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 development over the last two years has been the construction of a new manganese sulphate bath (Mn-bath) facility and this is covered in section 6 Major Facilities Maintenance and Development.

1 Radionuclide Source Based Fluence Standards

Measurements of neutron source emission rates using the Mn-bath technique have been performed at NPL since the 1960s in a small laboratory close to the site boundary. This area is scheduled for redevelopment, and a new facility has been constructed within the main neutron metrology building (the Chadwick Building). Despite some problems with aging equipment measurements have continued in the old facility up until April 2007 when the last measurement was made prior to the move of the equipment to the new facility.

Mn-bath measurements involve corrections for effects such as capture of thermal neutrons in the material of the source, leakage from the bath, etc. These are usually derived from neutron transport calculations. As part of an international exercise, organised by an EU supported Working Group (QUADOS), to investigate competencies in performing this type of calculation, NPL has participated in a comparison of Mn-bath correction calculations. The exercise involves calculating the uncertainties as well as the sizes of the corrections. This is one of several examples set by the QUADOS Group. A meeting will be held in October in Bologna to compare results.

One of the more significant corrections is for losses of neutrons to the $^{16}O(n,\alpha)$ reaction. In a review and revision of the corrections made a couple of years ago at NPL, ENDF/B-V cross sections for this reaction were replaced by ones from ENDF/B-VI, and the size of the correction increased, by amounts which depended on the source being measured, but which were comparable to the total uncertainty in the measurements (of the order of 1%). Very recently ENDF/B-VII files for this cross section have been released. These differ significantly from ENDF/B-VI and bring the results back towards the values derived with ENDF/B-V. These effects are still being investigated.

The work to investigate the influence of ^{250}Cf in typical ^{252}Cf sources was presented at the 10^{th} Neutron Dosimetry Symposium at Uppsala in the summer of 2006, and will be published in a special issue of Radiation Protection Dosimetry in 2007[1 #].

As part of a continuing programme to measure the characteristics of all the sources available for performing irradiations at NPL the anisotropy of emission was measured for a low

∗ Temporary staff covering for permanent staff on maternity or sabbatical leave.
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emission rate ^{241}Am-Be source in an X2 capsule. The ability of codes such as MCNP and McBend to calculate anisotropy effects has also been explored[2]. Data are now available at NPL for a wide range of X-type radionuclide source capsules and several different source types – ^{241}Am-Be, ^{241}Am-B, ^{241}Am-F, ^{241}Am-Li, and ^{252}Cf

2 Accelerator Based Neutron Fluence Standards

A 3.5 MV Van de Graaff accelerator is used to produce monoenergetic and thermal neutrons, and also to provide the primary neutrons for a simulated workplace field. The accelerator incorporates a pulser system for time-of-flight measurements. This facility has not been used for many years and one of the main activities over the last two years has been to revive this capability primarily to investigate target properties.

Difficulties were encountered in deriving a reliable timing signal from the beam pickup, and in obtaining a clean pulse from the beam pulser. Development work on these continues. Despite such problems, time-of-flight (ToF) spectra have been acquired for neutrons from the ^{7}Li(p,n) reaction, using both lithium glass and plastic as the neutron detector. Recently the system was put to a novel use when a group from Lancaster University tested a digital system for pulse shape discrimination in organic liquid scintillators. The effectiveness of their discrimination algorithm was successfully demonstrated by comparing its decision for each individual event with the ToF signal from the NPL system.

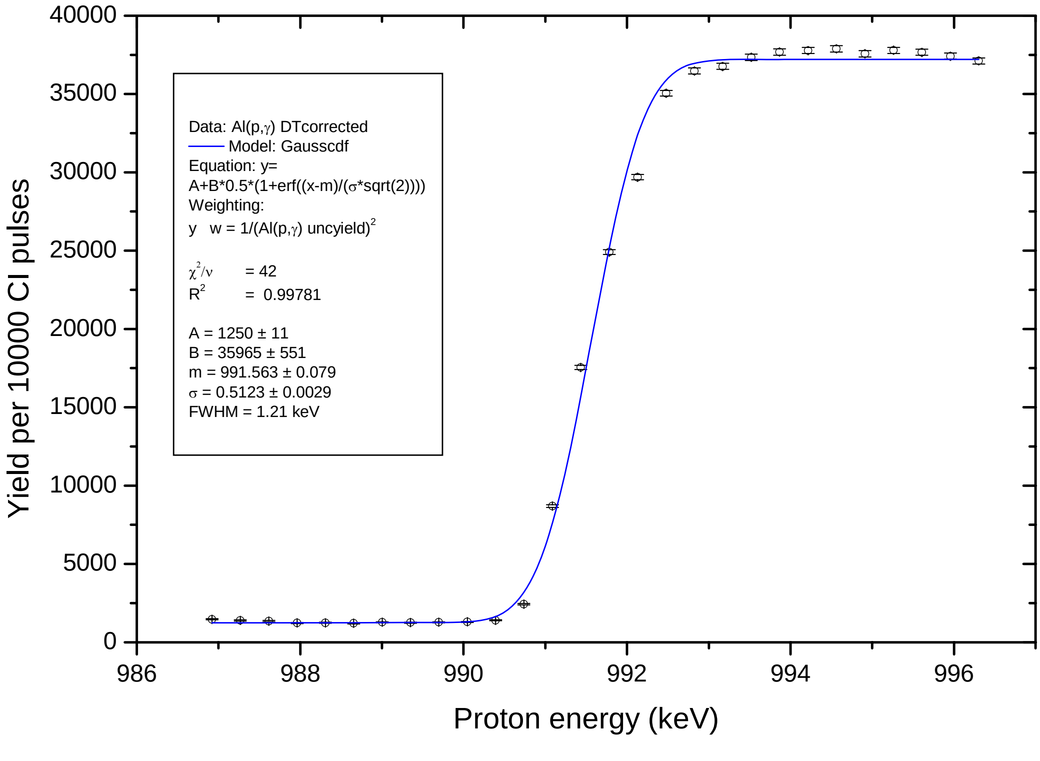

Figure 1. Fit of a Gaussian cumulative probability distribution to the yield curve for the ^{27}Al(p,γ) reaction at the 992 keV resonance.

An investigation has been undertaken of the shape of the ^{27}Al(p,γ) yield curve at the 992 keV resonance energy. This reaction is used regularly during the calibration of the energy-selection magnet of the accelerator. Because the resonance is very narrow the shape of the yield curve is a measure of the energy width of the proton beam. The yield was fitted to a

Gaussian cumulative probability distribution which should reproduce the data if the proton beam energy distribution is Gaussian. As may be seen from Figure 1 the fit is reasonable indicating that the assumption of a Gaussian shape is plausible. Yield curves from energy calibration measurements made over several years were fitted using this cumulative probability distribution and gave FWHM values for the proton beam which were of the order of 1.5 keV although these were somewhat variable with a value on one occasion of almost 3 keV.

One of the major problems in producing low energy neutron fields using the $^{45}Sc(p,n)$ reaction is that of obtaining suitable targets. The target layer must be sufficiently thick to provide a reasonable yield, but be thin enough to enable the resonance peaks of the reaction to be identified in a yield curve. Also, they must be deposited on a backing material which will allow high beam currents. NPL have experimented with tungsten and tantalum backings both having nominally 20 μg cm^{-2} thick layers of scandium deposited on them. The tungsten-backed targets appeared to give good yield curves in that the resonances could be identified from a plot of long counter count rates against proton bombarding energy, but the targets where not stable as they exhibited blistering after a single experimental run with beam currents of the order of 20 μA (Figure 2). The tantalum-backed targets were stable under long term bombardment but it proved very difficult to identify the peaks in the yield curve.

Figure 2. Tungsten-backed scandium target exhibiting typical blistering after proton bombardment

The tungsten and tantalum backed targets were obtained from different suppliers and it was decided to measure the thicknesses. This was done at the University of Surrey, using the Rutherford backscattering technique, which revealed that: a) the targets were not scandium metal but had oxidised to scandium oxide, b) the scandium in the tungsten-backed targets amounted to 20 μg cm^{-2} although the overall thickness was greater because of the oxygen present, c) the tantalum-backed targets were roughly 50% thicker. New tantalum-backed 20 μg cm^{-2} thick targets have been bought from the CEA in France and will be tried over the next few months.

When personal dosemeters are irradiated on phantom in monoenergetic neutron fields it is very common to mount as many dosemeters as possible over the phantom face. This is done to reduce the uncertainty in the measurement by improving the statistics. (The statistical uncertainty on the result from a single dosemeter can be large). The problem with distributing the dosemeters over the phantom face is that the energy and hence the dose equivalent varies, and so does the angle of incidence on the dosemeter. In the past a single dose equivalent, corresponding to that received by a dosemeter at the mid point of the phantom face, has been quoted. However, the variations for some irradiation conditions can be large (up to 40% in one recent case with neutrons incident at non-normal angles), and Excel spreadsheet software has been developed to calculate the dose variations.

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 neutrons are produced 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 vertical thermal beam extracted from a column within the block. Because the primary neutrons are produced using a nuclear reaction the thermal fluences can be varied, in the column from about $10\ cm^{-2}\ s^{-1}$ to more than $10^4\ cm^{-2}\ s^{-1}$, and in the hole between about $10^4\ cm^{-2}\ s^{-1}$ and $2 \times 10^7\ cm^{-2}\ s^{-1}$. The hole, which was originally designed for activating small foils, had a diameter of 90 mm, but this has recently been increased to 120 mm to accommodate larger items. It will now need to be characterised in terms of the fluence levels and variations across the hole and with height.

In order to provide thermal neutron dose equivalent standards not only the fluence but also the neutron spectrum needs to be known. Chopper measurements are not possible at the intensities available in the thermal column at NPL. However, estimates of the spectrum have been derived by assuming a Maxwellian shape for the neutron distribution and deriving an effective temperature from measurements with bare and cadmium covered gold foils. this involves the use of a relationship reported by Geiger and van der Zwan[3]. The effective temperature varies with height in the column and a series of gold foil measurements have been performed to determine this variation.

3 Comparisons and Demonstrations of Equivalence

A member of staff at NPL has been evaluating the key comparison CCRI(III)-K9 exercise involving circulation of an ^{241}Am-Be source for measurements of total emission rate. A draft A report has been written and circulated to the participants for comments. Account has been taken of all comments received and a preliminary draft B report of this exercise is now available.

Measurements for a EUROMET comparison of high energy (15 to 19 MeV) monoenergetic neutrons were performed in December 2004. There have been some difficulties characterising the exact spectral distributions for these fields, but the analysis of the NPL long counter and activation foil measurements is now complete and a report has been written.

The need for a thermal neutron fluence comparison was recognised by CCRI Section (III) some years ago. The initial proposal was to use ^{10}B-lined ionisation chambers. NPL acted as the pilot laboratory for this approach which, was eventually abandoned because of problems with the stability of the ^{10}B layers. A brief report on the pilot study was produced[4]. A revised approach, using ^{3}He proportional counters, has now been adopted and NPL have performed the measurements for this exercise in February 2007. A rough analysis of the data has been performed and the counters returned to the organisers, the PTB.

4 Neutron Spectrometry

NPL has two Bonner sphere sets, one with a spherical ^{3}He proportional counter as the central sensor, and the other with gold foils. The gold foil set has advantages in pulsed fields, and fields where the photon intensity is high, but suffers from a rather low sensitivity. Measurements of the response functions with monoenergetic neutrons are thus difficult, however, some work to improve the response functions has been undertaken by making measurements in the thermal pile facility and by extended calculations with both MCNP and the McBend code. The thermal measurements concentrated on the smaller spheres since these are the most important for determining the spectrum at low energies. For the 3", 4" and 5" spheres there was good agreement between measurements and calculations, but for the 2", 2.5", 3", and 3.5" spheres the measurements were about 12% lower than the calculations with the two codes which agreed well with each other see Figure 3. Although the fact that the two calculations agree might cast doubts on the measurements, it is difficult to see any experimental effect which would result in the correct value being determined for the 3", 4", and 5", spheres but incorrect values for the smaller spheres.

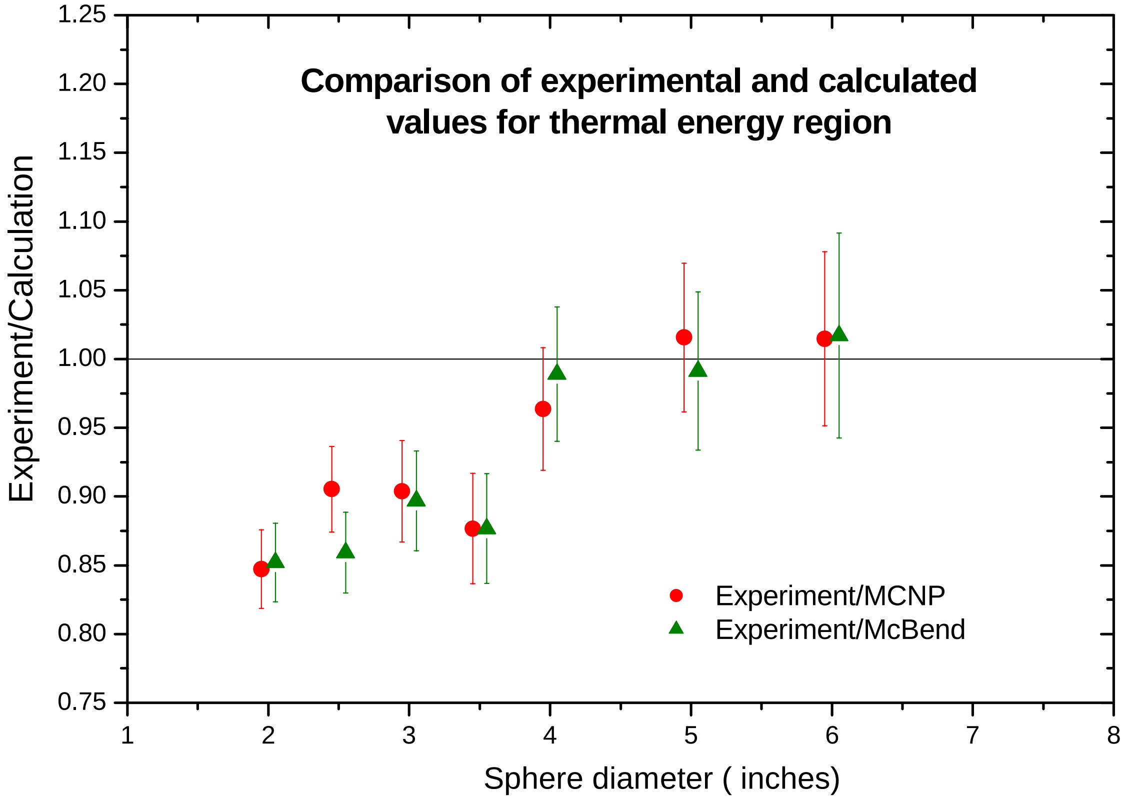

Figure 3. Results of measurements and calculations for the thermal response of the smaller spheres of the gold foil based Bonner sphere set.

The gold foils used in the Bonner spheres have a quite large diameter (~23 mm), in order to optimise the efficiency, and for the smallest spheres this may result in the responses not being completely isotropic. Calculations with MCNP showed that there was some anisotropy for the very smallest spheres at low energies but that the effect soon disappeared on going to larger spheres and higher energies – see Figure 4.

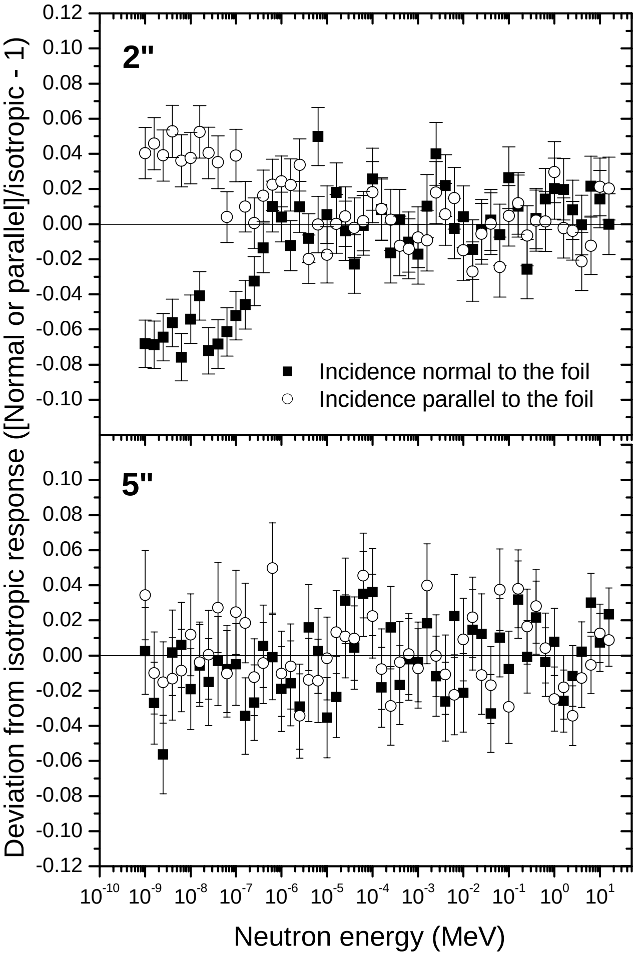

Figure 4. Gold foil based Bonner sphere responses for neutron incidence normal and parallel to the plane of the gold foil.

The gold foil based Bonner sphere was used to measure the neutron field around a hospital medical accelerator operating at an electron energy of 22 MeV, and the results together with a description of the work on the response functions were presented at the 10th Neutron Dosimetry Symposium[5].

Bonner spheres provide wide energy range measurements but poor resolution. Ideally in determining a neutron spectrum results from Bonner sphere measurements should be combined with those from high resolution instruments such as hydrogen recoil proportional counters or scintillators. The problem of combining these data to provide a best estimate of the spectrum is a formidable one although some progress in this area has been achieved and was reported at 10th Neutron Dosimetry Symposium[6].

In 2004 the cells containing the organic liquid scintillator on the NPL NE213 detectors were replaced because the old cells were showing a significant loss of performance due, it is thought, to an unusual aging process. The new response functions have now been measured at a range of monoenergetic neutron energies and common gamma reference energies, and response matrices are being constructed for each detector using the PTB codes NRESP7 and PHRESP. It has been observed that, following the installation of the new cells, the neutron-gamma discrimination signal from the Owens dynode network used on these detectors can fall below the threshold of the acquisition electronics for some gamma event energies, potentially distorting the gamma spectrum. The cause and impact of this is under investigation.

For high resolution neutron spectrometers n/gamma discrimination is often a requirement. Even in cases where it is not essential it is desirable in order to extend the measurements to lower neutron energies. Options for using digital electronics in such applications have been explored in particular for proton recoil proportional counters[7].

5 Neutron Dosimetry

The projects which fall under this heading are all aimed at improving measurements of dose equivalent for radiation protection. A project is presently underway to determine the photon dose equivalent in the fields from various radionuclide neutron sources. Several devices are being used for the measurements, all nominally neutron insensitive or with a very low neutron sensitivity. They are: a graphite ion chamber, Geiger Müller (GM) tubes, and electronic personal dosemeters. The data are in reasonable agreement although the results from different types of GM tubes show some initially unexplained differences. The reason for the disagreement is now thought to be the different dose equivalent responses of the different GM tubes. Although these devices are sold as "gamma compensated" detectors with a nominally constant dose equivalent response with energy the variation can be several tens of percent especially for low energy photons such as the 60 keV gammas from the americium in americium based (α,n) sources.

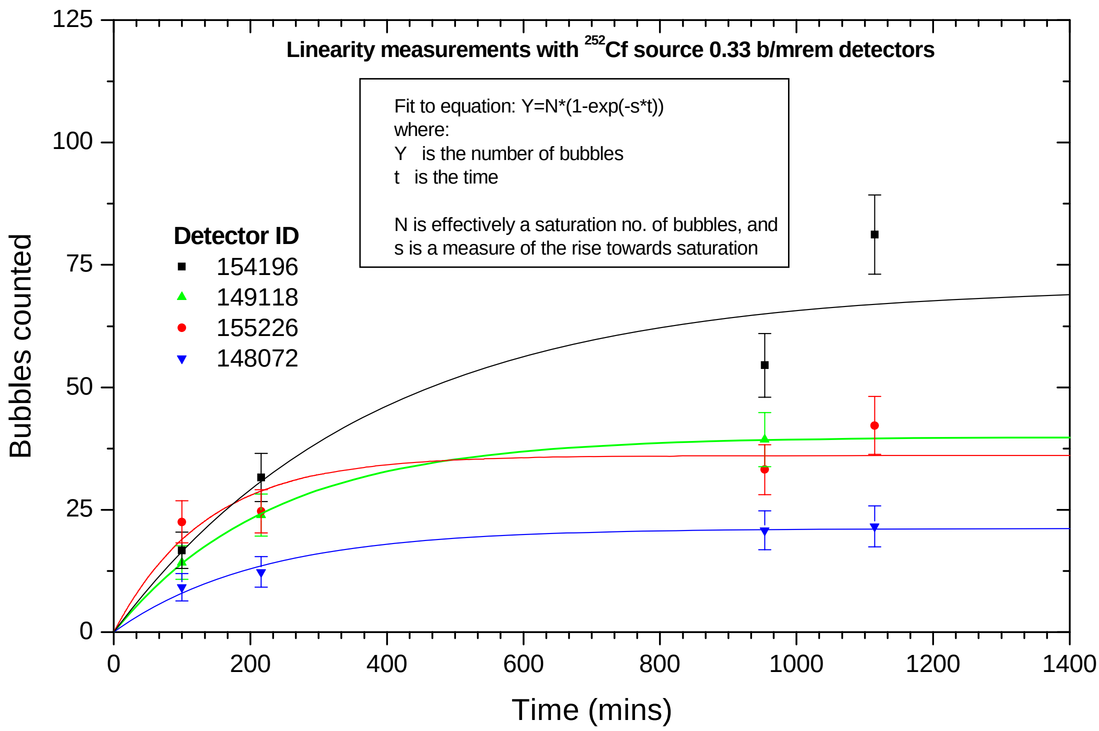

Figure 5. Number of bubbles seen after irradiation with ^{252}Cf neutrons for different periods of time

Work has continued on investigating the use of Bubble detectors to measure neutrons around medical LinAcs. Most of the results have, however, been negative rather than positive. Although supposedly insensitive to photons and therefore suitable for measuring neutron doses around therapy Linacs, bubble detector measurements made at the Kent Oncology Centre, Maidstone Hospital, demonstrated unequivocally that certain types of these devices (BD-100R series) *are* sensitive to photons at temperatures above ~23^{o} C. There is also a potential problem with non-linearity for these devices especially the least sensitive ones which are often used close to the isocentre. Measurements performed with a ^{252}Cf source showed that the number of bubbles did not increase linearly with irradiation time but showed definite saturation-like characteristics (Figure 5).

6 Major Facilities Maintenance and Development

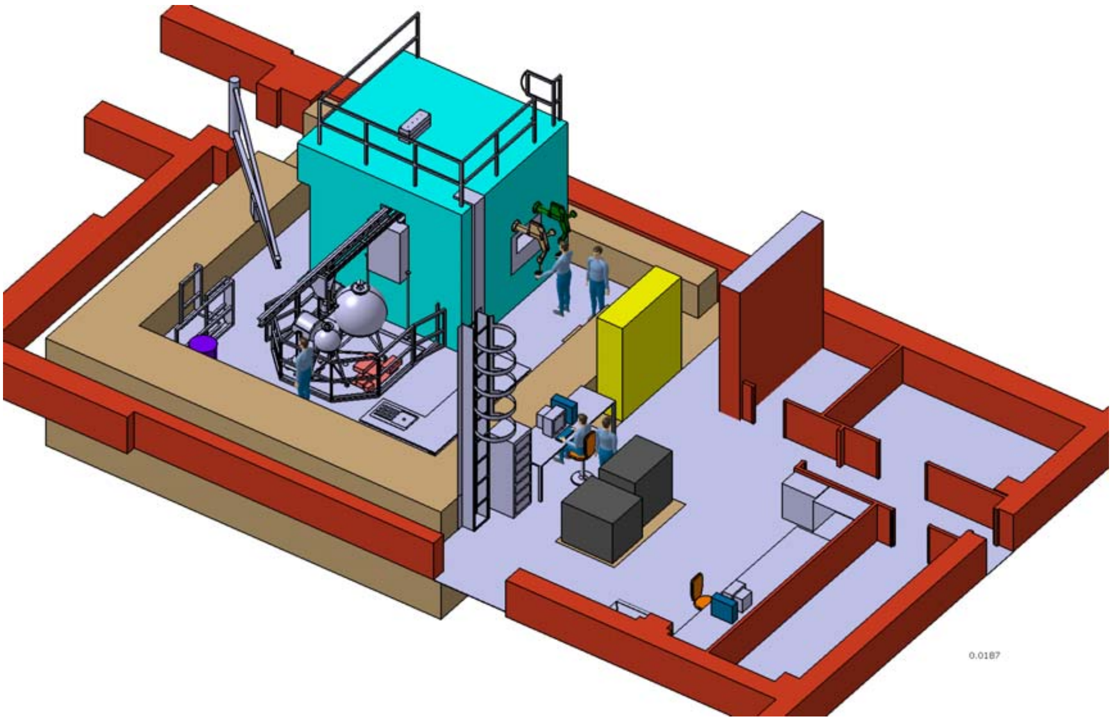

Figure 6. CAD representation of the new Mn-bath laboratory showing the control area (bottom right) and the bath lab (top left) with the small and large baths and the transfer system for transporting sources between the baths and the source cell.

During the spring and summer of 2007 the NPL manganese bath is moving into a new purpose built laboratory. The key features of the new facility, which is shown schematically in Figure 6. include:

- A concrete source handling cell with acrylic viewing window and two manipulators
- Four deep storage holes within the cell for storing neutron sources
- Two-axis transfer system for placing sources into either bath from the source cell
- Fully secure area for storing sources awaiting calibration
- Interlock system to prevent exposure to sources

The electronics and data capture and analysis software will also be upgraded during the move. Re-commissioning the system involves a large number of measurements and it is anticipated that the calibration of sources for outside customers in the Mn bath will resume in August or September 2007. Sources with outputs of 2.3×10^6 neutrons per second or lower can still be calibrated during this period using the moderator detector.

The 3.5 MV Van de Graaff has operated reasonably well over the last two years although there have been continuing minor problems with the pulser circuitry. New pulser boards have been designed and one has been installed in the high voltage terminal during the maintenance period in April 2007. It will be tested over the next few months.

Future work

Amongst our proposals for future work are:

- Continued development of primary neutron standards, including an increased range of monoenergetic neutron energies, improved target characterisation using time-of-flight, and development of the simulated workplace field facility.
- Improved reliability and usability of neutron spectrometers (for example by using digital electronics), and improved dosimetry in problematic locations such as high energy and directional fields.

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