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

Manganese bath

The manganese bath facility is the primary standard for neutron emission rate and incorporates a source cell with manipulators enabling high activity sources to be loaded and unloaded from containers and inserted into the manganese bath source transport system – see Figure 1.

![Figure 1: The large and small NPL manganese baths](image)

Long term stability is checked annually using a Ra-Be(\(\gamma\),n) photo-neutron source. The most recent measurement found it to be at borderline agreement with the historic mean and in agreement with the previous year’s measurement. The earlier decrease in emission rate thus seems to have been halted and has been attributed to the beryllium lid of the Ra-Be source becoming partially unscrewed. This decreased the emission rate as some of the target material (the outer beryllium capsule) was in a lower photon fluence from the inner radium pellet.
Other neutron sources have been routinely measured in the bath over a similar period and did not show any decrease in emission rate, other than that due to decay.

![Mn Bath Measurements of NASTRAE](image)

**Figure 2:** Graph of routine emission rate measurements of national standard Ra-Be(\(\gamma, n\)) photo-neutron source

### Photons from radionuclide neutron sources

A new set of GM tubes were used to measure the photon doses from radionuclide sources and neutron producing targets. This included a study of how the photon to neutron dose ratio varies with size for \(^{241}\)Am-Be sources. Measurements were made with the sources bare and under a 2 mm lead cap to shield the dominant 60 keV gamma from \(^{241}\)Am. Figure 3 shows that for the bare source measurements there were large variations with size in the photon to neutron \(H^*(10)\) ratio (23% to 122%), but when under a lead cap the ratio was constant at about 2.8% for all sources. This effect is due to the self-shielding of the 60 keV gamma ray. This depends on several factors such as the geometry of the source material, the thickness of the encapsulation and the americium to beryllium atomic density ratio.

Measurements of the dose rates from Cf sources also showed that the photon to neutron dose ratio changes from 5% for a 2 year old source to 25% for a 42 year old source.
The second part of the project was to measure the photon energy spectra of the neutron fields. A High Purity Germanium, HPGe, detector was used (see Figure 4). The measured spectra enabled many of the photon energies to be identified but to arrive at the actual photon energy spectra it was necessary to correct for spectral features due to photons escaping the detector before they have deposited all their energy and the detector efficiency. This was achieved by calculating the response of the detector to photons in 1 keV steps up to 10 MeV using the Monte Carlo code MCNP. The resulting response matrix was then used in an unfolding code, GRAVEL, more routinely used for interpreting data from neutron spectrometers.

One of the many interesting points revealed by this work has been the change in the spectrum from $^{252}$Cf sources as they age. Figure 5 shows that as a source gets older the continuum component of the spectrum, due to spontaneous fission of $^{252}$Cf and $^{250}$Cf, decreases and the discrete lines, from other longer-lived non-spontaneously fissionable isotopes of californium, $^{249}$Cf and $^{251}$Cf, and the fission product $^{137}$Cs, dominate.
2. Accelerator Based Neutron Fluence Standards

Photons in accelerator based neutron fields

Measurements of the photons produced from neutron-producing targets were made using the same GM tubes and HPGe detector mentioned above. Those from protons on a LiF target were of particular interest as this target is known to produce a high photon flux. In absolute terms the photon ambient dose equivalent varied little between the different neutron energies produced by a LiF target, although when expressed as a percentage of the neutron ambient dose equivalent it was found to vary from 14% at 70 keV down to 1.2% at 565 keV due to the different neutron yields at these energies. Ti(T) target measurements showed that the photon dose component is much smaller than for LiF targets – less than 0.1% of the neutron ambient dose equivalent at 1.2 MeV. At 16.5 MeV, using the DT reaction, the photon dose relative to the neutron dose was measured as 0.7%.

Figure 6: Unfolded photon spectra for 565keV from a LiF target (left) and 16.5 MeV from the DT reaction (right)

Spectral data have been measured and unfolded from all the monoenergetic neutron fields routinely used at NPL, and some examples of photon spectra from accelerator-based fields are shown in Figure 6. Some of the photons in the spectra are due to neutrons interacting in...
the room (e.g. 847 keV due to inelastic scatter in iron, and 2.2 MeV from thermal capture in hydrogen) and so will vary in intensity with position.

1 keV standards

A project has recently been completed to look into developing neutron standards at ~1keV. The $^{65}$Cu(p,n) reaction was chosen although it was necessary to use isotopically enriched targets to produce a satisfactory yield – about a factor of 2 higher than an unenriched target. Targets of 20, 30, and 40 $\mu$g cm$^{-2}$ were made and tested. Figure 7 shows that the 30 $\mu$g cm$^{-2}$ target produced the highest yield and the sharpest peak in the yield curve with a good gap between it and the next resonance. This is important as an irradiation with resonance neutrons requires the proton beam energy to be very stable. If the peak in the neutron yield curve is sharp it makes it easier to detect when the proton beam energy moves off resonance and to correct for this.

![Graph showing Cu(p,n) measurements](image)

Figure 7: Yield curve for neutrons produced using various copper targets

The reason why the 30 $\mu$g cm$^{-2}$ target is the optimum thickness is that the energy loss in the target is roughly equal to the width of the neutron energy distribution. Neutrons are thus produced at some point in the target by neutrons at all the energies covered by the neutron beam energy width.

Target study

Nominally monoenergetic neutron fields always have some component of neutrons at energies other than the designated monoenergetic value. This is due to scatter effects in the target, unwanted components in the neutron-producing target layer or its backing, etc. An investigation has been conducted into the possible sources of contaminant neutrons and their intensities with a view to drawing up a list of the most important targets to study. A literature survey of information on deuterium contamination in tritium targets came up with figures between 3% and 80%, but in general very little published in the open literature.
New deuterium and tritium targets have been acquired. Calculations have been performed of expected energy loss in the target layer of the new targets and calculations have also been performed of the expected target scatter contribution to the spectrum. This is the calculable part of the contaminant spectrum and results in a scatter background contribution extending from the energy of the main peak down to zero. The spectra from the targets will be measured using Time-of-Flight in the second year of the project.

3. Thermal neutrons

The thermal pile continues to be used regularly for reactor instrument pre-installation testing. There is no civil research reactor available in the UK to do this work.

NPL participated in the thermal neutron key calibration K8 with results which were significantly lower than the mean. A project to investigate the measurement of thermal neutrons at NPL has just been completed. Many of the parameters used in the analysis of gold foils at NPL, such as the beta efficiency and its associated \((1+K)\) correction, come from measurements made over 50 years ago at the GLEEP research reactor. A series of irradiations of different thickness foils in the cavity of the NPL thermal pile have been made to verify the parameters. The new measurements of the beta efficiency of the gold foils showed excellent agreement with the values from the measurements at GLEEP as shown in Figure 8. The region from 95 to 105 mg is of particular importance as the foils used for routine thermal measurements, including those for the key comparison exercise, are in this range.

![Figure 8: Comparison of new NPL gold foil beta efficiencies vs foil mass with those from GLEEP data](image)

The \((1+K)\) correction allows for the sensitivity of the beta counter to gamma rays. The size of the correction increases with increasing foil thickness and values are obtained from a fit to historic measurements. To test the validity of the \((1+K)\) correction a number of foils of different thicknesses were irradiated in the same location in the cavity of the NPL thermal pile. The activities of the foils should yield the same value for the neutron fluence (when
normalised to the fission chamber monitor, FC$_{L0}$). Figure 9 shows the values for the fluence obtained for each foil alongside the values obtained if the (1+K) correction is not applied. There is excellent agreement between the values for foils of all thicknesses when the (1+K) correction is applied.

Other potential problems with the key comparison measurements were also investigated. A check of the dead-time of the type of transfer instrument used in the comparison was performed and the original values validated. The effect of our limited knowledge of the temperature of the thermal energy distribution was also investigated but turns out to be unimportant because both the gold foils used to measure the fluences and the transfer instrument have the same cross section shape so uncertainties in the temperature cancel out.

4. Comparisons and Demonstrations of Equivalence

CCRI

NPL are piloting the supplementary comparison K9-AmBe-1 which will extend the traceability of neutron source emission rate measurements resulting from the earlier K9 comparison. Owing to insurmountable difficulties in sending the source to France it was not possible for LNHB to participate and the source has now been returned to China. The exercise will now be a bilateral one between NIM (China) and NPL. Both institutes have submitted their results to the BIPM so the evaluation of the exercise will proceed shortly.

EURAMET

EURAMET comparison #1104 of Bonner sphere measurements of different sized $^{241}\text{Am-Be}$ sources has concluded with the publication of the spectra in Nuclear Instruments and Methods. The spectra show clear differences at low energies from the current ISO 8529 spectrum (see Figure 10) and will form the basis of the revised standard. The participants
were NPL, INFN (Italy) and UAB (Spain) and the final spectra were obtained by combining the data sets from the three institutes and performing a single combined unfolding using both the UMG and FRUIT codes. The low energy part of the start spectrum used in the unfolding process came from a two-stage calculation involving SOURCES 4C and MCNP. This was merged with a high resolution measured spectrum to form the complete start spectrum.

![Comparison of unfolded 1, 10 and 15Ci spectra from Combined data set](image)

Figure 10: Bonner sphere measurements of the Am-Be energy spectra for 3 sizes of source compared with the ISO 8529 spectrum

### 5. Neutron Spectrometry

#### New Am-B and Am-F spectra

New energy spectra for $^{241}$Am-B and $^{241}$Am-F sources have been published based on Bonner sphere measurements unfolded using high resolution start spectra. Using the same approach adopted for the $^{241}$Am-Be measurements reported above, the start spectrum for each source was formed from a SOURCES 4C and MCNP spectrum at low energies and a high resolution measured spectrum at higher energies.

![Figure 11: Bonner sphere Am-B (left) and Am-F (right) unfolded spectra compared to other previously published spectra](image)
The new \(^{241}\)Am-B spectrum supports the already published spectra of Marsh et al. and Zimbal, but reduces confidence in the ISO 8529 spectrum. However in terms of derived quantities there is good agreement between all the available spectra. By contrast, the new \(^{241}\)Am-F spectrum is significantly different from those already published. The fluence to dose conversion coefficients derived from the new spectrum are 9% lower than currently accepted values, and the emission rates of \(^{241}\)Am-F sources measured by the manganese bath technique may need to be increased by up to 0.5%.

**New fast-neutron spectrometer**

A project is currently underway to design and build a fast-neutron spectrometer which is versatile and easily deployable. The first deliverable of this project is to select the main components of the spectrometer (scintillator, light sensor and digitiser) and assemble and test a prototype instrument. The new plastic scintillators that exhibit efficient pulse shape discrimination (PSD), e.g. that developed by Zaitseva et al.\(^{11}\), allowing neutron and gamma events to be distinguished from each other, are an attractive option for use in this portable instrument because of their improved robustness compared to liquid scintillators (which until recently were the only option for fast PSD). However, it was not obvious initially that the quality of their PSD would be adequate. Tests with a prototype detector have now shown good neutron / gamma separation from a plastic scintillator (see Figure 12), confirming this type as the favoured candidate.

![Figure 12: Scatter plot of the integrated charge (in arbitrary units) in pulses from a lithium-loaded PSD-capable plastic scintillator, exposed to a \(^{241}\)Am/Be source inside a polyethylene moderator. Separation into neutron and gamma events is clearly evident. The lithium loading produces a third group of events from the capture of thermal neutrons.](image)

Tests with NPL’s existing digitiser have revealed some issues with count rate dependence and linearity. Users of other digitisers report similar problems. Consequently there is no clear leading candidate and the decision on which to use is not straightforward.
6. Neutron Dosimetry

Cosmic ray dosimetry

Work has continued in the field of cosmic ray dosimetry however, with the cessation of inflight measurements at the end of 2012, attention turned to theoretical aspects of high energy dosimetry, primarily the development of a GEANT model of a TEPC. This is a particularly demanding simulation because any limitations in the physics models, particle production cross sections, stopping powers or w-values will result in discrepancies between the measured and modelled responses. Figures 13a and 13b show comparisons between TEPC measurements for 14.8 MeV and 252Cf neutrons and the GEANT model developed at NPL. The agreement, particularly for 14.8 MeV, is better than anything achieved before. Consequently the development of the model was presented as a poster at the “9th International Topical Meeting on Industrial Radiation and Radioisotope Measurement Applications”, held in Valencia in July 2014. The associated paper is currently undergoing peer review. Figures 13c and 13d show development work addressing the gamma response of the model, which has yet to be reported elsewhere.

Figure 13: Measured TEPC responses compared with GEANT4 simulations. (a) 14.8 MeV neutrons; (b) 252Cf fission neutron spectrum; (c) 241Am gamma spectrum (60 keV); (d) 60Co gamma spectrum (~1.2 MeV)

A meeting of EURADOS Working Group 11 (High Energy Radiation Fields) was hosted by NPL on 22nd and 23rd of September 2014, which included discussions on the current state of
solar particle event dosimetry, in particular the calculation of doses; there are still discrepancies of factors of three or more between methods.

Work in the field of anisotropic neutron dosimetry is progressing. A CASE Studentship at the University of Lancaster started in January 2013 with the aim of developing the concept of a directionally-sensitive neutron dosimeter presented at the NEUDOS-11 symposium in Cape Town in 2009.

**Improved tissue equivalence of active personal dosemeters**

Currently, all active personal neutron dosemeters are based on a silicon sensor. Unfortunately the way in which neutrons interact in silicon is significantly different from the way in which they interact in body tissue. This means that, although such devices can be calibrated for a particular neutron spectrum, they can, and do, misread significantly when used in a neutron field with a different spectrum. Novel materials and sensor technologies are being tested at NPL to measure their energy-dependent response to assess the degree of tissue equivalence.

**Neutron dosimetry based on radiobiology**

The risk from radiation exposure is currently estimated using the crude measure of energy deposited per unit mass (multiplied by various factors). Recently, studies have been undertaken to link risk to actual observable processes in biological cells, such as DNA strand breaks and the production of radicals and reactive oxygen species. Such studies may lead to proposals for new radiological quantities, along with new ways of measuring them, that replace the somewhat problematic concepts of dose equivalent and effective dose. These new quantities are in turn likely to lead to an increased emphasis on cell biology and on measurements on the nanometer scale.

A project is underway to report on what impact these developments are likely to have, on a timescale of 10 years or more, on the skills and facilities required for neutron standards in general and the NPL Neutron Metrology Group in particular. To date this has included participation in a EURAMET workshop where the neutron viewpoint was represented and valuable input was obtained from the European experts who were present as invited speakers or delegates. Other source material includes papers from the BioQuaRT project and from the neutron microbeam facility at Columbia University, USA.

### 7. Major Facilities Maintenance and Development

The NPL 3.5 MV Van de Graaff has worked well over the last two years with very few major breakdowns. One of the biggest issues is the need for very high beam currents, in excess of 70 µA, in order to provide the high intensity thermal neutron fields required for reactor instrument testing. The accelerator is capable of doing this, but the ion source does not last as long as in the past, and the accelerator tank has to be removed and the high voltage terminal inspected even for minor failures which prevent high beam current operation even though satisfactory low intensity beams can still be produced.

A study into the options for the future of accelerator-based capabilities has recently concluded. It reviewed the existing capabilities and requirements, compared the options for replacing the existing Van de Graaff and the logistics of installing a new accelerator.
References

Neutron Metrology Group, NPL
Publications since 2010


