

## Recent activities in neutron standardization at NMIJ/AIST

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### 1. Introduction

This report described recent activities on neutron standardization at the National Metrology Institute of Japan (NMIJ) in the National Institute of Advanced Industrial Science and Technology (AIST). We developed three monoenergetic neutron fluence standards at 2.5 MeV, 8.0 MeV and 24 keV, and calibration services respectively commenced in the beginning of fiscal years of 2007, 2008 and 2009. The development of 19 MeV monoenergetic neutron fluence standard is in progress and calibration services will commence in 2011. Our services on source-emitted neutron fluence standards for  $^{252}\text{Cf}$  and  $^{241}\text{Am-Be}$  were extended in the fiscal year of 2008 so that calibrations are also available in terms of ambient/personal dose equivalent. We started calibration services based on the Japan Calibration Service System (JCSS) for monoenergetic neutron fluence in the fiscal year of 2008 and for source-emitted neutron fluence in the fiscal year of 2009. The 2nd CIPM peer review was carried out in October 2008 and the ISO/IEC 17025 accreditation was updated in March 2009. We are participating in the key comparison CCRI(III)-K8 on thermal neutron fluence measurements. We made the measurements with transfer instruments in the year-end of 2006. A data analysis of the results is in its final stage. We are developing a new thermal neutron calibration field using a thermal neutron guided beam at the research reactor JRR-3M of the Japan Atomic Energy Agency (JAEA). Aiming at the establishment of the high-energy (20 - 100 MeV) neutron standard field, collaboration is in progress with the Tohoku University, the High Energy Accelerator Research Organization (KEK) and JAEA.

### 2. 8 MeV neutron field

This field will be helpful for the users who are interested in taking supplementary data on the response of neutron-measuring devices between 5.0 and 14.8 MeV, the ISO 8529-1 recommended energies for calibration. The 8 MeV neutrons are produced in the  $^9\text{Be}(\alpha,n)^{12}\text{C}$  reaction and accompanied by 3.5 MeV neutrons where 2.4 MeV  $^4\text{He}^+$  ions from the Van de Graaff accelerator impinges on a beryllium target [1,2]. The 8 MeV and 3.5 MeV neutrons respectively come from the transition to the ground and first excited states of their residual  $^{12}\text{C}$  nuclei. A polyethylene long counter is used as a neutron monitor during calibration. The 8.0-MeV neutron fluence was

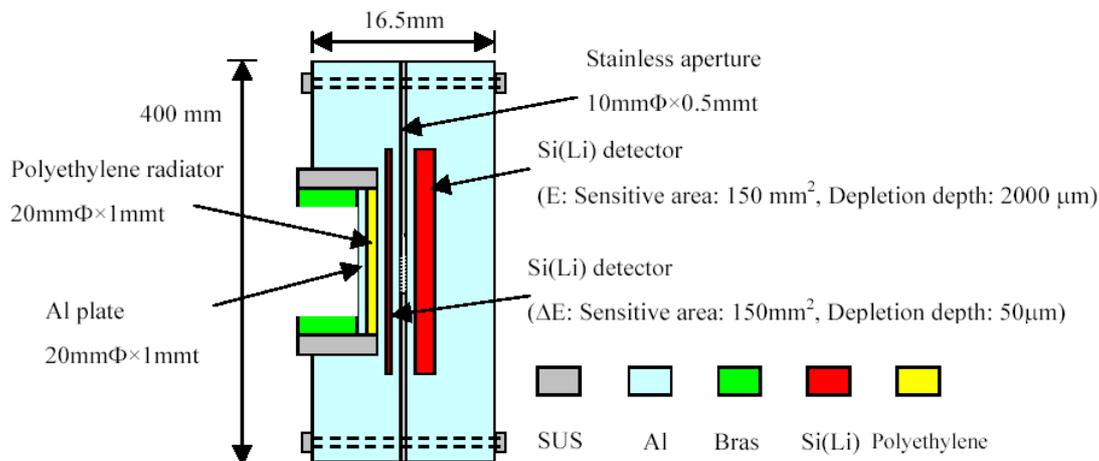


Figure 1: Schematic view of the Thick Radiator detector for 8.0 MeV

determined using a Thick Radiator (TR) detector in a similar manner as 2.5 MeV and 5.0 MeV. The TR detector used here has a dE-E configuration, as shown in Figure 1, incorporating a thin transmission-type Si(Li) detector (dE) of 50  $\mu\text{m}$  depletion depth between a 1mm thick polyethylene radiator disk and an Si(Li) detector (E) of 2 mm depletion depth. Coincidence counting between the dE and E detectors suppresses the events due to the unwanted reactions such as the Si(n,p) and Si(n, $\alpha$ ) reactions. The 3.5 MeV neutron fluence from the  ${}^9\text{Be}(\alpha,n){}^{12}\text{C}$  reaction was also evaluated from measurement with an NE213 scintillation detector. The calibration service at 8.0 MeV has been started from April 2008 in NMIJ.

### 3. 24 keV neutron field

24-keV monoenergetic neutrons are considered to be important as an intermediate energy region between the thermal and 144 keV standards, which are needed in the energy response measurement of neutron detectors and in the development of neutron dosimeters. This energy is also a reference energy point specified in ISO 8529-1. As shown in Figure 2, the 24 keV neutrons are obtained with iron filtering of a neutron beam from the  ${}^7\text{Li}(\text{p},\text{n}){}^7\text{Be}$  reaction near threshold energy. The proton energy was set to 1.890 MeV, 9 keV above the threshold energy of the  ${}^7\text{Li}(\text{p},\text{n}){}^7\text{Be}$  reaction so as to generate neutrons with a continuous distribution from 8 keV to 70 keV. The target consists of an approximately 20  $\mu\text{m}$  thick lithium layer evaporated on a tantalum disk. After the neutrons from the target were passed through a high purity iron plate with 120 mm diameter and 80 mm thickness, only 24.5 keV neutrons corresponding to the deep dip in the total cross section of  ${}^{56}\text{Fe}$  in this energy region were extracted as indicated in the calculated neutron spectrum shown in Figure 3. One important advantage in using the  ${}^7\text{Li}(\text{p},\text{n}){}^7\text{Be}$  reaction is the large cross section  $s$  as compared with those of the  ${}^{45}\text{Sc}(\text{p},\text{n}){}^{45}\text{Ti}$  reaction. The fluence rate per proton beam current of 1  $\mu\text{A}$  at 50 cm from

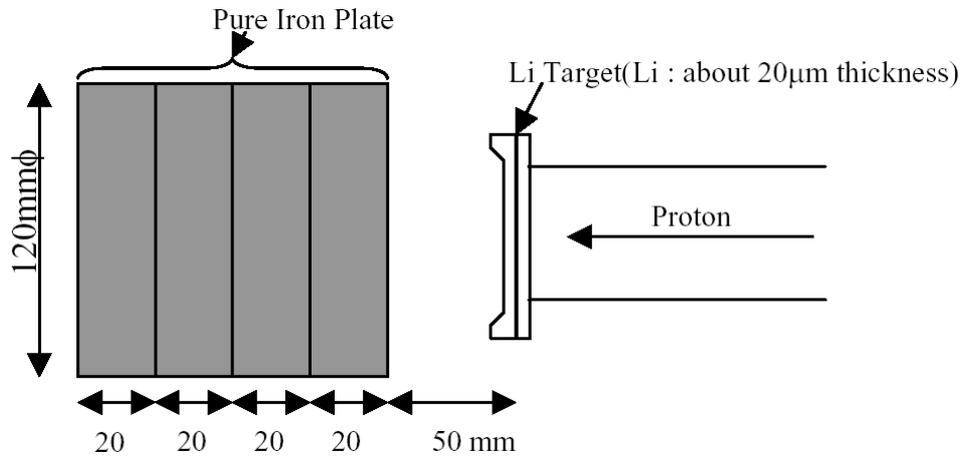


Figure 2: 24 keV neutron source by  ${}^7\text{Li}(p,n){}^7\text{Be}$  reaction and iron resonance filtering

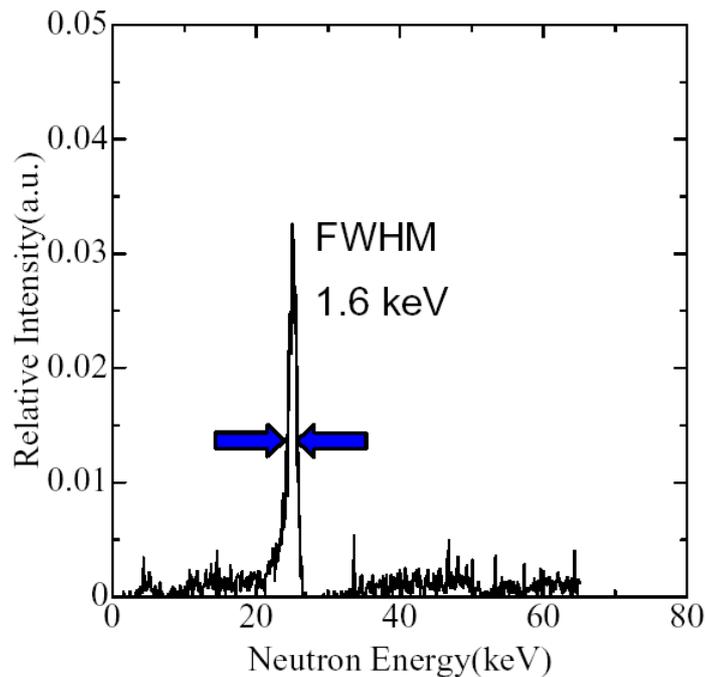


Figure 3: Calculated neutron spectrum transmitted through a high purity iron plate

the target is about 15 times larger than that from the  ${}^{45}\text{Sc}(p,n){}^{45}\text{Ti}$  reaction neutron source [3,4]. An additional advantage is that it is possible to generate stable neutron spectra that correspond to the total cross section curve of  ${}^{56}\text{Fe}$  and that is independent of a solid angle of a neutron detector.

A  ${}^3\text{He}$  cylindrical proportional counter (Reuter-Stokes RS-P4-0806: H1819) made of a stainless steel cylindrical tube filled with 0.4 MPa  ${}^3\text{He}$  and 0.2 MPa Ar gas was used to measure the absolute neutron fluence. The nominal sensitive region was 25.4 mm in diameter and 150 mm long. A 0.5-mm thick Cd plate covers the proportional counter so as to reduce the background due to the

thermal neutrons. The background due to neutrons scattered in the experimental room was determined by means of the shadow-cone method. The other backgrounds due to transmission of different energy neutrons and neutrons scattered in the target and the iron plate were determined using a titanium filter with a 20 mm thickness. Because the major isotope of the titanium,  $^{48}\text{Ti}$ , has a broad resonance around 24 keV in its total cross section. The calibration service at 24 keV will be started from April 2009 in NMIJ.

#### 4. Dose standards for source-emitted neutrons

In response to industrial needs, our calibration services on the neutron fluence from bare  $^{252}\text{Cf}$  and  $^{241}\text{Am-Be}$  sources were extended in the fiscal year of 2008 so that calibrations are also available in terms of ambient/personal dose equivalent. The related CMCs are seen later in Table 1. Neutron fluence is converted to ambient/personal dose equivalent using the conversion coefficients given in ISO 8529-3. To take account of uncertainties in the fluence spectrum, we introduce 1 % for  $^{252}\text{Cf}$  and 4 % for  $^{241}\text{Am-Be}$  as relative uncertainties ( $k=1$ ) of the conversion coefficient according to the recommendation of ISO 8529-2. If necessary, we will measure neutron spectra from our Am-Be sources to evaluate their own conversion coefficient for the spectra and to reduce the uncertainty.

#### 5. Japan Calibration Service System

The Japan Calibration Service System (JCSS) consists of the national standards provision system and the calibration laboratory accreditation system introduced by the amended measurement act enforced in November 1993. In the national measurement standards provision system, the Ministry of Economy, Trade and Industry (METI) designates national primary standards and NMIJ calibrates the reference standards of accredited calibration laboratories (i.e. secondary standards) with national primary standards. In the calibration laboratory accreditation system, calibration laboratories are assessed and accredited as accredited calibration laboratories to meet the requirements of the measurement act, relevant regulations and ISO/IEC 17025. Calibration laboratories are also required to periodically take assessment as well as proficiency testing. Calibration certificates with the JCSS symbol (see Figure 4) issued by accredited calibration laboratories assure the traceability to National Measurement Standards as well as a laboratory's technical and operational competence and are acceptable in the world through the ILAC and APLAC MRA.

We are renovating the traceability of neutron measurement in Japan by starting calibration services based on JCSS. On 15 May 2008, JCSS commenced on the monoenergetic neutron fluence standards at 144 keV, 565 keV, 5.0 MeV and 14.8 MeV. METI designated the primary standards and the secondary standards in the METI Notifications No.104 and No.106, respectively. Bonner spheres were specified as the secondary standards. Now one calibration laboratory intends to be a JCSS accredited calibration laboratory and plans to apply to IAJapan for accreditation. In



Figure 4: JCSS symbol

the beginning of March 2009, NMIJ performed the first JCSS calibration of the specified secondary standard of the calibration laboratory.

According to our original planning, JCSS is scheduled to start on thermal neutron fluence rate and neutron emission rate in the fiscal years of 2009 and 2011, respectively. In response to industrial needs, we decided to start JCSS calibration services on source-emitted neutron fluence for  $^{252}\text{Cf}$  and  $^{241}\text{Am-Be}$  as well. We changed the schedule so that JCSS will start on source-emitted neutron fluence in the fiscal years of 2009 and on thermal neutron fluence rate and neutron emission rate in the fiscal years of 2011.

JCSS on source-emitted neutron fluence was deliberated and approved in the METI measurement administration council on 4 November 2008. METI will designate the primary standards and the secondary standards in the METI Notifications in 2009. Flat response neutron detectors such as long counters will be specified as the secondary standards. So far, two calibration laboratories intend to obtain a JCSS accreditation on source-emitted neutron fluence.

## 6. CIPM Peer Review

Because five years have passed since the 1st peer review took place in 2003 and the peer reviewer, Dr. Horst Klein (PTB) assessed our calibration services on neutron standard, the 2nd peer review was held from 22 to 24, inclusive, October 2008. In the same way as the 1st peer review, the 2nd peer review was carried out jointly with the assessment of the quality management system by the International Accreditation Japan (IAJapan) of the National Institute of Technology and Evaluation (NITE) for all quantities on ionizing radiation including neutrons, (X,  $\gamma$ ,  $\beta$ )-ray dosimetry and radioactivity. The peer reviewer on neutron metrology was Dr. Miloslav Kralik (CMI).

In the peer review, 0 nonconformity were found but 1 concern and 5 comments were found on neutron metrology. Our corrective actions for them were approved by the peer reviewer on 2 March 2009. IAJapan accredited our calibration services under the ASNITE Accreditation Program (ASNITE: Accreditation System of NITE) on 10 March 2009. Our new CMC table shown in Table 1 will be posted for approval to be published in the Appendix C of the BIPM key comparison database (KCDB).

Table 1: Our new CMC table

Quantity	Calibration and Measurement Capabilities		
	Condition	Measurand Range	Expanded Uncertainty ( $k=2$ )
Fluence rate	Thermal	$5.0 \times 10 \text{ cm}^{-2} \text{ s}^{-1} - 1.0 \times 10^4 \text{ cm}^{-2} \text{ s}^{-1}$	2.8%
	144 keV	$9.0 \text{ cm}^{-2} \text{ s}^{-1} - 9.0 \times 10^2 \text{ cm}^{-2} \text{ s}^{-1}$	4.4%
	565 keV	$2.5 \times 10 \text{ cm}^{-2} \text{ s}^{-1} - 2.5 \times 10^3 \text{ cm}^{-2} \text{ s}^{-1}$	4.4%
	5.0 MeV	$1.0 \times 10 \text{ cm}^{-2} \text{ s}^{-1} - 1.0 \times 10^3 \text{ cm}^{-2} \text{ s}^{-1}$	6.2%
	14.8 MeV	$1.5 \times 10 \text{ cm}^{-2} \text{ s}^{-1} - 3.0 \times 10^3 \text{ cm}^{-2} \text{ s}^{-1}$	3.2%
	$^{252}\text{Cf}$	$8.0 \times 10^{-1} \text{ cm}^{-2} \text{ s}^{-1} - 8.0 \times 10^1 \text{ cm}^{-2} \text{ s}^{-1}$	3.6%
	Am-Be	$1.6 \text{ cm}^{-2} \text{ s}^{-1} - 7.4 \times 10^1 \text{ cm}^{-2} \text{ s}^{-1}$	2.8%
Personal dose equivalent rate	$^{252}\text{Cf}$	$1.2 \times 10^{-6} \text{ Sv h}^{-1} - 1.2 \times 10^{-4} \text{ Sv h}^{-1}$	4.1%
	Am-Be	$2.4 \times 10^{-6} \text{ Sv h}^{-1} - 1.1 \times 10^{-4} \text{ Sv h}^{-1}$	8.5%
Ambient dose equivalent rate	$^{252}\text{Cf}$	$1.1 \times 10^{-6} \text{ Sv h}^{-1} - 1.1 \times 10^{-4} \text{ Sv h}^{-1}$	4.1%
	Am-Be	$2.3 \times 10^{-6} \text{ Sv h}^{-1} - 1.0 \times 10^{-4} \text{ Sv h}^{-1}$	8.5%
Neutron emission rate	$^{252}\text{Cf}$	$1.0 \times 10^3 \text{ s}^{-1} - 3.0 \times 10^7 \text{ s}^{-1}$	3.2%
	Am-Be	$1.0 \times 10^3 \text{ s}^{-1} - 2.0 \times 10^7 \text{ s}^{-1}$	3.0%

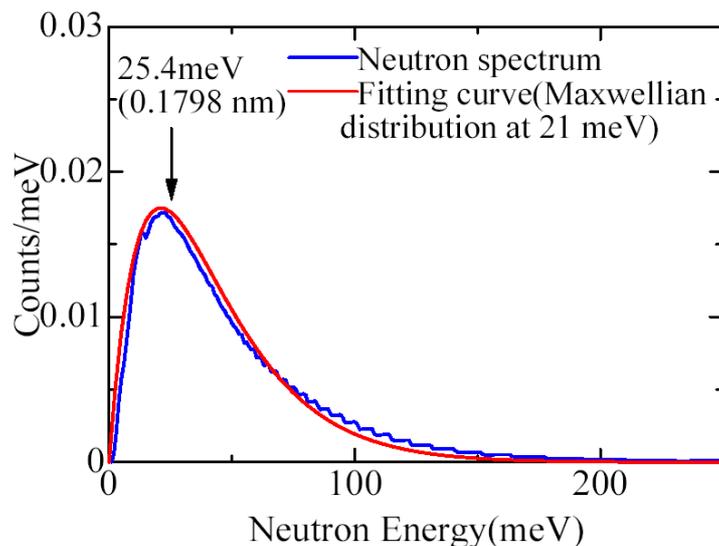


Figure 5: Neutron spectrum measured with the TOF method and fitting curve

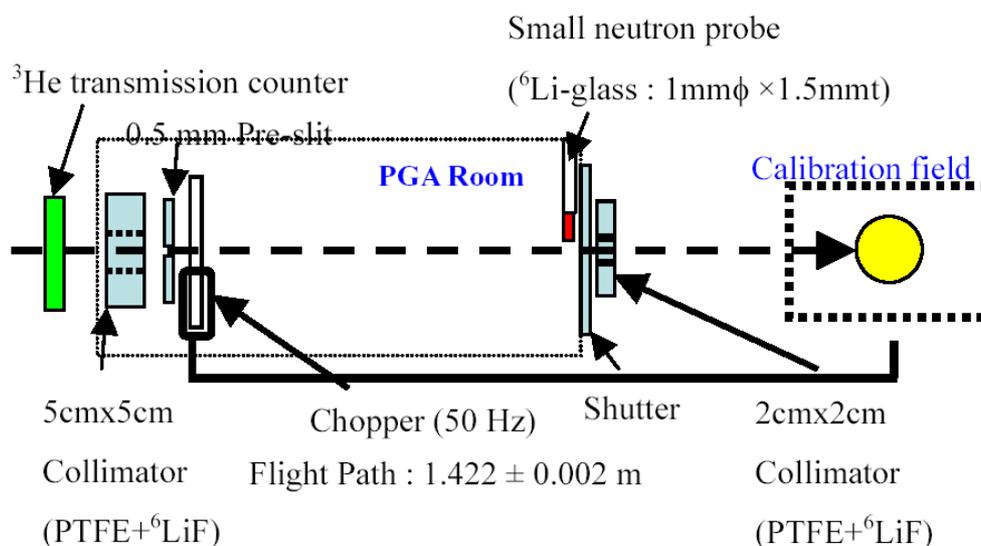


Figure 6: Experimental setup at JRR-3M

## 7. Intense thermal neutron calibration field using a research reactor JRR-3M

We have developed an intense thermal neutron calibration field using a reactor produced neutron beam in JRR-3M of the Japan Atomic Energy Agency. In this beam line, neutron induced prompt gamma ray analysis has been usually performed. Curved neutron guide after the reactor provides a pure thermal neutron beam. Neutron energy distribution with negligible contribution from epithermal neutrons was measured by means of a time of flight method with a mechanical chopper made of  ${}^6\text{LiF}+\text{PTFE}$  plate as shown in Figure 5. The flux of the chopped beam is about  $10^6 \text{ s}^{-1}$  and beam cross section is up to about  $4 \text{ cm}^2$ . We use a high purity Si detector with a  ${}^6\text{LiF}$ -evaporated

foil of a 5  $\mu\text{m}$  thickness as a neutron detector in the TOF measurement. A low-pressure  $^3\text{He}$  gas transmission counter and a small  $^6\text{Li}$ -glass scintillation probe with a plastic fiber were placed as a neutron monitor as shown in Figure 6. We have investigated a characterization of this field such as neutron spectrum, fluence rate, and beam stability. We will start the calibration service for the intense thermal neutrons by 2012.

#### 8. High energy neutron calibration field

We have developed measurement technique of high-energy neutrons above 20 MeV for establishing the high-energy neutron standard.

Fortunately, there are several high-energy neutron facilities such as TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) of JAEA, CYRIC (Cyclotron and Radioisotope center) of Tohoku University and RCNP (Research Center for Nuclear Physics) of Osaka University in Japan, which we can use. These facilities have been used for the nuclear data measurement of neutron-induced reaction, shielding benchmark experiments and characterization of neutron measuring devices. At TIARA and CYRIC Tohoku university, quasi-monoenergetic neutrons with an energy region from 20 to 90 MeV are produced using the  $^7\text{Li}(p,n)$  reaction and a proton beam from an AVF cyclotron [5,6]. In these facilities, a transmission plastic scintillator placed on the neutron beam line and fission chambers placed around the lithium target are used as a neutron monitor. Moreover, proton beam current on the beam dump from the accelerator is used as a secondary neutron monitor. However, the neutron fields of these facilities have different characteristics as shown in Table 2.

In these facilities, we have tested the neutron fluence measurement with a proton recoil telescope that consists of  $\text{CF}_4$  proportional counters and an NE213 scintillator, the neutron spectrum measurement with the TOF method and evaluation of low energy component with a bonner spectrometer.

We will compare results obtained from the experiments in TIARA and CYRIC. We will find several issues for evaluation technique of the characteristics on the neutron field and measurement precision.

We will also start tests in high-energy neutron field at RCNP. At TIARA and CYRIC Tohoku university, quasi-monoenergetic neutrons with an energy region from 250 to 390 MeV are produced using the  $^7\text{Li}(p,n)$  reaction and a proton beam from a ring cyclotron [7].

We will establish the high-energy neutron standard by 2015. These experiments have been performed in cooperation with the Tohoku University, KEK and JAEA.

Table 2: Comparison of the high-energy neutron fields

	dimensions	flux(n/cm <sup>2</sup> /s)
TIARA	19 m x 11 m x 6 m	1.2 x 10 <sup>5</sup> (5.2 m from the target)
CYRIC	11 m x 2 m x 5 m	10 <sup>7</sup> (0.7 m from the target)

**References:**

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