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

V.Gressier, L. Van Ryckeghem, B. Asselineau, R. Babut, J.F. Guerre-Chaley, V. Lacoste, L.Lebreton, A. Martin, H. Muller, G. Pelcot, J.L. Pochat

1 Changes in French metrology and IRSN organization

1.1 Re-organization of French metrology

The "Bureau National de Métrologie" BNM, which was French NMI does not exist anymore. The metrology activity is now performed by the LNE (Laboratoire National de métrologie et d'Essai) new french NMI. The National Laboratory Henri Becquerel (LNE-LNHB) is still the metrological institute for ionizing radiations. Concerning the neutron metrology, LNE-LNHB is in charge of the reference values related to the source activities whereas IRSN is in charge of the other quantities. IRSN, in that way, is "associated laboratory" to the LNE.

1.2 Re-organization of IRSN

Following IRSN internal re-organization, all IRSN neutron metrology activities are performed by the Laboratory of Neutron Dosimetry and Metrology (LMDN) of the External Dosimetry Department (SDE) of the Radiological Protection and Human Health Division (DRPH).

2 Progress since 2003

The only facility where IRSN proposed CMCs is the irradiator with radioactive sources AmBe and 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. Calibration using the neutron fields produced by all the other facilities will be integrated in a near future in the CMC list.

2.1 Radionuclide sources

2.1.1 AmBe source

The neutron energy distribution of the IRSN standard ^{241}AmBe(α,n) source was measured using a proton recoil liquid scintillator, BC501A, above 1.65 MeV [1]. The experimental data were compared to the recommended by ISO-8529 standard neutron energy distribution for an AmBe source and some significant discrepancies were observed as shown in Figure 1. Monte-Carlo simulations were then performed to investigate on the neutron source term in order to consider the different characteristics between the IRSN AmBe source and the one used to establish the neutron emission spectrum recommended by the ISO standard. The variation of the characteristics of the source did not explain the remaining discrepancies. A good agreement with the experimental results was observed when the theoretical neutron energy distribution from Geiger and Van der Zwan [2] was introduced in the study as new source term, as shown in Figure 1. These investigations showed that the ISO recommended AmBe distribution might not be well suited to represent the neutron energy distribution of all AmBe sources, and that the manufacturing of the sources might play a major role in the neutron fluence energy distribution. The ISO standard 8529-2 recommends to introduce a 4% uncertainty in spectrum averaged values of fluence-to-dose equivalent conversion coefficient. This study allowed to estimate more precisely this deviation due to this spectrum variation effect: 2,6% in the case of the IRSN source and for neutron energies above 1.65 MeV (energy domain which accounts for 85% of the total H*(10) value).

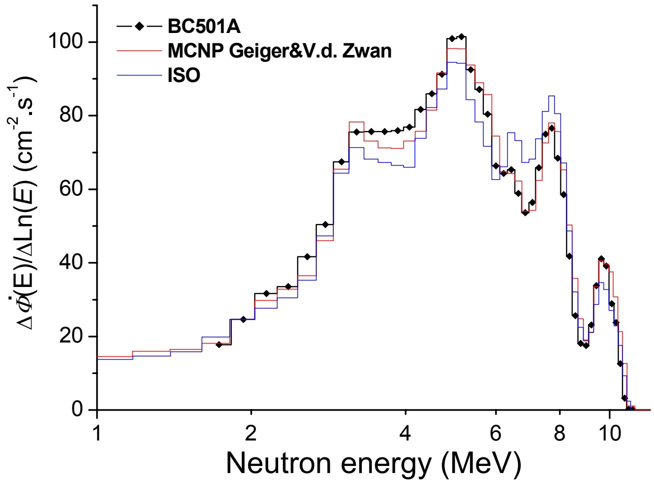

Figure 1. Experimental, calculated and reference neutron energy fluence distributions (the experimental measurements were performed at 150 cm from the source)

2.1.2 ^{252}Cf

The LMDN received a new ^{252}Cf source in April 2005 to replace the old one. The neutron activity has been determined by LNE-LNHB manganese bath method and the isotopic composition has been provided by the manufacturer. Neutron spectrometry with liquid scintillator and Bonner spheres will be performed and compared to MCNP calculation.

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

SIGMA consists of six Am-Be radioactive sources located in a 1.5 x 1.5 x 1.5 m^3 graphite moderator block. The SIGMA neutron field at the calibration position, situated at 50 cm from the west surface of the assembly, has been characterized over the whole energy range from thermal to 10.88 MeV [3]. The true thermal fluence was derived by two methods, one based on the activation of foils, the other using MCNP-4C Monte-Carlo simulation, leading to really quite comparable values. In addition, these two approaches allowed to determine the effective temperature of the neutrons in a large graphite block. At this temperature of 316.3 ± 5.0 K, the reference value for the true maxwellian fluence rate is 1513.2 ± 3% ($cm^{-2}.s^{-1}$).

Proton recoil spectrometers, in particular the standard BC501A scintillator, were used to measure the fast neutron fluence and deduce the AmBe sources density parameter for the Monte-Carlo simulation. Once the calculated fast neutron energy distribution adjusted to the proton recoil spectrum (Figure 2-b), a perfect agreement was observed between the BONNER SPHERE SYSTEM and MCNP4C neutron energy distributions above 0.5 eV (Figure 2-a). The agreement between all spectrometers results allows to consider also a global 3% uncertainty in the intermediate and fast energy domains.

The new characterization of the SIGMA neutron field makes this facility available for accurate calibration measurements of dosimeters and other measuring devices. In 2006, the 6 AmBe sources will be removed and replaced by new ones.

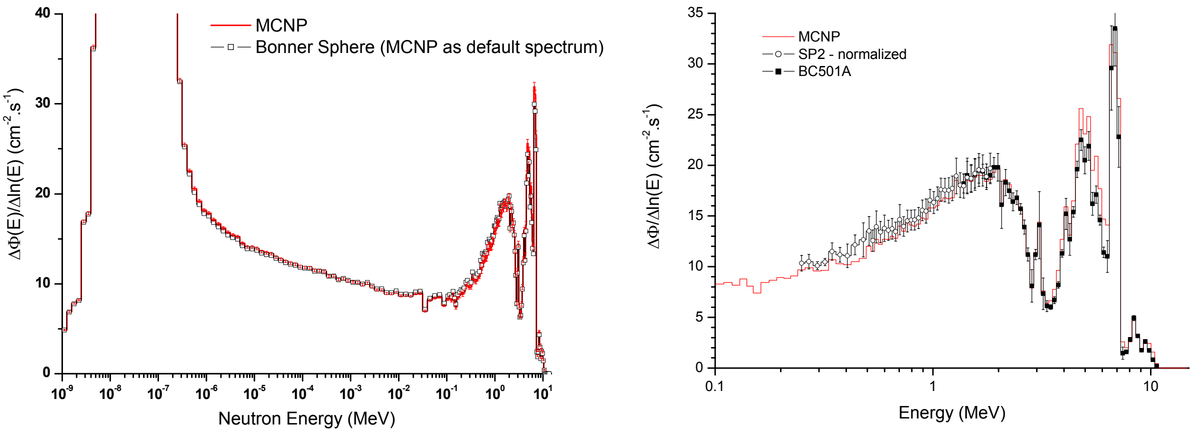

Figure 2. SIGMA fast neutron energy distribution measured with Bonner Sphere, liquid scintillator BC501A and SP2 proton recoil detectors and MCNP calculation for a 0.207 $g.cm^{-3}$ AmBe density.

2.2.2 CANEL

A new CANEL geometry has been recently designed, called CANEL/T400. This consists of a ^{238}U converter generating, from 3 MeV mono-energetic neutrons, a fission neutron spectrum which in turn is moderated by layers of iron and water placed inside a polyethylene duct.

An international measuring campaign, involving four laboratories with expertise in neutron metrology and spectrometry, was organized through a collaborative EUROMET project, n°670, to characterize the CANEL neutron field. The participating institutes are the National Physical Laboratory (NPL – United

Kingdom), the Physikalisch-Technische Bundesanstalt (PTB – Germany), the Autonomous University of Barcelona (UAB - Spain) and the IRSN [4].
Beside this campaign, intensive MCNP4C simulations were performed to determine the features of this neutron field [5].
During this campaign, four characterized Bonner sphere systems were used as well as proton recoil proportional spectrometers to provide detailed information in the energy region above 90 keV. A very good agreement was found between all the results of the measuring devices, but some discrepancies were observed in the fluence per monitor count compared to the MCNP calculation as shown in Figure 3.

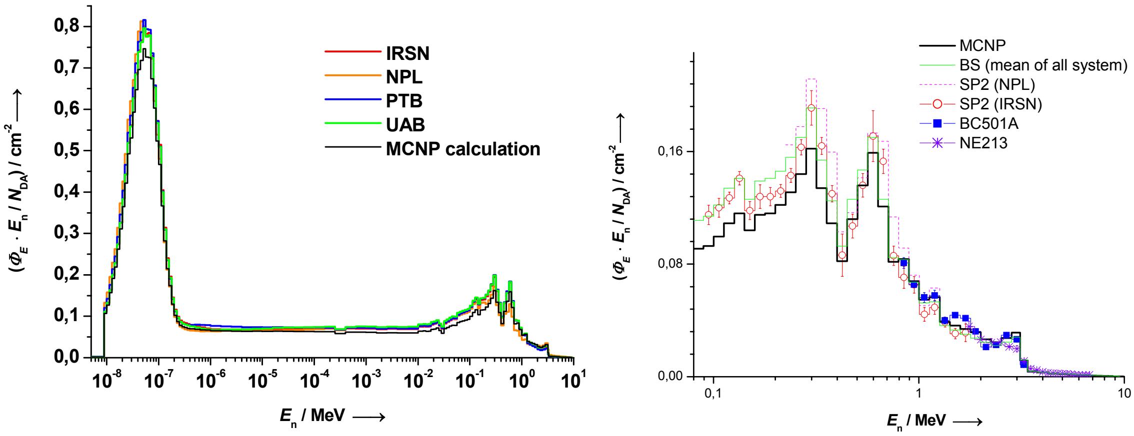

Figure 3. Left : Neutron energy distribution from MAXED unfolded Bonner sphere data and from MCNP calculation. Right : Comparison of the fast neutron fluence energy distribution, per monitor count, from the proton recoil detectors, Bonner sphere and MCNP.

The general shape of the neutron fluence energy distribution was validated but few discrepancies appeared in the intermediate and high energy regions, where neutrons play major role in dosimetric quantities. One of the main hypotheses was related to the quantity of water; therefore, the steel lens containing the water was dismounted to verify its dimensions as well as the volume of water. About 10% volume was missing, due to a leakage through a welding. Monte-Carlo simulation were further performed taking into account the measured volume of water, leading to a quite good agreement with the EUROMET experimental neutron spectrum in the intermediate and high energy regions, as shown in figure 1. The 10% discrepancy in the neutron fluence below 0.5 eV observed before the update of the geometry, was not fully explained, but reduced to ~7%. Concerning the ambient dose equivalent values, the new simulated result was in quite good agreement with the EUROMET reference value, due to the minor role played by the thermal neutrons in the dosimetric quantities. The updated geometry was then further used for the computation of the Hp(10).

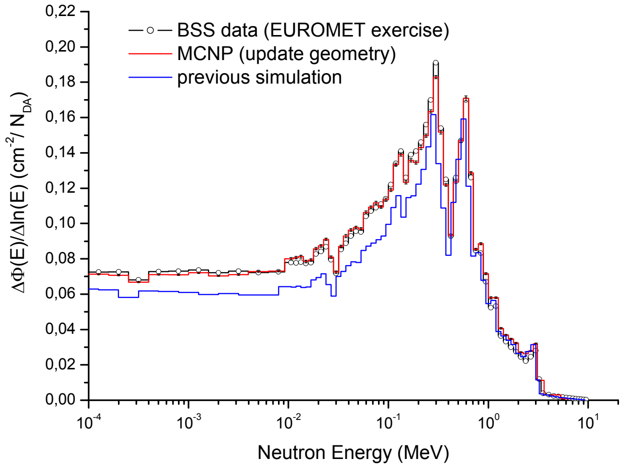

Figure 4. Neutron fluence energy distribution at CANEL/T400, from Bonner sphere measurements (average of four Bonner sphere systems results obtained during the EUROMET inter-comparison) and from MCNP simulations (updated geometry, with less water in the lens).

2.2.3 Hp(10) calculation

In the framework of the European project EVIDOS, irradiations of personal dosimeters were performed at CANEL and SIGMA. Monte-Carlo calculations were undergone to estimate the reference values of the personal dose equivalent at both facilities. The Hp(10) values were calculated for three different angular positions, 0°, 45° and 75°, of an ICRU slab phantom located at the position of irradiations [6].

To check the validity of the geometry and parameters introduced in the simulation input files, the calculated neutron fluence energy distribution had been beforehand compared to experimental reference spectrometry results. The calculated results can be considered as the most accurate values of the personal dose equivalent at these facilities since the use of Monte-Carlo code allowed to consider all the neutrons from any directions as well as the photons from neutron reactions.

Table 1: MCNP4C calculated values of the personal dose equivalent at CANEL/T400 and SIGMA, for three angular positions of a ICRU slab phantom at irradiation position.

	CANEL/T400	**SIGMA**
Angle (θ^{o})	$Hp(10,\theta)$ ($pSv.N_{DA}^{-1}$)	$Hp(10,\theta)$ ($\mu Sv.h^{-1}$)
0	120.2 ± 7.2	131.9 ± 4.7
45	97.9 ± 6.9	104.5 ± 3.3
75	42.2 ± 3.4	59.8 ± 1.9

2.3 AMANDE : Monoenergetic neutron fields

2.3.1 AMANDE facility

IRSN has acquired a new accelerator (named AMANDE as Accelerator for Metrology and Neutron Applications in External Dosimetry) to produce monoenergetic neutron fields within the energy range from about 2 keV up to 20 MeV. The neutrons will be created using the nuclear reactions between accelerated protons, deuterons and

thin targets like scandium, lithium, deuterium, tritium, as defined by ISO 8529 standard. The AMANDE facility is based on a 2 MV HVEE Tandetron accelerator, which has been installed end of 2004 in a new building.
Protons or deuterons are accelerated in the energy range 100 keV – 4 MeV, in a DC or in a pulsed mode, with an excellent energy resolution lower than 200 eV in DC mode (as shown during the installation test of the accelerator) and 4 keV awaited in pulsed mode. This latter mode will be used for time of flight experiments in open geometry.
AMANDE is housed in a new building consisting in two parts, one in concrete containing mainly the accelerator and control rooms, the other one being a 20 m x 20 m x 16 m experimental hall with metallic walls and a floor grating placed at 6 m above the ground over all the hall surface at the exception of a 6 meter-radius hole (the experimental area).
The neutrons are produced by nuclear interaction of accelerated charged particles (protons or deuterons) on thin targets of scandium, lithium (or lithium fluorite), deuterium or tritium in titanium. The targets are placed at the end of the beamline (7.2 m from the concrete floor) on a wobbling target holder with an air cooling system.
A full automated transport system place the detectors at any distance between 0,5 and 6 m and angle between -150° and +150° from the neutron-producing target. This system allows a ±180° rotation of the vertical axis of the detector holder for the study of the response of the devices with the direction of the neutrons. The precision on the position of the detector induces uncertainty on the neutron fluence to less than 0,1%.

2.3.2 Reference values

Energy

The mean energy of the monoenergetic neutron fields will be determined by two approach. First, the charged particle beam energy is well known from its calibration with nuclear reactions and several control loops at the deviation magnets guarantying an excellent energy stability. From kinematics and Target code, a calculated neutron energy distribution is obtained. This calculation is compared with time of flight measurements. These measurements are performed at distances from 0,5 up to 10 m depending on the neutron energy. As during the performance test of December 2004, pulse width of 0,8 ns for a 2 MeV proton beam has been obtained, an energy resolution lower than 1% could be achieved over almost the whole energy range.
For time of flight measurements, a 2”x2” BC501A liquid scintillators will be used above 1 MeV, and a plastic scintillator still to be determined at lower energies.

Fluence

In 2005 and 2006, the neutron fluence and energy distribution will be determined in the energy range 50 keV – 2 MeV by the use of two SP2 proton recoil proportional counters related to PTB neutron references. Above 800 keV, the BC501A liquid scintillators will be used. The neutron fluence will be determined by this set of detectors with an uncertainty of 3%.
In a near future (2007), a new designed long counter will be use to determine the reference fluence values over the whole energy range and a cylindrical

proportional counter will replace the SP2 counters, with a neutron-gamma discrimination allowing to measure down to a few keV.
Relative fluence measurements will be 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.

2.3.3 Scattered neutrons

MCNP-4C calculations have been performed to study for each "ISO-energy" and at several distances, the awaited integral scattered neutron contributions to fluence and dose equivalent of the neutrons scattered by the building (including in details all the elements present into the experimental hall, at the exception of the detector holder) alone and completed with the elements around the target (target backing, target holder).
In average, the scattered by building neutrons represents respectively 50 %, 70% and 90% of the total scattered neutron fluence or dose equivalent at 1 m, 2 m and 5 m. This study also showed that for distance from the target greater than 1 m, the use of metallic walls reduces significantly the background due to neutrons scattered by the building. At smaller distances, the scattering in the target holder and backing will dominate [7].

3 Neutron Spectrometry

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

3.1 Bonner sphere system

IRSN Bonner sphere system is based on the ortho-cylindrical ^{3}He proportional counter, type 0.5NH 1/1KI, manufactured by Eurisys Mesures. The total effective volume of the detector is 0.749 cm^{-3} where the ^{3}He gas is at a pressure of 800 kPa.
12 polyethylene spheres are available with diameter of 2.5" to 12". The five smallest spheres (up to 5'') are usually used bare and covered with a 1 mm thick cadmium shield to assess the thermal neutron fluence. The MCNP Monte-Carlo code was used to determine the response energy distribution of each polyethylene sphere. The Bonner sphere system was then characterized at mono-energetic and thermal neutron fields [8]. Measurements were done at the physikalisch-technische bundesanstalt (PTB) and national physical laboratory (NPL) standard laboratories, and at the newly characterized IRSN "SIGMA" facility.
The reference neutron fluence energy distributions were then folded with the response functions for comparison purposes with the experimental data. In almost all mono-energetic neutron field cases, a good agreement between experimental and calculated count rates was found (250 keV and 14.8 MeV were not considered as some variation of the neutron fluence per monitor count has been observed during the measurements at PTB), and some few percent discrepancies were observed in the thermal region. A correction was applied to the IRSN response functions to get an unfolded neutron spectrum in agreement with the reference

SIGMA neutron distribution. However, the investigation of the thermal part of the response function has still to be done.
This Bonner sphere system has been used during the EUROMET 670 exercise at CANEL and the results in fluence and dose equivalent were quite comparable to the other Bonner sphere systems (see Figure 3. left). At this time, the IRSN Bonner sphere system is used to provide spectrometric reference values for the EVIDOS measuring campaign [9].

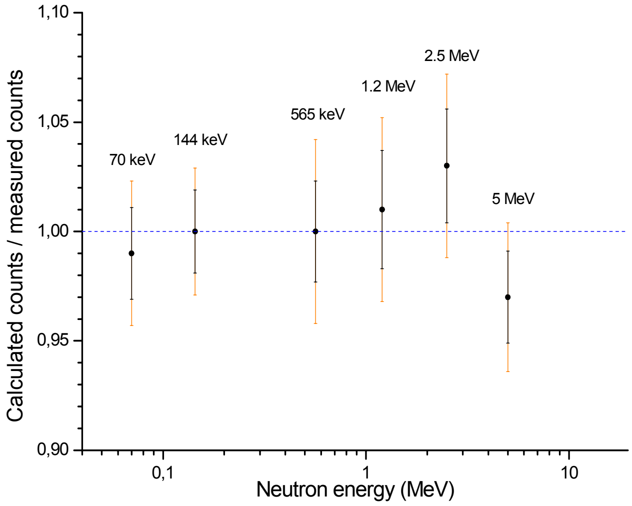

Figure 5. Mean ratio between the calculated and measured counts at standard monoenergetic neutron fields. The first uncertainty is the standard deviation of the ratio distribution on all the spheres, the second one is the quadratic sum with the uncertainty on the reference fluence.

3.2 Proton recoil response function simulation with MCNPX

Calculations with MCNPX of the proton recoil counters (cylindrical or spherical) response matrices are being investigated [10]. Comparison with PTB SPHERE and GNSR codes has shown some discrepancies mainly due to the choice of the stopping power in the gases. This new method of calculation will use a detailed description of the counters including wall and electric fields effect, and the influence of high energy neutrons trough (n,p) reactions in the wall.

3.3 Digital electronic

Complete digital electronic are being implemented on liquid scintillator BC501A. The performances that were obtained up to now are not so good than with standard analogical electronic but should be interesting in the future with the fast progress in this area.

4 Survey meter intercomparaison

Neutron survey instruments employed to determine ambient dose equivalent $H^*(10)$ at workplaces for radiation protection purposes are usually calibrated in the neutron field of easily accessible radionuclide sources: bare or D_2O-moderated ^{252}Cf source, or $^{241}Am/Be$ source.

Various European laboratories offer such calibration services, but a comparison exercise required for support of the quality management has never been organised, neither by CCRI(III) on international level nor by the technical committee for ionising radiation of EUROMET (TC-IR) on regional level. The main aim of the proposed supplementary comparison is to compare the calibration procedures the participating laboratories employ to determine the calibration factor N defined as

$$\mathbf{N = H^*(10) / M}$$

with: **H*(10)** the conventionally true value of the ambient dose equivalent as defined in 10 mm depth of the ICRU phantom [2], given in terms of [Sv]
M the instrument reading in counts, integrated and corrected for any disturbing effect, i.e. for dead time losses, non-linearity and scattered neutrons (room return neutrons)

The calibration factor N will be given in terms of [Sv] per detector count.

4.1 Transfer instrument to be calibrated

Two survey meters are chosen:

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

The two instruments, together with the electronic module needed for counting, is distributed by IRSN in sequence to all participants. After each calibration measurement the instruments shall be returned to IRSN for control measurements.

4.2 Partners

The participating laboratories are: SCK (Belgium), CMI (Czech Republic), PTB (Germany), NPL (United Kingdom), IAE (Poland), VNIIM (Russia), CIAE (China), KRISS (Korean), IRSN (France).

One month is allocated for each laboratory to perform all measurements. Including the time for transportation and the regular test measurements, two months is scheduled for each participating laboratory. In this way, the comparison may be carried out within two years after start.

4.3 Schedule

The comparison exercise began in September 2003 with measurements in the SCK, then to the CMI laboratory. But during the Russian measurements in VNIIM laboratory (St Petersburg, March 2004), it was observed an important deviation of the neutron sensitivity of the Studsvik with time. It was decided to send the device to the NPL for check tests and reparations. After a complete check and the

replacement of some electronic parts associated to the detector, a full new characterization was performed at NPL. All this work took more than one year and due to the changes generated in the response of the survey-meter, it was necessary to do again all the comparison exercise.
It will start again at the beginning of July 2005. A new invitation has been sent to all the participants (April 2005) in order to know their availability for the measurements in their respective laboratory. Some complementary tests are expected at IRSN during the June month before to send the two instruments to the first participant.
End of the exercise is now foreseen for end of 2007.

5 Future Works

Most of the future works should be performed through a scientific collaboration between NPL, PTB, UAB and IRSN.

5.1 AmBe energy distribution

In order to investigate in more details the AmBe energy distribution, international measurement campaign of several AmBe energy distribution will be performed. Beginning 2005, the PTB AmBe source energy distribution has been measured with IRSN BC501A liquid scintillator.

5.2 CANEL

The water leakage problems led to the replacement of the water lens by a 3.5 cm thick polyethylene plate. A new characterization both by experimental neutron spectrometry and MCNP simulation is being performed, as well as the study of the homogeneity in the calibration area.

5.3 Thermal energy range

There is a 7 % discrepancy between the thermal part of CANEL determined with the Bonner sphere (agreement between 4 systems used during EUROMET exercise) and the thermal part calculated with MCNP. These 7 % correspond to the bending of the calculated by MCNP response function of IRSN Bonner spheres in order to fit the SIGMA thermal energy distribution (below 0.5 eV). These discrepancies in the thermal energy ranges will be investigated by measurements performed with systems with different sensitivity to thermal neutrons at different reference thermal neutron fields.

5.4 Photon dosimetry and spectrometry

In order to determine the photon dose and energy distribution delivered by the neutron producing facilities, big efforts will be done in the future years in this domain.

Photon dose estimation at IRSN facilities (sources and simulated workplace neutron fields) will be performed by PTB in June 2005, using a well characterized Geiger-Müller counter.

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 code (and MCNPX may be used in addition for neutron sensitivity).
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.

5.5 AMANDE

The facility is now progressively starting, with a full characterization of the charged particles beam and neutron fields characterization in 2005-2006. To provide independent fluence references, IRSN will study a new design of long counter, optimizing with MCNP calculation the De Pangher geometry, to get the flattest response achievable between 2 keV and 20 MeV neutron energy. This detector should be available end of 2006 and will be calibrated first at the neutron reference fields of AmBe and ^{252}Cf.
To complete the set of reference detectors, a proton recoil telescope will be studied and designed. This study will be performed in the framework of a phD which should start in September 2005.
An international comparison at AMANDE is proposed for late 2007-2008.

5.6 Bonner spheres

In order to perform neutron spectrometry measurements in mixed n,γ fields where the photon component is high, for example at a 18 MV medical LINAC accelerator, the Bonner spheres will be used with passive detectors.

5.7 Digital electronic

Digital electronic will be implemented on proton recoil counters with neutron gamma discrimination possibility and also used for the time of flight measurements at AMANDE.

References

[1] N. Magalotti, V. Lacoste, L. Lebreton, V. Gressier, *Investigation of the neutron energu distribution of the IRSN ^{241}AmBe(α,n) source,* European workshop on individual monitoring of ionising radiation April 11 - 15, 2005, Vienna / Austria (to be published in RPD) (2005)

[2] K.W. Geiger, L. Van der Zwan, *Radioactive neutron source spectra from ^{9}Be(α,n) cross section data*, Nucl. Instrum. and Meth., 131, p 315-321 (1975)

[3] V. Lacoste, V. Gressier, H. Muller and L. Lebreton, *Characterisation of the IRSN graphite moderated Americium-Beryllium neutron field*, Radiation Protection Dosimetry 110 (1-4), p 135-139 (2004)

[4] V. Gressier, V. Lacoste, L. Lebreton, H. Muller, G. Pelcot, M. Bakali, F. Fernández, M. Tómas, N. J. Roberts, D. J. Thomas, M. Reginatto, B. Wiegel, and J. Wittstock, *Characterisation of the IRSN CANEL/T400 facility producing realistic neutron fields for calibration and test purposes*, Radiation Protection Dosimetry, 110 (1-4), p 523–527 (2004).

[5] V. Lacoste, V. Gressier, *Monte Carlo simulation of the IRSN CANEL/T400 realistic mixed neutron-photon radiation field,* Radiation Protection Dosimetry, 110 (1-4), p 123 – 127 (2004).

[6] V. Lacoste, V. Gressier, *Monte-Carlo simulation of the operational quantities at the realistic mixed neutron-photon radiation fields CANEL and SIGMA,* European workshop on individual monitoring of ionising radiation April 11 - 15, 2005, Vienna / Austria (to be published in RPD) (2005)

[7] V. Gressier, J. F. Guerre-Chaley, V. Lacoste, L. Lebreton, G. Pelcot, J. L. Pochat, T. Bolognese-Milstajn and D. Champion, *AMANDE: a new facility for monoenergetic neutron fields production between 2 keV and 20 MeV*, Radiation Protection Dosimetry, 110 (1-4), p 49–52 (2004).

[8] V. Lacoste, V. Gressier, J.-L. Pochat, F. Fernandez, M. Bakali and T. Bouassoule, *Characterization of Bonner sphere systems at monoenergetic and thermal neutron fields,* Radiation Protection Dosimetry, 110 (1-4), p 529–532 (2004).

[9] V. Lacoste, M. Reginatto, B. Asselineau, H. Muller, *Bonner Sphere Neutron Spectrometry at nuclear workplaces in the framework of EVIDOS project*, European workshop on individual monitoring of ionising radiation April 11 - 15, 2005, Vienna / Austria (to be published in RPD) (2005)

[10] R. Babut, *Simulation of the response energy distribution of cylindrical proton recoil proportional counter*, IRSN report SDE/04-57 (2004).