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

V.Gressier, V. Lacoste and L. Lebreton

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'Essai), 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 Progress since 2005

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 [Gre06].

2.1 Radionuclide sources

2.1.1 ^{241}Am-Be source

The neutron energy distribution of the IRSN standard ^{241}Am-Be source was measured using a proton-recoil liquid scintillator (BC501A) for neutron energies above 1.65 MeV. The experimental data were compared to the ISO 8529-2 [ISO1] recommended neutron energy distribution for the ^{241}Am-Be source and some significant discrepancies were observed within the energy interval from 3 to 6 MeV and around 8 MeV. These discrepancies contributed to a total fluence of 3% more than in the ISO distribution in the energy range between 2 and 11 MeV. This was estimated by taking the known source emission rate and anisotropy, using the ISO spectrum to calculate the fraction above 2 MeV, and comparing that value with the fluence as measured with the BC501A. Investigations undertaken to explain these discrepancies were published in a previous work [Mag07].
Within the framework of a collaboration between three national metrological institutes (PTB, Germany – NPL, United Kingdom and LNE-IRSN, France), the neutron energy distributions of different ^{241}Am-Be sources at each laboratory have been compared [Leb06]. The aim was to investigate reasons for the observed differences obtained with sources built by different manufacturers with different characteristics. Measurements performed at IRSN, PTB and NPL were made with the same detector system, IRSN-BC501A proton-recoil scintillator, and followed the

same procedure, thus allowing detailed comparisons to be made of the spectral shapes derived from each measurement.

Discrepancies observed for each source are shown in Figure 1. The differences are largest in two energy ranges, around 4 and 7 MeV. The way in which ^{241}Am-Be sources are made (mixture, cavity size) seems to have an influence on the neutron emission rate in these energy ranges. We can observe a general magnitude reduction in 3-9 MeV energy range when the activity or the volume of the source increases. One of the reasons might be a greater number of collisions of neutrons inside the source or in the capsule before escaping from the larger capsule that is required to contain the larger ^{241}Am activity. To confirm general magnitude reduction rather than differences in shape of the fluence energy distribution, each distribution has been normalised by the fluence integral between 2 and 11 MeV, as shown in figure 2. A good agreement is observed for all measurement, except for IRSN source. For this last, the significant differences in the composition compared to Amersham sources (PTB and NPL) can explain this behaviour.
In conclusion, measurements with a BC501A detector system on five ^{241}Am-Be sources point to there being some differences in the proportion to total fluence rate of high energy neutrons (above 2 MeV) depending of their activity or size. To understand the reasons for these discrepancies, we must go further in analysis with more detailed distribution obtained with different spectrometers and with comparisons between a greater numbers of ^{241}Am-Be sources. The link with chemical composition (mixture composition, size of cluster) of each source will be done.

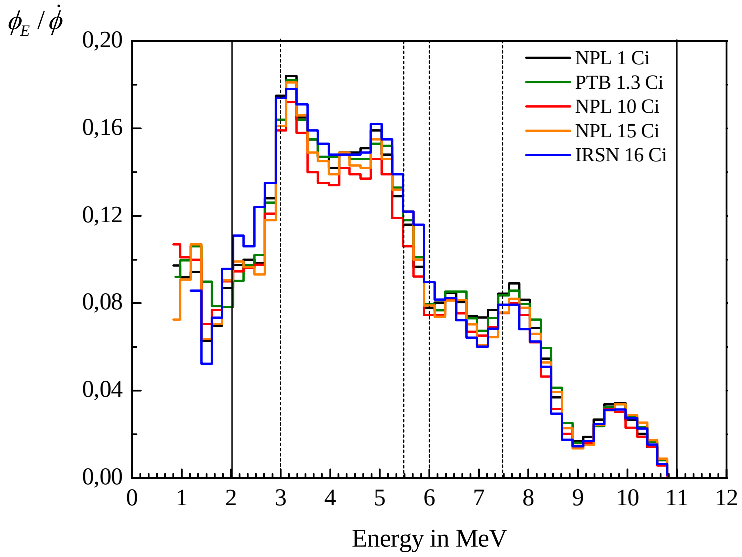

Figure 1: comparison of experimental neutron fluence energy distributions obtained for 5 different ^{241}Am-Be sources. The results are normalized by the total fluence rate at the measurement point.

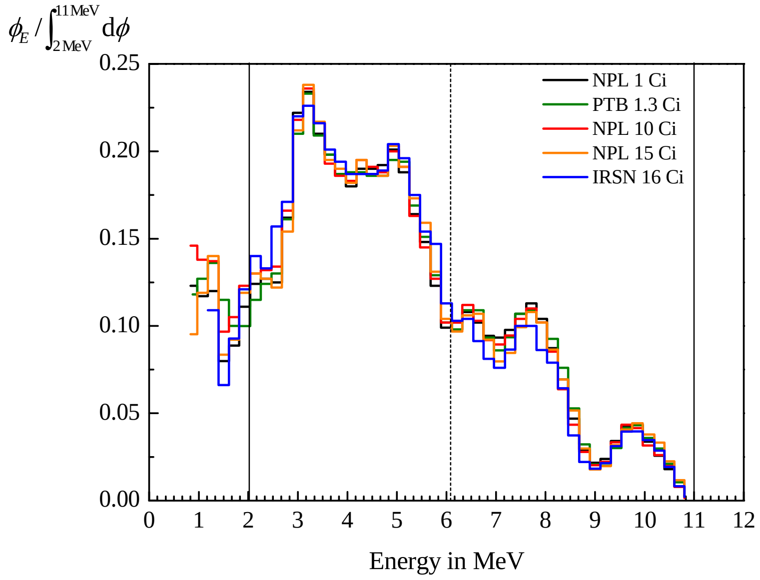

Figure 2: comparison of experimental neutron fluence energy distributions obtained for 5 different ^{241}Am-Be sources. The results are normalized by the fluence integrated between 2 and 11 MeV.

2.1.2 Source ^{137}Cs

As specified in the ISO 8529-2 standard in order 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) a 11 GBq ^{137}Cs source is being installed near the neutron source irradiator.

2.2 Simulated workplace neutron fields

The IRSN owns two facilities producing realistic mixed neutron-photon radiation fields, CANEL, an accelerator driven moderator modular device, and SIGMA, a graphite moderated americium-beryllium assembly. These fields are representative of some of those encountered at nuclear workplaces, and the corresponding facilities are designed and used for calibration of various instruments, like survey meters, personal dosimeters or spectrometric devices.

2.2.1 SIGMA

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}. The total neutron fluence rate at the position where calibration of instruments were usually performed, at 50 cm from the block, was 1767 cm^{-2}.s^{-1}, with a relative contribution of the neutrons of energies below 0.5 eV of 88.4%. Concerning the dose, the total neutron ambient dose equivalent rate was about 144 μSv.h^{-1} with a relative contribution of the thermal neutrons of about 44% [Lac04]. The occasion to improve the thermal components of the field was given since the ^{241}Am-Be sources

should be evacuated within the year 2007, due to the French regulations. A study, using Monte-Carlo simulations, aimed at determining the design of a new facility for the production of “pure” thermal neutron fields, based on IRSN existing facilities for minor investments [Lac06]. Two different configurations were investigated in details, one consisted in a ^{252}Cf source graphite moderated, the other one concerned a thermal field produced using the 400 kV accelerator coupled with the SIGMA graphite moderator assembly. These configurations were acceptable as reference thermal calibration field but the field produced with radionuclide source located at the centre of a moderator block of graphite of 240×240×240 cm^3 seemed more attractive for physical and convenient purposes.
During the year 2007, a ^{252}Cf source of 6.3 GBq (170 mCi) will be set-up in the nominal SIGMA graphite moderator block and the neutron field produced will be experimentally studied. This preliminary configuration will allow comparison with calculated data; the results from this preliminary configuration should help in the design of the final version of the new ^{252}Cf graphite moderated thermal field.

2.2.2 CANEL

An international measuring campaign, involving four laboratories with expertise in neutron metrology and spectrometry, was organized through a collaborative EUROMET project, n°670 in 2002 to characterize the CANEL neutron field. Some discrepancies between the experimental and calculated with MCNP results had been observed [Gre04]. One of the main hypotheses was related to the quantity of water contained in one of the moderator layers of the assembly [Lac05]. In 2005, CANEL was slightly modified by replacing the water lens by a 35 mm thick polyethylene plate. The new radiation field was characterized experimentally and by means of MCNP calculations. The preliminary results from Bonner spheres and proton recoil detectors are in agreement with the calculated neutron fluence energy distribution as shown in figure 3. Some discrepancies remain in the thermal energy domain.

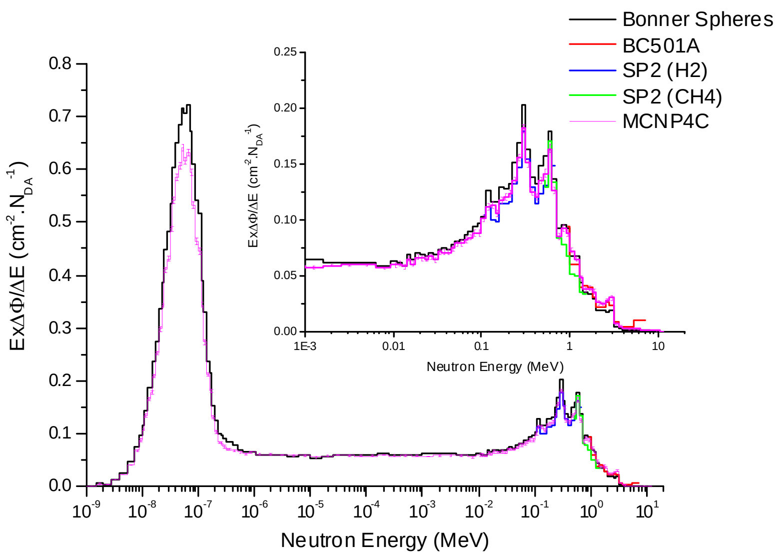

Figure 3 : Preliminary results of the neutron fluence energy distributions at 50 cm from the exit of the CANEL duct, from neutron spectrometers and MCNP calculations.

2.3 AMANDE : Monoenergetic neutron fields

2.3.1 AMANDE facility

AMANDE is IRSN new accelerator producing monoenergetic 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 [Gre04b]
Years 2005 and 2006 were dedicated to the accelerator facility equipment set-up as well as to the ion beam characterization in DC and pulsed mode.
From the latter study, it appears that AMANDE accelerator is able to accelerate protons and deuterons in an energy range going from 100 keV up to 4 MeV with an excellent energy resolution. The relative energy uncertainty is of 5×10^{-4} using a 90° magnet with a NMR teslameter, computer controlled slits with control loop on the accelerator high voltage and a set of two apertures. The ion beam energy spread is of 500 ± 80 eV and sub-nanosecond pulse widths have been measured in pulsed mode [Gre06b].

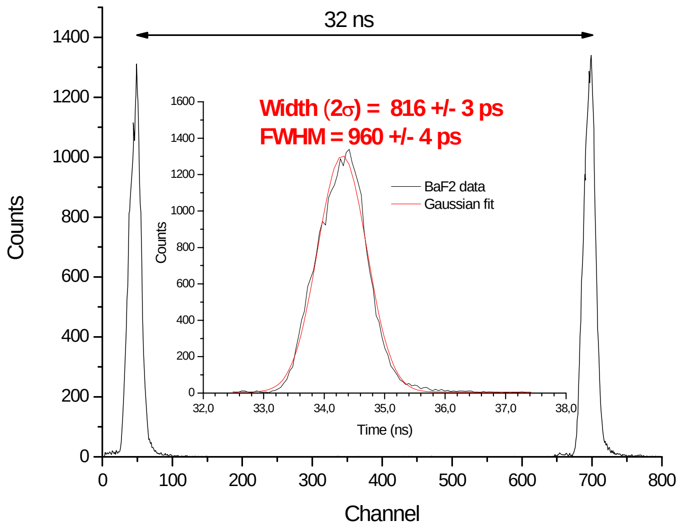

Figure 4 : Measurement of the pulse width at 2374 keV

In 2006, the neutron fluence energy distribution produced at all the ISO 8529-1 recommended energies above 100 keV have been measured by proton recoil spectrometers: two SP2 proton recoil proportional counters related to PTB neutron references in the energy range 50 keV – 2 MeV, and, a 2”x2” BC501A liquid scintillator above 800 keV. The neutron fluence is determined by this set of detector with an uncertainty between 3% and 5%. Relative fluence measurements are performed by the facility monitors, constituted by a second BC501A detector and two long counters, based upon ^{3}He counters inside De Pangher polyethylene shells.
Calibration of two passive Bonner sphere system has been performed in 2006 and since April 2007 the facility is open to users.

2.3.2 Reference long counter

Long counters are neutrons detectors whose main characteristics are a constant response over a relatively wide energy range (2 to 3 energy decades). A new system was studied and designed on geometry basis of existing long counters, using Monte-Carlo simulations. The intensive simulation work was undergone to optimize the geometry characteristics of a De Pangher long counter in the aim to obtain a quasi energy independent response function over almost 5 energy decades. In addition to the improvement of the response, the major interest to develop such a new device is to control all parameters in terms of dimensions, materials and components, in order to determine in a quite reliable way its neutron fluence response function. The preliminary calculated neutron fluence response function of the IRSN-PLC (new long counter) is shown in figure 1.

The new designed instrument will be considered as a standard system because its neutron fluence response function will be perfectly determined (from Monte-Carlo simulations) and in addition, a validation of the response will be performed at the standard neutron radioactive sources, ^{252}Cf and ^{241}AmBe, of the associated laboratory LNE-IRSN. This system will be used as standard neutron detector to determine the fluence at the mono-energetic neutron fields of the Amande facility.

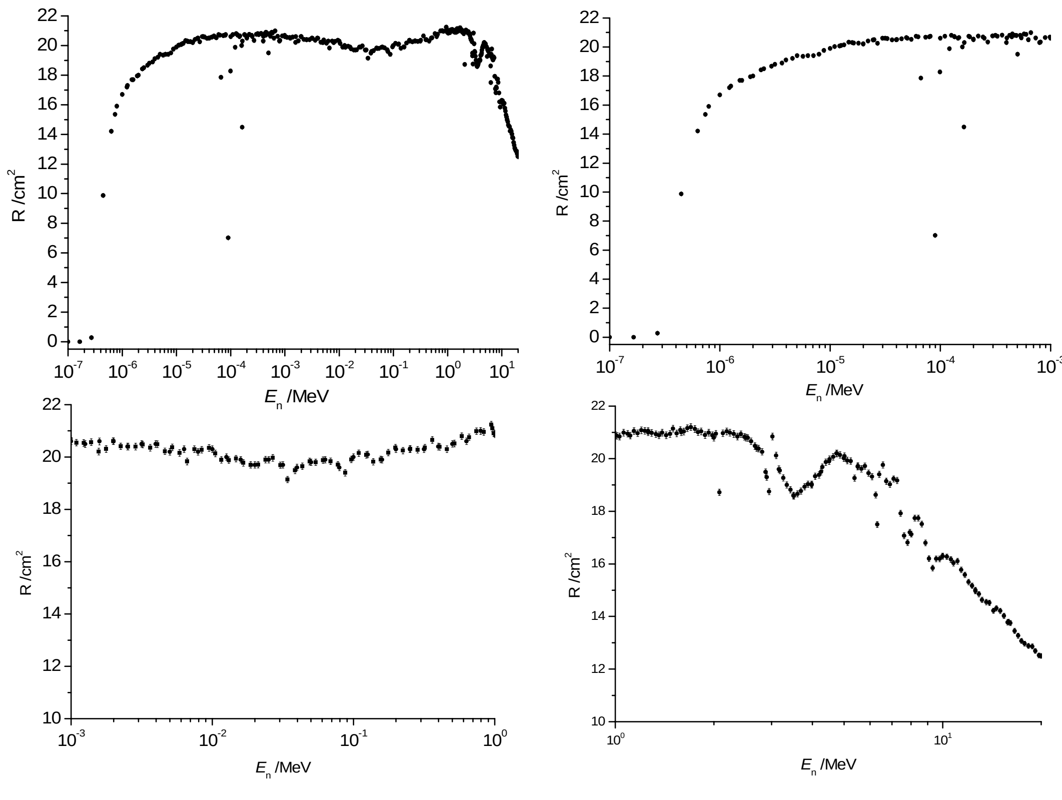

Figure 1 : MCNP calculated neutron fluence reponse function of the IRSN-PLC, represented over 0.1 eV and 20 MeV (top left), then detailed for three energy domains, 0.1 eV et 1 keV (top right), 1 keV and 1 MeV (bottom left) then 1 MeV and 20 MeV (bottom right) (preliminary results).

The IRSN-PLC will be made in June 2007. IRSN will participate to a comparison exercise of long counters fluence response, organized by the NPL end 2007.

3 Neutron Spectrometry

IRSN has several spectrometers used for fluence energy distribution characterization at workplaces (Bonner Spheres system) or at calibration facilities (3 spherical proton recoil counters SP2 and a liquid scintillator BC501A).

3.1 Bonner sphere system

The European standard IEC 60601-2-1 [IEC98], which recommends the determination of the secondary neutrons created by devices generating X-rays of more than 10 MeV has led the laboratory to extend its technical means for neutron spectrometry measurements in mixed neutron-photon radiation fields where the photon component predominates. A passive Bonner sphere system, based on activation of gold foils, was then developed in the laboratory [Amg06]. The fluence response functions of this system have been determined by Monte-Carlo simulations and validated at the AMANDE facility with mono-energetic reference neutron fields [Amg07]. This passive system allows measurements in environments where the active devices can not work properly due to pile-up, dead-time and CEM effects and perturbations.

3.2 Proton recoil response function simulation with MCNPX

Simulation with MCNPX of proton recoil counters (cylindrical or spherical) response matrices has been investigated. A method has been developed to take into account the electric fields effect into the counters using another code defining cells with specific gas amplification factors. Comparison with PTB SPHERE codes has shown some discrepancies mainly due to the wrong calculation in MCNPX of the stopping power in the gases for low energy protons [Bab06].
However, the use of MCNPX allows to take successfully into account the influence of high energy neutrons trough (n,p) reactions in the counter walls [Bab07].

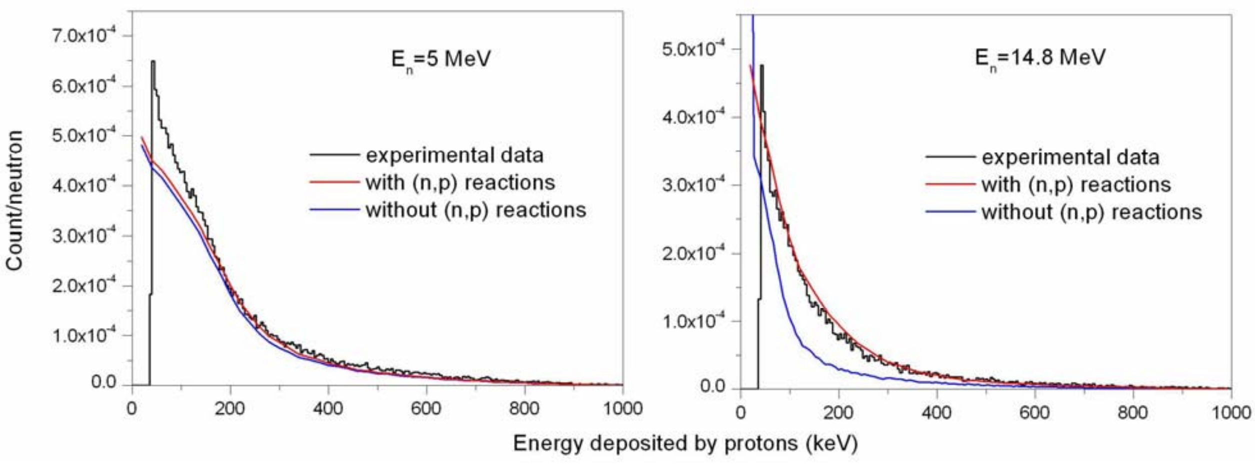

Figure 5 : Comparison between the MCNPX simulated response functions with and without (n,p) reactions in the counter walls and the experimental data obtained at the 5 MeV and 14.8 MeV AMANDE mono-energetic neutron field.

3.3 Digital electronic

IRSN uses a BC501A detector system to determine reference fluence energy distribution of neutron fields in the energy range between 1 and 20 MeV. The data acquisition chain associated to this detector consists of NIM standard electronic units which makes the system bulky and not easily movable. Recent developments in the field of numerical electronics offer the opportunity to use digital acquisition system which is compact, more convenient for measurements at workplaces and allows numerical data treatment.
Within the framework of collaboration between IRSN and the technological university of Cachan a new digital acquisition system was developed and tested with the BC501A. An Acqiris digital card with a data recording capability of 8 bits range at 4 GHz on two channels was chosen. Specific electronic and signal processing were developed to allow gain stability with high voltage feedback, and to obtain energy information with neutron/γ discrimination. The performances (energy resolution, threshold values, dead time and count rate) of the digital acquisition prototype were then compared with those of standard analogical acquisition. In the present state, the comparison results were not satisfying for metrology applications, but the use of new digital cards with 10 bits range is awaited to be fully competitive [Leb06b].

4 Status of the survey meter intercomparaison

This comparison is co-organised by IRSN with NPL and PTB institutes. The task of this exercise is to put a reference instrument at disposal of different primary laboratories participating at this exercise. Each participant had to calibrate this particular instrument in term of H*(10).

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)$_{/Cd}$.

Two survey meters were chosen as reference instrument:

- a spherical HARWELL monitor Mod. N91 n°70 provided by IRSN/LMDN and
- a cylindrical STUDSVIK monitor Mod. 2202 provided by NPL

The comparison exercise began in September 2003 but during one of the measurements in March 2004, it was observed an important deviation of the neutron sensitivity of the Studsvik with time. It takes more than one year to repair the Studsvick and due to the changes generated in the response of this survey-meter, it was necessary to do again all the comparison exercise. A new invitation has been sent middle 2005 and the participating laboratories are finally: SCK (Belgium), CMI (Czech Republic), PTB (Germany), NPL (United Kingdom), IEA (Poland), VNIIM (Russia), CIAE (China), KRISS (Korean), SMU (Slovakia) and IRSN (France).

This comparison should have been completed this year (IRSN, SCK, KRISS, CMI, IEA, SMU and NPL have performed their measurements), however some delay is awaited due to the fact that the Harwell N91 is facing a problem with its power supply.

5 Photon dosimetry

The knowledge of the photon dose contribution to the ambient dose equivalent in the neutron reference fields is of first importance for the calibration of photon sensitive neutron instruments.
Within the framework of a collaboration between Physikalisch-Technische Bundesanstalt (PTB) and IRSN, this photon dose contribution has been determined in the IRSN wide-spectrum neutron fields, i.e. in the two realistic mixed fields CANEL/T400 and SIGMA, as well as in the ^{241}Am-Be, ^{252}Cf and heavy water moderated ^{252}Cf radionuclide sources fields.
The measurements were performed with a calibrated PTB Geiger-Müller counter and were carried out with the bare detector and with the detector shielded by a cap which contained $^{6}Li_2CO_3$ to reduce the sensitivity of the detector to thermal neutrons. The sensitivity of the detector to fast neutrons was determined earlier and used to correct the measured readings.
The relative spectral photon distributions, required to convert the nominal value of air kerma to ambient dose equivalent, were calculated by IRSN using MCNP and measured with IRSN BGO and BC501 detectors, without photon induced by neutrons corrections, in a limited energy range (above 100 keV and 500 keV respectively).
The results showed that the photon contribution to the total ambient dose equivalent are of 3% for ^{241}Am-Be, 4% for ^{252}Cf, 9% for SIGMA, 12% for moderated ^{252}Cf and 20% for CANEL/T400 [Lan06].

6 Future Works (2007-2009)

6.1 AMANDE

In 2007, the commissioning of the pulsed mode is one of the major scientific programs of the facility.
The pulsed mode is used to perform time of flight measurements dedicated to the measure in an absolute way of the neutron energy of the monoenergetic fields.
The pulsed performances of the accelerator should be tested in the whole ion beam energy range, then time of flight measurements should be performed in the whole neutron energy range.
Neutron time of flight measurements will be performed using different devices depending on the energy. For instance, a BC501A liquid scintillator will be used for energy neutrons above 1 MeV. Below 1 MeV a new device as lithium glass or plastic scintillator has to be investigated.

In addition to the reference long counter, IRSN will use a cylindrical gaseous proportional counter, with neutron-photon discrimination, in order to determine the reference neutron fluence energy distribution between a few keV up to 2 MeV.
With these two instruments, low energy neutron fields produced with scandium or copper will be investigated.

A new BC501A liquid scintillator of smaller size will complete the reference detector set in order to be placed at the irradiation position with lower count rates than with the present BC501A leading sometimes to non-reasonable dead times.

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 will be done coupling MCNP and TARGET code within an interface allowing fast MCNP geometry modification of the detector transport system. Simulation will be compared in 2008 to experimental data from measurements performed with IRSN reference neutron spectrometers at several positions into the experimental hall.

6.2 Localization nucleus recoil detector system

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. These calibrations performed at PTB facilities can not be assumed as an absolute and direct way.
In order to avoid as most as possible these drawbacks, a new detector system is being developed. Based on the concept of recoil proton telescope system, the new instrument converts neutrons to recoil nucleus by elastic scattering and then measures the energy and angle of the recoil nucleus. The neutron energy is indeed directly related to the recoil nucleus energy. Recoil proton telescope systems are well known and used in several standard national laboratories but the efficiency of this kind of system is often very low (10^{-5}). The energy resolution depends on the solid angle of the recoil nucleus detector, leading to small detecting surfaces and to a compromise with the efficiency.
The aim of IRSN project is to develop a system with a higher efficiency, using a localization detector system. By this way it should be possible to increase the solid angle of detection and maintain a precise energy and fluence determination. This project is the aim of a PhD position supported by the LNE.

Two main solutions are investigated as function of the energy range. At high energy solution is to use localization detectors based on Si component associated to a neutron-proton polyethylene converter, to develop this project LMDN is in collaboration with CNRS-Ramses unit (radioprotection field), at low energy the solution investigated is to use a gaseous µ-time projection chamber (TPC). In this kind of detector the gas is used converter and allows to measure deposited energy as well as to track their direction. The energy range of application of such system is from around keV to 8 MeV depending on the nature and pressure of the gas, increasing even in the low energy direction the range of the proton recoil detectors presently used (SP2, BC501A). Hydrogen and ^{4}He are the most relevant gas for this

use. To develop the µ-TPC project LMDN has started collaboration with CNRS-MIMAC-He3 unit (Non baryonic dark matter research field).
An important work of simulation is in progress with MCNPX or GEANT4 codes for the two kinds of detectors (Si or µTPC). The determination of detectors efficiency, estimation of performances in energy and fluence measurements, estimation of uncertainties, choice of the best nature and pressure of the gas as function of the neutron energy range are essential steps for detector systems dedicated for neutron metrology. Experiments in front of AMANDE facility, already scheduled, will allow the validation in real condition of µ-TPC and localisation recoil proton telescope system performances in term of tracks and energy reconstruction.

6.3 Thermal field

A detailed experimental characterization of the neutron field produced by the modified SIGMA facility will be perform, by means of different neutron spectrometers (Bonner spheres, proton recoil detectors) and the thermal neutron fluence will be estimated with activation of gold foils [Gre05].
IRSN will take part to the comparison exercise of the thermal neutron fields, organized within the framework of the CCRI.

6.4 Bonner spheres

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 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 is being performed at in the framework of a PhD, of three years duration.

6.5 Quality control

In the two future years, intensive works will be performed in order to develop CMC’s for all neutron fields produced by IRSN.
This work will mainly be done following the ISO-17025 standard and will require several studies for reference traceability, uncertainty budgets and participations to international comparisons at CCRI or RMO level.

6.6 Am-Be energy distribution

The low energy part (below 1.65 MeV) of IRSN ^{241}Am-Be will be investigated with other spectrometers as Bonners spheres and/or SP2 spherical proportional counters.

6.7 Photon dosimetry and spectrometry

For photon spectrometry in mixed fields, IRSN intend to study the use of BGO detector as this detector allows to reach energies below 50 keV. A full work of characterization will be undertaken: the response matrix will be calculated using EGS4 or MCNPX codes.

Concerning the liquid scintillator BC501A, the response matrix of the photons induced by neutrons in the counters will have to be calculated following the calculation combination between MCNP and PhResp previously studied by T. Novotny at PTB.

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