Recent Developments in Neutron Metrology at the National Physical Laboratory

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The activities of the Neutron Metrology Group (NMG) cover several of technical areas, and a brief description of progress in each area is given below. The most significant developments over the last two years have been the move of the manganese bath into a new purpose built and greatly improved laboratory, and the increased demand for thermal neutron calibrations.

1 Radionuclide Source Based Fluence Standards

The new manganese bath facility for neutron source emission rate measurements was completed in 2007, with the first sources calibrated in early 2008. The new facility is a monumental improvement on the old one in terms of safety (radiation protection), security, working environment and general ease-of-use (see Figure 1).

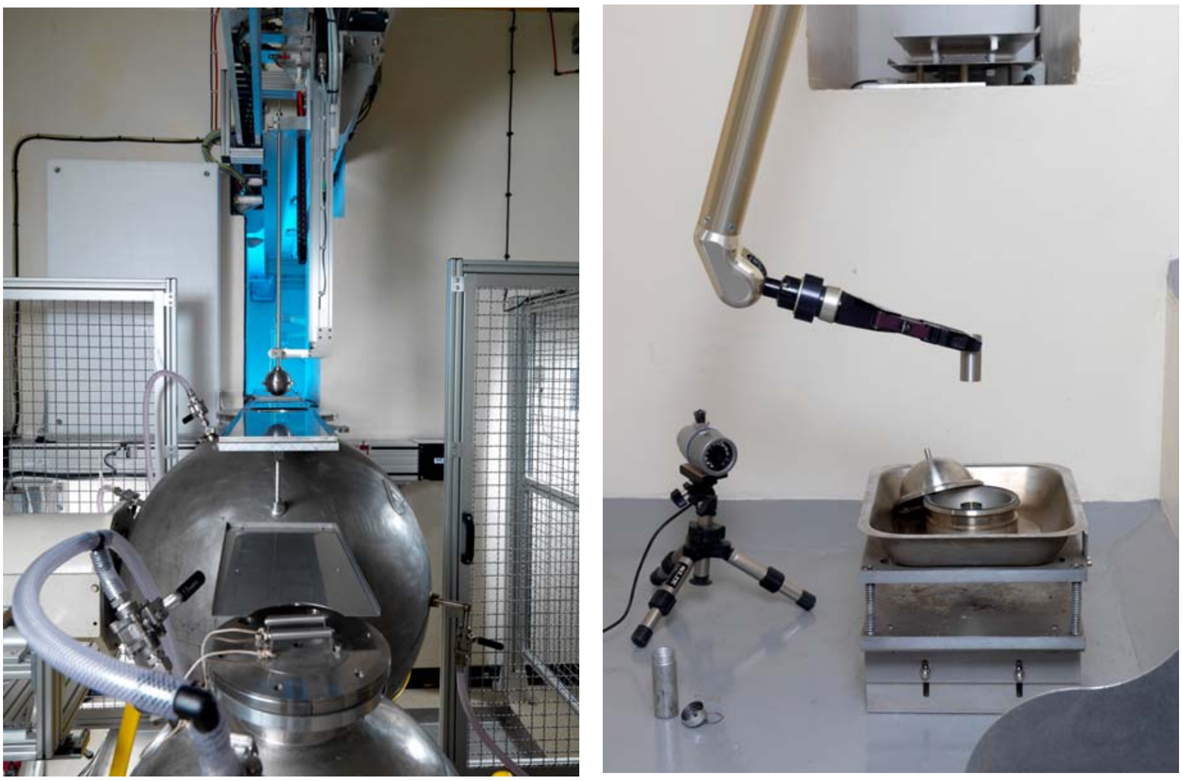

Figure 1: The source sphere being transferred into the large manganese bath (left), and source being loaded into the sphere using the manipulators in the source cell (right).

A series of measurements were performed to characterise the new facility with regards to the detection efficiency, solution concentration, impurities and effective flow rate. These showed only very minor changes from the old facility. To validate the new facility, emission rate

* Guest worker from China, started August 2008.

measurements of the National Standard ^{226}Ra-Be(γ,n) source and a 1 Ci ^{241}Am-Be source were made immediately before and immediately after the relocation. Comparing the results with the means of measurements made with the same sources going back almost 20 years shows that the small differences observed are well within the estimated uncertainties (see Figure 2). Further measurements were made with a 15 Ci ^{241}Am-Be source in order to validate the low efficiency counting channels. These demonstrated that the two channels agree to well within the uncorrelated uncertainties between them. Further details can be found in NPL Report IR 11[(1)#].

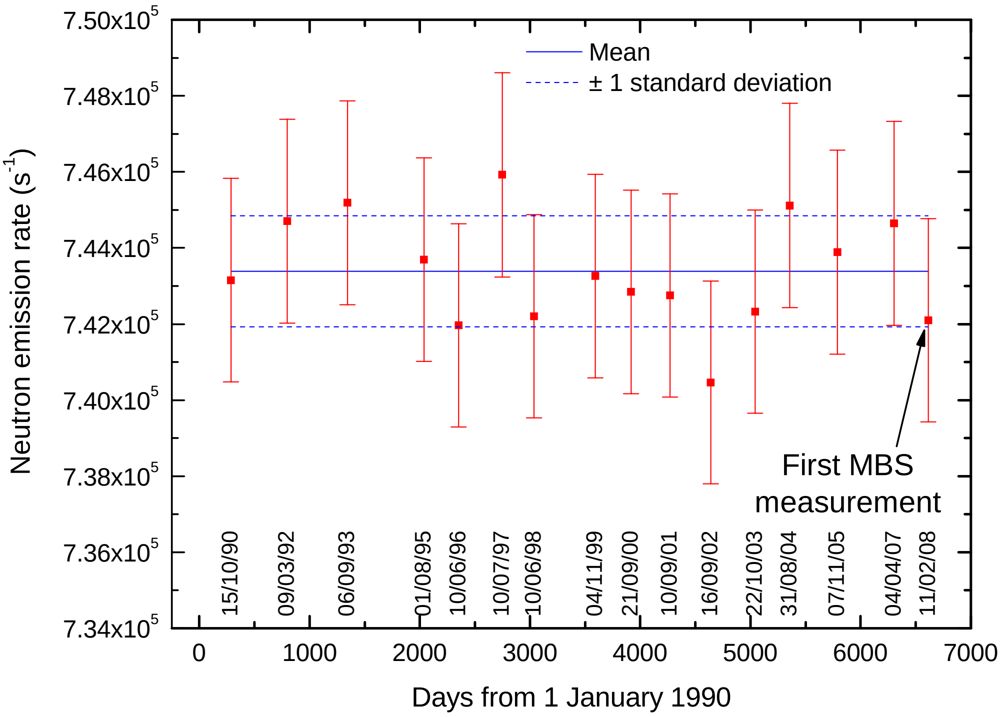

Figure 2: Annual measurements of the national standard ^{226}Ra-Be source, decay corrected to 1 January 1990 (error bars correspond to only the random uncertainty components at 1σ)

Until 2008 the NPL manganese bath facility had always been calibrated with reactor-activated ^{56}Mn, first from the GLEEP reactor at Harwell and more recently from the CONSORT reactor at Ascot. The active manganese was dissolved in sulphuric acid and the activity concentration of the liquid standardised at NPL. At the end of March 2008 the CONSORT reactor closed leaving the UK without a single research reactor. This situation had seemed likely for several years and so other options had been investigated, such as using a reactor in mainland Europe or using the ^{59}Co(d,αp)^{56}Mn reaction in a cyclotron in the UK, although they were not without significant problems.

Use of the NPL thermal pile had been discounted due to the far inferior thermal fluence of about 3×10^7 cm^{-2} s^{-1} compared to 2.4×10^{12} cm^{-2} s^{-1} at the CONSORT reactor. However if, instead of irradiating a small flake of manganese, a sample of the bath solution is irradiated then a comparable total activity can be achieved by irradiation in the NPL thermal pile due to the larger number of Mn nuclei in the solution. Tests have been carried out demonstrating

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that a 3 hour irradiation of approximately 250 ml of solution from the bath gives sufficient activity to calibrate the high and low efficiency channels in the manganese bath, and that a 10 ml sample of the solution contains enough activity to be standardised by an ionization chamber of the Radioactivity group at NPL. With an irradiation of 4-5 hours the activity can be standardised by absolute $4\pi\beta$-γ counting. The efficiencies derived from the tests agreed well with those using reactor-activated manganese. This new approach is far simpler and more cost-effective than any of the other options and so has been adopted as the preferred method for calibrating the manganese bath at NPL.

Investigations have continued into the variations of the O(n,α) cross section between different evaluations which presently accounts for a large uncertainty component when an ^{241}Am-Be source is calibrated in the manganese bath. A comparison of the O(n,α) capture fractions calculated using the different evaluations for the large NPL manganese bath with a ^{241}Am-Be source in a typical capsule (X3) are given in Table 1. The percentage differences in the O(n,α) capture fraction correspond to the same difference in the value obtained for the source emission rate, hence the difference between using ENDF/B-VI.0 and either ENDF/B-V or ENDF/B-VII.0 is 0.79%. Other evaluations were also tested (JEF2.2, JEF3.1, JENDL3.2) but these gave identical results, even as a function of energy, to other evaluations listed in the table.

Table 1: Oxygen (n,α) capture fractions for a ^{241}Am-Be source obtained using different cross-section evaluations

Library	ENDF/B-V	ENDF/B-VI.0	ENDF/B-VI.8	ENDF/B-VII.0	JENDL3.3
O(n,α) capture fraction	1.85%	2.64%	2.46%	1.85%	1.76%

There is a slight reduction when using ENDF/B-VI.8 instead of ENDF/B-VI.0, and a much greater reduction when using ENDF/B-VII.0. However, the remarkable agreement between the values obtained using ENDF/B-V and ENDF/B-VII.0 is not because the cross-sections agree as a function of energy. Figure 3 shows how the capture fraction breaks down over the energy range of interest. Although ENDF/B-VI.0 is greatest across almost the whole range, ENDF/B-VII.0 is the lowest from 4 to 5.5 MeV and ENDF/B-V is the lowest above 7 MeV. The ENDF/B-VI.8 cross-section has been renormalized downwards by 32% over the region 2.4 – 8.9 MeV in the ENDF/B-VII.0 evaluation and this is based on measurements of the ^{13}C(α,n)^{16}O reaction by Harissopulos et al(2).

Evaluations of the O(n,α) cross section rely heavily on measurements of the inverse ^{13}C(α,n)^{16}O reaction due to the limited amount of published data on the ^{16}O(n,α)^{13}C reaction. However, measurements have recently been made of the ^{16}O(n,α)^{13}C reaction by a team at IRMM(3). These showed good agreement with the ENDF/B-VII.0 values up to around 4.5 MeV. However above this energy their values are higher than those of ENDF/B-VII.0 and the disagreement increases with energy for each of the resonances up to 7.2 MeV, more in line with the values of ENDF/B-VI.0. It is hoped that a revised version of the ENDF/B-VII data can be produced from the IRMM measurements which should give an O(n,α) capture fraction in the manganese bath falling somewhere between the values obtained using ENDF/B-VI.0 and ENDF/B-VII.0 given in Table 1.

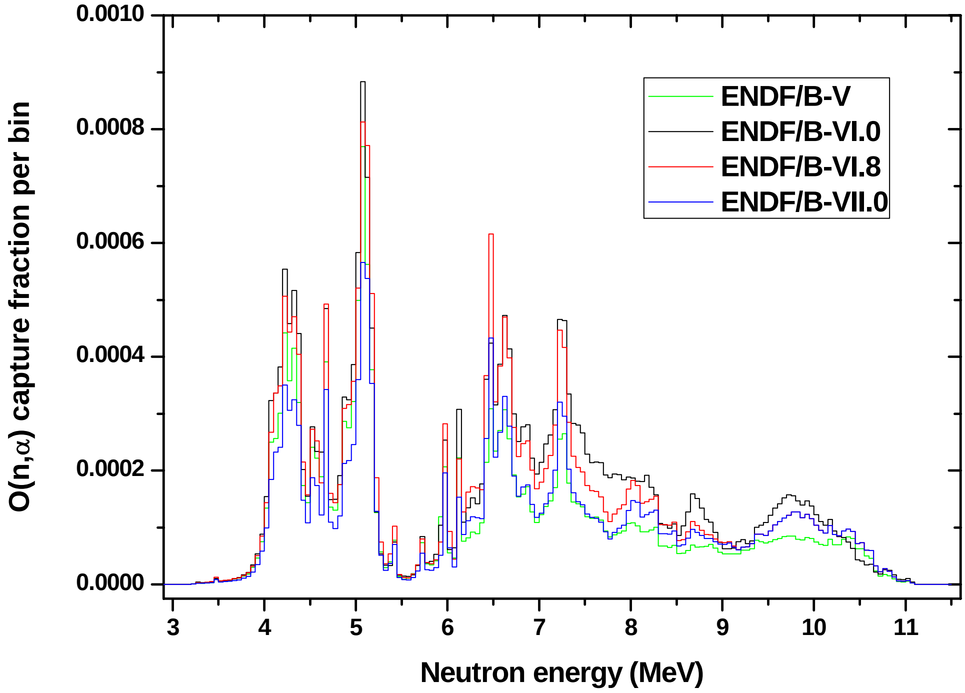

Figure 3: O(n,α) capture fraction versus neutron energy for different ENDF/B cross section evaluations

Much of the recent work in the manganese bath area was presented at the 7th International Topical Meeting on Industrial Radiation and Radioisotope Measurement Application in Prague in 2008, and will be published in a special issue of Applied Radiation and Isotopes in 2009(4).

NPL submitted a solution for the manganese bath problem of the CONRAD WP4 Uncertainty Assessment in Computational Dosimetry exercise and presented it at the workshop in Bologna in October 2007. There was good general agreement on the various capture fractions and correction factors between the majority of the 7 participants, NPL included. Four participants, NPL included, also investigated the uncertainties in the calculations due to several input parameters the most significant of which was the cross section libraries used. The proceedings are available from Eurados(5).

2 Accelerator Based Neutron Fluence Standards

The Sc(p,n) reaction can be used to produce monoenergetic neutrons in the range 8 to 24 keV, however, there are several problems with its use.

a) The neutrons are produced via resonances in the cross section so the proton beam energy has to be extremely stable.

b) The cross sections are very low and high beam currents have to be used to provide reasonable neutron fluences. These high beam currents create problems with blistering of the target backings, presumably due to hydrogen build-up.

c) Scandium target layers need to be of an optimum thickness to maximise the neutron fluence while still producing neutrons from only one of the closely spaced resonances. The optimum thickness depends on the energy width of the proton beam.

Recent work at NPL has investigated target backings, and it was found that tantalum, although not perfect, is far superior to tungsten in terms of not blistering. Rutherford backscattering measurements of scandium target thicknesses and composition have revealed that the targets, although deposited as the metal, very quickly oxidise. A Fortran program has been written to predict the yield curve, as measured with a long counter, as the proton energy is increased. This uses the thicknesses measured for the scandium layers and a value for the beam energy width (assumed Gaussian in shape). It requires data about the relative intensity of the various resonances and this information is only poorly known. Nevertheless, the program predicted the yield curve reasonably well although the resolution of the peaks in the measured yield curve was actually better than the program predicted. This implies either the targets were thinner than measured, or the beam width is narrower. Work on this reaction is continuing in collaboration with IRSN and PTB.

Work to provide monoenergetic 17 MeV neutron fluence standards at NPL using the T(d,n) reaction have shown that the low energy contamination is reasonably small for the newly acquired targets from Sodern in France. The yield from a matched un-tritiated target was in fact lower than the small contaminant field produced by the beam hitting the magnet control slits. The fluences will be measured using a long counter and the response of this instrument to 17 MeV neutrons was determined in a Euramet comparison of fluence measurements in the 15 to 19 MeV region. Some further work is planned in the near future to improve data on the long counter effective centre at this energy.

Problems with the Van de Graaff accelerator and its pulser system have restricted the running time available for time-of-flight work. However, data taken at NPL have been used in a paper on digital timing methods[6].

Historically, when performing calibrations of personal dosemeters on a phantom with monoenergetic neutrons the dosemeters have been limited to a small number positioned near the centre of the front face of the phantom. In this way the personal dose equivalent delivered to each dosemeter is almost the same. Increasingly, however, dosimetry services have wanted to maximise the information obtained in each experiment by maximising the number of dosemeters placed on the phantom. Figure 4 shows an extreme example of this where the dosemeters cover the surface. Although dosimetry services have in the past been told that the dose varies with position on the phantom face, and have been advised to make allowance for this effect, there is little evidence that the services were doing this, and so it has now become routine for this information to be provided on NPL certificates and for the personal dose equivalent to be quoted for each dosemeter separately.

Most irradiations are performed with the phantom positioned at the 0° angle as defined by the direction of the charged particle beam which produces the neutrons. This angle is chosen because the variation of the neutron fluence and energy is a minimum around this angle. Nevertheless, there is some variation, and four effects need to be considered.

1. The increased distance away from the mid point of the face. Allowance is easily made using the inverse square law.

2. The neutron energy changes with angle and hence a different fluence to dose equivalent conversion coefficient must be used. The variation of the energy can be derived from the kinematics.
3. The fluence changes with angle. Usually there is a decrease as the angle increases. This effect can be calculated from the angle dependence of the cross section[7].
4. The angle of incidence of the neutron is not normal to the dosemeter, except for one positioned at the centre of the phantom face, and the fluence to dose equivalent conversion coefficient appropriate for the actual angle of incidence should be used.

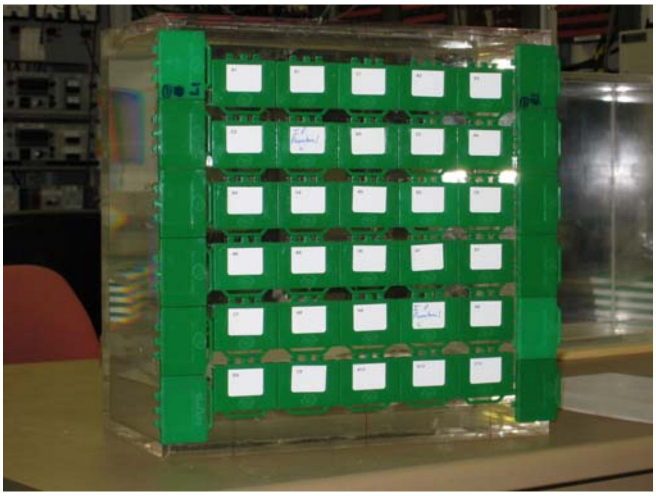

Figure 4. An ISO water phantom with the front face covered with passive personal dosemeters

An Excel workbook has been developed to calculate corrections to the 0^{o} value of the dose equivalent. A separate spreadsheet was developed for each ISO recommended energy[8], each sheet containing data for interpolating the conversion coefficients as a function of energy and angle for small variations about the value for the ISO energy and normal incidence. The extent of the variation is shown in Figure 5 which shows the effect of including the variation with position on the phantom face for an irradiation at the ISO energy of 144 keV. The X value indicates the horizontal position relative to the centre of the phantom, and the Y value the vertical position. The variation in the dose depends on the actual energy, but is often up to 20%.

Irradiations cannot always be performed at the 0^{o} angle, and when irradiations are performed at other angles the variation of the dose equivalent across the phantom surface is even more pronounced. Figure 6 shows the situation when the phantom is placed at 36^{o} in order to obtain 100 keV neutrons using the Li(p,n) reaction. The highest dose equivalent, which occurs for negative X values, i.e. for angles < 36^{o} can be up to 24% higher than the value for the centre of the phantom face, while the lowest dose equivalent can be up to 40% lower. The asymmetric variation occurs because the energy and the fluence, and hence the dose equivalent values, increase as the angle decreases, whereas the other two effects which influence the dose equivalent value result in a decrease as the X and Y values vary away from zero.

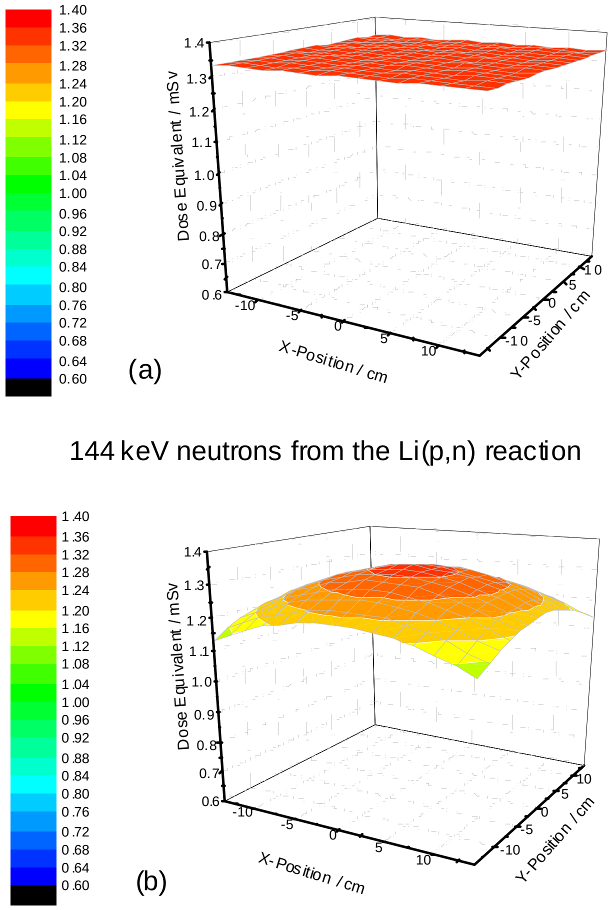

Figure 5. The top figure (a) shows the situation where the dose equivalent is assumed not to vary across the face of the phantom. It can be compared with the lower figure (b) where allowance has been made for all the effects which cause the dose equivalent to vary with position.

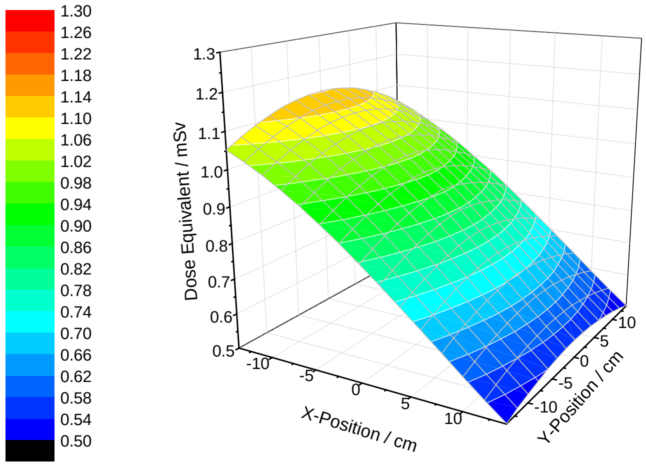

Figure 6. Variation of the dose equivalent across the surface of a phantom positioned at an angle of 36^{o} to a charged particle beam producing 144 keV neutrons at 0^{o} via the Li(p,n) reaction and 100 keV neutrons at 36^{o}.

3 Thermal neutrons

Thermal neutron fluence standards have been available at NPL since the facility, commonly known as the thermal pile, was set up in the late 1960s by Ryves and Paul(9). The pile consists of a large graphite block within which fast neutrons are produced by bombarding two beryllium targets, located on either side of a central irradiation cavity, with deuterons from the NPL 3.5 MV Van de Graaff accelerator. The graphite moderates these neutrons producing a well thermalized stable and uniform neutron fluence rate at the bottom of the central cavity. Figure 7 shows a schematic view of the pile.

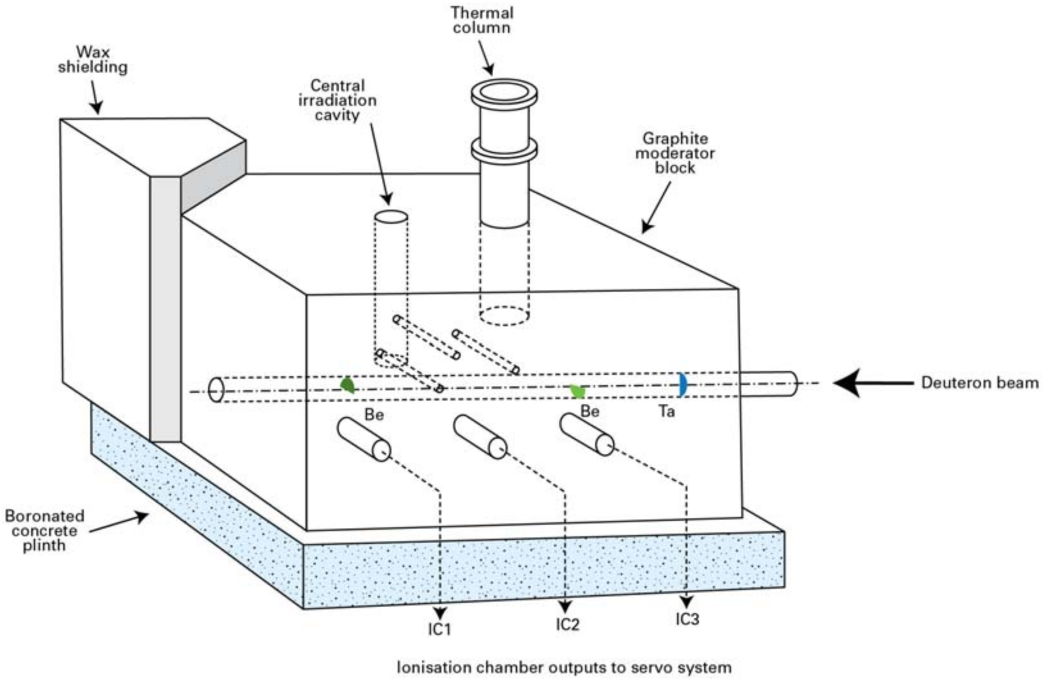

Figure 7. Schematic view of the NPL thermal pile

A servo-system which uses signals from three ion chambers located near the bottom of the pile, adjusts the deuteron beam to ensure the constant fluence rate. The cavity was designed for irradiating small devices to relatively high neutron fluences, up to 10^7 cm^{-2} s^{-1}. Larger devices which do not require such high fluence rates can be irradiated in the field of the thermal column shown to the left of the cavity in Figure 7 which extracts a broad 30 cm diameter beam from within the pile. This is where the SP9 counters for the CCRI key comparison of thermal fluences were irradiated.

The facility was originally designed and built to enable foil activation measurements to be made at the bottom of the cavity. There is no longer a demand for this work, and since the 1980s the facility has tended to be used only for irradiations in the column. With the closure of the last research reactor in the UK in March 2008 the thermal pile has found a new use in the testing of reactor instruments. Since some of these instruments are large devices the original 9 cm diameter central hole has had to be enlarged to 12 cm. The reactor instruments are cylindrical in shape and can be long, up to about 1 m, which means that the thermal fluence is not uniform over the active length of the instrument. A large amount of work has had to be performed to characterise the thermal fields in which these devices are irradiated measuring both the variation with height and the significant flux depression which occurs because of the large mass of the instruments.

Fluence rates at various positions on the surface of the instruments were measured using gold activation foils and these were related to the counts of a fission chamber monitor situated in the graphite near the bottom of the central cavity. Figure 8 illustrates the variation of the fluence with height above the bottom of the cavity and present results for several different devices in the hole.

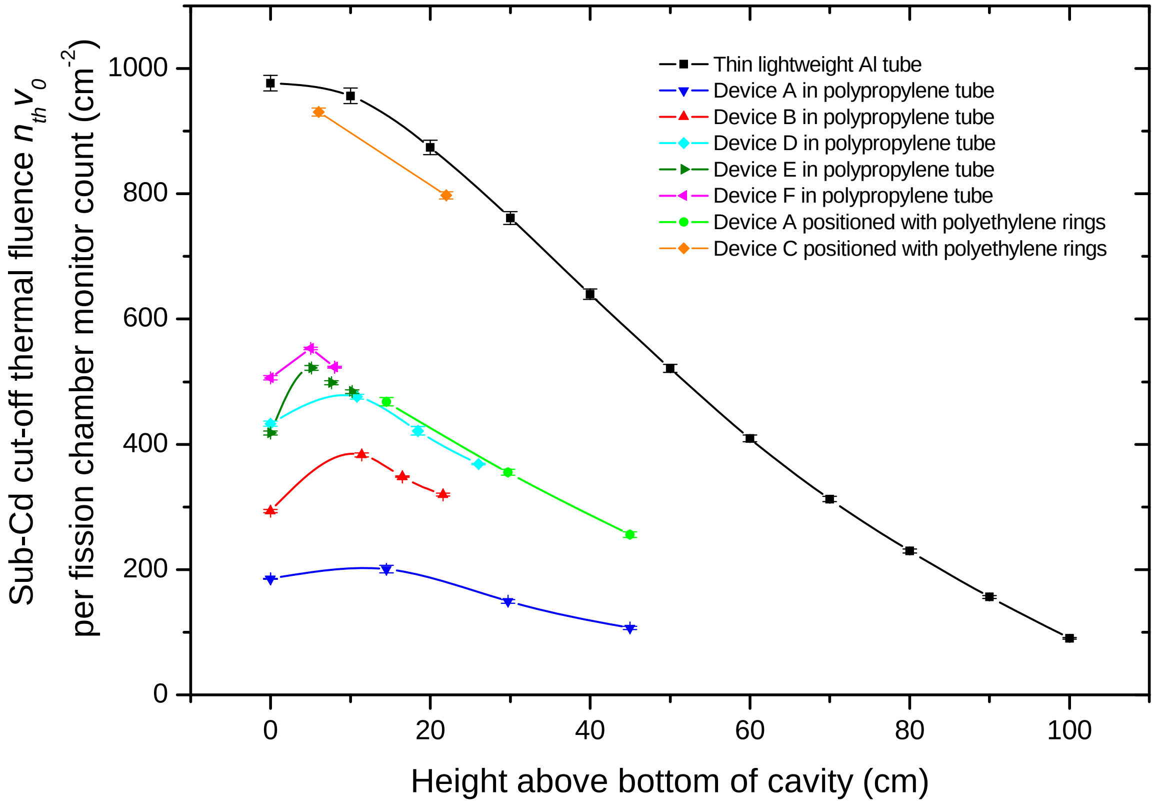

Figure 8. Variation of fluence per unit monitor count with height and type of device positioned in the central hole of the cavity

The thin aluminium tube measurements approximate to the situation of a zero mass device in the hole. The various instruments, labelled as devices A to F, have different masses, A being the largest and heaviest instrument and F the smallest and lightest. It is clear that the fluence decreases with height as expected, and that flux depression significantly reduces the fluence per unit monitor count when large devices are inserted in the hole. There were, however, some unexpected results. When the instruments were positioned in the hole inside a polypropylene tube to simplify handling (the instruments are heavy and have unwieldy cables attached) the fluence per unit monitor dropped by about a factor of two – see the results for Device A positioned using two small polyethylene rings compared to the result in the polypropylene tube. This was unexpected and is still not explained. Nevertheless, a series of successful instrument tests have been undertaken.

The cadmium ratio, R_{Cd}, is relatively high in the hole increasing from about 30 at the bottom to 140 at 50 cm. It is very much less dependant on what is positioned in the hole than the fluence, although large instruments do reduce the value, the minimum recorded being 11 for the bottom of the hole and the largest instrument.

4 Comparisons and Demonstrations of Equivalence

The evaluation of the key comparison of neutron source emission rate, CCRI(III)-K9.AmBe, is ongoing and a revised draft B report will be circulated to participants prior to the 18th CCRI(III) meeting.

A draft of the final report on the EUROMET Project 822 ‘Comparison of Neutron Fluence Measurements for Neutron Energies of 15.5 MeV, 16 MeV, 17 MeV and 19 MeV’ has been circulated to participants by the organisers, the PTB. NPL made measurements using a De Pangher long counter at all energies, and also aluminium activation foils at 16 MeV. There is very good agreement between the NPL results and those of the two other participants, IRMM and PTB, who both used proton recoil telescopes.

Measurements for Euramet project 936, a comparison of the performance of several long counters, were performed in June 2008. The three participants in the exercise are IRSN, NPL, and PTB. Long counters from IRSN and PTB were brought to NPL 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). To date the data has been analysed in terms of ratios of long counter readings, the next step will be to include the participants’ estimates of long counter responses in order to compare results for the fluences.

A detailed report has been written on NPL’s participation in the thermal neutron fluence key comparison, CCRI(III)-K8. It describes the measurement methods and presents the final results. The document, which is in the form of a formal NPL report, is presently going through the approval process at NPL.

5 Neutron Spectrometry

The low energy regions of the spectra of the two americium-based sources recommended by ISO(8) for use in calibrations (^{241}Am-Be and ^{241}Am-B) are poorly known. Good high-resolution data are available for the ^{241}Am-Be spectrum above 100 keV based on the work of Kluge and Weise(10). This is the basis of the ISO spectrum, and there is good agreement with a later measurement(11) covering the same energy region. However, below 100 keV the spectrum has not, until now, been measured, although ISO gives a recommendation based on the fact that, on theoretical grounds, very few low energy neutrons are to be expected in this region.

Figure 9 shows the results of recent measurements at NPL with a Bonner sphere set based on a spherical ^{3}He proportional counter as the central detector. The results are not yet finalised, but the spectrum unfolded using the ISO spectrum as *a priori* information, when is plotted on a linear energy scale (see the upper plot (a) in Figure 9), shows fewer neutrons above about 2 MeV and more neutrons below this energy. This is understandable as the NPL measurements were made for a physically larger source with a roughly 15 times higher output than the one whose spectrum is given by ISO so there is a higher probability of neutron scatter in the source producing lower energy neutrons.

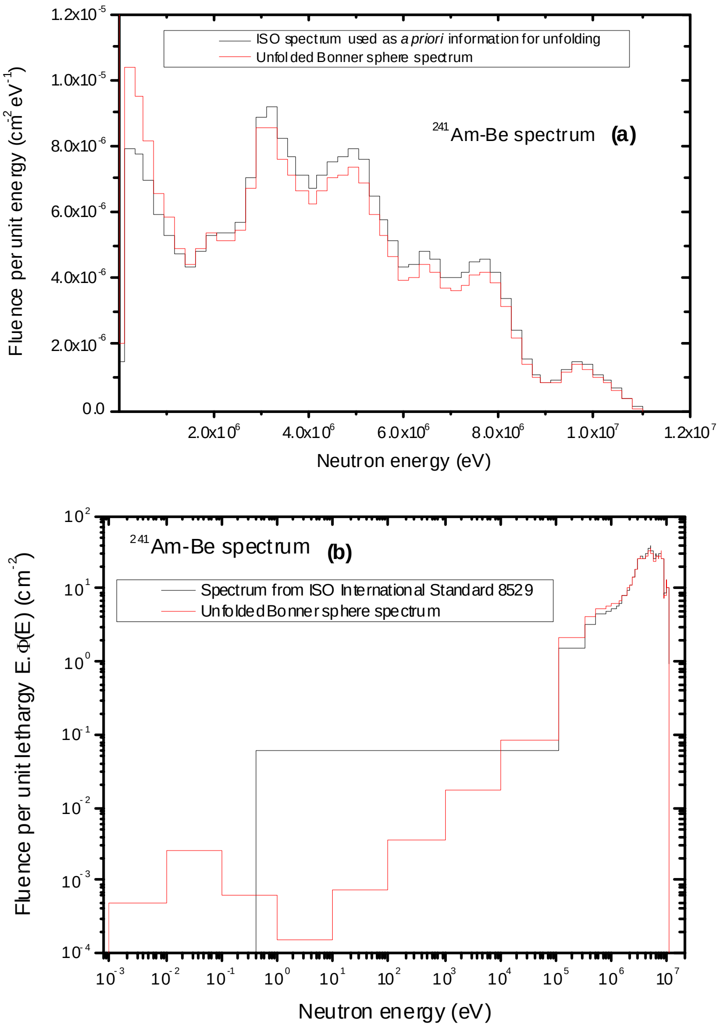

Figure 9. Results of Bonner sphere measurements of the ^{241}Am-Be spectrum showing the ISO recommended spectrum used as *a priori* information for the unfolding, and the unfolded spectrum.

When the data are plotted on a logarithmic energy scale, as in the lower plot in Figure 9, the spectrum in the low energy region is highlighted. It indicates that the ISO spectrum may not be very accurate in this energy region, but the ISO proposition that there are very few low energy neutrons, relative to the high energy ones, is fundamentally correct.

The other spectrum measured with Bonner spheres was that for an ^{241}Am-B source, and the situation here is a little different. The ISO spectrum, which appears to be based on the measurements of Lorch[12] published in 1973, does not agree with later measurements by Marsh *et al.*[11], and Zimbal[13], which agree with each other reasonably well. When the ISO spectrum was used as *a priori* data the Bonner sphere unfolding altered the *a priori* spectrum quite significantly. However when the spectrum of Marsh *et al*. was used as *a priori* information the unfolding barely changed the spectrum indicating that this spectrum is much more likely to be correct than that given by ISO. This is illustrated in Figure 10 which shows, the data of Marsh *et al*. used as *a priori* information, the unfolded Bonner sphere spectrum, which is almost identical, and also the ISO representation of this spectrum. These measurements add weight to the argument that the ISO ^{241}Am-B spectrum needs correcting.

The low-energy part of the spectrum is highlighted in the lower half of Figure 10. The Bonner sphere unfolding indicates even fewer neutrons than indicated by ISO.

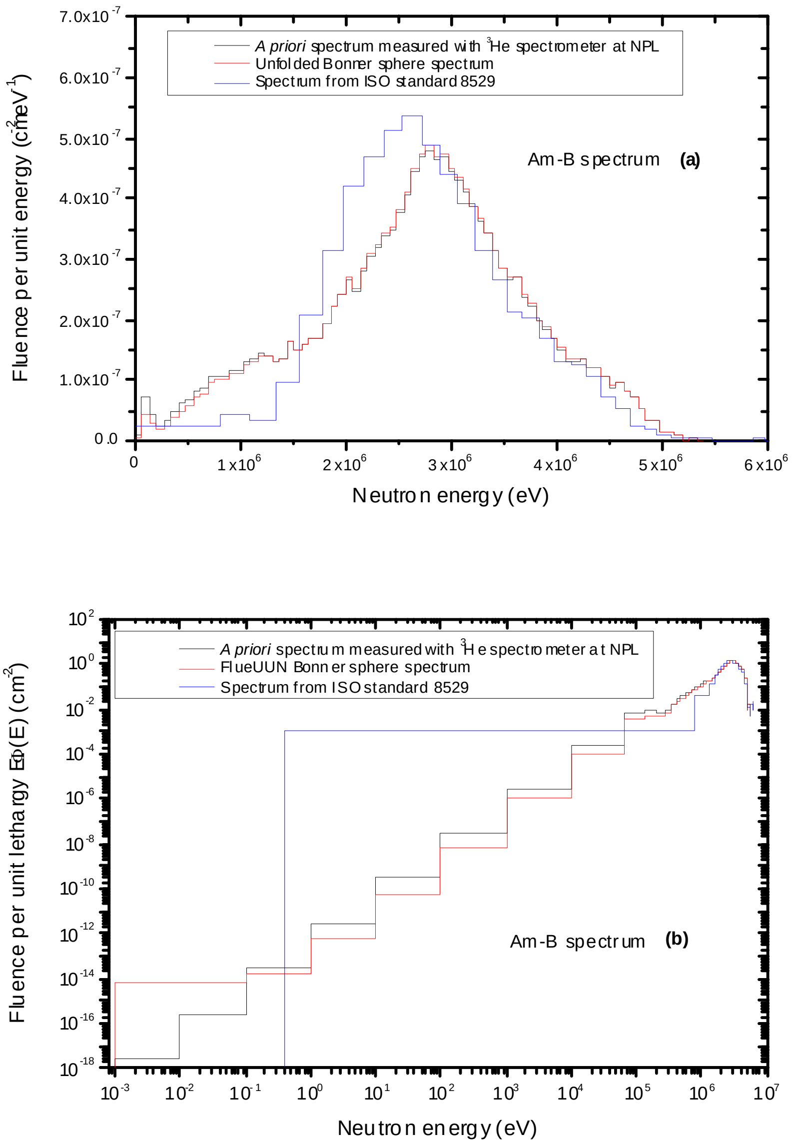

Figure 10. Results of Bonner sphere measurements of the ^{241}Am-B spectrum showing the *a priori* information used for the unfolding, the unfolded spectrum, and the ISO recommended spectrum for this source.

As part of NPL's investigation of the possibilities of digital signal processing a fast digital oscilloscope has been used to digitise the linear (Dynode 9) pulses from one of NPL's existing liquid scintillator spectrometers, with the aim of investigating pulse shape discrimination algorithms. The signal was found to be badly affected by a strong superimposed oscillatory component (Figure 11), and it was feared that the shape information had been lost. However, when the data were analysed by integrating the pulse over two different intervals and plotting one integral against the other, the pulses were seen to separate into two distinct populations (Figure 12), despite the poor quality of the signal.

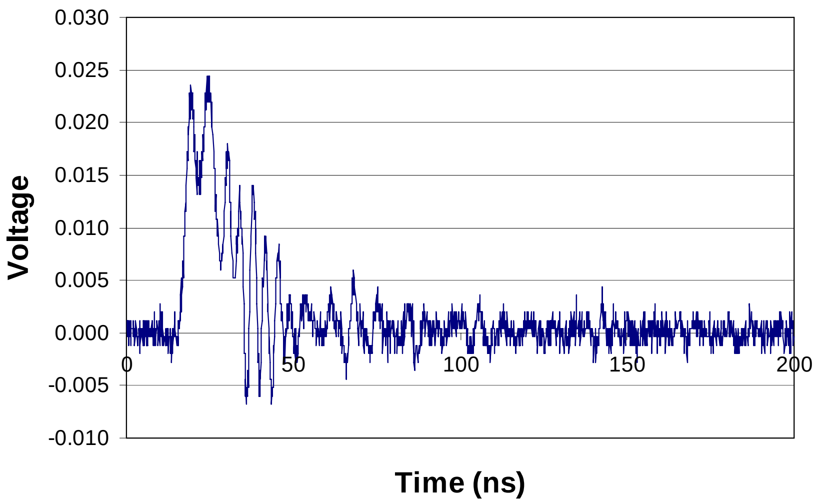

Figure 11. A typical linear pulse captured from the scintillation spectrometer.

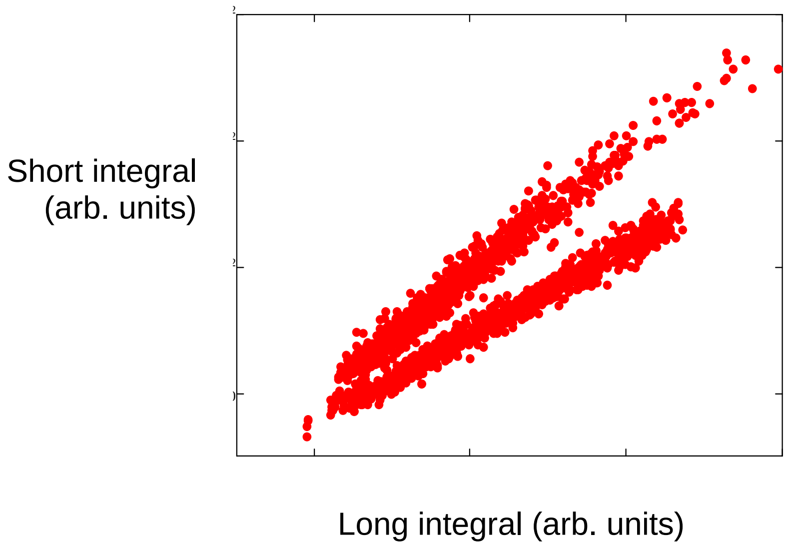

Figure 12. MathCAD plot showing Short vs. Long integrals for a number of spectrometer pulses (one dot per pulse). There is a clear separation into two populations, assumed to correspond to neutron events (upper branch) and gamma events (lower branch).

In view of the oscillatory nature of the linear signal from the existing spectrometer, plans have been brought forward to assemble a new spectrometer from components supplied by Photonis and Saint-Gobain. A miniature digitiser board has been ordered from Hybrid Instruments of Lancaster and will be used to investigate digital discrimination algorithms in conjunction with the new spectrometer.

6 Neutron Dosimetry

Work in the field of cosmic ray dosimetry is progressing on both experimental and calculational fronts. In-flight measurements have been performed in collaboration with QinetiQ on an executive jet flight from the UK to China, and also with TAG aviation on the Transpolar ’08 pole-to-pole challenge flight. Both sets of experimental data are being compared with route doses predicted by several codes. A long-term experimental collaboration with a major US airline and a UK-based start-up company is due to be announced in the near future as well. A basic GEANT model of a Hawk TEPC has also been developed, during a secondment from the University of Lancaster. The preliminary results are encouraging, and the model will be refined and developed in the future.

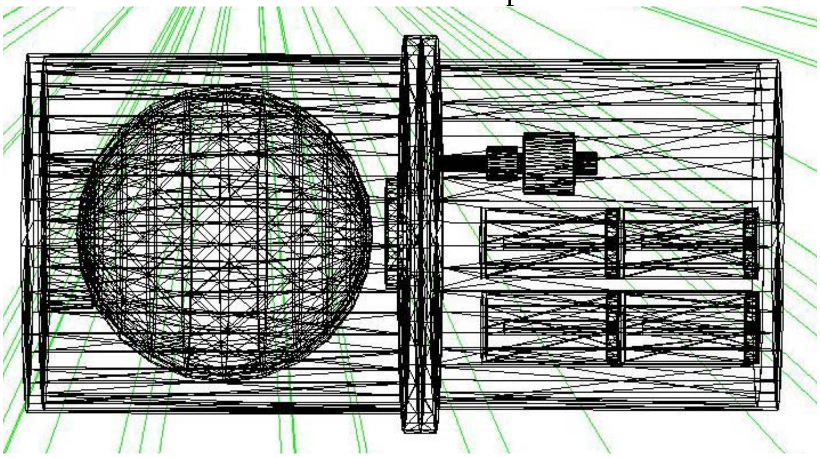

Figure 13 Wire mesh representation of the GEANT4 TEPC model illustrating the housing for the tissue-equivalent plastic sphere and support, 4x D cell batteries and gas filling valve. The green rays represent tracks from a neutron point source.

Work in the field of anisotropic neutron dosimetry is also under way, with a Monte Carlo based feasibility study into the development of a directionally-sensitive neutron dosimeter nearing its conclusion. It is expected that the results will be presented at the forthcoming Neutron Dosimetry Symposium scheduled for October 2009 in Cape Town.

7 Major Facilities Maintenance and Development

During December 2008 problems were experienced achieving high voltages on the terminal of the NPL 3.5 MV Van de Graaff. The indications were that the accelerator tube was beginning to fail. These items have a limited lifetime (~10 years). Visual inspection indicated that there were probably problems with some sections of the tube, and the experiment of running the accelerator without the tube confirmed that the problems stemmed from breakdowns in the tube. This was therefore replaced. During this relatively major operation the belt was inspected closely and, because there were definite signs of ware, a decision taken to also replace this component.

A new tube requires careful conditioning which took the better part of a month, however, the accelerator seems now to maintain high voltages well. At present there are some problems with obtaining a stable high beam current (~ 70 μA of deuterons) for running the thermal pile facility, but it is hoped that these can be resolved relatively quickly.

8 Future work

Amongst our proposals for future work are:

- Develop an integral absolute counting system for the Mn bath, possibly based on a Cerenkov-γ or γ-γ coincidence technique,
- Measurement of photon doses in monoenergetic neutron fields,
- Develop high energy neutron standards, possibly in collaboration with ISIS,

- Increase the fluence in the NPL thermal pile by changing the configuration or adding reflectors,
- Extend digital signal processing to new devices, e.g. plastic scintillators,
- Compare calculation and measurement of neutron doses to patients and staff around hospital linacs,
- 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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