Recent Developments in Neutron Metrology at the National Physical Laboratory

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The activities of the Neutron Metrology Group (NMG) cover several technical areas, and a brief description of progress in each area is given below.

1 Radionuclide Source Based Fluence Standards

Development work has continued with the new manganese bath facility for neutron source emission rate measurements. The CAD model that was created as part of the design of the new facility was converted into an MCNPX model using the program MCAM (see Figure 1).

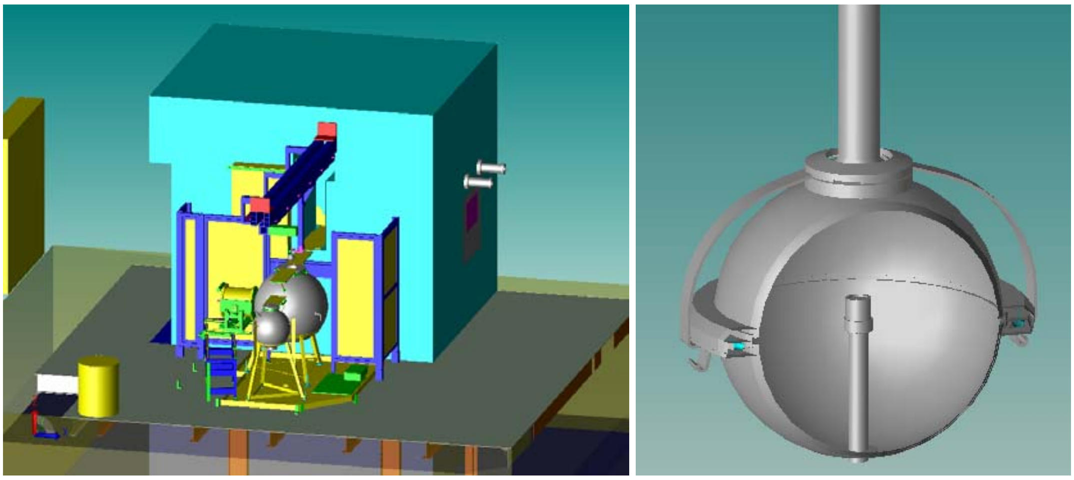

Figure 1: MCNPX model of manganese bath suite with blockhouse walls removed (left). Close-up of the source cavity, cut-away to show an X1 capsule inside (right)

The model was then used to make improved calculations of the correction factors and also to investigate the fraction of leakage neutrons that return to the bath. When considered together, the correction factors agreed well with those from an earlier model (see Table 1).

Table 1: Comparison of CAD model results with those from previous MCNP model ($N_H/N_{Mn} = 34.1922$, sulphur defined as 16000 not 16032)

	^{252}Cf, X1 capsule			^{241}Am-Be, X14 capsule		
	CAD	Previous	$\frac{\text{CAD}}{\text{Previous}} - 1$	CAD	Previous	$\frac{\text{CAD}}{\text{Previous}} - 1$
O & S capture loss - O	0.84%	0.83%	+0.3%	3.41%	3.41%	-0.05%
Source capture - S	1.85%	1.89%	-2.5%	2.16%	2.17%	-0.25%
Leakage - L	0.37%	0.35%	+7.7%	1.43%	1.35%	+6.0%
$\frac{1}{(1 - \text{O} - \text{S} - \text{L})}$	1.0316	1.0318	-0.02%	1.0752	1.0744	+0.08%

The leakage in particular is higher because the solution was a complete sphere in the previous model, whereas in the CAD model it is represented more realistically by including the upper planar surface which allows more neutrons to leak through the top of the bath.

The pre- and post-irradiation of the bath when a source is loaded into, or removed from, the bath was also calculated using the model (see Figure 2). The additional irradiation time is 0.4 s per loading or unloading which is much less than the estimated value in the previous facility of 12 s. The improvement is believed to be due to the improved source cell and the polyethylene shutter which is much better at shielding the bath from a source being loaded or unloaded from the cavity than was the case previously. This work was presented at NEUDOS11 and has been published in the proceedings in *Radiation Measurements*(1).

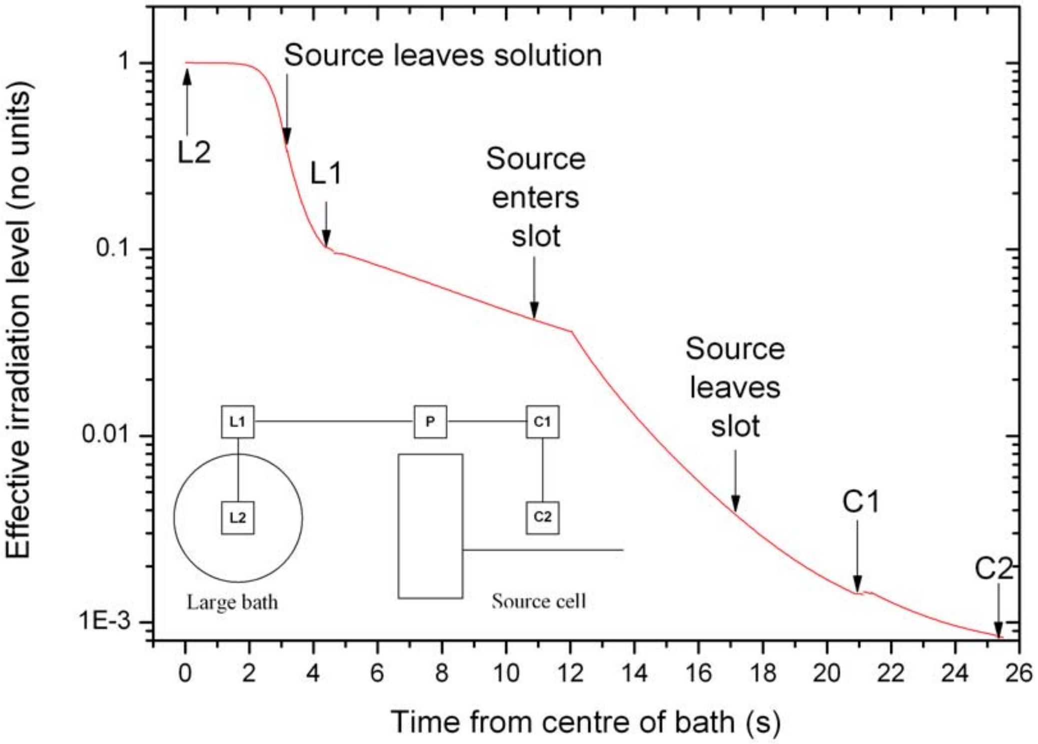

Figure 2: Effective irradiation level plotted against time from source leaving the centre of the bath

All the NaI detectors and electronics used in the manganese bath have been replaced with NIM-based systems. The emission rate of a 555 GBq (15 Ci) ^{241}Am-Be source was measured in both high and low efficiency channels to validate the new systems (see Figure 3).

A ^{226}Ra-Be photoneutron source (NASTRABE) is measured annually in the manganese bath as a stability check and until recently the variation in the measurements had always been within the random uncertainty. However, since the relocation of the bath in 2008 the emission rate of NASTRABE has shown a slow but steady decline (see Figure 4).

At the most recent measurement in November 2010 the rate was 1.3% lower than the mean rate before the relocation of the facility. The emission rates of all other sources have remained stable so either the emission rate of NASTRABE is genuinely decreasing or there is a problem with the manganese bath that only effects the ability to measure the emission rate of ^{226}Ra-Be photoneutron sources.

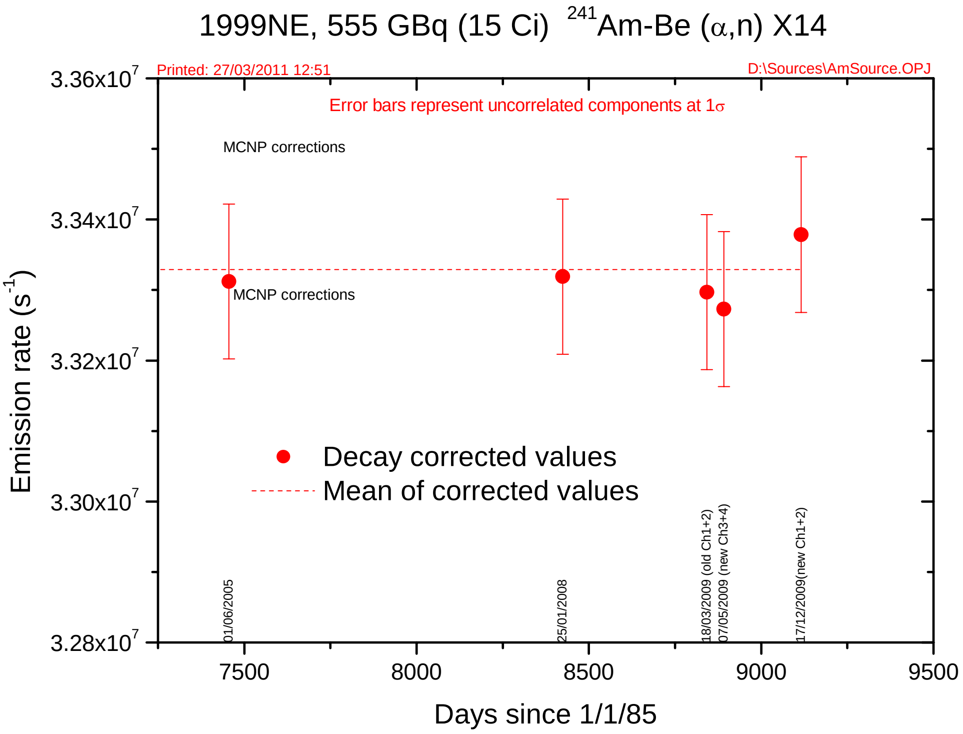

Figure 3: Emission rate measurements of 370 GBq ^{241}Am-Be source 1999NE made before and after the installation of new NaI detectors and electronics (error bars correspond to only the random uncertainty components at 1σ)

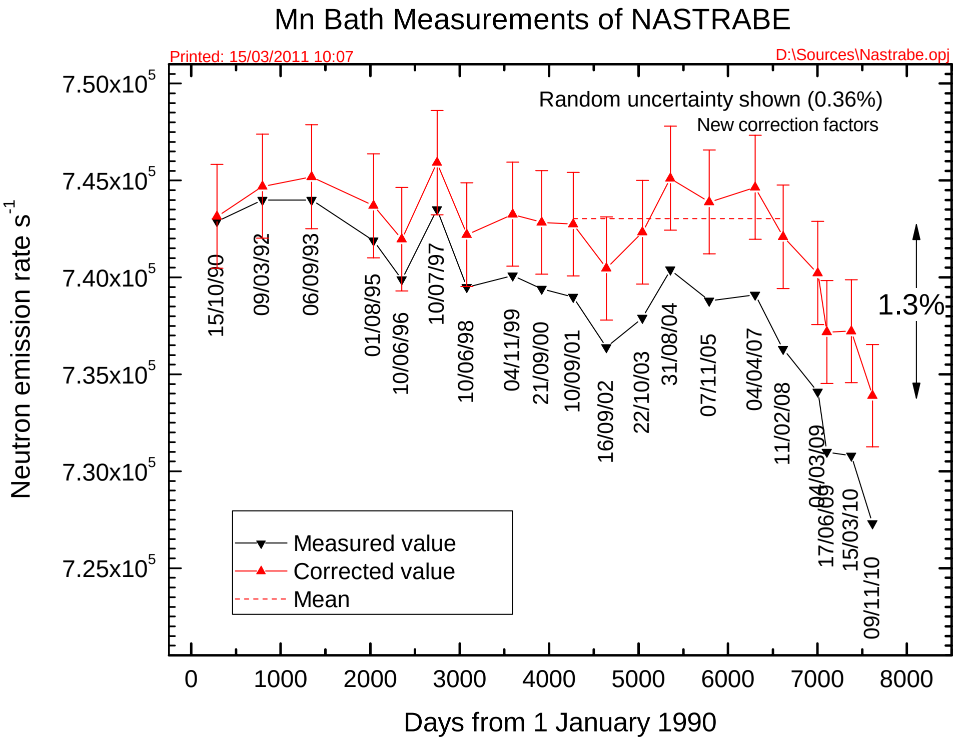

Figure 4: Annual measurements of the national standard ^{226}Ra-Be source, decay corrected to 1 January 1990 (error bars correspond to only the random uncertainty components at 1σ)

The source has the lowest neutron energy spectrum of any source routinely measured in the bath so the induced activity in the solution is far less distributed than for other sources. One possible explanation is that the mixing of the bath is not as effective as it once was leading to a reduced activity concentration passing through the loop to the NaI detector reservoirs. The emission rate of an ^{241}Am-Li source, which also has a low energy spectrum, was recently measured and found to be in excellent agreement with previous measurements.

In February 2011 the neutron group took receipt of a new high emission rate ^{252}Cf source, with a ^{252}Cf mass of just under 200 μg. The emission rate is approximately 4.6×10^8 s^{-1} which is a factor of 10 larger than the previous largest ^{252}Cf source at NPL. Apart from greatly speeding up irradiations, the high dose rates produced by the source open up work in the area of testing electronics for single event effects. A pneumatic transfer system has been designed which can fire the source from a storage cell to the low scatter area, and also to the manganese bath laboratory for regular re-calibrations.

2 Accelerator Based Neutron Fluence Standards

The ^{45}Sc(p,n) reaction has been studied extensively over recent years by the teams at IRSN, NPL, and PTB. It has now been used for customer calibrations including one at 3 keV.

Following the successful completion of EUROMET Project 822 ‘Comparison of Neutron Fluence Measurements for Neutron Energies of 15.5 MeV, 16 MeV, 17 MeV and 19 MeV’, monoenergetic neutron fluence standards at 17 MeV using the T(d,n) are now offered at NPL. More work needs to be done to characterise in detail the spectra from the NPL tritium targets. The primary monoenergetic fluence can be measured reasonably accurately, but the low energy component from scatter and unwanted reactions in the target still need to be quantified more accurately.

The time-of-flight system was used, in conjunction with the two-parameter data acquisition system recently installed in the accelerator control room, to measure time spectra from 5 MeV monoenergetic neutrons produced by the D(d,n) reaction.

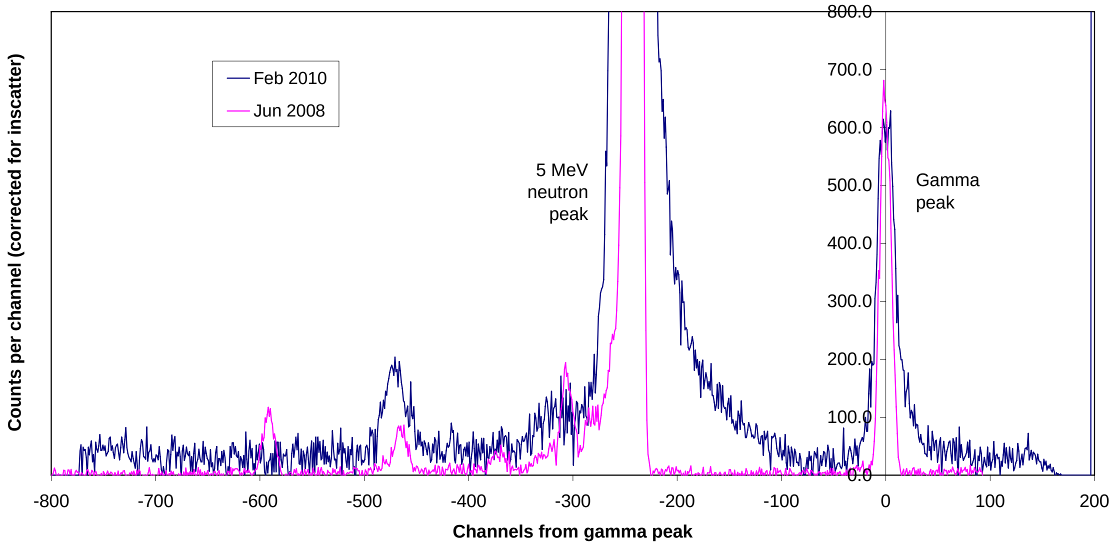

Figure 5. Time-of-flight spectra from 5 MeV monoenergetic neutrons, showing change in performance between June 2008 and February 2010. Note flight time decreases to the right.

The aim of this work was to investigate whether deuterium embedded in the target backing produces a significant fluence component at energies other than 5 MeV. However, problems were encountered with the consistency of the triggering from the beam pulse detector. These problems were similar to those observed in an earlier experiment in 2008, but of greater severity, as shown in Figure 1. (The timing spread is attributed to the trigger rather than to the neutron spectrum because the gamma peak, which should be narrow, is also affected.)

The problem is thought most likely to lie in the pulsing electronics in the top terminal of the Van de Graaff accelerator, and an investigation will be carried out during the 2011 shutdown scheduled for March/April.

Despite the poor timing resolution, a few small satellite peaks are evident in the timing spectrum, suggesting that the energy spectrum has some small components at other than 5 MeV. It is also interesting that the 2008 time spectrum has a peak at approximately -600 channels that is not present in the 2010 data. As the two spectra were acquired using the same target, it is unlikely that the peak is a genuine feature of the neutron spectrum. It could be explained as due to a spurious trigger that is significantly displaced in time from the genuine beam pulse but time-correlated with it.

Although build-up of a secondary target layer in the backing when bombarding deuterium targets with deuterons obviously occurs, there is no evidence of any increase in the fluence from these targets when they are only ever used for one neutron energy. The yield per unit monitor count, when using a current integrator as the monitor, has remained constant for the designated 5 MeV target since 1995 with a standard deviation of the readings of only 2.8%.

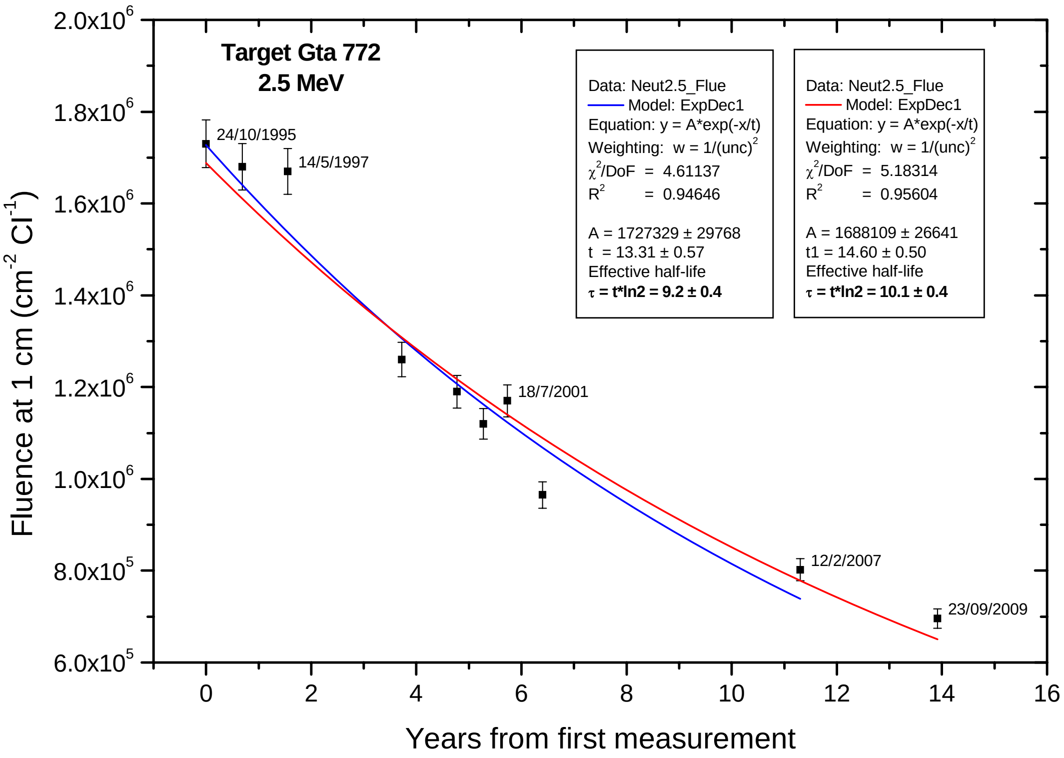

Figure 6. Yield of 2.5 MeV neutrons from a particular tritium in titanium target as a function of time.

The same is of course not true for the yield from a tritium target. This decays with a half-life which is usually shorter than the tritium half-life – see Figure 6. This is a well known phenomenon although the exact explanation is unclear. The obvious cause is the loss of tritium due to heating during bombardment, however, even during long runs of a many hours no visible drop in the yield is observed provided the usual precautions are taken, i.e. the beam current is kept low and is scanned on the target. From Figure 6 it is clear that the data do not fit the exponential decay curve particularly well, this may be due to inhomogeneity in the target layer with the beam striking different parts during different experiments. There is also a slight suggestion that the effective half life increases with time which would imply that the less well bound tritium is lost first and the more tightly bound tritium remains.

3 Thermal neutrons

Studies are underway investigating methods to boost the thermal neutron output from the NPL graphite pile, or to reduce the high energy component of the fields thus making them more truly thermal. The facility is accelerator driven and relies on high intensity deuteron beam currents from the 3.5 MV Van de Graaff – see Figure 7.

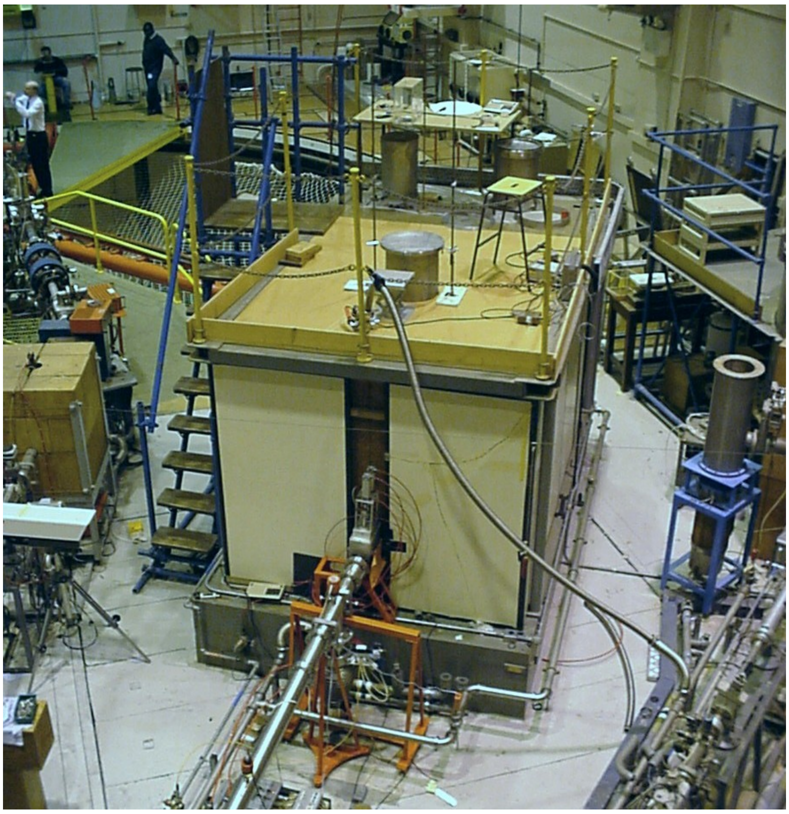

Figure 7. The NPL thermal pile

To date no real progress has been made on methods of boosting the field intensity (short of installing several high-power neutron generators around the pile). There has been some success, however, in identifying a method for reducing the high energy component, namely using protons rather than deuterons as the particles bombarding the beryllium targets in the pile. Although the neutron production per unit beam current of protons was measured to be roughly 20% that of deuterons, the fast neutron component from the proton irradiations was lower by a factor of two. The gamma field from the two different pile configurations was very similar for a given level of neutron production. Although the neutron production rates are considerably lower for proton irradiation of the thermal pile, it offers an interesting alternative for the calibration of sensitive neutron detection equipment since the higher beam currents needed allow finer control of the thermal neutron fluence rates and a smaller correction for the unwanted fast neutron component.

4 Comparisons and Demonstrations of Equivalence

The results of EURAMET project 936, a comparison of the performance of several long counters, were presented at NEUDOS11(2). The three participants in the exercise are IRSN, NPL, and PTB. Long counters from IRSN and PTB were brought to NPL in 2008 to compare with the two different long counters used at NPL. Measurements were performed with a range of radionuclide sources (^{241}Am-Be, ^{241}Am-B, ^{241}Am-F, ^{241}Am-Li and ^{252}Cf) and with monoenergetic neutrons (144 keV, 565 keV, 1.2 MeV, 5.0 MeV and 17 MeV).

The PTB and NPL De Pangher long counters was shown to be sufficiently alike that the NPL parameters can be applied to the PTB long counter although a 1.61% increase in the response needs to be made to allow for differences in the gas pressure of the central BF_3 tubes. Good agreement was obtained from the radionuclide source measurements, although they highlighted a possible normalisation issue with the NPL response calculations which should be resolved before the final report on the comparison is produced (see Figure 8). The monoenergetic results showed excellent agreement, particularly for the IRSN long counter which is an independently characterised instrument (see Figure 9).

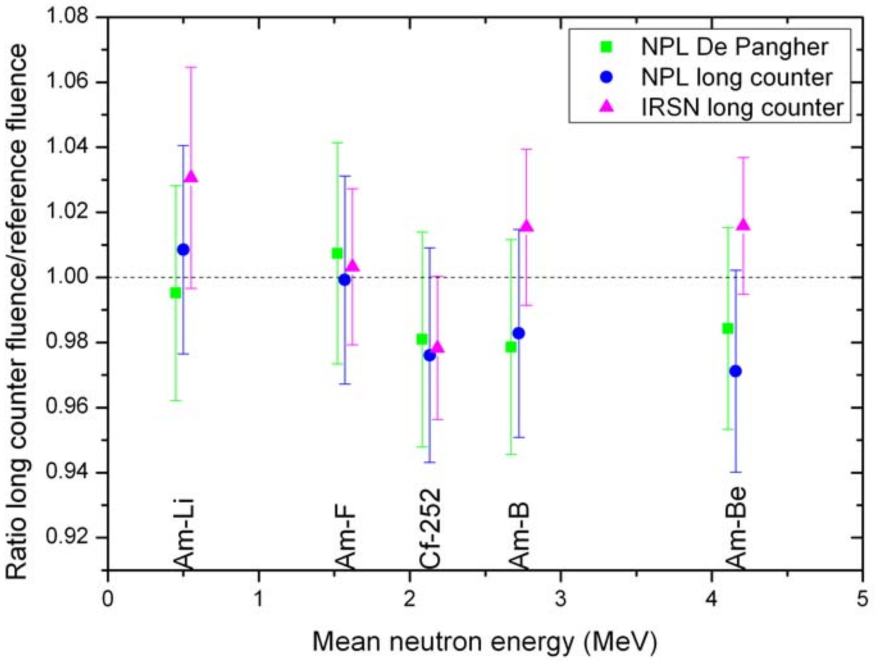

Figure 8. Ratio of participants' long counter fluences to the fluence determined from the emission rate and anisotropy factor for each radionuclide source (error bars represent the combined uncertainty at 1 σ)

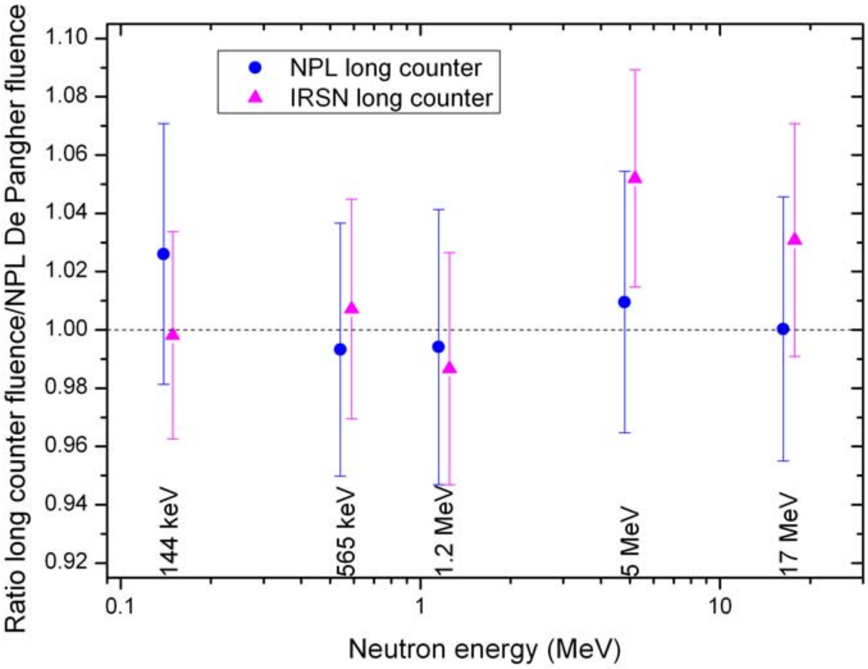

Figure 9. Ratio of participants' long counter fluences to the NPL De Pangher fluence for monoenergetic neutrons (error bars represent the combined uncertainty at 1 σ)

In February and March 2010 measurements were made at NPL as part of EURAMET project 1104: Comparison of the neutron spectra of reference neutron sources for the improvement of the ISO 8529 standard series. Representatives from INFN and UAB made Bonner sphere measurements of 37, 370, and 555 GBq (1, 10, and 15 Ci) ^{241}Am-Be sources, and also a ^{252}Cf source as a control (see **Figure 10**).

Figure 10. Setting up the UAB and INFN Bonner spheres in the low scatter facility at NPL

A preliminary analysis indicates that the spectrum from the NPL 37 GBq (1 Ci) ^{241}Am-Be source is completely consistent with the ISO 8529 spectrum, but those from the 370 GBq and 555 GBq (10 Ci and 15 Ci) ^{241}Am-Be sources have a greater component of lower energy neutrons than the ISO 8529 spectrum. This ties in with measurements made earlier by NPL on its sources.

5 Neutron Spectrometry

Over the years NPL has developed a number of neutron spectrometers including two Bonner sphere sets, one with an active central neutron detector and one which uses gold foils, a set of spherical hydrogen recoil counters, various scintillators, and a ^{3}He ionisation chamber spectrometer. These were developed both for both laboratory and field measurements.

The Bonner spheres are available for use and were in fact used in 2010 for measurements at a nuclear site. Some measurements were actually performed off-site where the neutron doses, although easily measurable with the Bonner spheres, were very low and a background subtraction had to be made. Rather surprisingly the background neutron dose rate at low altitudes (not much above sea level) has very rarely been measured. A number of mountain-top observatories have permanently mounted neutron spectrometers but ground level information is scarce.

The other devices available at NPL are high resolution spectrometers, and these are in various states of readiness for use. The hydrogen recoil counters are operational although their low efficiency tends to mean they can only be used in the laboratory. Following problems with the scintillator cell of the original NPL organic scintillator system new scintillator cells were acquired and these are being used primarily to investigate digital signal processing.

The ^{3}He spectrometer is operational, but concerns about the original response functions measured for the system when it was developed at Birmingham University have instigated some further investigation of this device. In particular codes to calculate the response matrix for this device are being investigated.

NPL has continued its investigations into digital signal processing (DSP) for organic liquid scintillators. One of the advantages of DSP is the potential to produce compact and portable analysis electronics for field use, but the sampling rate and resolution of such a device will inevitably differ from those available in non-portable lab-based units. It was therefore considered important to try a selection of neutron/gamma discrimination algorithms on such a portable unit, to assess their relative performance with data that are realistic for the intended application.

The compact digitiser used for this project was based on the TOM board(3), approximately 16 cm by 11 cm in size, produced by Hybrid Instruments. The ADC in this device takes up to 500 M samples / s, with up to 12 bits (reducing to 10.3 effective bits at maximum rate). For the purposes of this experiment, the on-board FPGA was programmed to send raw pulse shapes, comprising 128 samples taken at 2 ns intervals, to the host laptop by Ethernet link. The neutron detector was built from a Photonis XP2020 photomultiplier with transistorised divider chain, connected via a light guide to a 6 cm diameter by 6 cm MAB-1 cell of BC501A scintillator provided by Saint-Gobain. (See **Figure 11**.)

The detector was exposed to mixed neutron and gamma fields from three different sources: ^{241}Am-Be, ^{252}Cf, and D(d,n) reactions producing 5 MeV monoenergetic neutrons. In each case about 60000 events (about 50 MB) were recorded for off-line analysis. The three discrimination algorithms used in the assessment were Charge Comparison (comparing the pulse integrals during a long and short gate(4)), Pulse Gradient Analysis (based on the decrease in amplitude a fixed interval after the peak(5)), and a Model Pulse algorithm (where each pulse is compared to a model neutron and model gamma pulse, and identified according to which is the better match(6)). Discrimination performance was quantified in terms of a Figure of Merit (FoM), which was derived from the bi-modal probability distribution of the relevant discrimination parameter by measuring the separation of the peaks (one of which corresponds to neutrons and the other to gammas) and dividing by the sum of their widths.

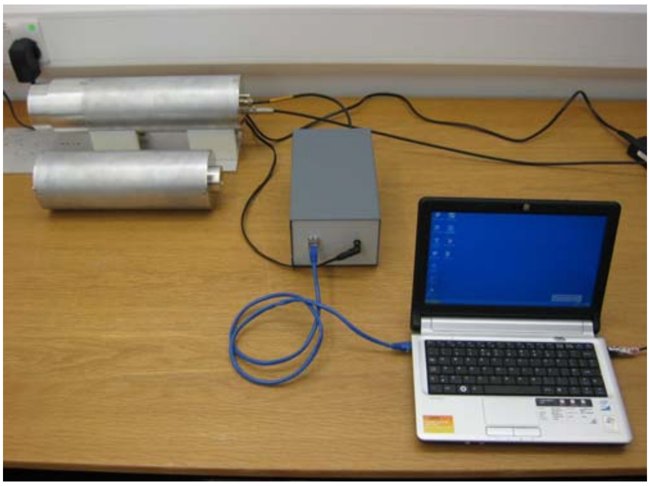

Figure 11. The hardware used in the study. Left, detectors (two shown but only the rearmost used here); centre, digitiser module; right, host laptop. The digitiser communicates with the laptop via an Ethernet link (blue cable).

Table 1. FoM values for optimised neutron / gamma discrimination algorithms (larger values indicate better discrimination).

Source	Charge Comparison	PGA	Model Pulse
^{241}Am/Be	1.13	1.10	0.93
^{252}Cf	1.38	1.01	0.93
5 MeV	1.35	0.91	0.98

The results are set out in Table 1, and show that the Charge Comparison algorithm has a small but measureable advantage. As it is also the simplest of the three, it is a strong candidate for incorporation into a compact digital scintillator system.

This work was presented at the NEUDOS-11 symposium in Cape Town in 2009, and has now been published in the proceedings(7).

6 Neutron Dosimetry

Work is continuing in the field of cosmic ray dosimetry. NPL is collaborating with SolarMetrics Ltd, QinetiQ, and a major US airline to fly a Hawk TEPC, a QinetiQ RaySure device and two Thermo EPDN-2s on a transpolar route roughly once every two days in the hope of recording the effects of a significant solar particle event. To date data has been collected on over 100 flights. Unfortunately the airline involved have yet to sign a contract to allow the project to be properly publicised or guarantee its continuation up to and beyond the next solar maximum, expected in the middle of 2013. Other avenues for flying instrumentation are being explored, as currently there is less than a 50% chance of instruments being airborne should such an event occur.

Work in the field of anisotropic neutron dosimetry is progressing. A Monte Carlo based feasibility study into the development of a directionally-sensitive neutron dosimeter was presented at the Neutron Dosimetry Symposium in Cape Town in 2009 and showed significant potential. Funding is being sought to start the process of developing a prototype device.

7 Major Facilities Maintenance and Development

Following replacement of the accelerator tube and the belt of the NPL 3.5 MV Van de Graaff in early 2009 the accelerator has worked very well with very few major breakdowns. The ion source presently in use providing substantial beam currents, in excess of 70 μA, down the central unanalysed beam. This is important for operating the thermal pile where high beam currents are needed to achieve the thermal fluence rates required by end users.

8 Future work

Work presently underway and proposed for the future include:

- Investigation of the corrections in activity foil fluence measurements
- Investigation of the possibilities of using organic diodes to measure neutron dose
- Measurement of photon doses in monoenergetic neutron fields,
- Develop high energy neutron standards, possibly in collaboration with ISIS,
- Extend digital signal processing to new devices, e.g. plastic scintillators,
- Develop a ‘white source’ (broad energy spectrum) time-of-flight system based on the Group’s 3.5 MV Van de Graaff accelerator, using either a thick Li - Al alloy target or a beryllium target a few millimetres thick.
- Continue to investigate directional effects in neutron personal dosimetry,
- Measure the effects of solar flares on cosmic ray doses during transpolar flights,
- Produce new simulated workplace fields.

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