Recent developments in neutron metrology at the Institute for Radiological Protection and Nuclear Safety (IRSN)

A. Allaoua, B. Asselineau, M. Benmosbah, M.A. Cognet, V. Gressier, V. Lacoste, V. Lamirand, L. Lebreton, S. Serre, L. Van Ryckeghem

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

For the moment, the only facility where IRSN proposed CMCs is the irradiator with radioactive sources ^{241}Am-Be and ^{252}Cf (moderated or not with a heavy water sphere). However, IRSN has several other facilities dedicated to neutron metrology and dosimetry, all situated at the Cadarache centre in the south of France.

2.1 Radionuclide sources

A 11 GBq ^{137}Cs source has been installed near the neutron source irradiator to verify that the response of the instrument to be calibrated are not highly sensitive to photons (their contribution representing a few percent of the neutron dose equivalent).
The test point is located at 75 cm from the centre of the ^{137}Cs source. At this point, the ambient neutron dose equivalent is of about 750 μSv/h with ^{241}AmBe source and the photon dose of about 1700 μSv/h with the ^{137}Cs source.

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 150×150×150 cm^3 on side. In total, the neutron sources strength was about 1.9×10^8 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. We used this as an opportunity to improve the thermal components of the field. As a first step, a 6.3 GBq (170 mCi) ^{252}Cf source will be set-up at the center of the existing graphite moderator block and the neutron field

produced will be experimentally studied. This new facility will provide 95% of neutrons with energy below 0.5 eV generating 88% of the neutron total dose equivalent. The AmBe sources evacuation was foreseen in 2007, with the arrival of the new source end of 2007. Several difficulties induced however large delays. The first difficulty was an impossibility to evacuate the ^{241}AmBe in reasonable delays. At the end of 2007 a container was built to store the sources in the SIGMA experimental hall. This container is a block composed of polyethylene and lead. It was designed by MCNP simulation in order that the residual radiation field coming out this block generates only negligible contribution in fluence and dose to the new thermal neutron field. The 6 ^{241}AmBe sources were transferred into this storage container in February 2008 for an undefined period.
The second encountered difficulty was the ^{252}Cf delivery, foreseen initially middle of 2008. Unfortunately, the selected provider (QSA Global) did not deliver the source due to the fact that the nuclear reactor used to create these sources in USA stopped its activity. The delivery was postponed to more than one year. Therefore we looked after a new supplier and the ^{252}Cf source should be delivered by the end of 2009 in the best case. Once the source set-up in the graphite moderator block done, a complete characterization the neutron field will be performed. Different neutron spectrometers will be used to determine the neutron fluence energy distribution over the whole energy range, from thermal to a few MeV. The thermal neutron fluence rate will be determined by activation of gold foils. All these delays postponed our participation to the CCRI-K8 comparison exercise foreseen initially in 2008. We hope to be able to participate to this inter comparison exercise by mid of 2010.

2.3 AMANDE : Mono-energetic neutron fields

2.3.1 AMANDE facility

AMANDE is IRSN new 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 new building.
Years 2007 and 2008 were dedicated to:

- calibration of neutron dosimeters, spectrometers and reference instruments, mainly for IRSN but also for French companies as Saphymo or MGPI.
- commissioning of the time of flight measurements above 1 MeV
- first studies of the ^{45}Sc(p,n) reactions

A quality control is being established for the calibration at AMANDE facility following the ISO-17025 standard and requiring several studies for reference traceabilities and uncertainty budgets. In the two future years, intensive works will be performed in order to develop CMC's for the monoenergetic fields and prepare our participation to the next CCRI comparison exercise on fluence in monoenergetic neutron fields, which could be organized at AMANDE facility.

2.3.2 Reference long counter

In 2006, the laboratory has undertaken a study related to the optimization of the geometry of a long counter, to determine the neutron fluence reference values at the AMANDE facility. These studies were performed by using Monte-Carlo simulations, and on basis of a De Pangher long counter nominal geometry. The materials, the dimensions and the geometry were optimized to get a flat neutron response energy distribution, over a wide energy range, from a few eV to a few MeV. The calculated response function for a high pressure ^{3}He proportional counter (10 atm) is shown in Figure 1. In 2007, the device was built (Figure 2**Error! Reference source not found.**) and first measurements were performed at the radionuclides sources and AMANDE mono-energetic neutron fields to determine the effective centers and the fluence response at several neutron energies. Additional measurements were performed at AMANDE to determine the dead time of the ^{3}He proportional counter. The dead time value was estimated to 19.3 μs.

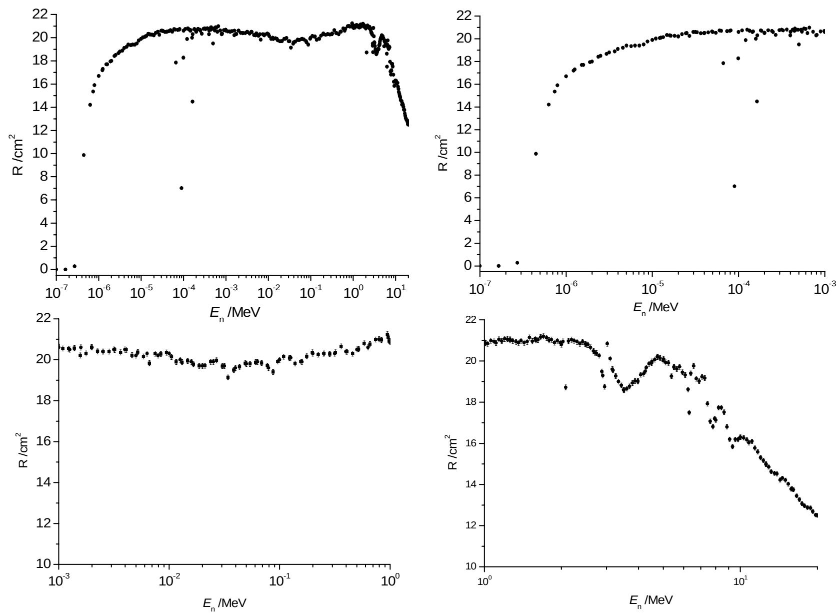

Figure 1: Response function of the IRSN long counter, calculated for a 10 atm ^{3}He proportional counter. The response is calculated for 242 neutron energies and is shown over the whole energy range from 0.1 eV and 20 MeV, then from 0.1 eV to 1 keV, from 1 keV to 1 MeV and from 1 MeV to 20 MeV.

Figure 2 : IRSN long counter on its support, manufactured on basis of the geometry defined by Monte-Carlo study

The effective center values of the IRSN long counter were calculated and experimentally determined at the AMANDE facility. A good agreement is observed overall the neutron energy range, from 144 keV up to 17 MeV, as shown in Figure 3.

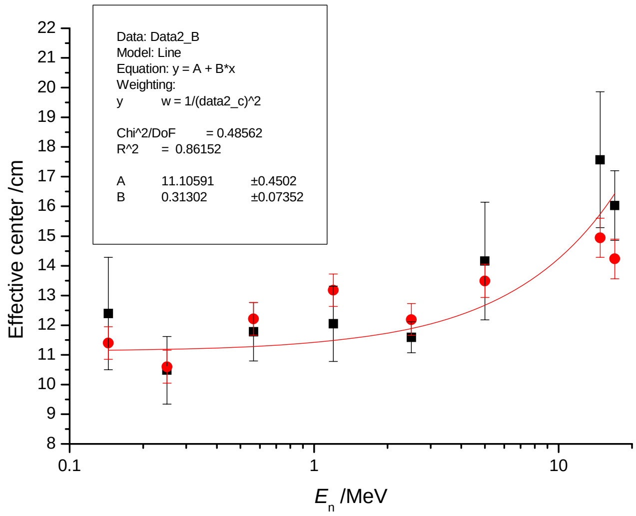

Figure 3 : Experimental and calculated effective centers as a function of the neutron energy. The squares correspond to the experimental data.

As mentioned before, measurements at the IRSN radionuclides sources, ^{252}Cf and ^{241}Am-Be, were performed to determine the efficiency of the device. These measurements are mandatory to know whether a scaling factor should be applied to the calculated response function. The measurements were performed at several distances, with then without shadow cone. The corrected count rates were compared to the calculated ones, obtained by Monte-Carlo simulations using ISO spectra for both the sources. Some discrepancies were observed, from 12% to 20 %, for the ^{241}Am-Be and ^{252}Cf sources respectively. By increasing the gas pressure by 10% (for instance) the discrepancies were decreased by about 6%. Further investigations using Monte-Carlo simulations are in progress to explain these discrepancies, and new measurements at the IRSN radionuclides sources facility are foreseen in April 2009 to check the previous experimental data.

In June 2008, the laboratory has participated together with the NPL and the PTB, 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 radionuclides sources. The set-up of the four devices used during this measuring campaign is shown on Figure 4.
Measurements were performed at the following neutron fields:

- ^{252}Cf, ^{241}AmBe, ^{241}AmB, ^{241}AmLi and ^{241}AmF,
- 144 keV, 565 keV, 1.2 MeV, 5 MeV and 17 MeV mono-energetic neutrons

The analysis of the data is in progress but preliminary results (ratio of the IRSN and NPL data) show a rather good behavior of the IRSN long counter with the neutron energy.

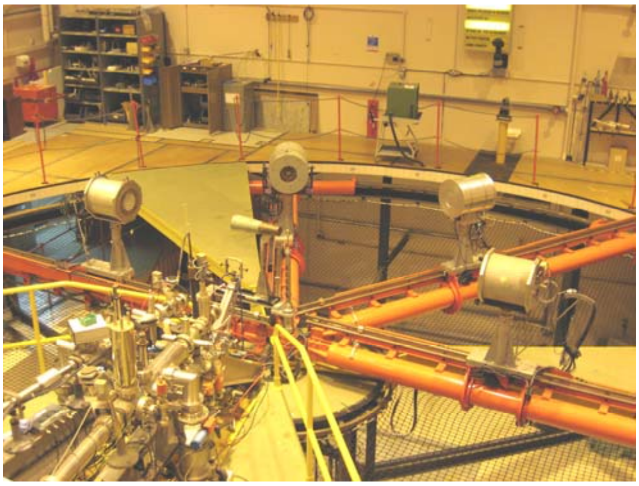

Figure 4 : « EURAMET 936 » at NPL, in June 2008. From left to the right of the picture, the PTB De Pangher long counter, the NPL long counter, the IRSN long counter and the NPL De Pangher long counter

2.3.3 Time of flight measurements

The pulsed mode is used to perform time of flight (ToF) measurements dedicated to the measurement in an absolute way of the neutron energy of the mono-energetic fields. The ToF method consists in measuring the time between the creation of the neutrons in the target and their interaction point in a detector situated up to 10 meters from the target. By this way, the neutron energy will be directly related to the time and distance standards. These measurements are performed using different detectors, all with fast time responses, as a BC501A liquid scintillator for neutron energy above 1 MeV.

The pulsed performances of the accelerator have been tested in the whole ion beam energy range, then time of flight measurements have been performed with a 2'' × 2'' BC501A liquid scintillator for energy neutrons above 1 MeV. To establish the energy references by ToF method, studies have been done to estimate the influence of different parameters to the energy resolution: neutron energy, flight path length, thickness of the neutron producing targets, pulse width of the ion beam, thickness of the detector, electronics, uncertainty on the time calibration, etc.
All these parameters were taken into account in the analysis method using simulations with Monte-Carlo codes: TARGET and NRESP developed by the PTB. The main encountered difficulty was related to the time shape of the ion beam pulse which has to be precisely measured during the experiments to be implemented in the simulation of the energy resolution. This time shape is obtained from the photon peak in the ToF spectrum with care on the discrimination level in the settings of the electronics.

The experimental data are consistent with the theory whereas some studies are still needed especially to improve the total uncertainty. Moreover, for energy below 1 MeV, new devices as lithium glass, BaF2 or plastic scintillator are being investigated.

2.3.4 Nucleus recoil telescope

IRSN proton recoil reference detectors (SP2 proportional counter and BC501A liquid scintillator) can not be assumed as primary detector systems. In SP2 case, the gas pressure inside the detector is not precisely determined. For BC501A, the composition and the reaction cross section of each component are not well known. For these two protons recoil detector systems the neutron fluence energy distribution is determined using unfolding code for which the response function of the system is needed. The response function is obtained by simulation code taking into account geometry, composition, reaction cross section, gas pressure or specific parameters defined through experimental calibration at PTB. Thus, the absolute neutron energy distribution at AMANDE can not be obtained.
For this purpose, the development of new detectors acting as Recoil Nuclear Telescopes (RNT) has been started.

The RNT principle is based on the detection of the recoil nucleus emitted by the elastic scattering of the neutron on a thin converter foil. The simultaneous measurement of the nucleus energy and recoil angle leads to the initial neutron energy. For some existing Recoil Proton Telescope (RPT) used in this energy, 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 0.001%).

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). The helium 4 gaseous µ-time projection chamber (µ-TPC ^{4}He) will be dedicated to the lowest energies (2 keV – 5 MeV). This study is in collaboration with CNRS-MIMAC-He3 unit (Non baryonic dark matter research field).
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. First preliminary measurements in front of AMANDE facility were realised in 2008 demonstrated the proof of principle of these novel detectors for neutron metrology. Experiments already scheduled in 2009, with both prototypes, will allow the recoil nuclei tracks detection and recoil nuclei energy reconstruction to access to the absolute neutron energy measurements.

2.3.5 Simulation of the neutron fluence energy distribution

In order to take into account as best as possible the scattered neutrons contribution to the fluence and dose equivalent, a complete detailed simulation of AMANDE experimental hall has been done coupling MCNP and TARGET code within an interface allowing fast MCNP geometry modification of the detector transport system. Simulations are being compared to experimental data from measurements performed with IRSN reference neutron spectrometers at several positions into the experimental hall. The impact of the scattered neutrons to the fluence to dose conversion factors is also being investigated.

2.3.6 Investigation of neutron reaction for low energy monoenergetic neutron fields

The response of a neutron detector varies with neutron energy. The experimental determination of detector response functions is usually performed at facilities producing monoenergetic neutron fields. For the commonly used nuclear reactions, monoenergetic neutrons are obtained between 120 keV and 20 MeV in the ion beam direction (0°). Lower neutron energies are available at other angles. However, use of these angles presents problems because the neutron energy and fluence vary across the detector surface. They also involve an increase in the relative scattered-neutron contribution. An alternative solution is to use different nuclear reactions. Despite the resonant structure of its cross-section with incident proton energy, the ^{45}Sc(p,n) reaction appears to be one of the most favourable candidates as it allows the energy range to be extended down to 8 keV at 0° with sufficient neutron yield.
A complete study of the scandium reaction is being undertaken within the framework of a European scientific cooperation between IRSN, NPL, PTB and IRMM. Preliminary measurements of cross-section, fluence and isotropy in the centre of mass have been conducted at AMANDE facility. The next step of this work would be dedicated to the selection of the most suitable backing for scandium targets. Indeed, due to the low cross-section of the reaction (< 2mb), targets must sustain ion beam currents up to 50µA for several hours. Scandium targets with Al, Ta, Mo, Ag, W and Pt backings will be tested under irradiation at the NPL facility. Their composition, homogeneity and thickness are analysed at CENBG AIFIRA facility by Rutherford backscattering spectrometry (RBS) before and after irradiation. The results of this study as well as calibration measurements of survey meters performed at NPL and IRSN facilities at 8 and 27 keV with the ^{45}Sc(p,n) reaction will be reported at NEUDOS 11 in October 2009. Further studies are planed, including an investigation of cross-section variation with angle and proton energy, time of flight and photon measurements, and the possibility of using other nuclear reactions.

3 Neutron Spectrometry

IRSN has several spectrometers used for fluence energy distribution characterization at workplaces (Active and passive Bonner Spheres system) or at calibration facilities (spherical proton recoil counters SP2 and liquid scintillators BC501A).

3.1 High energy Bonner spheres system

The study and design of a spectrometer for high energy neutrons is of great interest for IRSN which has to face to an increasing demand concerning the characterization of high energy neutron fields. Several requests were received from the medical facilities, to perform measurements around accelerators used in proton-therapy. Other requests concerned the atmospheric neutron environment characterisation for single event upsets applications and the characterization of neutron fields around high energy research accelerators, for the design of radiological protections. The study and the development of this new spectrometry system for high energy neutron are being performed in the framework of a PhD, of three years duration.

The PhD thesis started with a bibliography work related to the characteristics of the high energy neutron fields encountered in various domains (research, medical accelerators, aeronautics, etc). The main characteristics concerned the total neutron fluence rate, the energy range of the neutron spectrum, the other particles in the fields. A short study then consisted in the review of all existing high energy neutron spectrometry systems used worldwide for any type of applications.
In a further step, we have determined the type of thermal neutron counters which may be used with a multisphere spectrometer suited for low fluence rate and high energy neutrons. All this information led to the definition and design of the spectrometer. Meanwhile, studies by Monte-Carlo simulations of the response functions and the estimate of the associated uncertainties of the system are undergone. This is one of the most difficult parts of the work; indeed, the high energy field is composed of high energy neutrons and of many different types of charged particles (protons, charges pions); secondary neutrons can be created by the interactions of the high energy neutrons and charges particles with the nuclei of atoms of the materials of the spectrometer. Therefore, the knowledge of the charged particles distributions which may be present in the measured field is a pre-requisite to estimate the secondary neutrons and subtract them from the measurements. The uncertainties associated to the response functions are then being studied in details, considering different parameters: the variations of the evaluated nuclear reactions cross-sections which may differ according the databases, the differences between high energy nuclear models, and the variations of composition and densities of the materials.
Once available, the system has been experimentally characterized, ie the calculated neutron fluence response energy distributions have been experimentally validated for a few neutron energies; for that, calibration of the the system was performed at the IRSN radionuclides facility, using ^{241}Am-Be standard neutron sources. For measurements with higher than 20 MeV neutron energies, the neutron fluence responses were experimentally determined at The Svedberg Laboratory (TSL) quasi-mono-energetic neutron fields. The good agreement between the experimental results and the reference values provided by the laboratories will indicate the good behaviour of the system and validate the calculated response functions. Different measurement campaigns will be organized at different period within the year to determine the variation of the neutron spectra and the acceleration factors with atmospheric and climate conditions.

3.2 ROSPEC characterization

The ROSPEC device is a multi-detector system which has been designed by Bubble Technologies Industries (BTI at Chalk River, Ontario, Canada) to assess neutron spectra, and hence neutron dose quantities, at workplace fields. It is made up of six gaseous proportional counters that detect neutrons via the elastic (n,p) scattering (four hydrogenous counters) and with the ^{3}He(n,p)T reaction (two ^{3}He filled counters). For energy and fluence calibration purposes, measurements were performed with the AMANDE facility which provides monoenergetic neutron radiation fields from 2 keV to 20 MeV. Two kinds of experiments were carried out. First, the ROSPEC was used in its rotational mode for the ISO energies. Then, each detector was irradiated with the spectrometer stopped with all the available energies. The energy values of the neutron beam were given thanks to calculations with the TARGET code. A BC501-A liquid scintillation spectrometer provided the fluence values for energies beyond 1.2 MeV, a methane-filled SP2 counter from 800 keV to 1.4 MeV and a H_2-filled SP2 counter from 144 keV to 800 keV. Reference data for 70 keV monoenergetic neutrons were given by the IRSN Long Counter. Results showed that the ROSPEC device was in agreement with the absolute neutron fluences within 10 %. Moreover, the new energy calibration factors are in good agreement with those derived by BTI. Further investigations aim to determine the response matrices of the spectrometer in order to perform the unfolding of the spectral data. The response matrices are being calculated using MCNPX code. Low energy new MCNPX modules for Stopping Powers calculation for protons and alpha particles have been developed within the framework of a collaboration between Los Alamos laboratory and IRSN. The purpose of this study is to carry out a better pulse height distributions treatment than the one done by the current SPEC4 code and to enable a high resolution neutron spectrometry in the energy range between 50 keV and 4.5 MeV.

4 Status of the survey meter intercomparaison

The EUROMET.RI(III)- S1 comparison is co-organised by IRSN, NPL and PTB institutes. This exercise consists in calibrating in H*(10) a reference instrument (survey-meter) sent to all the participating national metrological institutes.
According to the recommendations of the ISO 8529 standard, the measurements are performed in the following neutron fields:

- ^{241}Am-Be ,
- ^{252}Cf,
- (^{252}Cf+D_2O)$_{/Cd}$.

The first part of this comparison exercise implied 6 laboratories (IRSN, SCK, KRISS, CMI, IEA, SMU) between 2005 and 2007. These participants obtained consistent results in terms of ambient dose equivalent calibration detailed in an intermediate report sent to the participants in 2008.
A second and last part of this comparison should be organised in 2009-2010.

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