Developments in neutron metrology at the Institute for Radiological Protection and Nuclear Safety in 2009 and 2010

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1 Introduction

The institute for radiological protection and nuclear safety (IRSN) is in charge of the neutron fluence and dose equivalent quantities for the LNE (Laboratoire National de métrologie et d'Essais), the French National Metrological Institute.

IRSN laboratory for neutron metrology and dosimetry (LMDN) disposes of several facilities producing reference neutron fields and performs neutron spectrometry with several types of instrument in order to determine these quantities with the best achievable accuracy.

2 Neutron reference fields

Up to now, the only facility where IRSN proposed CMCs is the irradiator with radioactive sources ²⁴¹Am-Be and ²⁵²Cf (moderated or not with a heavy water sphere). However, IRSN has several other reference facilities dedicated to neutron metrology and dosimetry, all Located on the Cadarache site, in the South of France.

2.1 Radionuclide sources

The neutron irradiator facility consists in radionuclides sources of ²⁴¹Am-Be and ²⁵²Cf. In 2010, the traceability of the bare sources to the NPL references has been checked with a Berthold survey meter, this control being performed every 5 years.

2.2 Thermal neutron field

The SIGMA facility consisted of six 241 Am-Be neutron sources, of 0.56TBq each, which were located in a graphite moderator block of $150 \times 150 \times 150 \text{ cm}^3$ on side. In total, the neutron sources strength was about $1.9 \times 10^8 \text{ s}^{-1}$. As explained in the last report, the more than 10 years old 241 Am-Be sources should be evacuated, due to the French regulations, stopping the use of this facility beginning of 2006. We used this as an opportunity to improve the thermal components of the field. As a first step, it was decided to place a 6.3 GBq (170 mCi) 252 Cf source at the center of the existing graphite moderator block resulting in 95% of neutrons with energy below 0.5 eV generating 88% of the neutron total dose equivalent. However, it has not been possible in the last years to get the 252 Cf source with reasonable cost and delay. This situation resulted in the cancellation of IRSN participation to the CCRI key comparison K-8 as announced in 2009. Since the last CCRI meeting, IRSN

decided to no longer consider the ²⁵²Cf source option and to build an accelerator based-moderated neutron source to create the thermal neutron field. This new facility will however not be available before 2014.

2.3 AMANDE : Mono-energetic neutron fields

AMANDE is IRSN accelerator producing mono-energetic neutron fields within the energy range from a few keV up to 20 MeV. Neutrons are created using nuclear reactions between accelerated protons, deuterons and thin targets like scandium, lithium, deuterium, tritium, as defined by ISO 8529-1 standard. The AMANDE facility is based on a 2 MV HVEE Tandetron accelerator, which has been installed at the end of 2004 in a low scatter building.

Related metrological activities in 2009 and 2010 concern:

- the finalization of the characterization of IRSN long counter, used as measurement reference standard for neutron fluence at AMANDE facility
- the development of a measurement reference standard for neutron energies between 1 MeV and 20 MeV using time of flight method
- the further development of two nucleus recoil telescopes for both energy and fluence reference determination
- investigations of the ⁴⁵Sc(p,n) reactions
- several studies for reference traceabilities and uncertainty budgets in order to establish a quality control following the ISO-17025 standard (foreseen in 2012) and to prepare the participation to the next CCRI comparison exercise on fluence in monoenergetic neutron fields

2.3.1 PLC long counter

IRSN long counter (PLC) is an optimized geometry of the De Pangher long counters to get a flat neutron response energy distribution, over a wide energy range, from a few eV's to a few MeV's [Lac10a]

Measurements at the IRSN radionuclides sources, ²⁵²Cf and ²⁴¹Am-Be, were performed to determine the scaling factor to be applied to the calculated response function. A comparison was then performed between the fluence determined by the PLC and IRSN proton recoil spectrometers (SP2 gaseous proportional counters and BC501A liquid scintillator, traceable in 2001 to the PTB neutron references).

The obtained results were in good agreement within the experimental uncertainties as shown in the table 1 [Lac10b].

Table 1: Comparison between the fluence per monitor count determined in several monoenergetic neutron fields by IRSN proton recoil counters (SP2 and BC501A) and IRSN long counter (PLC). All measurements were performed at 0°, using the shadow cone method and the uncertainties are given for a coverage factor of 1.

E_n (MeV)	Φ_{ref}/M (SP2/BC501A)	<i>Ф/М</i> (PLC)	Φ/Φ_{ref}
0.18	8.68 ± 0.36 cm ⁻²	8.73 ± 0.21 cm ⁻²	1.006 ± 0.048
0.64	5.54 ± 0.19 cm ⁻²	5.39 ± 0.11 cm ⁻²	0.974 ± 0.041
3.19	0.55 ± 0.02 cm ⁻²	0.55 ± 0.01 cm ⁻²	1.013 ± 0.037
4.50	1.24 ± 0.04 cm ⁻²	1.17 ± 0.02 cm ⁻²	0.948 ± 0.037

IRSN participated with the PLC to the EURAMET 936 inter comparison exercise. This exercise consisted in the comparison of long counters results after measurements at the NPL Van de Graff mono energetic neutron fields and radionuclide sources. An excellent agreement was obtained between all the participants [Rob10], validating the characterization of the PLC. During the K11 comparison exercise, the IRSN long counter will be used to provide the reference values of the neutron fluence.

2.3.2 Time of flight measurements

The reference measurement standard for neutron energy above 1 MeV is based on the time of flight (ToF) method, using a 2"x2" BC501A liquid scintillator as fast neutron detector. To extract as accurately as possible the mean energy from the experimental neutron ToF distribution, a simulated energy distribution is fitted to the measurements. This simulation includes a detailed analysis of the energy resolution, taking into account all the contributions influencing the ToF measurements, i.e. the energy spread of the ion beam, the thickness of the neutron-producing target, the energy variation on the detector front surface, the time distribution of the beam pulse, the time distribution due to the detector and the time spread due to the electronics (bin width, jitter, etc).

Comparison with neutron energy calculated from kinematics leads to the conclusion that the ToF method at the AMANDE facility can be considered as a reference measurement standard for neutron energies between 1 MeV and 20 MeV, with a relative uncertainty lower than 1.5%, as shown in the figure 2 [Cog10].



Figure 2: Relative difference between experimental and calculated (by kinematics) ToF values.

The neutron energy range available with this method will be extended down to a few kiloelectronvolts or lower in a further study, using Li-glass and/or plastic scintillators.

2.3.3 Nucleus recoil telescope

Since 2007, IRSN is developing two new systems for the measurement of the energy and fluence parameters of neutron fields in the framework of two collaborations with the CNRS (National Scientific Research Center).

The RNT principle is based on the detection of the recoil nucleus emitted by the elastic scattering of the neutron on a converter. The simultaneous measurement of the nucleus energy and recoil angle leads to the initial neutron energy. For some existing Recoil Proton Telescope (RPT), the recoil angle is fixed and the energy of the scattered proton is measured in a narrow solid angle generated by a small area detector. The precision on fluence is of 3 to 5%, depending on the neutron energy. However, due to the small acceptance angle, the efficiency is quite low (about 10⁻⁵). To improve this method, it is necessary to enhance the efficiency by increasing the detection solid angle. The main idea is to use localization detector systems in order to get full spatial information on the nucleus scattering angle. Because of a large energy range of AMANDE neutron fields, two systems are needed.

A proton recoil telescope using CMOS sensor (CMOS-RPT) is studied, in collaboration with CNRS-Ramses unit (radioprotection field), for measurements at high energies (5 MeV - 20 MeV). This system is composed of a polyethylene converter foil, three pixellised CMOS detector layers (pixels with a pitch of 30 μ m) with a thickness of 50 μ m to reduce the energy threshold and a Si(Li) diode to measure the proton recoil nucleus energy.

A gaseous μ -time projection chamber (μ -TPC) will be dedicated to the lowest energies (2 keV - 1 MeV). This study is in collaboration with CNRS-MIMAC unit (Directional non baryonic dark matter research field). The μ TPC using a gaseous neutron converter provides therefore a very low energy detection threshold, several mixture may be used (e.g. C₄H₁₀, He⁴+ C₄H₁₀). The primary electrons-ions pairs produced by nuclear recoil in the TPC are detected by drifting the electrons to the grid of a bulk micromegas. The electrons move towards the grid in the drift space and are projected on the anode thus allowing getting information on X and Y coordinates. Anode present a 10 by 10 cm² active area, segmented in pixels with a pitch of 350 µm. In order to reconstruct the third dimension Z of the recoil, a selftriggered electronics has been developed. It allows performing the anode sampling at a frequency of 50 MHz. The Z coordinates is then obtained with the drift time, providing the electron drift velocity is known.

Simulations of the two systems were performed with the transport Monte Carlo code MCNPX, to choose the components and the geometry, to optimize the efficiency and detection limits of both devices or to estimate performances expected (efficiency about 10^{-3} for µTPC and 10^{-4} for CMOS RPT and energy resolution about 5-10%).

Measurements campaigns at AMANDE facility are performed (around two weeks per year) to test and optimize the two prototypes. Both detectors will be used finally as primary measurement standards for the characterization of the mono-energetic neutron fields of AMANDE facility.

2.3.4 Investigation of neutron reaction for low energy monoenergetic neutron fields

A study on the low energy monoenergetic neutron fields (below 100 keV) is in progress within the framework of a European scientific cooperation between IRSN, NPL, PTB and IRMM. The main part of this study is focused on the 45 Sc(p,n) reaction.

<u>Investigation of suitable reactions for low energy neutron reference fields</u> production

The variation of the neutron yield with ion beam energy from the neutron threshold up to about 50 keV has been measured at IRSN AMANDE facility for the ${}^{45}Sc(p,n)$, ${}^{65}Cu(p,n)$, ${}^{51}V(p,n)$, ${}^{57}Fe(p,n)$ and ${}^{37}Cl(p,n)$ reactions and is presented in the figure 3. ${}^{51}V(p,n)$, ${}^{57}Fe(p,n)$ and ${}^{37}Cl(p,n)$ reactions have very low neutron yields that are not suitable for any practical calibration of neutron measuring devices. The first resonance of the ${}^{65}Cu(p,n)$ reaction corresponds to a proton energy below the monoenergetic threshold. A second neutron group of 0.116 keV exists at 0° with a fluence production corresponding to about 9% relatively to the 1.17 keV resonance (Böttger et al., 1989). In addition, the yield is 17 times lower than for the 8 keV scandium resonance. Therefore the ${}^{45}Sc(p,n)$ is the best suited reaction to produce low energy neutron reference fields.



Figure 3: Count rate, for 1 keV ion beam energy loss in the target, for the IRSN long counter placed at 0° and 66 cm from the target for respectively ${}^{45}Sc(p,n)$, ${}^{65}Cu(p,n)$, ${}^{51}V(p,n)$, ${}^{57}Fe(p,n)$ and ${}^{37}CI(p,n)$ reactions, as a function of the proton beam energy.

• ⁴⁵<u>Sc(p,n) reaction</u>

This reaction exhibits two resonances suitable for neutron metrology and calibration of neutron devices. The corresponding neutron energies at 0° emission angle are respectively 8 and 27 keV, the latter one being proposed in the CCRI(III)-K11 instead of the 24 keV of the previous K1 comparison.

Investigations were led at the NPL in order to determine what backing is the most suited for scandium targets. Indeed, due to the low cross-section of the reaction (< 2mb), targets must sustain ion beam currents up to 50μ A for several hours. Tantalum was selected as the best choice [Lam10a].

The study of the cross-section variation with angle performed at NPL has confirmed the neutron isotropic emission in the centre of mass as illustrated in the figure 3.



Figure 3: Variation of the neutron fluence, determined with two different long counters at NPL facility, as a function of the forward emission angle (up to 70°) in the laboratory frame of reference, for the 8.15 and 27.4 keV resonances of the ⁴⁵Sc(p,n) reaction and comparison with the theoretical variation in the case of an isotropic emission in the centre of mass.

The variation of this cross section with the proton energy is under study at AMANDE facility as well as at AIFIRA facility from CENBG (CNRS-Bordeaux), offering a beam energy spread of only 100 eV. Further studies are planned, including time of flight and photon measurements, cross section determination at the two resonances. Once evaluated, the cross-section data of this study will be implemented in the TARGET or NEUSDESC Monte-Carlo simulation softwares.

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