Recent Developments in Neutron Metrology at the National Physical Laboratory

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The activities of the Neutron Metrology Group (NMG) at NPL fall into six topic areas: 1. Radionuclide Sources Based Standards, 2. Accelerator Based Neutron Fluence Standards, 3. Comparisons and Demonstrations of Equivalence, 4. Neutron Spectrometry, 5. Neutron Dosimetry, and 6. Major Facilities Maintenance and Development. This report concentrates on areas 1, 2, 3, and 6 which are of greater relevance than 4 and 5 to CCRI(III) activities.

1 Radionuclide Source Based Fluence Standards

Routine activities which fall under this heading include the maintenance and development of the manganese sulphate bath, and routine measurements of the radionuclide sources used to provide fluence standards at NPL.

The work to investigate the influence of ^{250}Cf in typical ^{252}Cf sources, outlined in a previous report to CCRI(III)[1], has been completed and the results presented in an NPL Report[2#]. Sources which are nominally ^{252}Cf in fact contain varying amounts of ^{250}Cf, and because of the longer half-life of this isotope, its influence increases with time, introducing problems if the emission rate of the source is calculated, at some time after a calibration, simply using the ^{252}Cf half-life. For very old sources there is the additional complication of the presence of ^{248}Cm produced as the daughter of ^{252}Cf although this is negligibly small for most sources.

Table 1. Measured original weight fractions for ^{250}Cf and ^{252}Cf in four NPL sources

Source identifier	Nominal weight at manufacture	Date of manufacture	^{250}Cf percentage by weight (%)	^{252}Cf percentage by weight (%)
CVN 10/54	10 μg	1971	15.8 ± 0.3	84.2 ± 0.3
4774 NC	1.7 mg	1982	16.9 ± 1.5	83.1 ± 1.1
4775 NC	100 μg	1982	17.5 ± 3.0	82.5 ± 2.2
4193 NC	10 μg	1988	18.8 ± 5.4	81.2 ± 4.2

Data on the typical fractions of ^{250}Cf and ^{252}Cf were derived from repeat measurements made for several NPL sources over a number of years (going back in one case to 1972). The typical fractions at the date of manufacture include in some cases uncertainties for the exact date of manufacture, but are all reasonably similar, as shown in Table 1. It should be noted that the weight fractions given in Table 1 add up to 100%, i.e. they refer only to ^{250}Cf and ^{252}Cf, and ignore other isotopes which either have long half-lives and small or zero spontaneous fission branches (^{249}Cf and ^{251}Cf), or have short half lives and small initial weights (^{253}Cf and ^{254}Cf), so that in all cases their presence is negligible in conventional sources. The values in Table 1 differ somewhat from the one set of figures made available by the manufacturer in 1987, for a particular batch of californium shortly after production, where the relative weights of ^{250}Cf and ^{252}Cf were reported to be 8.9% and 91.1% respectively.

Most NPL reports and some papers are available on the NPL website in pdf format.

Figure 1 gives a graphical representation of the percentage error which would arise in estimating the emission rate of a typical source, i.e. one with similar weight fractions to those shown in Table 1, if the ^{252}Cf half-life is used to correct for decay. Predictably the error is greater the longer the time since the last calibration, but it also depends strongly on the age of the source. A recommendation is made that the source emission rate is measured at regular intervals and the data fitted to a function involving the activities and half-lives of both ^{252}Cf and ^{250}Cf, and that this function is used to predict the emission rate as the source gets older.

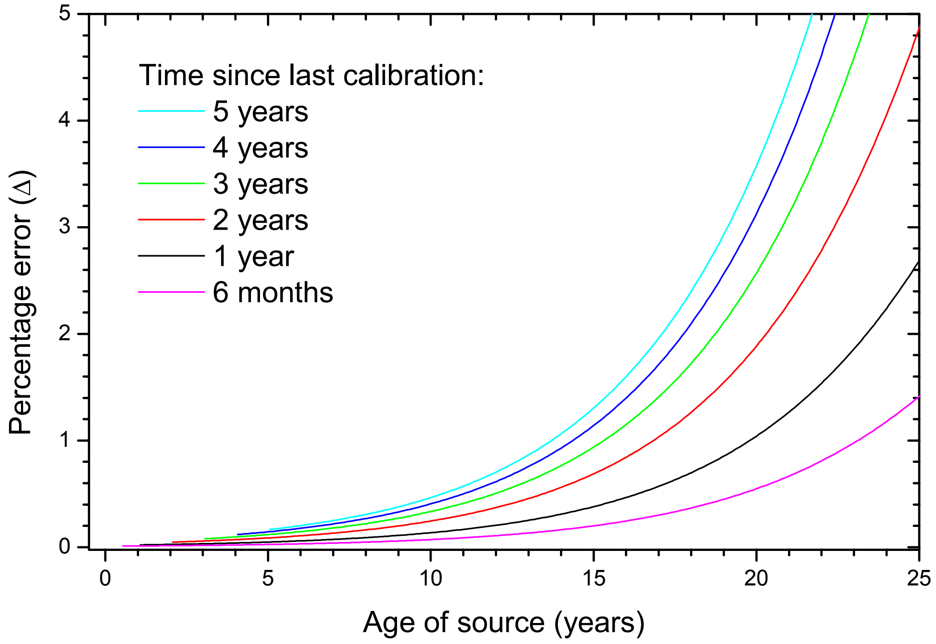

Figure 1. Percentage error in the neutron emission rate of a typical californium source when correcting for decay using the ^{252}Cf decay constant.

2 Accelerator Based Neutron Fluence Standards

The Neutron Metrology Group at NPL have a 3.5 MV Van de Graaff, and this is used to produce monoenergetic neutrons, thermal neutrons, and also to provide the primary neutrons for a simulated workplace field. The most significant developments over the last two years have been to knowledge of the effective centres of the long counters used to measure monoenergetic fluences, to knowledge of the thermal field characteristics, and to the characterisation and testing of the simulated workplace field (see section 5).

To use a long counter to measure monoenergetic neutron fluences two quantities need to be known; the efficiency and effective centre of the instrument, both as a function of energy. Following on from work to improve knowledge of the efficiency[3], efforts have been made recently to obtain better data on effective centres. Measurements of the effective centre are possible, by taking data with the instrument at a whole series of distances from a point source and fitting the results to a function of $(d + r)^{-2}$ where d is the distance from the source to a reference point on the long counter, and r is the unknown effective centre depth. However, these measurements tended to be inconsistent when repeated, and not to agree when done by different people or different institutes. A calculational approach was devised where an ideal experiment, conducted in vacuum, was simulated by performing MCNP calculations for a large number of source to detector distances and analysing the results as if they were experimental data. This has the advantage over real measurements of avoiding the need to correct for room and air scatter. Also, the calculations can be done for a large number of energies. The initial calculated results, however, showed large and unrealistic structures. These were eventually identified as being artefacts introduced into the approach by correlation effects between the calculations at different distances. When these were removed, by a random number approach to choosing the first random number, a much more reasonable curve was obtained for the effective centre as a function of energy. The only structures occur in places where they would be expected because of structure in the cross sections of the materials used in the construction of the long counters (mainly in carbon). The results have been published in an NPL Report[4], and are shown for a De Pangher long counter in Figure 2 together with experimental results from various sources and also the empirical curve, derived by Hunt in the late 1970s, and used for many years at NPL.

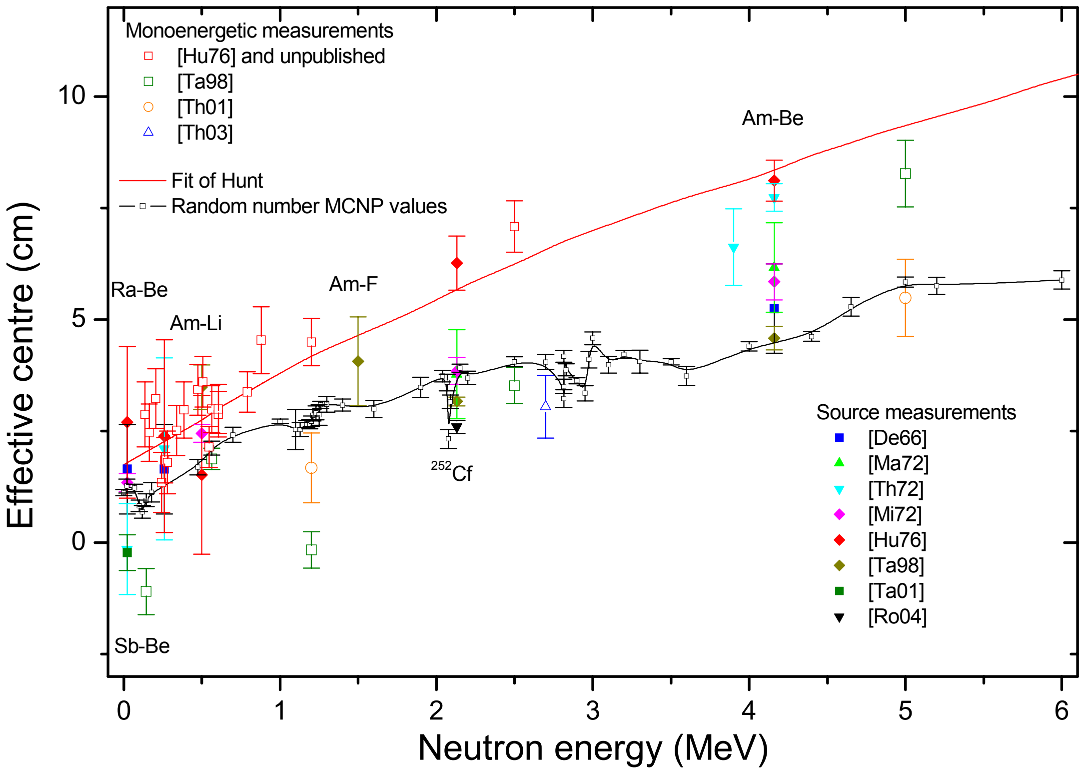

Figure 2. Effective center measurements for a De Pangher long counter compared with the empirical curve of Hunt and the calculations of Roberts et al[4]. All the monoenergetic data are from NPL, as are the source measurements of Hunt [Hu76], Tagziria [Ta01] and Roberts [Ro04].

It is clear from Figure 2 that the measured data, except for those of Hunt [Hu76] agree better with the new calculations than the old empirical fit curve. Nevertheless, the agreement is not as good as might be expected. Some of the monoenergetic results of Tagziria [Ta98] may be questionable because of the use of beam currents which were too high, but there are also other disagreements, for example for neutrons from an ^{241}Am-Be source. This should be one of the most straightforward measurements because of the constant and high source emission rate. An analysis, reported in reference 4, for a spherical device, where the effective centre should be at the centre of the sphere, casts some light on the potential problems of making the measurements. It only requires a rather small percentage error in the in-scatter correction at each distance for the effective centre result to be significantly distorted. Such small percentage errors are very likely when using shadow cones to perform these corrections because of the difficulty of setting up the cones to give perfect shadowing. These effects are unimportant for a single fluence measurement when the overall in-scatter correction is small, but may combine to distort the answer for an effective centre measurement.

The thermal neutron facility at NPL is based on primary neutron production by the d + Be reaction using a deuteron beam from the Van de Graaff accelerator. The use of two 'D' shaped targets, both of which are struck by the deuteron beam, allows neutrons to be produced at two places within a large graphite block where they are moderated to provide thermal fields. Thermal irradiations can be performed either in a hole at the centre of the block, or in a thermal beam extracted from an area within the block. Over recent years the beam has been used much more frequently than the central hole. Traditionally the thermal fluence quoted on certificates has been the Westcott fluence, nv_0, where n is the neutron density and v_0 a velocity of 2200 m s^{-1}. This quantity is well understood by reactor physicists and people concerned with foil activation experiments, but not by people requiring thermal neutron calibrations of dosemeters or survey instruments. The relationship between the Westcott fluence and the 'true' fluence, $n\bar{v}$, for the NPL thermal beam has been investigated using gold foil activation both bare and under cadmium cover. From this information fluence to dose equivalent conversion coefficients for the thermal beam have been derived[5].

A programme of measurements has been undertaken to investigate the optimum position for a shadow cone when performing scatter corrections. The final results of this work are being compiled as an NPL report.

3 Comparisons and Demonstrations of Equivalence

For the comparison exercise involving circulation of an ^{241}Am-Be source for measurements of total emission rate, NPL has made two measurement using the manganese bath technique. The results of these indicate that there have been no changes whatsoever in the source emission rate between the first and second measurements made in October 1999 and July 2004 respectively. A report on these measurements is almost complete. Measurements have now been made by all participants, [NPL(UK), VNIIM(Russia), CMI(Czech Republic), KRISS(Korea), CIAE(China), NIST(USA), IRD(Brazil), and LNHB(France)] although the Korean and Chinese labs have requested that they receive the source for a second time because of difficulties with the original measurements. The source is currently in Korea and is expected to be sent to China next month (May).

A brief report on NPL's experience as pilot laboratory for the recently abandoned CCRI thermal comparison exercise with ^{10}B ionisation chambers has been written and should be available soon. A Euromet comparison of the calibration of area survey instruments by European Labs is currently underway being organized mainly by IRSN with some assistance from NPL. The NPL measurements for the two instruments involved have been performed, although problems with one instrument (the Stydsvik 2202D) are delaying the exercise.

4 Neutron Spectrometry

NPL has a number of neutron spectrometers, developed mainly for characterising workplace fields, but also used for characterising calibration fields. They consist of: two Bonner sphere sets, one with a spherical ^{3}He proportional counter central sensor, and the other with gold foils as the central sensors; a set of three SP2 spherical hydrogen recoil counters (1, 3, and 9 atm.); two NE213 scintillators (one 6 cm diameter by 6 cm thick, and the other 2 cm diameter by 2 cm thick); and a ^{3}He gridded ion chamber.

During the last two years the two NE213 scintillators have had to be replaced because of severe degradation of the resolution. The replacement scintillators (BC 501A) are presently being characterised. An investigation of n/γ discrimination for hydrogen recoil spectrometers using digital signal processing techniques has shown some promise, although further work needs to be done to optimise the signal collection and processing[6]. An accident to the NPL 9 atm. SP2 hydrogen recoil counter has meant it had to be replaced. A new detector (called an SP10) has been acquired from Centronic and is presently being evaluated. New response function data for the gold foil based Bonner sphere set have been calculated using MCNP, and are presently being compared with the original response functions based on ANISN calculations and a very limited number of measurements.

Several of the spectrometers were used in the Euromet spectrometry exercise to characterise a simulated realistic field at the IRSN CANEL facility at Cadarache.

5 Neutron Dosimetry

The projects which fall under this heading are all aimed at improving measurements of dose equivalent for radiation protection. Work over the last two years has included a project to investigate the use of superheated drop (bubble) detectors - particularly in mixed fields, to investigate a dual instrument approach to improving area survey measurements – the two instruments being a conventional moderator-based instrument and a TEPC, and to improve understanding of TEPC detectors. The main area of application for TEPCs has been in cosmic ray dosimetry, and a large body of flight data has been accumulated, amounting now to over 800 commercial flights, including a significant number in the southern hemisphere a region where coverage has hitherto been lacking.

A simulated workplace field, designed to replicate the types of low-energy fields encountered around reactors, has been developed and characterised at NPL over recent years. Over the last year this facility has been used to investigate the responses of various devices including: neutron spectrometers, area survey instruments, and personal dosemeters. The survey instrument and dosemeter results exhibited readings that were up to a factor of 6 wrong. Some of the most interesting data were obtained using two commercial spectrometry systems, the ROSPEC and the Transportable Neutron Spectrometer (TNS). The initial results are shown graphically in Figure 3 and in numerical form in Table 2.

Ratio	Fluence (cm)	H*(10)
ROSPEC/NPL	1.55	1.13
TNS/NPL	0.90	0.97

Table 2. Results of spectrometric measurements at simulated workplace field facility

Two features of the spectrometric results which are evident from Figure 3 are the rather good agreement in the region from about 50 keV upwards, and the decidedly poor agreement at lower energies. The NPL spectrometric estimates are based on MCNP calculations and Bonner sphere measurements. The TNS employs spherical hydrogen recoil counters and a liquid scintillator in the region above 50 keV, while the ROSPEC employs similar hydrogen recoil counters and a methane recoil counter. Below 50 keV the TNS uses two BF_3 counters, one covered with a cadmium layer, while the ROSPEC uses two ^{3}He detectors one covered with a materiel containing boron as a thermal neutron absorber.

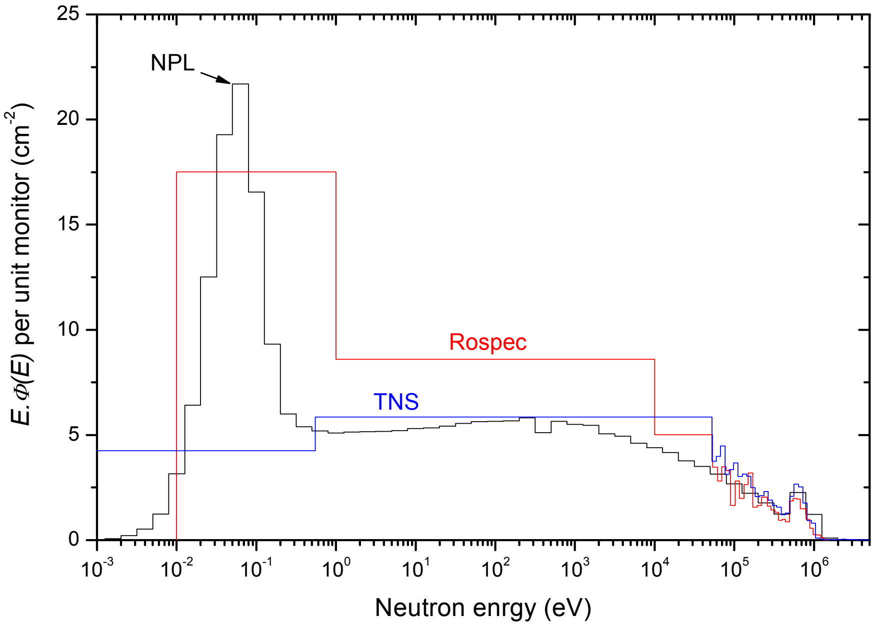

Figure 3. Results of spectrometric measurements with a ROSPEC and a Transportable Neutron Spectrometer (TNS) at the NPL simulated workplace field facility

Investigation of the results indicated possible problems with the angular dependence of the response of the TNS detectors used for the low energy part of the spectrum, and with the calibration of the detectors in the case of ROSPEC. On the basis of the simulated field results and of results from measurements of the thermal response of the ROSPEC elsewhere, the value was adjusted and a revised analysis made. The results are shown in Figure 4. The tests to date have highlighted the usefulness of the simulated field facility as a test bed for instrument performance in regions where good standard fields are difficult to provide.

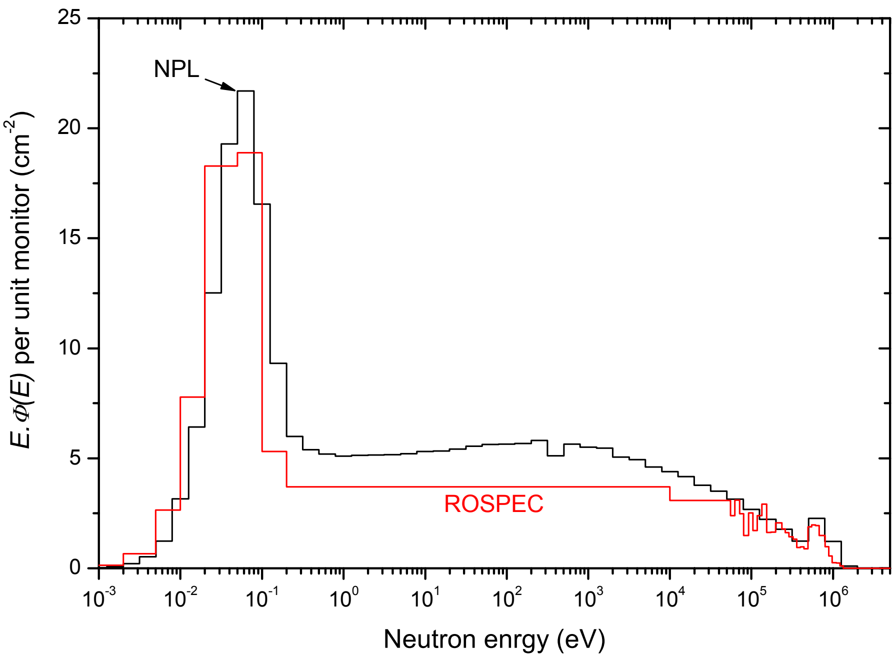

Figure 4. Revised analysis of ROSPEC measurements of simulated workplace field.

6 Major Facilities Maintenance and Development

Projects under this heading involve the maintenance and development of the Van de Graaff accelerator and measurement facilities. The main development in this area has been the reinstallation of the pulser system in the Van de Graaff. This has been a fairly major undertaking since the pulser has not been used for the last 15 years. The characteristics of the system are an un-bunched pulse width of about 12 ns, which is reduced to about 3 ns on bunching. The basic frequency, F, is 2.5 MHz (400 ns between pulses), although lower frequencies, F/2, F/4, F/8 etc down to F/128 are available in principle. A first time-of-flight measurement was performed last year and a programme of development and utilisation of this capability will commence shortly.

Future work

Work has just started on projects to:

- Investigate photon components in standard monoenergetic and radionuclide neutron fields.
- Investigate target characteristics, such as thickness, to improve knowledge of the mean neutron energy for monoenergetic fields and the neutron energy width.
- Provide further information on variations in the thermal fields in the NPL beam facility at different points in the beam.
- Produce standardised neutron fields in the energy region between 15 and 20 MeV.

References

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6 N. P. Hawkes, *Pulse Shape Discrimination in Hydrogen Proportional Counters by Digital Methods*, DQL-RN 007, 2005.