

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). The present status of our accelerator-based neutron field is described mainly on the development of monoenergetic neutron fluence standards [1-3]. Collaboration is in progress with the Japan Atomic Energy Agency (JAEA) aiming at the establishment of the high-energy (20 - 100 MeV) neutron standard field. We have also carried out studies on relative calibration of the neutron source emission rate between different source types [4] and on tiny thermal neutron sensors with gamma-ray suppression capability. In October 2006, the calibration and measurement capabilities were published for our standards on the quantities of fast monoenergetic neutrons fluence, thermal neutron fluence rate and neutron emission rate in the Appendix C of the BIPM key comparison database (KCDB). We are participating in the key comparison CCRI(III)-K8 on thermal neutron fluence measurements. We made the measurements with transfer instruments from November to December 2006 and returned them to the pilot laboratory in January 2007.

2. Monoenergetic neutron fluence standards

Figure 1 shows the irradiation room for our monoenergetic neutron fluence standards. The Van de Graaff accelerator is used to produce 144 keV, 565 keV and 5.0 MeV neutrons and the Cockcroft-Walton accelerator is used for 14.8 MeV neutrons. Calibration is performed on 0 deg direction to the accelerated beam lines from these accelerators. The CMCs for these energies are registered in the KCDB. The dimensions of the room are 11.5 m x 11.5 m x 11.5 m. The target assembly and the measuring instruments are placed on an aluminum-grating floor supported at mid-height of the room to reduce room-return neutrons. We have been preparing for new neutron standard fields for 2.5 - 3 MeV, 8.0 MeV and 27 keV neutrons. The development of a 19 MeV neutron standard field is underway. We are just installing a tritium target on the auxiliary beam line from the Van de Graaff accelerator for this purpose.

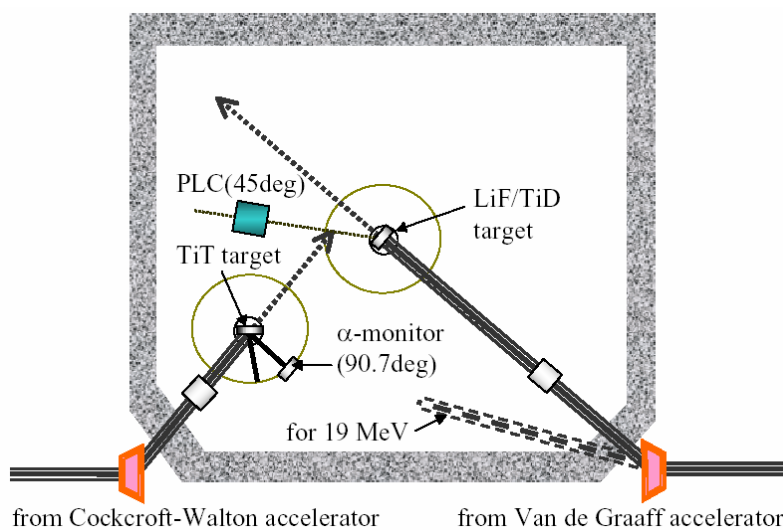


Figure 1: Schematic drawing of the neutron source

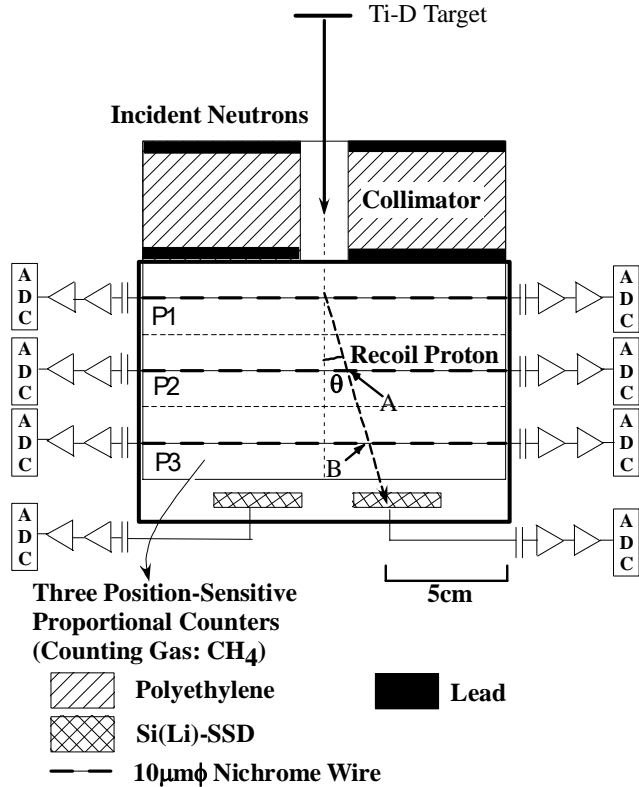


Figure 2 : Schematic drawing of the proton recoil spectrometer

2.1. 2.5 - 3 MeV neutron field

Neutrons are generated in the $D(d,n)^3He$ reaction by bombardment of a deuterium-titanium target with a 230 keV deuteron beam from the Cockcroft-Walton accelerator. The 2.5 and 3 MeV neutrons are respectively emitted to the 0 and 89.3 degrees with respect to the deuteron-beam direction. During calibration, the neutron fluence is monitored with a Bonner sphere consisting of

a spherical ^3He proportional counter of 33 mm diameter mounted at the center of a 9.5-inch diameter polyethylene spherical moderator. Absolute measurement of the neutron fluence was performed using a proton recoil neutron detector called TR (Thick Radiator) detector, which consists of a 0.5 mm thick polyethylene radiator disk mounted in front of a silicon surface barrier detector of 300 μm depletion depth. Neutron spectra were measured using a newly developed recoil proton spectrometer, shown in Figure 2, composed of three position sensitive proportional counters with methane gas as a radiator and counting gas, two lithium drifted silicon surface barrier detectors and a collimator made of polyethylene and lead [2]. The gamma rays existing in the field were also characterized using a liquid organic scintillation detector. Further details were presented at the international workshop on Fast Neutron Detectors and Applications (FNDA2006) and described in ref [3]. ISO 8529-1 gives 2.5 MeV as one of the recommended energies for determining the response of neutron-measuring devices. The calibration service at 2.5 MeV has started from April 2006 in NMIJ.

2.2. 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. The 8 MeV and 3.5 MeV neutrons respectively come from the transition to the ground and first excited states of their residual ^{12}C nuclei. A polyethylene long counter is used as a neutron monitor during calibration. Absolute measurement of the 8.0-MeV neutron fluence was performed in the same manner as 2.5 MeV. The TR detector used here consists of a silicon surface barrier detector of 2 mm depletion depth with a 1 mm thick polyethylene radiator. Neutron spectra were measured and the fractions of the accompanying 3.5 MeV neutrons were clarified with the recoil proton spectrometer used for 2.5 MeV as well as a liquid organic scintillation detector. Further investigation is in progress on techniques to subtract the contribution of the 3.5 MeV neutrons in actual calibrations. The gamma rays existing in the field were also characterized in the same manner as 2.5 MeV. Further details were presented at the 10th symposium on Neutron Dosimetry (NEUDOS10) and will be published in ref [4].

2.3. 27 keV neutron field

Monoenergetic neutrons of 10-30 keV 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. Neutrons were produced from the $^{45}\text{Sc}(p, n)^{45}\text{Ti}$ reaction by bombarding a scandium target with a proton beam from

the Van de Graaff accelerator at NMIJ/AIST. The proton energy was set to 2.926 MeV, 18 keV above the threshold energy of the $^{45}\text{Sc}(p,n)^{45}\text{Ti}$ reaction so as to generate neutrons with an average energy of 27.4 keV in the 0 degrees direction.

A ^3He cylindrical proportional counter (Reuter-Stokes RS-P4-0806: H1819) made of a stainless steel cylindrical tube filled with 0.4 MPa ^3He 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.

Relative measurement for the 27-keV neutrons was also performed using a Bonner sphere (BS), consisting of a spherical ^3He proportional counter 33 mm in diameter mounted at the center of a 241-mm diameter polyethylene spherical moderator, calibrated at our 144 keV standard neutron field.

For the 27 keV neutrons, the fluences obtained with the proportional counter and the BS agree within the uncertainties. The counting statistics, the gas pressure, the geometry, the background neutron contamination and the calculated efficiency were considered in the evaluation of the uncertainties. For the 144 keV neutrons, the fluence obtained from the proportional counter is in good agreement with that obtained from the standard calibration. This indicates that the measurements in the 144 keV standard neutron field supported the reliability of the data analysis procedure and the experimental conditions for each detector. Further details were presented at the 10th symposium on Neutron Dosimetry (NEUDOS10) and will be published in ref [5].

3. High energy neutron calibration field

There is a need for neutron fluence standards of 20-100 MeV energy regions since these neutrons are very important for radiation protection against the cosmic rays and around the facilities producing high intensity energetic neutrons such as J-PARC. Fortunately, we have several high-energy neutron facilities in Japan and we are examining their feasibility to the standard field.

Takasaki Ion Accelerators for Advanced Radiation Application (TIARA) of JAEA is one of the promising candidates. At TIARA, proton beam from an AVF cyclotron (K=110) is transported to a ^7Li target and quasi-monoenergetic neutrons are generated by the $^7\text{Li}(p,n)^7\text{Be}$ reaction, which are led into an irradiation room through a cylindrical collimator, 3 m in thickness and 11 cm in diameter. Dimensions of the irradiation room are 19 m x 11 m x 6 m. This field has been used for a lot of studies such as nuclear data measurements and shielding benchmark experiments.

Dr. Yoshiaki Shikaze et al. at JAEA investigated the neutron beam profile using the imaging plates with a polyethylene converter to measure the recoil protons produced in the converter. TOF measurements were performed with organic liquid scintillation detector. They also developed a

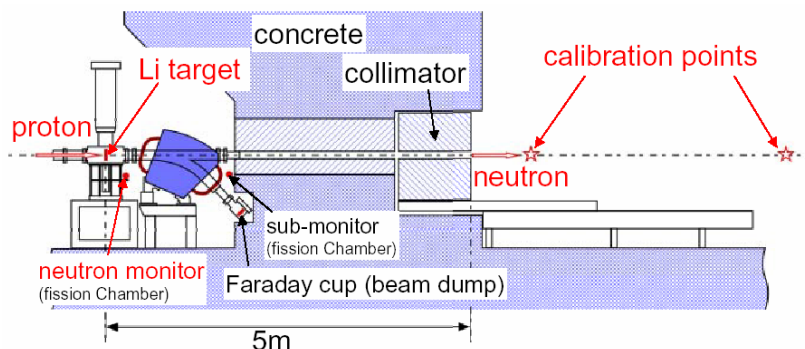


Figure 3 : High energy neutron field at TIARA

new recoil proton telescope to determine the neutron fluence. Collaboration is in progress with them to use this field for calibration purposes of 45, 60 and 75 MeV neutrons.

4. Relative calibration of the neutron source emission rate between different source types

We provide national standards for neutron emission rates from ^{252}Cf and $^{241}\text{Am-Be}$ sources. In calibration measurements, we use our standard graphite pile and perform relative calibration to the standard $^{241}\text{Am-Be}$ source calibrated at NPL. Our calibration method was proposed and established by our predecessor, Dr. T. Michikawa in the early 1980s [6]. We have performed supplementary experiments for this method and examined the physical mechanism by calculating the spectral fluence of the neutrons thermalized inside the graphite pile using the MCNP4C code.

$^{241}\text{Am-Be}$ or ^{252}Cf sources were placed at the center of the graphite pile, 230 cm wide, 190 cm deep and 190 cm high, and profile measurement of the neutron fluence was performed with a gold activation method. Gold foils, 25 mm thick and 20 mm in diameter, were placed in cadmium or aluminum covers of 1 mm thickness and embedded in the graphite pile. The neutron emission rates of the neutron sources were calibrated with the manganese bath method. The induced activities of the gold foils were determined by absolute measurement using the $4\pi\beta\text{-}\gamma$ coincidence counting equipment, the primary standard of radioactivity in NMIJ.

Figure 4 shows the saturated activities induced by the thermal neutrons in the graphite pile obtained for $^{241}\text{Am-Be}$ and ^{252}Cf neutron sources, which were normalized by the foil weight and the source neutron emission rate. The $^{241}\text{Am-Be}$ source had a broader spatial distribution than the ^{252}Cf source. The reason may be that $^{241}\text{Am-Be}$ neutrons need more collisions against carbon atoms to thermalize themselves, since the mean neutron energy of $^{241}\text{Am-Be}$ sources is about twice that of ^{252}Cf sources. Figure 4 also reveals that there is good agreement in the normalized activities between $^{241}\text{Am-Be}$ and ^{252}Cf neutron sources at the 90-cm position.

The neutron transport in the graphite pile was simulated with the MCNP4C code. The results are consistent with the tendency exhibited by experiments. The neutron spectral fluence distributions were characterized using the formula, $\Phi(E)=\Phi_t E/(kT)^2 \exp(-E/kT) + \Phi_n \Delta/E$ where T , Φ_t and Φ_n

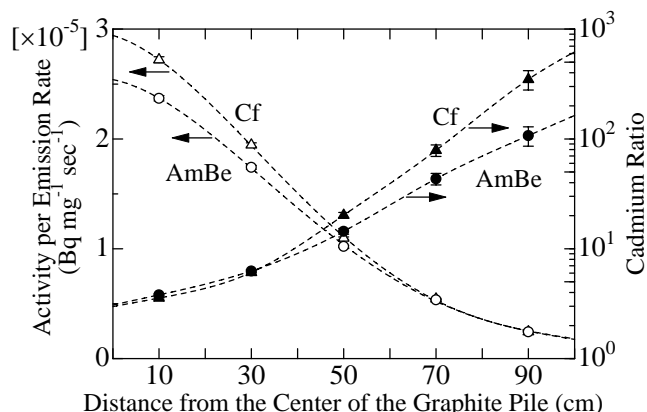


Figure 4 : Spatial distributions of saturated activities and the cadmium ratios in the graphite pile.

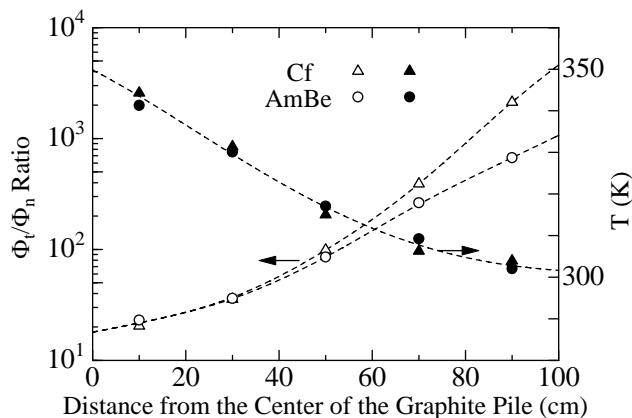


Figure 5 : Profiles of the neutron temperatures and the thermal to nonthermal neutron fluence.

represent the neutron temperature, the thermal and nonthermal neutron fluence and Δ is the joining function. The results shown in Figure 5 show that all neutrons have been thermalized with almost the same neutron temperature at 90 cm, which could explain the agreement between the saturated activities at 90 cm shown in Figure 4.

We tried to determine neutron emission rates by relative measurement between reference and sample sources of different source types. The relative measurement was performed just by comparing the counting rate of a thermal neutron monitor at the 90 cm distance from the neutron source placed at the center of the graphite pile. The successful results were obtained which agreed within 1 % as shown in Table I.

5. Novel small-sized neutron detector based on a ⁶Li-glass scintillator

Precise measurement of a spatial distribution of a thermal neutron fluence rate is necessary in studies on characterization of a thermal neutron field and neutron capture therapy. Then we have developed a small-sized neutron detector with optical fibers [7]. We suggested a gamma-ray suppression method for the small-sized thermal neutron detector.

Table I : Summary of the calibration results.

Sample sources	Reference sources			NMIJ Mn-Bath
	(c) Am-Be	(d) Ra(α)-Be	(e) Cf	
(a) Am-Be ($\times 10^5$ /s)	2.357 \pm 0.014	2.353 \pm 0.014	2.372 \pm 0.014	-
(b) Pu-Be ($\times 10^6$ /s)	4.648 \pm 0.028	4.640 \pm 0.028	4.677 \pm 0.028	-
(c) Am-Be ($\times 10^7$ /s)	-	1.127 \pm 0.007	1.137 \pm 0.007	1.129 \pm 0.007
(d) Ra(α)-Be ($\times 10^6$ /s)	1.626 \pm 0.010	-	1.636 \pm 0.010	1.623 \pm 0.010
(e) Cf ($\times 10^6$ /s)	8.864 \pm 0.054	8.849 \pm 0.054	-	8.920 \pm 0.054

Sample sources	Reference sources		NPL Mn-bath
	(f) Am-Be	(g) Cf	
(f) Am-Be ($\times 10^6$ /s)	-	8.992 \pm 0.046	9.006 \pm 0.072
(g) Cf ($\times 10^7$ /s)	2.343 \pm 0.019	-	2.339 \pm 0.012

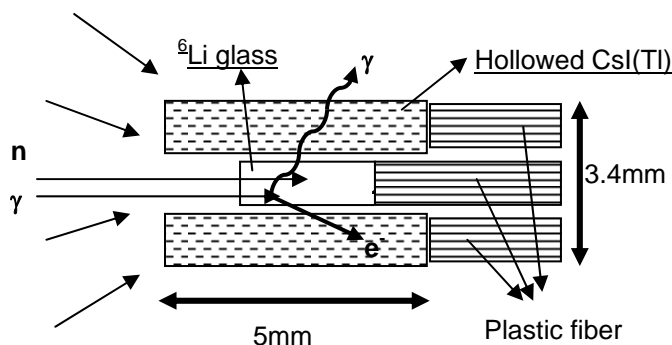
Figure 6 : Schematic drawing of the γ -ray suppression type small-sized thermal neutron detector.

Figure 6 shows a schematic drawing of the detector. The detector is a ^6Li -glass scintillator centered in a 3.4-mm outer diameter by 5.0-mm hollow CsI(Tl) scintillator. Transmission of scintillation light between the ^6Li -glass and the CsI(Tl) is interrupted by an aluminum foil with a thickness of about 40 μm . Plastic optical fibers are used for transmission of the scintillation light from the ^6Li -glass and the CsI(Tl) scintillators. The scintillation light from the CsI(Tl) is collected by summing up the signals from the several fibers.

In the detector, most of the secondary electrons escaping from the glass travel to the hollow CsI(Tl) scintillator through the aluminum foil. A part of secondary electrons produced in the CsI(Tl) scintillator also observed with the glass. The electron energy is deposited in the ^6Li glass and the CsI(Tl) scintillators, simultaneously. Finally, signals due to the gamma rays are rejected by an anti-coincidence method.

6. Future work

Calibration services will be started on the fluence standards for the monoenergetic neutrons of 8 MeV, 27 keV and 19 MeV, respectively in the fiscal years of 2007, 2008 and 2010. We have already established source-emitted neutron fluence standards for ^{252}Cf and $^{241}\text{Am-Be}$, which are currently provided in terms of the quantity of neutron fluence as seen our CMC list in the KCDB. In response to industrial needs, we will extend our services on these standards in the fiscal year of 2007 so that calibrations are also available in terms of dose quantities, e.g. ambient or personal dose equivalent. We also schedule to start calibration services based on the Japan Calibration Service System (JCSS) for the standards on the quantities of fast monoenergetic neutrons fluence, thermal neutron fluence rate and neutron emission rate respectively in the fiscal years of 2007, 2008 and 2010. The system will help to improve the traceability of neutron measurement in Japan since calibration laboratories are assessed and accredited to meet the requirements of ISO/IEC 17025.

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