
intensity	SEC	5.09×10^{13}	$9.56{ imes}10^{12}$	$1.51{\times}10^{13}$
Ratio of the results	ICT/SEC	1.178	1.120	1.018

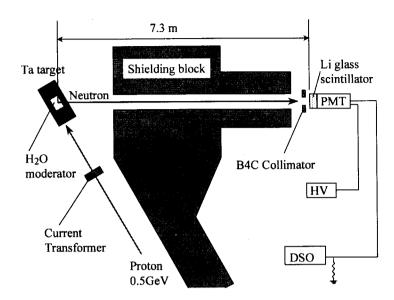


Fig. 1: Schematic drawing of examine experiment arrangement at KENS

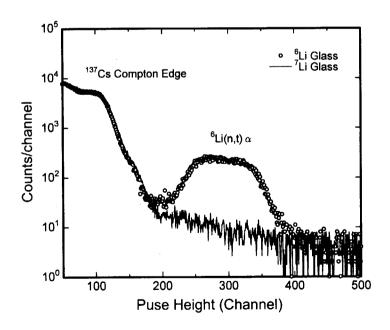


Fig. 2: Pulse height distribution of enriched ^7Li (GS30) and ^6Li (GS20) glass scintillators for the γ rays and neutrons of ^{137}Cs and Am-Be source.

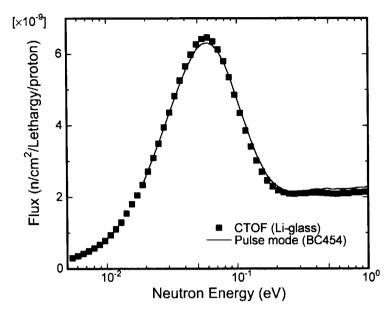


Fig. 3: Comparison of the neutron spectra obtained by the CTOF and pulse mode technique.

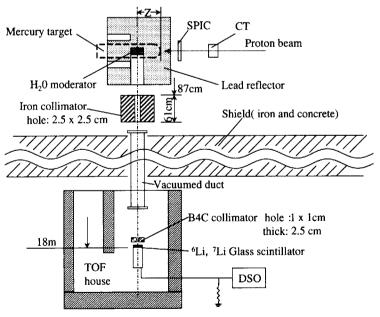


Fig. 4: Schematic drawing of experiment arrangement at AGS

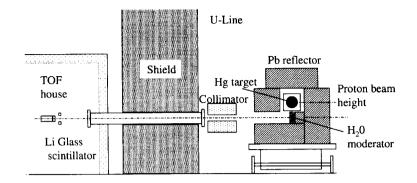


Fig. 5: Schematic drawing of cross sectional view of experiment arrangement on perpendicular plain for the beam direction at center of moderator.

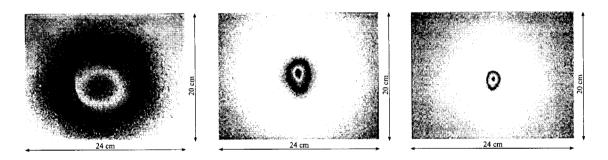


Fig. 6: Two-dimensional distribution of the incident protons obtained by imaging plate technique. Left, center and right figures show the results for 1.94-, 12- and 24-GeV protons, respectively.

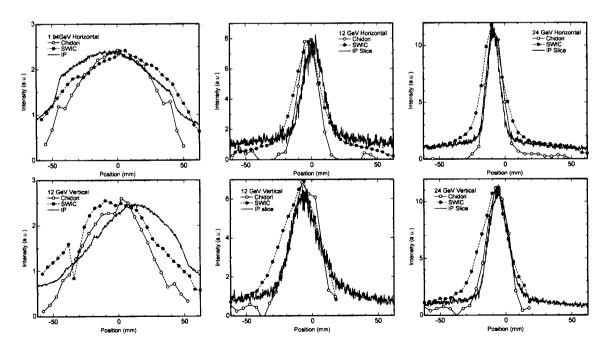


Fig. 7: Comparison of the results of beam profile obtained by CHIDORI, imaging plate and SWIC. Left, center and right figures show the results for 1.94-, 12- and 24-GeV protons, respectively.

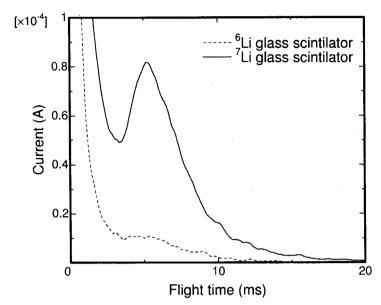


Fig. 8: Time-of-flight spectrum obtained by CTOF technique in AGS experiment.

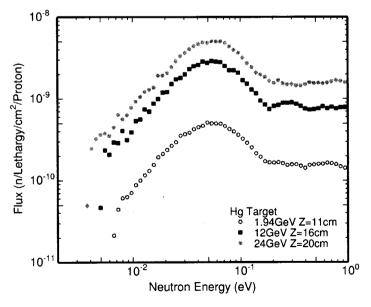


Fig. 9: Measured neutron energy spectrum per incident protons for 1.94-, 12- and 24-GeV protons.

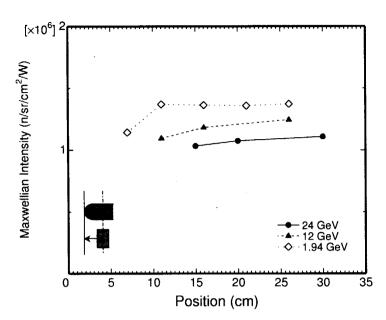


Fig. 10: Integrated intensity over Maxwellian per incident power of protons for each proton energy and target position. Horizontal axis represents distance between the front surface of the target container and center of moderator.

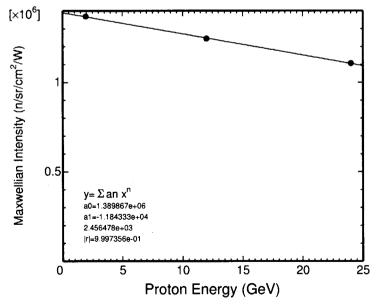


Fig. 11: Dependence of the peak neutron intensity over Maxwellian per incident power of protons.