A Portable Neutron Spectrometer for Dosimetry in Mixed Radiation Environments

by

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Abstract

Neutron spectrometry is necessary in the workplace around nuclear facilities in order to provide accurate assessments of neutron dose equivalent to those who are occupationally exposed. Knowledge of the neutron fluence spectrum may be used to correct for deficiencies in the energy dependent response of devices used as personal or area survey dosemeters. However, neutron spectrometry is not routinely performed largely due to the limitations of the existing instruments, such as long data acquisition times and complex data analysis to unfold the spectrum after the measurement.

A portable system that could indicate the approximate neutron energy spectrum in a short time would be extremely useful in radiation protection. A composite scintillator, consisting of lithium gadolinium borate (LGB) crystals in a plastic scintillator matrix, is being tested for this purpose. A prototype device based on this scintillator and digital pulse processing electronics has been calibrated using quasi-monoenergetic neutron fields at the low-scatter facility of the UK National Physical Laboratory (NPL). Energies selected were 144, 250, 565, 1400, 2500 and 5000 keV, with correction for scattered neutrons being made using the shadow cone technique. Measurements were also made in the NPL thermal neutron field. Pulse distributions collected with the digitiser in capture-gated mode are presented, and the fluence response of the spectrometer derived. For comparison, neutron spectra were also collected using the commercially available Microspec N-Probe from Bubble Technology Industries, which consists of an NE213 scintillator and a $^3$He proportional counter.
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Chapter 1

Introduction

1.1 Background
The aim of neutron dosimetry for radiation protection is to provide a method of assessing the magnitude of the detrimental health effects of exposure to neutron radiation. Two objectives must be achieved in order to achieve this:

1. a quantity must be identified that is a reasonable measure of the biological detriment caused by neutron radiation, and

2. a method must be found for the measurement of this quantity that is suitable for routine use in the workplace.

The first task is by far the more complicated as our understanding of the biological effects of ionising radiation in general, and neutron radiation in particular, is not complete. The second objective is easier to solve as it involves only the physics of neutron interaction with matter, our knowledge of which is rather more developed than it is for the biological response.

The purpose of a dosemeter then is to determine and record, as accurately as possible, the radiation dose received by the exposed person. Neutron dosimetry is difficult because no single instrument or dosemeter can measure over the entire range of neutron energies encountered. It is therefore important to have knowledge of not only the energy response of the neutron dosemeter of interest, but also knowledge of the neutron fluence energy spectrum in the area where the dosemeter is to be used. With
this knowledge, it may be possible to correct for the imperfect response of the
dosemeter and arrive at a better estimate of the received dose.

Neutron fluence spectra are the fundamental descriptors of neutron fields. From
fluence spectra, operational quantities such as personal and ambient dose equivalents
and fluence-averaged or dose-equivalent-averaged mean energies can be calculated.
These are the quantities that characterise a neutron fields from a radiation protection
perspective. Neutron fluence spectra can be used to predict the behaviour of a
dosemeter with a known energy response in that field.

Neutron fields in workplaces in the nuclear fuel cycle, in nuclear power generation,
and in areas near medical accelerators or where radionuclide sources are used, span
energies from thermal (∼0.025 eV) to 20 MeV. During routine operations in many
workplaces the neutron doses received by personnel are very low. Occasionally,
however, it may be necessary to enter areas where neutron doses are more significant
in order to carry out maintenance. It is important to know how the neutron dosemeter
will function in both environments.

Considering all these facts, there is obviously a motivation to perform neutron spectrometry in the workplace in order to characterise the neutron field for the provision of accurate dosimetry. In practice very few spectrometry measurements are carried out in the workplace due to the financial expense of the equipment, the time consuming nature of the measurements and the complexity of the data analysis.

1.2 Thesis Outline
This thesis explores the issues surrounding the use of neutron spectrometry for
radiation protection. The objective was to find a measurement technique that would
bypass some or all of the traditional obstacles to such measurements in the workplace.

In chapter 2 the basic physics of neutron interactions is explained and methods for exploiting this physics for neutron detection and spectroscopy are outlined. The design and operation of commonly used neutron dosimeters is covered here.

Chapter 3 describes the basis of radiation protection, the definitions of quantities and the relationships between them, with particular emphasis on neutron dosimetry. Typical neutron dosimeters, personal and area survey, are described and their energy response explained. Neutron spectra for a variety of workplaces are presented to show the requirement for spectrometry in radiation protection.

In terms of detection efficiency and energy resolution achieved, proton recoil detectors are superior to all other types of neutron spectrometer. The use of gas-filled proportional counters and organic scintillators for proton recoil spectrometry is further examined in chapter 4, which describes the development and operation of the Transportable Neutron Spectrometer (TNS) currently used by the MOD. Spectrum measurements with the TNS have been made in neutron fields at different irradiation facilities in the UK. The results are discussed and conclusions drawn about TNS performance. Recommendations for the improvement of the system as a whole are made.

Chapter 5 explains the principle of capture-gated neutron spectrometry and how this may have advantages for measurements in mixed radiation fields often encountered in the workplace. Capture-gated spectrometers are sometimes known as total absorption spectrometers, and it is this criterion for detection that avoids the need for spectrum unfolding. Recent developments in capture-gated spectrometry are reviewed and new scintillator material for neutron detection, lithium gadolinium borate (LGB), is introduced and its applicability for spectrometry discussed. Initial experiment results
are presented in this section, and simulations are performed using the Monte Carlo radiation transport code MCNP to predict the response of the LGB spectrometer.

Chapter 6 details the monoenergetic and thermal neutron irradiations of LGB performed at the National Physical Laboratory (NPL). These establish the detection efficiency and energy resolution of the LGB spectrometer. Fluence and dose responses for LGB are derived and compared with modelling predictions.

Chapter 7 describes measurements made with the LGB spectrometer in radiation fields around two reactors, the Low Flux Reactor (LFR), Petten, The Netherlands, and the NEPTUNE test reactor operated by Rolls Royce, Derby, UK. At NEPTUNE the objective was to characterise the neutron field at different positions in the reactor hall to identify location that would be representative of the workplace field around power-generating nuclear reactors.

Chapter 8 summarises the work performed and draws conclusions, suggesting routes for further development of the LGB spectrometer.
2.1 Neutron Interactions with Matter

Since the neutron is uncharged it is unaffected by the Coulomb force that dominates the interactions of electrons and heavy charged particles with matter. Neutrons interact with the nuclei of the material instead, with even neutrons of very low energy (eV or less) being able to initiate nuclear reactions. The neutron may emerge from these reactions with altered energy or direction, or may be absorbed completely by the nucleus with the emission of one or more secondary radiations. All neutron interactions may then be classified as belonging to one of two broad types: scattering or absorption.

2.1.1 Elastic Scattering

In an elastic scattering reaction, the incident neutron transfers some of its initial energy to the target nucleus. All the transferred energy appears as kinetic energy. There is no change in the internal energy state of the nucleus, and so no secondary radiations are emitted. The transfer of energy between the neutron and the nucleus can therefore be described simply by the laws of conservation of energy and momentum, and is determined by the scattering angle between them. Light elements are most efficient at slowing neutrons down by elastic scattering. Materials with high hydrogen content such as water, polythene, or concrete are therefore preferred.
2.1.2 Inelastic Scattering
If the energy of the incident neutron is sufficiently high, the target nucleus may be raised to an excited state in the collision. The excited nucleus usually returns to the ground state very quickly through emission of a gamma ray. This is known as inelastic scattering. Momentum is conserved but kinetic energy is not.

2.1.3 Radiative Capture
Radiative capture is usually the most probable reaction for slow neutrons. The incident neutron is absorbed by the target nucleus, which gains excitation energy equal to the binding energy of the neutron plus its initial kinetic energy. The compound nucleus formed will most likely lose this excitation energy by the emission of a gamma ray, returning to its ground state. The process is referred to as radiative capture, or \((n,\gamma)\) reaction.

2.1.4 Prompt Disintegration
Prompt disintegration is a term covering reactions where a nucleon or combination of nucleons is emitted from the compound nucleus formed by the absorption of a neutron. For fast neutrons, many reactions such as \((n,p)\), \((n,\alpha)\), or \((n,2n)\) become possible. Often there will be a threshold energy that the incident neutron must exceed for the reaction to take place. Slow neutrons can undergo these types of reactions in certain light nuclei, as we will discuss later. Another special case is nuclear fission, which can be initiated in certain isotopes by slow neutrons.
2.1.5 Cross-Section

For neutrons of a particular energy, the probability of undergoing a particular reaction with a target nucleus is a constant. This probability per nucleus is referred to as the microscopic cross-section, $\sigma$, and is measured in barns, where 1 barn is equal to $10^{-28}$ m$^2$. The cross-section is a function of neutron energy. A cross-section exists for each type of reaction possible, for example an elastic scattering cross-section $\sigma_{\text{elastic}}$, an inelastic scattering cross-section $\sigma_{\text{inelastic}}$, a radiative capture cross-section $\sigma_{\text{capture}}$ etc.

The total cross-section, $\sigma_{\text{total}}$, is just the sum of all the individual reaction cross-sections.

$$\sigma_{\text{total}} = \sigma_{\text{elastic}} + \sigma_{\text{inelastic}} + \sigma_{\text{capture}} + \ldots$$

(2.1)

Multiplying the microscopic cross-section $\sigma$ by the number of nuclei $N$ per unit volume results in the so-called “macroscopic” cross-section $\Sigma$.

$$\Sigma = N\sigma$$

(2.2)

The macroscopic cross-section has dimensions of inverse length ($m^{-1}$), and so $\Sigma$ has the physical interpretation of probability per unit path length for the specific process, which makes $\Sigma_{\text{total}}$ the probability per unit path length for any interaction to occur. The neutron mean free path $\lambda$ is the inverse of the total macroscopic cross-section.

$$\lambda = \frac{1}{\Sigma_{\text{total}}}$$

(2.3)
Introducing the concept of neutron flux (or more correctly neutron fluence rate) $\phi$, as being the number of neutrons crossing unit area per unit time ($m^{-2} s^{-1}$). Then for neutrons of a particular energy, the product of the neutron flux and the macroscopic cross-section for that reaction gives the reaction rate per unit volume

$$reaction\ rate\ per\ unit\ volume = \phi(E)\Sigma(E)$$ (2.4)

The cross-section of many nuclear reactions exhibit a steady decrease with increasing neutron energy that is inversely proportional to the neutron velocity. Such reactions are said to have a $1/v$ cross-section. Obviously, such reactions are more likely to occur with slow neutrons.

2.2 Neutron Sources

Radioisotopic sources recommended for routine calibration of neutron dosemeters are discussed by international standard ISO 8529 [ISO, 1989], which contains information on the neutron energy spectrum per unit source strength that can be expected from these sources. (The actual energy spectrum of emitted neutrons is somewhat dependent on details such as the source encapsulation.) Spectra for the sources mentioned in ISO 8529 are plotted in the following section. It should be noted that all these sources produce 'hard' spectra, that is the mean spectral energy is rather high, and are therefore not in general very representative of neutron fields encountered in the workplace.
2.2.1 Spontaneous Fission

Transuranic elements that have an appreciable spontaneous fission rate can be useful neutron sources as between 2 and 4 neutrons are emitted in each fission event.

The most common spontaneous fission neutron source is $^{252}$Cf, which has a half-life of 2.65 years and a yield of 0.116 neutrons s$^{-1}$ per Bq; that is $2.3 \times 10^{12}$ neutrons s$^{-1}$ per gram of $^{252}$Cf.

Neutrons produced in fission have a continuous neutron energy distribution that peaks between 1 and 2 MeV and can be approximated by the expression

$$\frac{dN}{dE} = E^{1/2} \exp(-E/T)$$  

(2.5)

where $T$ is a constant defining the most probable neutron energy for the isotope in question. $T$ equals 1.3 MeV for $^{252}$Cf. The disadvantages of $^{252}$Cf sources are the short half-life, which may mean the source needs replacing regularly, and moderately high gamma ray emission. Figure 2.1 shows the ISO reference spectrum for a $^{252}$Cf source.
Figure 2.1 Reference spectrum for a $^{252}\text{Cf}$ neutron source from ISO 8529.

The ISO also recommends the use of a heavy water ($\text{D}_2\text{O}$) moderated $^{252}\text{Cf}$ source for the calibration of neutron instruments. To produce the reference field, the $^{252}\text{Cf}$ source is contained within a 30 cm diameter sphere of heavy water with a shell of cadmium metal 1 mm thick. The purpose of the cadmium is to stop the emission of any neutrons that were thermalised in the heavy water. The $\text{D}_2\text{O}$ moderated $^{252}\text{Cf}$ spectrum is softer, that is it has a lower mean energy, than for bare $^{252}\text{Cf}$, which makes it somewhat more representative of real workplace spectra. The ISO reference spectrum for $\text{D}_2\text{O}$ moderated $^{252}\text{Cf}$ is shown in Figure 2.2.
2.2.2 Radioisotope (α,n) Sources

The stable isotope of beryllium, $^9\text{Be}$, has a neutron with a binding energy of just 1.7 MeV. That is easily overcome by the average energy of an α particle from radioactive decay. It is therefore possible to construct a self-contained neutron source by mixing an alpha-emitting isotope with the target beryllium (other isotopes can be chosen as the target, $^{10}\text{B}$ and $^{11}\text{B}$ for example, but the maximum yield is obtained with beryllium). The reaction proceeds as follows

$$^4\alpha+^9\text{Be} \rightarrow ^{12}\text{C}+^{1}\text{n}$$  \hspace{1cm} (2.6)
with a Q-value of +5.71 MeV. Suitably energetic and long-lived alpha-emitters include $^{226}$Ra (alpha energy 4.78 MeV, half-life 1602 years), $^{238}$Pu (alpha energy 5.48 MeV, half-life 87.4 years), and $^{241}$Am (alpha energy 5.48 MeV, half-life 433 years). The later has become the most commonly used isotope due to the combination of useful neutron yield (approximately 70 neutrons s$^{-1}$ per MBq of $^{241}$Am), ready availability and low gamma ray emission. The long source half-life means replacement will not be an issue, but can create disposal problems.

The energy spectra of neutrons emitted from all alpha/Be sources are similar, showing a complicated structure that can be explained in terms of the excitation state of the $^{12}$C product nucleus and the slowing down of $\alpha$ particles before the ($\alpha$,n) reaction. The distribution is strongly peaked between 3 and 8 MeV and neutrons in excess of 10 MeV are produced. The energy spectrum of the emitted spectrum is somewhat dependent on the size of the source, since neutron scattering, (n,2n) reactions in beryllium, or fission reactions will be more probable in larger sources. The mean energy for a $^{241}$Am/Be source is 4.5 MeV. Figure 2.2 shows the ISO reference spectrum for an $^{241}$Am/Be source.
2.2.3 Photoneutron Sources

A radioisotope that emits a sufficiently energetic gamma ray to liberate a neutron from a suitable target nucleus can form another source of neutrons. Only $^9$Be and $^2$H are of practical use as target nuclei for such photoneutron, or $(\gamma,n)$, sources.

\[ ^9\text{Be} + \gamma \rightarrow ^8\text{Be} + ^1\text{n} \]
Q-value: -1.666 MeV \hspace{2cm} (2.7)

\[ ^2\text{H} + \gamma \rightarrow ^1\text{H} + ^1\text{n} \]
Q-value: -2.226 MeV \hspace{2cm} (2.8)

An advantage of photoneutron sources is that the neutrons produced will be nearly monoenergetic. This follows from the fact that the gamma rays are monoenergetic.
and that the neutron energy is only slightly dependent on the angle of emission. For example, $^{24}\text{Na}$ emits a gamma ray of 2.76 MeV, which produces a neutron of mean energy 967 keV when absorbed by a $^{9}$Be nucleus. The disadvantage is the very large gamma ray activities necessary to produce useable neutron yields. In the example case given above, the yield is 34 neutrons s$^{-1}$ per MBq Ci of $^{24}\text{Na}$, with a half-life of only 15 hours.

2.2.4 Nuclear Reactions from Accelerated Charged Particles

Many nuclear reactions are available for neutron production, but, as only alpha particles are available from radioisotopes, reactions involving incident protons, deuterons or the like, require a particle accelerator. This has its disadvantages, but allows the creation of reasonably monoenergetic neutron beams of almost any energy by selection of the incident particle type and energy, of the target material, and the angle at which the emitted neutron is observed.

When energetic nuclear particles are incident on a target, there is a certain probability that a nuclear transformation will occur. A typical nuclear reaction may be written as

$$X + a \rightarrow Y + b \quad \text{or} \quad X(a, b)Y$$

where $a$ is the projectile (incident particle), $X$ the target nucleus, $b$ the emitted light particle and $Y$ the residual nucleus.
The total yield of neutrons produced in a nuclear reaction is proportional to the flux of incident particles and the number of target nuclei exposed to the incident beam, or

\[ F = N \sigma \phi A t \]  \hspace{1cm} (2.10)

where \( F \) is the neutron yield (per second), \( N \) is the number of target nuclei per centimetre cubed, \( \sigma \) is the reaction cross-section (in centimetres squared), \( \phi \) is the incident particle flux (per centimetre squared per second), \( A \) is the area of the particle beam, and \( t \) is the thickness of the target.

The production of neutrons through the bombardment of suitable target with isotopes of hydrogen is attractive since the net energy gain by the reaction is quite large.

For the D-D reactions,

\[ ^2H + ^2H \rightarrow ^3He + ^1n \hspace{1cm} \text{Q-value} = +3.269 \text{ MeV} \] \hspace{1cm} (2.11)

\[ ^2H + ^3H \rightarrow ^3He + ^1p \hspace{1cm} \text{Q-value} = +4.033 \text{ MeV} \] \hspace{1cm} (2.12)

and for the D-T reaction,

\[ ^2H + ^3H \rightarrow ^4He + ^1n \hspace{1cm} \text{Q-value} = +17.590 \text{ MeV} \] \hspace{1cm} (2.13)
The stopping and scattering of the deuterons in thick targets can cause a definite change in the neutron energy of emission angle. Below 500 keV deuteron energy, the neutron energy for the D-D and D-T reactions, with thin or thick targets, can be approximated by the simple formula

\[ E_n(E_d, \theta) = E_n \sum_{i=1}^{n} E_i \cos^i \theta \]  

As the incident deuteron energy does not need to be very high to overcome the Coulomb barrier in the D-D and D-T reactions, these can be used in small, sealed-tube "neutron generators". Deuterium ions are accelerated through a potential of 100-300 kV, which is small compared to the Q-value of the reaction and therefore the neutrons produced are nearly monoenergetic (around 3 MeV for the D-D reaction and 14 MeV for the D-T reaction). With a 1 mA beam current, a yield of \(10^9\) neutrons s\(^{-1}\) is possible from a deuterium target, and \(10^{11}\) neutrons s\(^{-1}\) from a tritium target.

Other charged particle induced reactions have negative Q-values or targets with a higher atomic number, and hence a higher Coulomb barrier. A large accelerator such as a cyclotron or Van der Graaff is needed to supply the much higher incident particle energy necessary to utilise such reactions. Commonly used reactions are \(^9\)Be(d,n), \(^7\)Li(p,n), and \(^3\)H(p,n).

### 3.2.5 Research Reactors

Nuclear reactors can produce extremely high neutron fluxes that can be accessed by placing samples in irradiation tubes near the reactor core, or by extracting a beam of...
neutrons through the reactor biological shielding. The energy spectrum will be a moderated fission spectrum that is a fission peak between 1 and 2 MeV, a thermal peak described by the Maxwell distribution, and a continuous slowing down distribution between them.

It is possible to produce a quasi-monoenergetic beam of neutrons from a reactor by use of a filter. The technique uses deep relative minima in the total cross-section of the filter material at distinct energies. Hence, neutrons of this energy are preferentially transmitted through the filter. Examples include minima at 2 keV in scandium, 24 keV in iron and aluminium, and 144 keV in silicon.

### 2.3 Reactions used for Neutron Detection

In this section methods for the detection of neutrons without any attempt to measure the neutron energy are discussed. The next section will deal with the subject of neutron spectrometry.

Important factors to be considered in the design of a neutron detector are the size and geometry of the detector, reaction cross-section and Q-value, the isotopic abundance of the target nuclide, and the range of the reaction products within the detector. Detector efficiency will be decided by the size of the reaction cross-section and the isotopic abundance of the target nuclide. The cross-section should be as large as possible to give good detection efficiency. For the same reason, the target nuclide should have a high natural isotopic abundance, or be readily available in an artificially enriched form. Since cross-sections for neutron interactions in matter are strongly energy dependent, often approximated as being inversely proportional to neutron energy, many reactions favour the detection of slow neutrons over fast.
Furthermore, measurements are often made in mixed fields of neutrons and gamma rays. Discrimination against gamma rays can be performed simply by pulse height analysis if the Q-value of the neutron capture reaction, and hence the kinetic energy of the reaction products, is high. Unfortunately, higher kinetic energy means longer range within the detector medium. If the active volume of the detector is not large compared to these ranges, some of the reaction products will not deposit their full energy and detector performance may suffer.

2.3.1 Boron Reaction – $^{10}\text{B}(n,\alpha)$

A widely used reaction in neutron detection is the $(n,\alpha)$ capture reaction in $^{10}\text{B}$, which has a cross-section of 3840 barns for thermal neutrons (0.025 eV) and shows a $1/\nu$ dependence.

\[
^{7}\text{Li} + ^{1}\text{n} \rightarrow ^{7}\text{Li} + ^{4}\text{He} \quad \text{Q-value} = +2.792 \text{ MeV (ground state)} \quad (2.15)
\]

\[
^{7}\text{Li} + ^{1}\text{n} \rightarrow ^{7}\text{Li}^* + ^{4}\text{He} \quad \text{Q-value} = +2.310 \text{ MeV (excited state)} \quad (2.16)
\]

The $^{7}\text{Li}$ nucleus is left in the excited state 94% of the time, but promptly returns to the ground state by emission of a 0.48 MeV gamma ray.

Probably the most common type of slow neutron detector is the boron trifluoride (BF$_3$) tube. This is a cylindrical proportional counter filled with BF$_3$ gas, usually highly enriched in $^{10}\text{B}$. 

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2.3.2 Helium Reaction – \(^{3}\text{He}(n,p)\)

Despite its high cost, \(^{3}\text{He}\) gas is also widely used in neutron detection because of its high thermal neutron (n,p) capture cross-section of 5330 barns. Again the cross-section shows a \(1/\nu\) dependence.

\[
^{3}\text{He} + ^{1}\text{n} \rightarrow ^{1}\text{H} + ^{1}\text{p} \quad \text{Q-value} = +0.764 \text{ MeV} \tag{2.17}
\]

As a proportional gas, \(^{3}\text{He}\) is superior to BF\(_{3}\), and so can be operated at much higher pressures. Together with the larger cross-section, this makes \(^{3}\text{He}\) counters preferable where maximum detection efficiency is required. However, the lower Q-value makes gamma ray discrimination harder.

2.3.3 Lithium Reaction – \(^{6}\text{Li}(n,\alpha)\)

Another common reaction for neutron detection is (n,\(\alpha\)) capture in \(^{6}\text{Li}\). The thermal neutron cross-section, at 940 barns, is considerably less than for the \(^{10}\text{B}\) reaction, but this must be weighed against the higher Q-value and the product tritium nucleus being in the ground state. The cross-section displays \(1/\nu\) dependence.

\[
^{6}\text{Li} + ^{1}\text{n} \rightarrow ^{3}\text{H} + ^{4}\alpha \quad \text{Q-value} = +4.78 \text{ MeV} \tag{2.18}
\]

No stable lithium-containing proportional gas exists, but there do exist scintillators that utilise lithium for neutron detection, for example, lithium iodide (LiI(Eu)), mixtures of lithium and ZnS(Ag) scintillator, and lithium-containing glasses.
Table 2.1 summarises the details of these common slow neutron detection reactions, while Figure 2.3 shows their cross-section.

Table 2.1 Reactions commonly used in neutron detection.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Reaction</th>
<th>Q-value (MeV)</th>
<th>Cross-section (barns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{10}$B</td>
<td>$^{10}$B(n,α)$^7$Li</td>
<td>+2.792 (ground state) +2.310 (excited state)</td>
<td>3840</td>
</tr>
<tr>
<td>$^3$He</td>
<td>$^3$He(n,p)$^1$H</td>
<td>+0.764</td>
<td>5330</td>
</tr>
<tr>
<td>$^6$Li</td>
<td>$^6$Li(n,α)$^3$H</td>
<td>+4.78</td>
<td>940</td>
</tr>
</tbody>
</table>

Figure 2.4 Cross-sections of the three most commonly used slow neutron detection reactions, data taken from ENDF-B/V1.
2.4 Fast Neutron Spectrometry

2.4.1 Neutron Moderation

All the neutron-induced reactions already discussed suffer from the reduction in cross-section with increasing neutron energy when used for the detection of fast neutrons. One method of improving the fast neutron detection probability for detectors based on these reactions is to surround the detector with a layer of moderating material so that an incident fast neutron might lose a significant fraction of its energy before arriving at the detector. This is the principle used in most neutron survey instruments such as the spherical moderator design of Leake [Leake, 1966], or the cylindrical moderator design of Andersson and Braun [Andersson, 1964].

This approach can also be used to determine the neutron energy spectrum, as first described by Bramblett, Ewing, and Bonner [Bramblett, 1960], who used a small \text{Li}_2\text{Eu} scintillator with a series of polythene moderating spheres of different diameters. These are now generally known as Bonner spheres after one of the authors, or multisphere spectrometers. Each sphere has a unique energy dependent response curve that may be measured experimentally, but is now more likely to be determined by neutron transport calculations. From the count rate measured by each individual sphere, and using knowledge of their response, it is possible to “unfold” the energy distribution of the neutron field. The process of unfolding spectra will be discussed later. Figure shows the response function for a set of Bonner spheres belonging to the UK National Physical Laboratory taken from the SPKTBIB catalogue [SPKTBIB, 1996]. The neutron detector used is a spherical $^3\text{He}$ proportional counter (Centronics
The responses were calculated with the Monte Carlo radiation transport code MCNP and normalised to experimental measurements.

![Response functions for the NPL Bonner sphere set based on a $^3$He detector.](image)

As the response functions are not sharply peaked and overlap considerably, the resolution of the unfolded spectrum is not very high. However, this is not a significant problem if the results are used to determine neutron dose as the fluence-to-dose conversion coefficients vary smoothly as a function of energy.

### 2.4.2 Fast Neutron Scattering

Fast neutron spectrometry can be performed using the simplest of reactions: elastic scattering of neutrons by light nuclei. The scattered neutron transfers some fraction of its energy to the target nucleus, which then recoils from the collision producing
ionisation in the detector medium. Hydrogen is the most useful target and, as the hydrogen nucleus is merely a proton, detectors based on this reaction are called proton recoil detectors.

Conservation of energy and momentum in the centre-of-mass coordinate system leads to the following equation for the energy of the recoil nucleus

\[
E_R = \frac{2A}{(1 + A)^2} (1 - \cos \Theta) E_n
\]  

(2.19)

where \( A \) is the mass of the nucleus, \( E_n \) is the incident neutron energy, and \( \Theta \) is the neutron scattering angle in the centre-of-mass system. We convert to the laboratory coordinate system, where the target nucleus is initially at rest, using the transformation

\[
\cos \theta = \sqrt{\frac{1 - \cos \Theta}{2}}
\]  

(2.20)

This gives the final relation for the recoil nucleus energy as

\[
E_R = \frac{4A}{(1 + A)^2} (\cos^2 \Theta) E_n
\]  

(2.21)
The energy transferred to the recoil nucleus is therefore uniquely determined by the scattering angle, and we see that the neutron can impart anything from nearly zero (in a grazing collision) up to some maximum

\[
E_{r\text{max}} = \frac{4A}{(1 + A)^2} E_n
\]  

(2.22)

(in a head-on collision). It should be noted that the maximum recoil energy decreases with increasing target nucleus mass. For hydrogen nuclei, since the neutron and the proton are of almost equivalent mass, the neutron can transfer all its energy in a single collision. Table gives the maximum fraction of incident neutron energy that can be transferred in a single collision for a variety of target nuclei, demonstrating why light nuclei only are of interest in recoil detectors.

Table 2.2 Maximum fractional recoil energy for neutron elastic scattering from different nuclei.

<table>
<thead>
<tr>
<th>Target Nucleus</th>
<th>Atomic Mass, A</th>
<th>Maximum Fractional Recoil Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^1)H</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>(^3)H</td>
<td>2</td>
<td>0.889</td>
</tr>
<tr>
<td>(^4)He</td>
<td>3</td>
<td>0.750</td>
</tr>
<tr>
<td>(^4)He</td>
<td>4</td>
<td>0.640</td>
</tr>
<tr>
<td>(^12)C</td>
<td>12</td>
<td>0.284</td>
</tr>
<tr>
<td>(^16)O</td>
<td>16</td>
<td>0.221</td>
</tr>
</tbody>
</table>

Hydrogen has another advantage as a recoil detector medium in that the elastic scattering reaction is isotropic in the centre-of-mass coordinate system over the energy range \(E_n < 10\) MeV. Therefore, the expected proton recoil energy distribution is a simple rectangular shape extending from zero to the full incident neutron energy.
Detection Efficiency

For a device based on the nuclear recoil principle where only one species of nucleus is present, the intrinsic efficiency is given by

\[ \varepsilon = 1 - \exp(-N\sigma_s d) \] (2.23)

where \( N \) is the number of target nuclei, \( \sigma_s \) is the elastic scattering cross-section for these nuclei, and \( d \) is the path length through the detector for incident neutrons. Proton recoil detectors often have carbon nuclei present in combination to hydrogen, and the effect of neutron scattering from carbon must also be accounted for. If we neglect the possibility of multiple scattering, the counting efficiency of such a detector is given by

\[ \varepsilon = \frac{N_H \sigma_{H}}{N_H \sigma_H + N_C \sigma_C} \left\{ 1 - \exp\left[-\left(N_H \sigma_{H} + N_C \sigma_C\right) d\right] \right\} \] (2.24)

where the subscripts H and C refer to the separate values of the quantities defined above for hydrogen and carbon. Cross-sections for neutron elastic scattering from hydrogen and carbon are shown in Figure 2.6.
2.5 Neutron Scintillation Detectors

2.5.1 Scintillation Mechanism in Organic Scintillators

The fluorescence mechanism in organic substances is based on transitions in the energy level structure of individual molecules. This means that, unlike inorganic scintillators, the fluorescence can be observed whatever the physical state of the substance.

Many practical organic scintillators come from the family of organic molecules having a π-electron structure. Figure 2.7 illustrates the general properties of the π-electron structure. A molecule absorbs energy by exciting the electron configuration into any one of the numerous excited states. Singlet states (spin 0) are labelled as $S_0$,
$S_1, S_2 \ldots$ in the figure, while triplet states (spin 1) are labelled $T_1, T_2, T_3 \ldots$. A suitable organic scintillator has an energy spacing of 3 – 4 eV between the $S_0$ and $S_1$ states, with narrower spacing between the higher states. Each state is further subdivided into a series of finely spaced levels (of the order of 0.15 eV) corresponding to the vibrational states of the molecule. A second subscript is used to denote these sublevels, for example $S_{100}$ denotes the lowest vibrational state of the electronic ground state. [Knoll, 1989]

Compared to thermal energies (0.025 eV), the spacing of these vibrational states is quite large. Therefore, at room temperature, almost all the molecules will be in the $S_{10}$ state. Molecules will absorb kinetic energy from a passing charged particle by promoting electrons into the higher states. The higher singlet states will quickly (in the order of picoseconds) de-excite down to the $S_1$ electronic level through radiationless internal conversion. Any remaining excess vibrational energy (e.g. $S_{11}$ or $S_{12}$) will also be quickly lost through interactions with neighbouring molecules. The result is a population of molecules in the $S_{10}$ state is formed in a very short time, and the principle scintillation light comes from transitions from the $S_{10}$ state and the electronic ground state $S_0$. This is termed the prompt fluorescence. The prompt fluorescence intensity at time $t$ following excitation is given by

$$I = I_0 \ e^{-\tau t} \quad (2.25)$$

where $\tau$ is the fluorescence decay time for the $S_{10}$ level. In most organic scintillators $\tau$ is only a few nanoseconds, which gives them excellent timing properties.
The triplet states can become populated through a transition called intersystem crossing. The lifetime of the triplet states is long compared to the singlet states, perhaps as much as $10^{-3}$ seconds, and so radiation emitted in transitions from $T_1$ to $S_0$ appears as delayed light. This is termed phosphorescence. Since the $T_1$ state lies below the $S_1$ state, the wavelength of the phosphorescent light will be longer than the fluorescent. Some molecules in the $T_1$ state may be excited into the $S_1$ state again, with the subsequent emission of delayed fluorescent light.

The energy level diagram also explains how organic scintillators can be transparent to their own light. All the fluorescence transitions, with the exception of $S_{10}$ to $S_{00}$, are lower in energy than the minimum excitation energy, and so there is little overlap between the absorption and emission spectra, and therefore little self-absorption.

The fraction of all incident charged particle energy that is converted into measurable light is called the scintillation efficiency. There are other modes of deexcitation available to excited molecules that do not involve the emission of light. All such processes are referred to as quenching. Impurities in the scintillator are an important source of quenching, for example the presence of dissolved oxygen in liquid organic scintillators.
2.5.2 Types of Organic Scintillator

Pure Organic Crystals

An example of a pure organic scintillator is stilbene. Large crystals are expensive and liable to damage from thermal and mechanical shock. The light output of recoil protons in stilbene is dependent on the direction of the charged particle track with respect to the crystal axis, known as the anisotropy effect. The anisotropy must be taken into account during the calculation of detector response functions for use in spectrum unfolding.
Liquid Organic Solutions

Dissolving an organic scintillator in a hydrogen-rich solvent can produce a very efficient neutron detector working by the proton recoil method. Excitation energy is readily transferred between molecules before deexcitation occurs, and can pass by this mechanism from the bulk solvent to the one of the efficient scintillator molecules. It is common to find a wavelength shifting material added to the mixture also, either to minimise self-absorption or to match the spectral response of the photomultiplier tube. This is achieved by absorbing the primary scintillation light and re-emitting it at a longer wavelength.

The solvent component is often very inexpensive, making liquid scintillator very cost effective in large volume applications. There is no anisotropy effect in a liquid scintillator, which simplifies the unfolding process compared to stilbene.

As dissolved oxygen greatly reduces the scintillation efficiency, the scintillator must be sealed in a container from which most of the oxygen has been purged. Liquid scintillator mixtures are available commercially pre-sealed in glass containers or aluminium with a glass window for coupling to the PMT. A common example of an organic liquid scintillator is NE213 by Nuclear Enterprises (now manufactured as BC-501A by Bicron).

Recipes for efficient scintillators have largely been developed through experimentation. About 0.5 per cent of fluorescent compound is dissolved in the bulk solvent, usually an aromatic compound such as toluene or xylene. The wavelength shifter, if required, is present at around ten to one hundred parts per million. Of course, the solution must be stable, clear and colourless. The passage of ionising...
radiation through the organic liquid first excites the bulk solvent. In a suitably chosen solvent this is a relatively efficient process. It results in a distribution of excited energy states that are relatively stable against decay by either radiation or solvent quenching. The lifetime for non-radiative transfer of energy is very short, however. Thus the energy travels between molecules until it is trapped in the scintillating solute. In competition with internal and self-quenching the solute radiates a portion of the energy as light. If a wavelength shifter is present in the solution it will strongly absorb the scintillator light and reemit it at a longer wavelength. This entire process usually takes place in a time of the order of a few nanoseconds. Proper names and the common abbreviations for some of the most widely used scintillator substances are listed in Table 2.3.

<table>
<thead>
<tr>
<th>Name</th>
<th>Compound</th>
</tr>
</thead>
<tbody>
<tr>
<td>α-NPO</td>
<td>2-(1-naphthyl)-5-phenyloxazole</td>
</tr>
<tr>
<td>PPO</td>
<td>2,5 diphenyloxazole</td>
</tr>
<tr>
<td>POPOP</td>
<td>1,4 di-[2-(5 phenyloxazolyl)]-benzene</td>
</tr>
<tr>
<td>BBO</td>
<td>2,5 di-(4-biphenyl)-oxazole</td>
</tr>
<tr>
<td>PBD</td>
<td>2-phenyl-5-(4 biphenyl)-1,3,4 oxadiazole</td>
</tr>
<tr>
<td>Octoate</td>
<td>2-ethyl-hexanoate</td>
</tr>
</tbody>
</table>

**Plastic Scintillators**

If the solvent into which the organic scintillator is dissolved can be polymerised, it is possible to make a solid solution having similar detection properties as the liquid scintillator. Again, plastic scintillators are a very cost effective solution for large volume detectors, and have the added advantage of ruggedness. In addition they can be fabricated in novel, application specific shapes, such as rods, cylinders or flat...
sheets. A commonly used example of a plastic scintillator is NE102 from Nuclear Enterprises, which is now manufactured as BC-404 by Bicron.

2.5.3 Light Output of Organic Scintillators

Only a fraction of the kinetic energy lost by a charged particle in the scintillator will be converted into fluorescent light, the remainder being dissipated non-radiatively in molecular vibrations and heat. The fraction of the particle energy converted (the scintillation efficiency) depends on the particle type and its energy. For electrons, the response is linear to particle energies above 125 keV. Heavy charged particles, such as protons or alpha particles produce less scintillation light than electrons of equivalent energy, and show a non-linearity up to much higher energies.

The response of organic scintillators to charged particles may be described by a relation between the fluorescent energy emitted per unit path length, dL/dx, and the specific energy loss for the charged particle, dE/dx, called Birk’s formula. An important assumption is that highly ionising particles cause damage to molecules along their tracks, which leads to quenching and a lowering of scintillation efficiency. The density of damaged molecules along the track is taken to be directly proportional to the ionisation density. Also assuming that, in the absence of quenching, the light yield is proportional to energy loss, Birk’s formula may be written as

$$\frac{dL}{dx} = \frac{S}{1 + kB} \frac{dE}{dx}$$  \hspace{1cm} (2.26)
where $S$ is the normal scintillation efficiency, and $B$ and $k$ are proportionality constants representing the degree of molecular damage and the fraction that will lead to quenching.

For reliable spectrometry it is vital to know the pulse height function $L(E)$ and the pulse height resolution function $dL/L(L)$, which are characteristic of each individual detector. Measurements with photon sources in the energy range $40 \text{ keV} < E_r < 1.6 \text{ MeV}$ yield channel positions $P_c$ of the Compton energies $E_c$ that follow the linear relationship

$$P_c = G(E_c - 5\text{ keV}) \quad (2.27)$$

with the calibration factor $G$ in terms of channels per electron energy and the light output $L = (E_c - 5 \text{ keV})$. It is possible to determine $G$ to an uncertainty of less than $0.5\%$ in this energy range. A linear extrapolation is generally used, although it has been shown that light output from electrons does become nonlinear at higher energies. The light output functions for recoil protons and alpha-particles are strongly nonlinear, and are dependent on the size and construction of the scintillator. It is therefore advisable to determine the light output for individual detectors to ensure accuracy in the unfolded spectra. Figure 2.8 shows the light output function for protons, alpha-particles and carbon nuclei in NE102 plastic scintillator.
Figure 2.8 Light output versus particle energy for recoil protons, alpha-particles and carbon nuclei in the plastic scintillator NE102.

The pulse height resolution $dL/L$ as a function of the pulse height $L$ is derived from the calibration measurements with photon sources and monoenergetic neutrons. The data points obtained for the light output corresponding to the Compton energy and the maximum recoil proton energy can generally be parameterised according to

$$
\frac{dL}{L} = \left( A^2 + \frac{B^2}{L} + \frac{C^2}{L^2} \right)^{1/2}
$$

(3.28)

where the coefficients represent an uncorrelated superposition of contributions due to:

1. the geometry effect of the position dependent light transmission,
2. the statistical variation of the production and multiplication of photoelectrons.

3. the noise of the analog dynode chain of the photomultiplier.

![Figure 2.9 Example of the resolution function dL/L for an organic scintillator.](image)

2.5.4 Photon Spectrometry

Organic scintillators respond to photons as well as neutrons, and so can be used to measure gamma-ray spectra. The response to photons is easily studied with common radionuclide sources having a few, well separated, photon energies up to 2 MeV, e.g. $^{137}$Cs, $^{22}$Na, $^{65}$Zn, $^{60}$Co and $^{88}$Y. Higher photon energies can be produced through nuclear reactions, e.g. the mixed n-$\gamma$ field of $^{241}$Am/$^{9}$Be(\alpha,n)$^{13}$C $^\ast$ (4.44 MeV) with pulse shape discrimination to select only the gamma rays. However, being made up of low atomic number elements, photon interaction in organic scintillators is almost entirely through Compton scattering and so the measured pulse height distribution has
no full energy peak, only a Compton edge and continuum. Obtaining the incident
gamma-ray spectrum therefore requires the use of unfolding techniques.

Photon response functions can be simulated using computational methods and much
work in this area has been done at the Physikalisch Technische Bundesanstalt (PTB),
the German national standard laboratory. For photon energies below 2 MeV and linear
dimensions of the detector higher than the mean electron range, the GRESP code
[Dietze, 1982a] may still be used. However, the more recent PHRESP code [Novotny,
1997], based on the EGS4 program [Nelson, 1995] in combination with the PRESTA
routine [Bielajew, 1987], represents a considerable improvement. The simulated
spectra can then be folded with a Gaussian function representing the pulse height
resolution to yield a smooth distribution that can be fitted to the experimental data. In
this way, the calibration factor G and the pulse height resolution at the Compton edge
are determined. More importantly, the factor used to scale the simulated spectrum to
the experimental one is the photon fluence $\Phi_{\gamma}$.

### 2.5.5 Neutron Spectrometry

The response to mono-energetic neutrons is not so easily determined, requiring the
use of a particle accelerator. Two approaches can be taken; either produce a series of
mono-energetic fields through the use of different neutron producing targets and
charged particle beam energies, or use the time-of-flight method with a nanosecond-
pulsed charged particle beam.

Again, the experimental measurements are used to support computer simulations of
the detector response. For neutron energies up to 20 MeV the NRESP code [Dietze,
1982] has been used very successfully, while the SCINFUL code [Dickens, 1988]
allows simulations up to 80 MeV. Neutron scattering from hydrogen make the
dominant contribution to the response function. Despite the fact that a neutron may
transfer up to 24% of its energy, the contribution from recoil carbon nuclei is
negligible due to the very low light output of heavy ions in the scintillator. Carbon
scattering affects the response function instead because of the probability of multiple
interactions on carbon and then hydrogen. It is therefore necessary to simulate the
neutron transport as realistically as possible. As before, the fit of the simulated
response to the measured spectrum yields the light output in terms of equivalent
electron energy, the pulse height resolution for the maximum proton recoil energy and
the neutron fluence $\Phi_n$.

Since the proton recoil edge in the response function is closely related to the
corresponding neutron energy ($E_p = E_n$), the proton light output function should be
determined as accurately as possible to establish a reliable energy scale for the
unfolded neutron spectrum.

From the simulations and fitting procedure the response matrix for the detector may
be calculated in any group structure required. The unfolding of the measured pulse
height spectra is performed on the basis of these calculated response functions.

2.5.6 Pulse Shape Discrimination

In most situations the neutron spectrum measurement will take place in a mixed
radiation field, i.e. there will be gamma-rays present as well. Since the organic
scintillator is responsive to both neutrons and photons a method must be found of
separating signals caused by gamma-rays from the signals caused by neutrons. The
difference in the light output characteristics of organic scintillators for heavy charged
particles and electrons allows this to be done by examining the shape of pulses from the photomultiplier.

There are two dominant techniques used in pulse shape discrimination with radiation detectors: zero-crossing and charge comparison. The former relies on passing the pulse through a shaping network to produce a bipolar shape, for which the time the pulse crosses zero is independent of pulse amplitude, but dependent on pulse shape and rise time instead. In charge comparison independent measurements are made of the integrated charge over two different time regions of the pulse. The Link Analytical 5010 and 5020 models [Adams, 1978] used in the TNS systems, are based on charge comparison, using the difference between a long and a short pulse integrating gate to make the distinction between gamma ray and neutron induced events. The Link PSD is no longer under manufacture, although there are still many in service in laboratories worldwide.

Experimental [Wolski, 1995] and theoretical [Cao, 1998] comparisons have been made between the two techniques. Both find slightly in favour of the zero-crossing method, but Cao and Miller qualify this with the observation that charge comparison is easier to optimise and suffers less from electronic noise.

Alternatively, a dual parameter approach to pulse shape discrimination can be taken. Information must then be collected on both the amplitude and rise time of each signal pulse, and this information placed in an array of memory locations in which one axis corresponds to pulse amplitude and the other to rise time. The resulting distribution is similar in form to the output of the Link 5020 in its display mode, being a two-dimensional pattern representing the frequency of particular combinations of amplitude and rise time.
The quality of n/γ discrimination may be measured by defining a figure-of-merit FOM

\[
FOM (L) = \frac{DIST_{\gamma}}{FWHM_{\gamma} + FWHM_n} \quad (2.29)
\]

Where DIST is the separation distance and FWHM\(_{\gamma}\) and FWHM\(_n\) are the full widths at half maximum of the photon and neutron pulse shape spectra correlated to the pulse height \(L\).

### 2.6 Neutron Spectrum Unfolding

Neutron spectrometry does not usually provide details of the incident neutron fluence spectrum directly. Rather, the measurements provide numerical data that has to be unfolded prior to yielding the required energy distribution. Unfolding problems can generally be separated into two cases: multi-channel unfolding and few channel unfolding. In multi-group unfolding, the number of measurement channels, \(n\), is of the same order as the desired number of energy groups, \(m\), in the unfolded spectrum. For example, multi-channel unfolding would be used with an NE213 scintillator, with the number of measurement channels, \(n\), equal to the number of channels in the pulse height distribution recorded on the MCA. In few-channel unfolding, the number of measurement channels is much less than the number of energy groups in the unfolded spectrum. An example would be unfolding data from a set of Bonner spheres, where \(n\) equals the number of spheres in the set.
For the multi-channel case, the fundamental equation may be written as:

\[ \Psi(E') = \int_0^{\infty} R(E', E) \Phi(E) dE + \epsilon(E') \tag{2.30} \]

where \( \Psi(E') \) is the pulse height distribution measured by the detector, \( R(E', E) \) is the detector response function, \( \Phi(E) \) is the incident neutron energy spectrum and \( \epsilon(E') \) is an error term. In the discrete form, the integral is replaced by a sum and the number of bins used in the discretisation of \( E \) is of the same order as the number of bins used in the discretisation of \( E' \).

In the few-channel case the equation may be written as:

\[ N_k = \int_0^{\infty} R_k(E) \Phi(E) dE + \epsilon_k, \quad k = 1, \ldots, n \tag{2.31} \]

where \( N_k \) is the set of Bonner sphere readings, \( R_k(E) \) is the response function for the \( k^{th} \) sphere, \( \Phi(E) \) is the incident neutron energy spectrum and \( \epsilon_k \) is the error term. In the discrete form, the integral is replaced by a sum and the number of bins used in the discretisation of \( E \) is much larger than the number of spheres, \( n \).

In principle, the mathematical solution, \( \Phi \), results from an unfolding procedure based on matrix inversion, but obtaining a unique and physically significant solution requires taking into account errors and statistical fluctuations in input data, response functions and detector readings, as well as rounding errors in the matrix inversion process itself, leading to large oscillations and particularly negative values in the solution neutron spectrum. To obtain a solution it is necessary to introduce additional
information to complement the measurement data. This additional information depends on the formulation of the unfolding method, but a good unfolding method should produce a unique solution that is not unphysical, not unstable and provides some estimate of the uncertainty.

In most real Bonner sphere unfolding problems, the number of energy intervals exceeds the number of detectors, i.e. \( M < N \). As a result, the system of equations is ill-conditioned (or under-determined), and the alternative idea is therefore to look for an ‘acceptable’ solution \( \Phi' \) which minimises the deviations between the measured values \( M_i \) and the values \( M_i' \) calculated from the solution \( \Phi' \), i.e. which minimises the expression

\[
\sum_{i=1}^{M} \left( \frac{M_i - M_i'}{M_i} \right)^2
\]  

Un fortunately, this least-squares condition does not solve the other problems mentioned. Several methods have been developed in different codes where, thanks to some a priori information, solutions are forced to have certain additional properties, including non-negativity and some ‘degree of smoothness’. The following procedures are found in the literature.

Regularisation. The general condition of minimising the above equation is modified by additional terms taking into account several constraint conditions such as least mean squares errors, a certain degree of smoothness and some similarity to an
assumed spectrum. This method is applied in the codes LOUHI 78, LSL-M2 and STAY'SL.

Iteration. From a guessed spectrum, detector readings $M_i$ are calculated and iteratively compared to the actual detector readings $M_i$ to modify the guessed spectrum in an appropriate way. The iterative procedure is stopped as soon as the similarity criteria are fulfilled. In the SAND-II code, this method has the advantage of the simplicity of computation because a response matrix inversion is not required. Several versions of this code have been developed which differ by the algorithm used in the sequence of iterations.

Parametrisation. In this technique, it is assumed that the solution spectrum $\Phi$ can be expressed as the sum of several ($N_m$) functions $\psi_i$ ('model spectra') of known shapes but unknown amplitudes $a_i$:

$$\Phi_{\psi_j} \Delta E_j = \sum_{i=1}^{N_m} a_i \psi_{\psi_j} \Delta E_j, \quad j = 1, ..., N \quad (2.33)$$

Combining this with the discrete form of the unfolding problem, and assuming that the responses of detectors to functions $\psi_i$ are also known, a system of equations is derived which has to be solved for determining the coefficients $\alpha_i$, with similar problems as discussed previously, i.e. ill-conditioning and instability due to experimental statistics.

Monte Carlo unfolding. If sufficient a priori information on the particle spectrum is not available, the least-square codes tend to perform poorly. This drawback was overcome by the development of unfolding codes utilising Monte Carlo techniques.
The MIEKE code, the SPECAN/UNFANA code, and the MAXED code are based on the principle of maximum entropy. UNFANA and MAXED can be used as an alternative analytical approach to the Monte Carlo procedure.

In addition to the aforementioned codes, several computer programs have been developed for unfolding ‘few channels’ (e.g. from multi-sphere systems or threshold detectors), and/or multi-channel pulse-height distributions (e.g. from proportional counters or NE213 scintillators). The HEPRO package by Matzke collects a series of computer codes elaborated and tested in specialised laboratories. The package contains the ‘least-squares type’ codes DIFMAZ and GRAVEL, a modified version of SAND-II codes. Other codes, such as BUNKI, and FERDOR/FORIST also belong to this type of code, for ‘few-channel’ and ‘multi-channel’ unfolding, respectively. A recent overview of unfolding codes has been given by Matzke.
Chapter 3

Neutron Dosimetry

3.1 Quantities and Definitions

Quantifying the effect of radiation on the human body is an extremely complex task that is under constant refinement. Radiation protection recommendations require that combined knowledge of the biological effects of radiation, radiation measurement and nuclear physics be framed in a clear and consistent set of protocols that can form the basis of national and international legislation. The quantities involved have therefore evolved gradually over the years as new and improved data becomes available.

Basic quantities to be used in radiation protection are defined by the International Commission for Radiological Protection (ICRP) and the International Commission on Radiation Units and Measurements (ICRU), and their recommendations form the basis for UK law [HSE, 1999].

![Diagram showing the relationship between radiation protection quantities defined by the ICRP and ICRU.](image-url)

Figure 3.1 Diagram showing the relationship between radiation protection quantities defined by the ICRP and ICRU.
3.2 Basic Physical Quantities

Basic physical quantities for characterisation of radiation fields must be defined at every point in the field and be directly measurable. Three basic physical quantities are defined by the ICRU: fluence, kerma and absorbed dose.

3.2.1 Fluence

The fluence, $\Phi$, is the quotient of $dN$ by $da$, where $dN$ is the number of particles incident on a sphere of cross-sectional area $da$.

$$\Phi = \frac{dN}{da}$$  \hspace{1cm} (3.1)

The unit of fluence is $m^{-2}$.

3.2.2 Kerma

The kerma, $K$, is the quotient $dE_r$ by $dm$, where $dE_r$ is the sum of the initial kinetic energies of all charged ionising particles liberated by uncharged ionising particles in a volume element of mass $dm$,

$$K = \frac{dE_r}{dm}$$  \hspace{1cm} (3.2)

The unit of kerma is the joule per kilogram ($J \text{ kg}^{-1}$), and is given the special name gray (Gy).
3.2.3 Absorbed Dose

The absorbed dose, $D$, is the quotient of $d\bar{E}$ by $dm$, where $d\bar{E}$ is the mean energy imparted by ionising radiation to matter of mass $dm$.

$$ D = \frac{d\bar{E}}{dm} $$ (3.3)

The unit of absorbed dose is the joule per kilogram ($J/kg$), and is given the special name gray (Gy).

3.3 Protection Quantities

The basic quantities are not suitable for use in dose limitation purposes because different radiations (e.g. photons, electrons, neutrons, protons) cause different biological effects at the same dose level, and different organs and tissues in the human body have different radio-sensitivities. Instead, the ICRP have defined a pair of protection quantities to take account of these two issues: equivalent dose, for individual organs and tissues; and effective dose, for the whole body.

3.3.1 Equivalent Dose

The equivalent dose, $H_{T,R}$, is the absorbed dose in an organ or tissue multiplied by the relevant radiation weighting factor,

$$ H_{T,R} = w_R D_{T,R} $$ (3.4)

where $D_{T,R}$ is the absorbed dose averaged over the tissue or organ $T$, due to radiation type $R$, and $w_R$ is the radiation weighting factor for radiation type $R$. The radiation weighting factor takes account of the greater biological damage caused by high LET
radiations, such as neutrons, protons or alpha particles, compared to low LET radiations, like photons and electrons. Values are given in Table 3.1

<table>
<thead>
<tr>
<th>Radiation type, $R$</th>
<th>Radiation weight factor, $w_R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photons, all energies</td>
<td>1</td>
</tr>
<tr>
<td>Electrons and muons, all energies</td>
<td>1</td>
</tr>
<tr>
<td>Neutrons</td>
<td></td>
</tr>
<tr>
<td>&lt; 10 keV</td>
<td>5</td>
</tr>
<tr>
<td>10 keV to 100 keV</td>
<td>10</td>
</tr>
<tr>
<td>100 keV to 2 MeV</td>
<td>20</td>
</tr>
<tr>
<td>2 MeV to 20 MeV</td>
<td>10</td>
</tr>
<tr>
<td>&gt; 20 MeV</td>
<td>5</td>
</tr>
<tr>
<td>Protons, other than proton recoils, energy &gt; 2 MeV</td>
<td>5</td>
</tr>
<tr>
<td>Alpha particles, fission fragments, heavy nuclei</td>
<td>20</td>
</tr>
</tbody>
</table>

Table 3.1 Radiation weighting factors defined in ICRP 60 for different types of radiation.

When the radiation field is composed of more than one type of radiation, the equivalent dose is the sum of the different components:

$$H_T = \sum w_x D_{T,R}$$  \hspace{1cm} (3.5)

The unit of equivalent dose is the joule per kilogram ($J \text{ kg}^{-1}$), which is given the special name **sievert** (Sv).
3.3.2 Effective Dose

The effective dose, \( E \), is the summation of the equivalent doses in a particular tissue multiplied by the appropriate tissue weighting factor.

\[
E = \sum_T w_T H_T
\]  

(3.6)

where \( H_T \) is the equivalent dose in tissue \( T \) and \( w_T \) is the tissue weighting factor for tissue \( T \), which accounts for the relative stochastic detriment resulting from radiation exposure of that tissue. The unit of effective dose is the joule per kilogram (J kg\(^{-1}\)), which is given the special name Sievert (Sv).

Effective dose requires consideration of the geometry of irradiation, and so separate sets of conversion coefficients are defined for anterior-posterior (AP), posterior-anterior (PA), right lateral (RLAT), left lateral (LLAT), rotational (ROT) and isotropic (ISO) irradiation conditions. (Rotational geometry describes a field where the body is irradiated by a broad beam and rotated about its vertical axis.) Figure 3.2 shows effective dose as a function of energy plotted for the different types of irradiation geometry.
3.4 Operational Quantities

The protection quantities outlined above are not in fact directly measurable. In principle, they can be calculated if the irradiation geometry and radiation field characteristics are known with sufficient accuracy. For routine monitoring, the ICRU has instead defined a set of operational quantities that can be measured and which give a reasonable estimate of the protection quantities. For neutron radiation the relevant operational quantities are personal dose equivalent, $H_p(10)$, directional dose equivalent, $H'(d,\Omega)$, and ambient dose equivalent, $H^*(10)$. All three operational quantities are based on the concept of dose equivalent, $H$ (not the same as equivalent dose), which is defined as the product of $Q$ and $D$ at a point in tissue, where $D$ is the absorbed dose and $Q$ is the quality factor at that point.
Again the unit of dose equivalent is the joule per kilogram (J kg⁻¹), or Sievert (Sv).

The quality factor Q is defined as a function of the unrestricted linear energy transfer (or linear collision stopping power), L, in water. For a charged particle, L is the quotient of dE by dl, where dE is the mean energy lost by the particle, due to collisions with electrons, in traversing a distance dl through a material.

\[ L = \frac{dE}{dl} \]  

L is measured in joules per meter (J m⁻¹), but it is usual for it to be expressed in eV per some convenient unit of length, such as keV µm⁻¹. The ICRP specified the Q(L) function for water in ICRP Publication 60 [ICRP. 1991]. Then in 1993 the ICRU published new data on the stopping powers of protons and alpha particles, which required an updating of the Q(L) relationship. The results of this update are included in ICRP 74 [ICRP. 1996] and ICRU 57 [ICRU, 1998].

### 3.4.1 Supplementary Definitions

The operational quantities described below make use of certain theoretical concepts in their definitions that should be explained first:

- **Real field**: The real field is the actual field to be measured unperturbed by any measuring instrument.

- **Expanded field**: The expanded field has the same fluence, directional and spectral distribution as the real field at the point of reference, but over the whole volume of interest.
Expanded and aligned field: The expanded and aligned field has the same fluence and spectral distribution as the real field at the point of reference, but over the whole volume of interest and with a unidirectional distribution.

These concepts are illustrated in Figure 3.3.

3.4.2 Ambient Dose Equivalent

The ambient dose equivalent, $H^*(d)$, at a point in a radiation field is the dose equivalent that would be produced in the corresponding expanded and aligned field, in the ICRU sphere at a depth $d$, on a radius opposing the direction of the aligned field. For strongly penetrating radiations, like neutrons, the reference depth $d$ is 10 mm. Weakly penetrating radiation, like electrons, have recommended reference depths of 0.07 mm for the skin and 3 mm for the eye.
Ambient dose equivalent is the quantity recommended for area monitoring, and its definition means that for measurement the radiation field should be uniform over the whole instrument and that the instrument have an isotropic response.

### 3.4.3 Directional Dose Equivalent

The directional dose equivalent, $H'(d, \Omega)$, at a point in the radiation field, is the dose equivalent that would be produced by the corresponding expanded field, in the ICRU sphere at a depth $d$, on a radius in a specified direction $\Omega$. Recommended reference depths for strongly and weakly penetrating radiations are the same as for ambient dose equivalent. In addition, an irradiation direction $\Omega$ must be given, with reference to a specified coordinate system.

### 3.4.4 Personal Dose Equivalent

The personal dose equivalent, $H_p(d)$, at a point in the radiation field is the dose equivalent in soft tissue at an appropriate depth $d$, below a specified point on the body. Again, recommended reference depths for strongly and weakly penetrating radiations are the same as for ambient dose equivalent.

Personal dose equivalent is the operational quantity associated with dosimeters worn on the body by individuals. Figure shows a plot of ambient dose equivalent $H^*(10)$, personal dose equivalent $H_{p,\text{slab}}(10,0^\circ)$ and effective dose for AP irradiation geometry. Notice that all the functions rise most quickly in the energy region from around 10 keV to 1 MeV.
Figure 3.4 Ambient dose equivalent $H^*(10)$, personal dose equivalent $H_{p,\text{slab}}(10,0^\circ)$ and effective dose for AP geometry taken from ICRP 74.

The operational quantities still do not agree completely with the protection quantities, as shown in Figure 3.5, which shows the ratio of effective dose for AP irradiation geometry to ambient dose equivalent $H^*(10)$ and personal dose equivalent $H_{p,\text{slab}}(10,0^\circ)$. Notice that $H^*(10)$ overestimates $E(\text{AP})$ below about 1 eV, underestimates $E(\text{AP})$ in the range 1 eV – 40 keV, and slightly overestimates $E(\text{AP})$ from 40 keV to around 1 MeV. Overall, it may be said that $H^*(10)$ provides a conservative estimate of effective dose.
3.5 Requirement for Spectral Dosimetry

Biological damage caused by neutrons is highly dependent on the energy of the neutrons incident on the body. As indirectly ionising particles, neutrons can undergo many possible reactions in human tissue. Fast neutrons deposit their energy in tissue mostly by elastic scattering from hydrogen. Thermal neutrons are then captured by hydrogen with the release of a 2.2 MeV gamma ray. Other activation products may be created in the body by neutron capture. Monte Carlo methods form the basis for estimating the dose equivalent as a function of neutron energy, the fluence to dose conversion factors. Thus it is important to know the energy spectrum of the neutron radiation field the person is being exposed to.
The ideal neutron dose equivalent meter would have an energy dependent response that exactly followed the fluence-to-dose equivalent curve published in the ICRP recommendations. No real instrument matches this function perfectly, and of course, all instruments require calibration in fields with a well known energy spectrum. Therefore, neutron spectrometry is vital to inform and improve radiation protection practices.

Commonly used calibration fields for neutron instruments, such as those from $^{241}$Am-Be or $^{252}$Cf, are known to be considerably harder (e.g. to exhibit mean spectral energies that are greater), than those around nuclear reactors. Even the ISO reference spectrum for heavy-water moderated $^{252}$Cf is not sufficiently representative of the soft spectra encountered around heavily shielded nuclear plant. The result of this dissimilarity between calibration and workplace spectra can be considerable inaccuracy in dosimeter readings. Figure 3.6 shows a comparison between the energy spectra the three commonly used calibration fields, $^{241}$Am/Be, $^{252}$Cf and D$_2$O moderated $^{252}$Cf, and two spectra measured around a pressurised water reactor (PWR). All the spectra were taken from the SPKTBIB database published by NPL (Naismith, 1996).
Figure 3.6. Neutron spectra taken from SPKTBIB showing the dissimilar nature of calibration and occupational fields.

It is possible to estimate a correction factor to apply to dosimeter readings using spectral data from measurements carried out in the workplace, information on the dose equivalent energy response of the neutron dosimeter, and knowledge of the field used for its calibration. Such a procedure is carried out to correct the energy response of the CR-39 track-etch neutron dosimeter [Barlow, 1997] used by the Defence Radiological Protection Service (DRPS), shown in Figure 3.7. As calibration of CR-39 is carried out with $^{241}$Am/Be sources, and occupational fields tend to be softer with a significant fraction of low energy neutrons, it is likely that dosimeters will significantly underestimate the dose equivalent under these circumstances. So called ‘local correction factors’ (LCFs) are derived by DRPS using energy spectra supplied by the TNS system for the occupational and calibration fields to which the dosimeter

\[ E_d(E) \]
was exposed. Account must also be taken of the irradiation geometry in this instance since the response of the CR-39 dosimeter varies with the orientation of the dosimeter with respect to the direction of the incident radiation.

![Graph showing dose equivalent response of CR-39](image)

*Figure 3.7. The energy dependent dose equivalent response of CR-39.*

The contribution to dose from each of the 44 energy groups generated by TNS is calculated for both the calibration and workplace spectra. These 44 energy groups and their respective dose contributions are then consolidated into seven broader energy groups for which CR-39 angular and energy response characteristics have been derived. Additionally, the contribution to dose by thermal neutrons, to which CR-39 is insensitive, must be taken into account. Then application of the LCF corrects for the departure of the workplace field from that used for calibration and gives a better estimate of dose equivalent.
The Nuclear Enterprises Mk 7 NRM neutron survey instrument is closely based on the spherical neutron dosimeter developed by Leake [Leake, 1966], and is widely used in the routine assessment of neutron dose equivalent rates. The instrument consists of a spherical $^3$He proportional counter (Centronics type SP9) as the thermal neutron detector at the centre of a 20.8 cm diameter polyethylene moderating sphere.

As the energy response of the system to thermal and epithermal neutrons is higher than ideal, a perforated cadmium absorber surrounds the $^3$He detector to shape the response curve. Despite this, a large over response remains in the keV region and an under response arises at higher energies. An under response is more serious for dosimetry as this could lead to an underestimate of dose. The aim of dosimetry is to conservatively overestimate dose to ensure that dose limits are not exceeded. The departure from the ideal response (i.e. following the ICRP fluence to dose equivalent curve) approaches a factor of five at around 10 keV and therefore serious overestimates of dose rate can be expected for very soft spectra, while under prediction will occur for spectra with large components above about 0.5 MeV. The spectral characterisation of typical environments on plant is thus highly desirable in order that routine dosimetry can be carried out with a high level of confidence. The SPKTBIB catalogue can be used to fold together the energy dependent response of instruments with neutron energy spectra and this is demonstrated in Figure 3.8. A Harwell 0949 has been taken as the instrument most closely resembling the Mk7 NRM, and its response relative to the ICRP fluence-to-dose curve is shown for the three commonest calibration fields. This graphically illustrates how choice of calibration field can affect instrument performance.
Figure 3.8. Response function of a Mk7 NRM folded with energy spectra for the three most common calibration fields to show the actual response compared to ICRP74 fluence-to-dose equivalent curve.
4.1 Introduction
Currently spectrometry measurements for the Naval Nuclear Propulsion Program are made using a system called the Transportable Neutron Spectrometer (TNS) developed by the UK Atomic Energy Authority (AEA) under Ministry of Defence (MOD) contract in the mid-eighties [Armishaw, 1986]. Only a prototype and three commercial versions were produced. One belongs to the Defence Radiological Protection Service (DRPS) and the other two belong to BAe Systems Marine Ltd. The DRPS system is used in the determination of correction factors for their CR-39 personal neutron dosimetry service. BAe Systems use the TNS in the commissioning of nuclear submarines to verify shielding calculations.

4.2 Review of TNS Development
Neutron spectrometers to be used in radiation protection dosimetry must cover a wide dynamic energy range (thermal to tens of MeV), should have an isotropic response and a neutron detection efficiency large enough for the investigation of neutron fields with a dose equivalent rate below 100 μSv/h. As spectrometry is generally performed in mixed fields with photon fluences up to a factor of 100 higher than neutron fluences, the spectrometers must either be insensitive to photons or capable of discriminating between neutron and photon induced events. Finally, extreme environmental conditions (temperature, humidity etc.) must to some extent be considered.
The TNS uses six different detectors to characterise the neutron field from thermal energies up to 10 MeV. Unfolding the proton recoil distributions from four separate detectors produces a neutron energy spectrum in 42 lethargy groups. There are three spherical proportional counters (Centronics type SP2, radius 2 cm) filled with hydrogen at nominal pressures of 100, 300 and 1000 kPa to cover the three overlapping energy ranges 50 – 300 keV, 83 – 600 keV and 0.3 – 1.4 MeV. An NE213 organic liquid scintillator (3.7 cm³ active volume) covers energies above 1 MeV, employing a Link Analytical 5020 pulse shape discriminator to reject gamma ray induced events. In addition, the thermal and epithermal fluence rates are measured by two boron-trifluoride counters (Harwell type 5EB70), one being covered with a cadmium sheath. The energy ranges covered by each detector are shown in Table.

<table>
<thead>
<tr>
<th>Detector type</th>
<th>Energy Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>NE213 scintillator (3.68 cm³)</td>
<td>$E \geq 1\text{ MeV} - 10\text{ MeV}$</td>
</tr>
<tr>
<td>1000 kPa SP2 (3000 V)</td>
<td>300 keV – 1.4 MeV</td>
</tr>
<tr>
<td>300 kPa SP2 (2000 V)</td>
<td>83 keV – 600 keV</td>
</tr>
<tr>
<td>100 kPa SP2 (1600 V)</td>
<td>50 keV – 300 keV</td>
</tr>
<tr>
<td>BF₃ (Cadmium covered)</td>
<td>$0.55\text{ eV} – 50\text{ keV}$ (Epithermal)</td>
</tr>
<tr>
<td>BF₃ (Bare)</td>
<td>$E \leq 0.55\text{ eV}$ (Thermal)</td>
</tr>
</tbody>
</table>

The TNS meets most of the requirements mentioned earlier, covering as it does the energy range from thermal to 10 MeV, and being sensitive down to dose equivalent rates of around $10\mu\text{Sv/h}$ (corresponding to fluxes in the order of 10 to 100 cm⁻²s⁻¹). A requirement not previously stated, but important in the confines of submarine reactor compartments, is compactness of the probe unit. For this reason the detectors are
packed in close proximity to one another, the NE213 and its photomultiplier forming the centre axis with the SP2 and BF$_3$ counters arranged around it, as pictured in Figure. This compact design undoubtedly perturbs the neutron field it is measuring to some extent, but the effect is almost certainly so small as to be masked by the uncertainties in the unfolding process. Another effect of the design may be 'cross-talk' between detectors when a fast neutron is detected in one of the proton recoil counters after previously scattering in another. This scenario is unlikely because the detection efficiency of the hydrogen-filled SP2 counters is very low, and so cross-talk may be discounted.

![Diagram of TNS probe unit](image)

Figure 4.1 TNS probe unit, showing the arrangement of the different detector types and their associated electronics.

Regarding gamma sensitivity, the SP2 and BF$_3$ counters are relatively insensitive to gamma rays and, other than adjustment of their amplifiers low-level discriminator, no action is taken to preclude gamma ray induced events. For the NE213 scintillator active gamma event rejection is mandatory and provided by the Link Analytical Pulse
Shape Discriminator, with a gamma rejection efficiency of up to $10^4$ gammas per detected neutron claimed by Link [Adams, 1978]. The Link 5020 model used in the TNS systems, is based on charge comparison, using the difference between a long and a short pulse integrating gate to make the distinction between gamma and neutron events. When connect to an oscilloscope in its display mode, it shows a two-dimensional pattern representing the frequency of particular combinations of amplitude and rise time. The user adjusts controls to achieve the best possible separation of neutron and gamma ray pulses on the screen. The Link 5020 is then set to gate the ADC only when the pulse is from a neutron event. The TNS literature suggests an operational limit of 10:1 gamma dose to neutron dose.

4.2.1 NE213 Scintillator

Again the principle of operation is neutron induced proton recoils. In the organic scintillator the resultant detector pulse is the product of light output from molecules excited by the passage of the protons, as collected by a photomultiplier. Organic liquid scintillators such as NE213 are often used for the detection of fast neutrons mainly because of their high detection efficiency, good timing resolution and excellent n-γ pulse shape discrimination properties.

For the purpose of neutron spectroscopy the detection efficiency due to the incident neutrons must be known. However, it has to be considered that the efficiency of the detector strongly depends on the energy of the incident neutrons and on the bias level that has been selected for the registration of pulses from the scintillation detector. A calibration therefore requires measurements in accurately known monoenergetic neutron fields for several neutron energies. If a spectrum unfolding method is to be
applied for neutron spectroscopy, then a set of response functions – pulse height spectra from incident monoenergetic neutrons – has to be determined.

Organic scintillators chiefly contain hydrogen and carbon and a large fraction of the pulse height spectrum is due to recoil protons from neutron scattering. The neutron-proton scattering cross section in the centre of mass system is nearly isotropic for neutron energies below 5 MeV. The energy distribution of the recoil protons in the laboratory system is therefore rectangular in shape up to an energy equal to the incident neutron energy. However, even for monoenergetic neutrons below 5 MeV, the real pulse height distribution at the output of a scintillation detector deviates strongly from this simple shape due to many effects, i.e. multiple scattering of neutrons, non-linear light output functions for charged particles and pulse height resolution effects.

A theoretical calculation of a response function can be made by means of the Monte Carlo method. The histories of many neutrons are followed from the neutron source to the detector by simulating all reactions of the neutrons inside or near the detector and calculating the light output induced by these reactions. Only a correction for non-linear light output from recoil protons is made by the TNS NE213 computer code, i.e. there is no treatment of wall effect or multiple scattering in the detector. The justification for this is the small volume of NE213 used (3.68 cm$^3$), which makes multiple scattering unlikely. The neutron fluence spectrum is then calculated by simple differentiation of the pulse height spectrum, after the previous correction, and formatted for input into the RADAK unfolding code. Figure XX is a pulse height distribution for 5 MeV incident neutrons collected at NPL using an NE213 detector from the TNS.
Figure 1 Pulse height distribution from an NE213 scintillator for 5 MeV incident neutrons measured at NPL.

4.2.2 SP2 Counters

Gas-filled proton recoil proportional counters have been widely used for measuring neutron energy spectra in the range keV up to a few MeV. Neutrons entering the counter are elastically scattered from hydrogen in the fill gas. The resulting recoil protons lose their kinetic energy by atomic ionisation and excitation along their tracks. For monoenergetic neutrons, ideal proton recoil detector response of a rectangular energy distribution would only be observed if all protons lost their entire energy within the counter volume, if the W-values for ionisation were independent of proton energy and if gas amplification of the ionisation electrons were constant over the whole detector volume. Indeed the W-values are constant across the energy range considered, but the other two considerations are not usually met. Some protons will strike the counter wall before depositing their entire energy – wall effect. Gas amplification is not constant over the whole counter volume, as the electric field along the anode drops at the ends, where the wire is fixed – end-effect. The wall effect
effectively determines the upper limit for the energy range covered by a particular counter, while the lower limit is set by interference from gamma ray interactions mainly in the counter wall.

The spherical proportional counter design originates with the work of Benjamin [Benjamin, 1968]. The spherical cathode design was chosen in an attempt to make the detector response more or less isotropic (independent of the direction of the incident neutron). Careful design of the anode supports kept the electric field, and hence gas amplification, near constant along its length despite the spherical cathode. However, there are still "polar cap" regions near the wire supports where gas amplification is reduced. Together these regions are estimated to be around 10% of the counter volume. Figure 2 shows a cut through of the SP2 detector design.

![Diagram of the SP2 detector design](image)

Figure 4.2 The spherical proton recoil counter design by Benjamin.

Response functions for the SP2 detectors in TNS are calculated by the PRC (proton recoil counter) program. This calculates the shape of recoil proton distributions for groups of monoenergetic neutrons taking into account the wall effect in the SP2 counter. The method used is the analytical technique devised by Snidow and Warren
Snidow, 1967], and the program includes range data for hydrogen, methane and argon fillings. Comparisons carried out by Kemshall [Kemshall, 1973] between measured and calculated proton recoil distributions showed disagreement, and so a table of empirical correction factors is incorporated into the program to account for this. A set of response functions for a 100 kPa (1 atm) hydrogen filled counter taken from the work of Kemshall are shown in Figure. The slopes calculated by the Snidow algorithm are shown by the solid lines, together with the corresponding experimental data points. The dashed lines are linear least squares fits to the experimental data, from which the empirical correction factors, CFE, are determined.
Figure 4.3 Response function for a 100 kPa (1 atm) hydrogen filled SP2 counter: solid line, calculated by Snidow algorithm; symbols, measured data; dashed line, linear least squares fit to measured data.
4.3 Detector Response Functions

Neutrons of energy $E_n$ can produce, through elastic scattering with hydrogen, recoil protons of energy $E_p$, where $0 \leq E_p \leq E_n$. That is $E_{p,\text{max}} = E_n$. This gives us a rectangular distribution for the recoil protons.

In a perfect proton recoil spectrometer, the response function for unit fluence of neutrons of energy $E_n$ is given by:

$$H(E_p) = \frac{K \sigma(E_n)}{E_n}$$  \hspace{1cm} (4.1)

where $K$ is the number of hydrogen atoms in the detector; $\sigma(E_n)$ is the cross section for hydrogen-neutron elastic scattering for neutrons of energy $E_n$; and $H(E_p)$ is the number of protons per MeV at energy $E_p$ to $E_p + dE_p$ due to neutrons of energy $E_n$. The magnitude of neutron energy that can be detected depends on the stopping power of the filling gas in the counter and the dimensions of the counter.

In reality, the system is not perfect; and the distribution departs from this ideal shape. For example, there are ‘wall-effects’ – i.e. recoil protons interact with the detector walls and do not deposit all their energy in the detector gas to create a measurable signal.

An analytical expression developed by Snidow takes account of this effect in a spherical counter. The expression for the fraction of recoil protons of energy $E_p$ to $E_p + dE_p$ due to neutrons of energy $E_n$ is given by:
\[
\frac{C(E_p)}{C_j} = \left( \frac{F(E_p)}{E_n} + \frac{1}{E_n} \int_{E'=E_p}^{E'} \frac{dR}{dE_p} (E' - E_p) P \left[ R(E') - R(E' - E_p) \right] dE' \right) \frac{1}{\sigma(E_n)}
\]  

(4.2)

where \(C_T\) is the total number of recoil protons; \(R(E)\) is the range of protons of energy \(E\); \(F(E_p)\) is the fraction of protons of energy \(E_p\) that are stopped in the detector. If we define \(P(L)\), the probability that protons will travel a distance between \(L\) and \(L+dL\) to the detector wall, as follows:

\[
P(L) = \frac{3}{4a} \left[ 1 - \frac{L^2}{4a^2} \right] dL
\]  

(4.3)

then expression (3) is obtained by integrating \(P(L)\) as shown:

\[
\int_{L=R(E_p)}^{L_{up}} P(L) dL
\]  

(4.4)

where \(a\) is the radius of the detector. Equations 3 and 4 highlight the requirement for accurate proton range data.

In the energy ranges of interest here, the response function would be rectangular if the detector were perfect, as indicated by Equation 2. If one defines a correction factor, \(CF\), as follows:

\[
CF = \frac{\text{No. of pulses at } E_{p,max}}{\text{No. of pulses at } \frac{1}{4} E_{p,max}}
\]  

(4.5)
then, for a perfect system, CF = 1. Correction factors determined from response functions calculated using Equation 4 deviate from 1, and this deviation increases with increasing 'range over radius', R/a, where R is the range of the maximum energy proton produced by neutrons of energy E\textsubscript{n} (i.e. E\textsubscript{p,max} = E\textsubscript{n}). Further more, it was found that these calculated response functions differed significantly from measured ones. In the SPEC4 code, calculated response functions are modified by ratios of correction factors determined from measured response functions (CFE; E for experimental) to those determined from calculated response functions (CFS; S for Snidow); these ratios are referred to as CFE:CFS.

4.4 Unfolding the Neutron Spectrum

The neutron fluence above E\textsubscript{max}, the maximum energy analysed, must be measured by some other means, calculated or otherwise specified. The recoil proton distribution due to this fluence is calculated following Equation 4.1. These data are subtracted the distribution measured by the counter – i.e. the contribution due to neutrons above E\textsubscript{max} is subtracted. The remaining pulse height distribution is then unfolded to give the neutron fluence spectrum. In the SPEC4 unfolding program, slopes of response functions are used; this is said to prevent oscillations in the unfolded spectrum.

For the set SP2 counters in TNS, the spectrum unfolded for the highest energy counter serves as input for the next-highest energy counter. Thus, it is likely that uncertainties will be largest for the lowest energy counter (100 kPa) because its data will rely on the cumulative results from the other two counters. Unfortunately, this counter also has the lowest detection efficiency and so the poorest counting statistics. For the 1000 kPa counter, the neutron spectrum above E\textsubscript{max} is supplied from the result of the NE213 unfolding.
For each proportional counter it is assumed that the number of counts in a specific channel of the multichannel analyser segment, is proportional to the number of recoil protons having an energy corresponding to that channel. The proton recoil pulse height distribution is given by:

\[ M_i = \sum_{j=1}^{m} H_j^i \phi_j, \quad i = 1 \ldots 256 \]  \hspace{1cm} (4.6)

where \( M_i \) is the number of counts or recoil protons in channel \( i \) due to neutrons in energy groups \( 1 \) to \( m \); \( \phi_j \) is the neutron fluence in energy group \( j \) (\( \text{cm}^{-2} \)); and \( H_j^i \) is the response function contribution to channel \( i \), that is the number of recoil protons in channel \( i \) produced by monoenergetic neutrons in energy group \( j \). The neutron energy in group \( j \) exceeds or equals the proton energy in group \( i \).

The user is presented with two options for performing the spectrum unfolding. The first is the early code for the analysis of proton recoil spectra in proportional counters, SPEC4 [Kemshall, 1973]. The second is a more advanced code called RADAK [Grimstone, 1997] that constrains the solution spectrum and provides some estimation of the uncertainty in the solution. RADAK falls under the category of least squares solutions, using a weighted first difference smoothing method to avoid spurious structure in the solution spectrum.
4.5 Measurements made with TNS

4.5.1 Laboratory $^{241}$Am/Be sources

The spectrum of an unshielded $^{241}$Am/Be source was measured at HMS Sultan under typical laboratory conditions where the contribution from room return scattered neutrons would be high. The source positioned at the centre of the 5m x 5m x 3m room, 1 metre off the floor with the TNS probe positioned 1 metre away. The resulting pulse height distributions were unfolded by the RADAK program and compared with the ISO reference spectrum [ISO, 1989] spectrum for $^{241}$Am/Be, as shown in Figure. Agreement is close at the highest energies with the TNS spectrum reproducing the peaks in the ISO spectrum quite well. However, the spectrum starts to deviate significantly below 5 MeV, which has been attributed to room return of scattered neutrons.

![Figure 4.4 AmBe spectrum recorded by TNS with ISO reference spectrum for comparison.](image)
4.5.2 CONSORT Research Reactor

Measurements were also carried out at the CONSORT research reactor, operated by Imperial College, London, on one of the horizontal experimental beamlines, with and without the first biological shield plug removed. Figure 4.5 shows the spectra with the plug removed and in place, respectively. The dose-rate at the TNS probe position was measured using a calibrated Leake type dosimeter (Mk7 NRM) and a gamma monitor (Nuclear Enterprises PDR2). With the shield plug removed the neutron dose-rate observed was 200 μSv/h with a substantial gamma-ray background of 170 μSv/h. With the shield plug in place the shape of the spectrum did not noticeably change, but the dose-rates were reduced significantly, with recorded values of 70 μSv/h for neutrons and 120 μSv/h for gamma rays. A further measurement was performed on the reactor top-plate with the shield doors slightly ajar. Dose-rates measured here by the area monitors were 220 μSv/h for neutrons and approximately 2mSv/h for gamma rays. This is in excess of the 10:1 gamma dose to neutron dose ratio, beyond which the pulse shape discriminator may fail to operate correctly. Indeed, RADAK was unable to unfold a meaningful spectrum from the NE213 data.
4.5.3 Rolls-Royce NEPTUNE Test Reactor

The neutron spectrum of the $^{252}$Cf start-up source installed in the NEPTUNE zero-power test reactor (Rolls-Royce, Derby, UK) was measured in situ. The RADAK unfolded spectrum is shown in Figure 6 together with the standard ISO spectrum for $^{252}$Cf. Major deviation from the ISO spectrum is seen on the low energy side of the peak, caused by the presence of highly scattering materials in the confined space near the source. This spectrum was measured in a considerable gamma ray background, around 500 $\mu$Sv/h according to reactor based instrumentation.
Figure 4.6 TNS measured spectrum for the $^{252}$Cf start-up source at the NEPTUNE reactor, with the ISO reference spectrum shown for comparison.

4.5.4 $^{252}$Cf measurement at DRaStaC

To establish that room return was responsible for the deviation from ISO standards in the previous californium source measurement (Figure 6), in the absence of suitable shadow cone measurements, a second spectrum for $^{252}$Cf was taken at the Defense Radiological Standards Centre (DRaStaC) low scatter calibration facility. The DRaStaC facility meets the requirements outlined in the ISO standard and is UKAS (NAMAS) accredited. This means that the neutron sources used have had their emission rate determined by the National Physical Laboratory, and the Long Counter used to measure the delivered fluence is also traceable to the NPL Long Counter. (The DRaStaC Long Counter is a transfer instrument.)
The dimensions of the irradiation room are 9.1 m x 5.5 m x 4.4 m, which is sufficiently large to meet the ISO requirement that the room return contribution to the dosemeter reading is less than 40%. The source, in its irradiation position, is located on the centre line of the room 3.5 m from the nearest wall and 2 m off the ground. The source control apparatus, support structures for detectors etc. are all made from aluminium alloy with hollow struts and so should not significantly perturb the neutron field. The source used was a $^{252}$Cf source with an emission rate of 2.11 x $10^7$ s$^{-1}$ certified on 26 July 2000 (by measurement at NPL. For the measurements reported here the source emission rate had decayed to 2.00 x $10^7$ s$^{-1}$, which equates to an ambient dose equivalent rate of 220.6 μSv h$^{-1}$ at 1 m. The scatter correction for this source is reported by DRaStaC as 19% normalised at 1 metre.

The spectrum recorded by TNS is shown in Figure 4.7, again with the ISO spectrum for reference. Clearly this spectrum matches the ISO spectrum more closely than Figure 4.6, with fewer low energy scattered neutrons contributing. TNS still underestimates the fluence at higher energies, and shows a shift in the peak energy of the distribution. This effect could be the result of a systematic error in the energy calibration procedure for the NE213 scintillator, based on the $^{137}$Cs Compton edge position.
Figure 4.7 TNS measured spectrum for $^{252}$Cf at DRaStaC, with the ISO reference spectrum shown for comparison.

### 4.6 Discussion and conclusions

Table 4.2 shows a comparison between dose equivalent rates calculated from the TNS measured spectra and Mk7 NRM readings. Also shown are gamma dose rates for the measurements at NEPTUNE and CONSORT. Ratios from the two instruments show TNS tending to estimate neutron dose equivalent 10 – 20 % higher than the Mk7 NRM. Such a variation might well be expected given the known limitations in the energy response of Leake type dosimeters. The TNS result should be more reliable as the dose equivalent is being calculated from the actual energy spectrum of the neutron fluence. In fact, the TNS spectrum can be used to calculate a correction factor for the Mk7 NRM reading in a particular neutron field.
Table 4.2 Summary of neutron dose rates measured by TNS and Mk7 NRM, and gamma dose rates measured by NE PDR1 for the measurements described above in Section 4.5.

<table>
<thead>
<tr>
<th>Spectrum Description</th>
<th>Mk7 NRM Neutron Dose-rate (µSv/h)</th>
<th>TNS Neutron Dose-rate (µSv/h)</th>
<th>Ratio TNS: Mk7 NRM</th>
<th>Gamma Dose-rate (µSv/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>221Am/Be</td>
<td>30</td>
<td>35 ± 8</td>
<td>1.166</td>
<td>-</td>
</tr>
<tr>
<td>252Cf (NEPTUNE)</td>
<td>-</td>
<td>2580 ± 56</td>
<td>-</td>
<td>100</td>
</tr>
<tr>
<td>252Cf (DRaStaC)</td>
<td>250</td>
<td>298 ± 2</td>
<td>1.192</td>
<td>-</td>
</tr>
<tr>
<td>Beamline without shield plug</td>
<td>200</td>
<td>229 ± 14</td>
<td>1.145</td>
<td>170</td>
</tr>
<tr>
<td>Beamline with shield plug</td>
<td>70</td>
<td>76 ± 21</td>
<td>1.086</td>
<td>120</td>
</tr>
</tbody>
</table>

For the DRaStaC spectrum, TNS records a neutron dose equivalent rate of 298 µSv/h. Using the given scatter correction figure (calculated in accordance with ISO 8529) of 19% for 252Cf at 1 metre, the scatter corrected dose rate for the TNS spectrum is 250 µSv/h, which is in agreement with DRaStaC data for the source used [Danyluk, 2001].

The results show that the TNS is capable of producing high resolution spectra for assessment of dose equivalent even in mixed fields where the gamma dose exceeds that from neutrons provided the ratio of gamma dose to neutron dose does not exceed 10:1.

Validity of Snidow and Warren Responses

The PRC program takes account of wall effect only; there is no treatment of electric field distortion at the ends of the anode or finite counter resolution. Therefore, it cannot be relied upon to provide response functions for any new counters. The use of empirical corrections made with other detectors in the past is suspect as performance can vary considerably between individual counters. Long-term stability for hydrogen-filled counters appears good, but has not been properly assessed. Counter resolution strongly depends on gas purity, and gas pressure is an important parameter in
subsequent analysis, and therefore these factors will be considered in future work on SP2 responses.

Proton range data hard coded into the PRC/RADAK package dates from 1963 and more accurate range data is now available. New values for protons stopping power and range in hydrogen, methane and argon have been published by the ICRU publication [ICRU, 1993], which would affect calculated response functions.

There are several deficiencies identified in the operation of the TNS. Responses for the SP2 counters calculated by the Snidow algorithm are not reliable as it only corrects for the wall effect and not electric field distortions. Furthermore, the factors used to correct the disagreement between empirical and calculated responses are taken from published measurements with a different set of detectors, and were not confirmed to be applicable to the TNS detectors. More accurate response functions for SP2 type counters have been calculated by the SPHERE code [Knauf, 1998] from PTB in Germany, which includes treatment of electric field distortions near the ends of the counter anode as well as the wall effect. All calculated responses should be verified with monoenergetic irradiations in well-characterised fields such as those available at the UK National Physical Laboratory (NPL).

The assumption of a near-ideal response function for the NE213 detector is perhaps justified in such a small scintillator cell, but this cannot be checked by calculation with codes such as NRESP [Dietze, 1982] because of the non-cylindrical cell shape. Mono-energetic irradiations should be performed in order to confirm the light output parameters, and to find the pulse height resolution of the detector for folding with the response function.
Once accurate detector responses for the current system have been established, the performance of the RADAK code against some more widely used or recently developed unfolding codes could be determined, e.g. UNFANA [Weise, 1995] in the HEPRO package [Matzke, 1994], or the MAXED [Reginatto, 1998] maximum entropy approach for proton recoil counters [Reginatto, 2002].

The key to making neutron spectrometry a routine part of health physics work in the field is to improve reliability and portability of instruments. As most of the weight in the TNS is made up of NIM electronics, this will be achieved through the use of modern digital signal processing to perform data acquisition and analysis. Work on such a system is being performed at Lancaster University [Weaver, 2002] in collaboration with BAe Systems Ltd., the main user of TNS.

4.7 BTI Microspec-2 N-Probe

During the course of this work the Nuclear Department at HMS Sultan acquired a Bubble Technology Industries (BTI) Microspec N-Probe. This is sold as a simple spectrometer suitable for use by non-specialist technicians in the workplace.

4.7.1 Basic Description

The Microspec-2 N-probe is manufactured by Bubble Technology Industries (BTI) as part of its Microspec range of portable instruments. The N-Probe is designed around two detector types, an NE213 organic liquid scintillator and a helium-3 counter, the latter being covered by a ‘boron shield’. They are housed one above the other in a conveniently portable unit, together with their high voltage supply circuits and rechargeable battery. This probe unit connects to the Microspec analyser unit, consisting of an amplifier and 256 channel MCA married to an Atari Portfolio palm-
top computer, which perform the spectrum unfolding and displays the results. The operating software is stored together with the acquired data on a 1 Mb memory card specific to the Atari Portfolio. Data can be transferred to a PC or printer via a parallel port connection on the analyser unit. Also included in the package purchased was a small (2" x 2") NaI detector for measuring environmental gamma rays in the range 60 keV to 3 MeV, known as the E-probe. This provides gamma ray spectra and dose information, including radioisotope identification from a peak library. Figure 4.8 is a photograph of the N-Probe.

Figure 4.8 The BTI Microspec N-Probe, probe unit consisting of NE213 scintillator and boron-shielded $^3$He counter, and analyser unit with palmtop computer.

4.7.2 Detailed Description of Detectors and Operation

The two detectors in the N-Probe cover different energy regions of the neutron spectrum. The NE213 scintillator covers the energy range 800 keV to 20 MeV, and the $^3$He detector from thermal to 800 keV. As NE213 is also sensitive to gamma rays,
a pulse shape discrimination circuit is built into the system in order to distinguish between neutron and gamma ray induced events. This can be disabled to allow a calibration check using a gamma source ($^{22}$Na recommended), but no other user control is available. Pulse shape discrimination between neutrons and gamma rays is not possible much below 1 MeV, and this sets the lower energy limit for operation of the NE213 to the 800 keV quoted. The NE213 scintillator volume is 2” in diameter by 2” thick. This gives a higher detection efficiency at the expense of complicating the response function with multiple neutron scattering events in the detector becoming significant. Response functions for this detector have been measured by BTI using accelerator produced monoenergetic neutron fields.

The region of the spectrum below 800 keV is covered by the $^3$He counter. Of course, this is primarily a slow neutron detector, but the cross-section for the $^3$He(n,p) reaction does extend to higher energies. Surveying the literature shows several authors have attempted to unfold the incident neutron spectrum from $^3$He with qualified success. Calculations of $^3$He detector response functions are extremely complex, leading to large uncertainties that distort the unfolded spectrum. A further problem comes from the presence of thermal neutrons in the measured field. These will be detected with such high efficiency that pile-up becomes a problem, obscuring detail in the fast neutron spectrum. BTI have tackled these inherent problems in two ways. Firstly, the unfolded spectrum from the N-Probe $^3$He counter consists of only 3 energy groups (thermal – 10 keV, 10 keV – 500 keV, and 500 keV – 1 MeV). (The highest energy group obviously overlaps with the lowest NE213 group, but how this is handled in the unfolding algorithm is not known.) Secondly, the over-response to thermal neutrons is reduced by the inclusion of a ‘boron shield’ around the $^3$He detector. According to BTI, this has been carefully designed to optimise the detectors.
energy response by the inclusion of partial and full penetrations through the shield. Calibration of the \(^3\)He detector is not routinely required, but can be verified by exposure to thermal neutrons to produce the recognisable peak in the spectrum at 764 keV (the Q-value of the \(^3\)He(n,p)\(^3\)H reaction).

N-Probe has two modes of operation: spectrometry mode showing pulse height distributions from the two detectors recorded in different segments of the MCA, or dosimetry view showing a real time view of dose rate and cumulative dose. In spectrometry mode, when acquisition has finished, the neutron energy spectrum is unfolded at the touch of a key, requiring no extra input from the user. A neutron energy spectrum from thermal to 20 MeV in 18 energy groups is produced. Details of the unfolding algorithm used are not divulged by the manufacturer, however, data files for the NE213 light output and response functions for monoenergetic neutrons are stored externally to the actual unfolding software and may be examined. Unfortunately, certain information regarding the structure and scaling of this data is omitted in the files, making interpretation difficult. Dosimetric quantities are calculated from the spectrum using the desired fluence-to-dose conversion factors. These are read from a simple text file, and so can be updated as necessary.

4.7.3 Example Spectrum

The spectrum of a 100 mCi (3.7 GBq) \(^{241}\)Am/Be test source has been measured at HMS Sultan using the N-Probe. The measurement was performed in a large room with the source suspended 1.5 m off the floor to reduce the contribution from room-scattered neutrons and the N-Probe positioned 1 m away. Data was accumulated for 4 hours. The result is shown in figure 1 below, together with the ISO reference spectrum for Am/Be (normalised to equal fluence in the highest energy group).
Figure 4.9. N-probe spectrum for a 100 mCi Am/Be source compared to the ISO reference spectra for $^{241}$Am/Be (ISO 8529-1).

The N-Probe spectrum reproduces the features of the $^{241}$Am/Be reference spectrum well enough, the resolution is quite low. A large number of neutrons are present in the intermediate and slow energy regions due to the effect of room scatter. The lower energy portion of the spectrum is not covered in detail; just three energy groups are contributed by the $^3$He counter. Clearly this is not sufficient in such a vital area of interest for neutron dosimetry. Also, the accuracy of N-probe spectral data in these groups can be questioned, as in the paper by Devine et al. [Devine, 2002]. He compared MCNP simulated spectra for a variety of neutron fields with those measured by N-probe. In all cases close resemblance was found in the high-energy portion, but results often disagreed markedly in the lower energy groups.
Chapter 5

Lithium Gadolinium Borate

5.1 Capture-gated Spectrometry

The response functions of organic liquid or plastic scintillators have already been discussed in previous sections. The pulse height spectrum produced by monoenergetic neutrons in a typical organic or plastic scintillator consists of a broad continuum from zero up to the equivalent of the full neutron energy. This continuum is the distribution of recoil proton energies produced in single or multiple elastic scatterings of the fast neutrons from hydrogen in the detector. Most fast neutrons enter the detector and then escape after having deposited a variable fraction of their energy, and so a broad continuum is the result rather than a peak. The outcome of this is the need to use unfolding methods to arrive at the original neutron spectrum.

For incident monoenergetic neutrons, an ideal neutron spectrometer would give a pulse height distribution that was a single, narrow peak. It is possible to approach this ideal situation somewhat by an alternative method of operating organic scintillators. Take for example a plastic scintillator that has been loaded with a small percentage of a nuclei with a high capture cross-section for thermal neutrons, such as $^6\text{Li}$ or $^{10}\text{B}$. A fast neutron that undergoes enough collisions in the detector may become sufficiently moderated to have a high likelihood of capture by the thermal neutron absorber, i.e. it does not escape but is captured as a slow neutron in the scintillator. In slowing down the neutron will have produced a series of recoil protons whose energy must sum to be the incident neutron energy. A neutron in the MeV energy range is travelling extremely fast (e.g. 4.6% of the speed of light for a 1 MeV neutron), so all the recoil protons are produced within a very short time period,
typically less than 50 ns. This is a very short time compared to typical electronic pulse shaping times and so the total light from the recoil protons will add together to give a single output pulse. If only these “full energy deposition” events could be selected and all others were rejected, the pulse height spectrum would be a single peak in the corresponding to the incident neutron energy.

The method of selection is to actually detect the capture of the thermalised neutron within the scintillator medium. Once the neutron has lost its energy through these multiple collisions, it will diffuse through the scintillator until it escapes or is captured. If there is a sufficient concentration of say boron, the high cross-section for capture in $^{10}\text{B}$ will predominate, and almost all thermalised neutrons will be captured by $^{10}\text{B}$ nuclei. The Q-value for capture in $^{10}\text{B}$ is 2.3 MeV, and this energy will be deposited locally by the reaction products to give a second light pulse in the scintillator. The average time separation between the light produced by the recoil protons and the light produced by the boron capture products is the mean diffusion time of thermal neutrons in the boron-loaded scintillator, and would be of the order of $10 – 20 \mu$s. The characteristic signal to indicate that the fast neutron deposited its full energy in the detector would be this double pulse signature, first a light pulse from proton recoils and then a light pulse from capture in the thermal neutron absorber. Pulse height selection could also be applied to the second pulse to require that its energy corresponds to a window around the Q-value of the reaction. Neutrons that enter the detector, create one or more recoil protons, and escape are rejected because a second pulse does not occur as in the case of full energy deposition.

Spectrometers of this type can be constructed using, as in the example above, boron-loaded plastic, boron-loaded liquid scintillator (which allows for pulse shape discrimination against incident gamma rays), or heterogeneous combinations of
plastic scintillator and $^6$Li loaded glass scintillator plates. In the latter case, the neutron capture leads to deposition of 4.78 MeV in the glass scintillator. The different decay times of the plastic scintillator and the lithium glass allow pulse shape discrimination techniques to be used to differentiate between proton recoil and capture pulses, permitting a more specific identification of the full-energy absorption event.

5.2 Lithium Gadolinium Borate Scintillator

Lithium gadolinium borate (hereafter referred to as LGB) was originally produced as a laser material, but, as lithium, gadolinium and boron all possess isotopes having large neutron cross-sections, it soon attracted attention as a possible high efficiency neutron detector [Czirr, 1999]. Another attractive feature is that all three elements have low cross-section isotopes also, making the scintillator highly adaptable.

The new material, Li$_6$Gd(BO$_3$)$_3$ activated with Ce$^{3+}$ has a refractive index of 1.66, which is close enough to that of organic polymers ($\sim$1.6) to allow their use as binding materials for powdered LGB scintillator. In this way large area thermal neutron detectors for neutron counting can be made.

Similarly, for neutron spectroscopy the LGB can be dispersed in a plastic scintillator (Bicron BC404, refractive index 1.58) and the resulting scintillator matrix remains fairly transparent to its own light. The particular isotopic composition used is $^6$Li$_6$^{154}Gd(BO$_3$)$_3$;Ce, since this gives the highest light output.

Fast neutrons scatter elastically from hydrogen in the plastic scintillator producing recoil protons. If the incident neutron is sufficiently moderated in this or multiple proton collisions, it may be subsequently captured by one of the large (slow neutron) cross-section isotopes. Considering the macroscopic cross-section as the neutron slows down, this is most probable in $^6$Li. Therefore, fast neutron detection and discrimination against other events occurs on the basis of this “double pulse” signal; a
proton recoil pulse followed by a $^6\text{Li}$ capture pulse. Figure 5.1 illustrates the detection process in the LGB capture-gated spectrometer.

Figure 5.1 Illustration of the detection process in the LGB capture-gated spectrometer.

The detector is, of course, sensitive to slow neutrons as well as fast. Examination of the macroscopic cross-sections shows neutron capture to be most probable in gadolinium below around 0.3 eV, and in lithium-6 above this. Therefore, Photogenics apply the principle that this composition is sensitive to thermal neutrons through capture in Gd, and to epithermal neutrons through capture in $^6\text{Li}$. However, the distinction does not really exist as both cross-sections are competing for neutron capture over all energies.
5.3 Description of LGB Spectrometer

A prototype of the LGB spectrometer was obtained from Photogenics for evaluation as a replacement for the NE213 scintillator in the TNS. The detector is a cylinder of the composite plastic and powdered lithium gadolinium borate scintillator, diameter 2" (5.08 cm) and length 1.5" (3.81 cm) encased in an aluminium can and coupled to a Burle photomultiplier and Canberra pre-amplifier base.

Signals from the detector are fed to the custom built Photogenics digitiser box. This consists of a fast 8-bit ADC (30 MHz, 100 MS/s) and logic control circuits on a printed circuit board that captures the shape of each pulse digitally and makes decisions about the originating event – did it come from a recoil proton, a Compton electron or capture in lithium-6 or gadolinium-157. Logic decisions are made by a fully programmable gate array (FPGA). If the event was determined to be a recoil proton due to fast neutron (n, p) scattering, the coincidence window for capture gated counting is turned on. The shape of the pulse waveform is sampled every 10 ns and assigned a value in the range 1 to 256 (actually pulses saturate at a value of 247 with the current electronics). The coincidence window stays open for 10 μs.

Events that are tagged as coming from fast neutrons are fed to a program on the PC via an RS-232 serial link. This program, “COMREAD”, writes the event details to a file for later analysis offline by another program called “SCOPE” that allows individual events to be examined and scatter plots of pulse parameters produced. Three of the four views available in SCOPE essentially demonstrate the pulse shape discrimination performed by the digitiser, while the fourth shows the distribution of pulse heights for both proton recoil and 6Li capture.

The double pulse technique is also referred to as “capture-gating”, since the proton recoil signals are being gated to the output on the condition that they are each
followed by a capture pulse. Figure 5.2 shows a recoil proton pulse captured by the waveform digitiser. The logic circuits on the digitiser board recognise this pulse shape as being from a recoil proton by means of its duration (width), which should be greater than for a gamma ray induced event. The pulse height is proportional to the energy of the recoil proton. This pulse has a width of 9 channels (FWHM), corresponding to 90 ns (each channel is 10 ns wide) and this is a mean value for recoil proton pulses with this system.

![Graph](image)

**Figure 5.2** Digitised waveform of a recoil proton pulse in the LGB spectrometer. Each channel is 10 ns wide.

A characteristic pulse from $^6$Li capture that does not occur in the window of coincidence with a proton recoil is counted as an epithermal neutron event, the justification being that, until about 0.3 eV, the cross-section for $^6$Li capture is larger than for capture in gadolinium. Neutron capture in lithium-6 proceeds via the reaction $^6$Li(n,α)$^3$H, with the release of 4.78 MeV. This produces a large, slow pulse from the
photomultiplier that is easily recognised. Figure 5.3 shows such a pulse as captured by the waveform digitiser.

![Figure 5.3 Characteristic pulse from LGB for neutron capture in lithium-6.](image)

Since the reaction products (an α-particle of energy 2.05 MeV and a triton of energy 2.73 MeV) have a very short range in the scintillator, we expect all their energy to be deposited in the scintillator volume. Therefore, the $^6$Li pulses form a peak whose width is mainly determined by the light collection from the scintillator and the statistics of electron multiplication in the scintillator. The $^6$Li capture peak areas lie in the range from 2000 ± 400, and their width is 25 ± 7 channels (250 ± 70 ns). Figure 6.4 is a plot of $^6$Li capture pulse heights and a fitted Gaussian distribution. This suggests an energy resolution at FWHM of 30%, which is in agreement with Photogenics [Czirr, 2002].
Neutron capture in gadolinium does not produce any heavy charged particles, but a cascade of gamma rays averaging 8 MeV in energy is emitted. These gamma rays will deposit some of their energy in the scintillator before escaping, and on average this will amount to 2 MeV. A single channel analyser set to look at a narrow window around 2 MeV would therefore give a measure of the number of captures occurring in gadolinium, and this can be related to the thermal neutron fluence.

The waveform digitiser has an LCD that displays event counting information in real time. There are four counting channels in all and they are described here using the labels attached to them by Photogenics. Fast neutron (capture-gated) events are displayed in the first counting channel. Neutron captures that occur in $^6$Li outside of the delayed coincidence window are counted as epithermal neutron detection on the basis of macroscopic cross-sections discussed earlier. The characteristic signal of
neutron capture in gadolinium is recorded as thermal neutron detection by the same argument. The fourth channel records the number of gamma ray events discarded by the pulse shape discriminator.

5.3.1 LGB with $^7$Li and $^{10}$B

Later a sample of LGB with the isotopic composition $^7$Li$_6$Gd($^{10}$BO$_3$)$_3$:Ce was acquired. The capture-gating for this version is done by delayed coincidence capture in $^{10}$B, which gives less light since the reaction has a Q-value of 2.31 MeV and has heavier reaction products. Potentially, the higher cross-section of the $^{10}$B reaction will make this version of the spectrometer more efficient than the $^6$Li version.

5.4 Initial Testing

Measurements have been performed using $^{241}$Am/Be sources to test the operation of the system. Figure 5.5 shows a proton recoil pulse height distribution from one of these measurements for the $^6$Li version of LGB. This distribution was collected using a 1 Ci (37 GBq) $^{241}$Am/Be source placed in a “neutron howitzer” (an aluminium drum filled with wax having a central hole into which the source is lowered, and beam ports exiting at half height). It contains 3069 individual proton recoil events collected in 30 minutes. The energy bins were chosen with a width of 5 pulse height units, but this is arbitrary and depends on the level of detail required. The detector response diminishes to zero at pulse heights of around 25-30 due to the neutron detection threshold set at around 1 MeV. This threshold is necessary because pulse shape discrimination between neutron and gamma ray events in the plastic scintillator is not possible below this energy.
Figure 5.5 An Am/Be pulse height distribution measured by the $^6$Li version of LGB.

Figure 5.6 shows the proton recoil pulse height distribution collected by the $^{10}$B version of the detector for the howitzer spectrum. The distribution shows the same general features, but it contains only 844 events collected over the same time (30 minutes). This is despite the fact that the $^{10}$B version should have a higher detection efficiency due to the $^{10}$B capture cross-section being larger than that for $^6$Li capture. The reason for this reduced efficiency is the pulse shape analysis parameters of the waveform digitiser being optimised for the $^6$Li version. No adjustments can be made to the pre-set parameters in the digitiser at present.
Figure 5.6. Proton recoil distribution for an $^{241}\text{Am}/\text{Be}$ source measured by the $^{10}\text{B}$ version of LGB.

These pulse height distributions cannot be interpreted any further since the detectors have not been calibrated, i.e. their detection efficiency and energy resolution is unknown. The next stage in the evaluation of the LGB spectrometer must be to perform monoenergetic neutron irradiations of a precisely known fluence at an accredited calibration facility.
Chapter 6

Monoenergetic Irradiations at NPL

6.1 The NPL Low Scatter Facility

Calibrations of the fluence response of the lithium gadolinium borate spectrometer (LGB) were performed in the low-scatter facility associated with the 3.5 MV Van de Graaff accelerator in the Chadwick Building at the National Physical Laboratory. Neutrons are produced by accelerating beams of protons or deuterons on to targets mounted at the centre of the irradiation area. The low-scatter facility (or Main Bay) is a very large room (23 metres long, 17 metres wide and 18 metres high) and the irradiation area lies at one end where the floor has been removed to further reduce scatter. The pit, as it is known, is 6 metres in radius and looks down into the basement, the floor of which is 6 metres below. There is a circular platform called the pit circle 1.5 metres below the target position at the end of the Van de Graaff beamline. Detectors for calibration are mounted on stands attached to small carriages on rotatable arms that pivot about the centre of the pit circle. Low-density walkways allow access to the detector positions. A photograph of the main bay is shown in Figure 6.1.

In this work, three different neutron-producing targets were employed with six different charged particle beam energies to produce monoenergetic neutrons at 144 keV, 250 keV, 565 keV, 1.2 MeV, 2.5 MeV and 5.0 MeV. The quoted energies are for positions at 0° to the proton or deuteron beam direction, and all the measurements were made along this line.
The beam parameters, target properties and resulting neutron field characteristics are given in Table 6.1 together with uncertainty values where appropriate. This data is taken from the official NPL Calibration Certificate.

The neutron fluence delivered during an irradiation run is determined by the use of the NPL long counter. The defining characteristic of a long counter is that its detection efficiency versus neutron energy is almost a constant, i.e. it has a flat energy response. The NPL long counter follows a common design of a long BF$_3$ tube in a cylindrical moderator, designed so that only neutrons incident near the centre of the long counter face are likely to be detected. As the BF$_3$ tube is a slow neutron detector, incident fast neutrons must first be moderated. The average distance a neutron penetrates into the counter before being moderated increases with energy, but if the BF$_3$ tube and
moderator are long enough the probability of detection does not depend strongly on neutron energy. An initial measurement of the fluence at a fixed position along the 0° axis was made before any instrument irradiations. The long counter reading is corrected for air and room in-scatter by means of a shadow cone measurement, and for air out-scatter by calculations using the known oxygen and nitrogen cross-sections. A further correction is made for dead-time effects, after which the neutron fluence at the effective centre of the long counter can be derived using the known response of the device. The fluence response of the long counter has been measured using radionuclide neutron sources whose emission rate has been measured absolutely in the NPL manganese sulphate bath. Further confirmation of the response function comes from measurements made with a proton recoil telescope for the neutron energy range from 2.5 MeV to 5.5 MeV, and from the associated target activity technique over the neutron energy range from 0.5 MeV to 1 MeV. These response function measurements are supported by extensive calculations made recently by NPL, and by Monte Carlo simulations to determine the energy dependent effective centre of the device.
Table 6.1 Charged particle beam parameters, target properties and resulting neutron field characteristics. (All energies and target thicknesses, i.e. energy loses, are in keV.)

<table>
<thead>
<tr>
<th>Neutron Producing Reaction</th>
<th>(^7)Li(p,n)</th>
<th>D(d,n)</th>
<th>T(p,n)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incident Particle Energy (E_p)</td>
<td>2296±3</td>
<td>1830±3</td>
<td>2018±3</td>
</tr>
<tr>
<td>Target Thickness</td>
<td>5.6±1.4</td>
<td>113±40</td>
<td>40±14</td>
</tr>
<tr>
<td>Angle of Fluence Measurement ((0^\circ)*</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Neutron Energy (E_n) ((0^\circ)*</td>
<td>565±3</td>
<td>5000±22</td>
<td>1200±8</td>
</tr>
<tr>
<td>Full Width of Neutron Energy Distribution</td>
<td>6.1±1.5</td>
<td>121±43</td>
<td>41±14</td>
</tr>
<tr>
<td>(E_n) (5°)</td>
<td>564±3</td>
<td>4990±22</td>
<td>1196±8</td>
</tr>
<tr>
<td>(E_n) (10°)</td>
<td>559±3</td>
<td>4960±21</td>
<td>1183±8</td>
</tr>
<tr>
<td>(\frac{\partial E_n}{\partial E_p}) ((0^\circ)*</td>
<td>1.086</td>
<td>1.069</td>
<td>1.024</td>
</tr>
</tbody>
</table>
Table 6.1 continued.

<table>
<thead>
<tr>
<th>Neutron Producing Reaction</th>
<th>$^7\text{Li}(\text{p,n})$</th>
<th>$\text{T}(\text{p,n})$</th>
<th>$^7\text{Li}(\text{p,n})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incident Particle Energy $E_p$</td>
<td>1938±3</td>
<td>3296±3</td>
<td>2018±3</td>
</tr>
<tr>
<td>Target Thickness</td>
<td>6.4±1.4</td>
<td>28±10</td>
<td>6.4±14</td>
</tr>
<tr>
<td>Angle of Fluence Measurement ($\theta^\circ$)</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Neutron Energy $E_n$ ($\theta^\circ$)</td>
<td>144±4</td>
<td>2500±6</td>
<td>250±4</td>
</tr>
<tr>
<td>Full Width of Neutron Energy Distribution</td>
<td>9.3±2.0</td>
<td>28±10</td>
<td>7.6±1.7</td>
</tr>
<tr>
<td>$E_n$ ($5^\circ$)</td>
<td>143±4</td>
<td>2492±6</td>
<td>248±4</td>
</tr>
<tr>
<td>$E_n$ ($10^\circ$)</td>
<td>140±4</td>
<td>2470±6</td>
<td>245±4</td>
</tr>
<tr>
<td>$\frac{\partial E_n}{\partial E_p}$ ($\theta^\circ$)</td>
<td>1.452</td>
<td>1.006</td>
<td>1.233</td>
</tr>
</tbody>
</table>

* The angle is measured between the direction of the neutrons and the direction of the charged particle beam.

# The neutron energy quoted is an estimate of the mean value.

The reported uncertainties are based on standard uncertainties multiplied by a coverage factor, $k=2$, providing a level of confidence of approximately 95%.
As the instrument irradiations are performed at 0° also, it is not possible to make a simultaneous measurement of the fluence with the long counter. Instead there are two systems used to monitor neutron production and relate the long counter measurement to the instrument irradiation. The first of these is a current integrator measuring the beam current on the target, and the second is the ‘SLAB’ detector mounted on the Main Bay wall. The ‘SLAB’ detector consists of several BF$_3$ proportional counters embedded in three layers at different depths within a slab of polyethylene moderator. The long counter measurement essentially acts as a calibration for these two monitors on the day of the irradiations. On instrument irradiations, the fluence could then be calculated by applying the inverse-square law for distance from the target and a small correction for air out-scatter. For each irradiation, the effect of room and air in-scatter on the instrument reading was determined using the shadow cone technique, illustrated in Figure 6.2.

![Shadow Cone Diagram](image)

**Figure 6.2** An illustration of the shadow cone technique for determining the contribution to a measurement from scattered neutrons.
6.1.1 Target Scattering Effects

The neutron fields produced for these calibrations are only nominally monoenergetic. A small fraction of neutrons will have been scattered in the target assembly and therefore have lower energies than the primary neutrons, with an energy spectrum extending from the primary energy down to zero. The fluence measured with the long counter will include these target-scattered neutrons since the instrument is designed to have a response that is nearly constant over the energy range of interest.

A Monte Carlo study of scattering effects in the target assembly has been carried out by NPL to estimate the number of scattered neutrons and their energy distribution. A value for the total target-scattered fluence at each energy is shown in Table 6.2. It indicates the extent to which the neutrons are not completely monoenergetic.

<table>
<thead>
<tr>
<th>Neutron Energy (keV)</th>
<th>Target-scatter as a percentage of the total fluence</th>
</tr>
</thead>
<tbody>
<tr>
<td>144</td>
<td>1.1</td>
</tr>
<tr>
<td>250</td>
<td>2.7</td>
</tr>
<tr>
<td>565</td>
<td>0.9</td>
</tr>
<tr>
<td>1200</td>
<td>4.2</td>
</tr>
<tr>
<td>2500</td>
<td>2.4</td>
</tr>
<tr>
<td>5000</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Table 6.2. Estimates of the contribution to the total fluence from neutrons scattered within the target assembly. Data taken from NPL Calibration Certificate. (No uncertainties are given as this is the result of a Monte Carlo simulation.)
6.1.2 Neutrons from the Target Backing

Backing materials used to support the thin neutron-producing target layers have been chosen to minimise any neutron production in the backing. In most cases the contribution is negligible, however, in the case of tritium targets a small number of neutrons are produced by proton reactions in the titanium layer into which the tritium has been absorbed. The contribution has been measured by means of a blank target having a layer of titanium but no absorbed tritium. The contributions measured with the long counter were (as a percentage of the total fluence produced) 0.6% for 1.2 MeV neutron production and 2.9% for 2.5 MeV neutron production. The energies of neutrons produced in the titanium will be lower than the primary neutron energy.

6.2 Lithium Gadolinium Borate Spectrometer Irradiations

Both versions (\(^{6}\text{Li}\) capture and \(^{10}\text{B}\) capture) of the LGB spectrometer were irradiated for all six neutron energies. For each measurement, the spectrometer was held vertically in a clamp so that the centre of the detector was level and in line with the centre of the neutron producing target. The exact distance from target to detector was measured each time. Two irradiations were then performed: one with and one without the shadow cone in place. By subtracting the result of the shadow cone measurement from the result of the un-shadowed measurement, the effect of only those neutrons directly impinging on the detector can be determined. The results obtained for each measurement consist of the counting data in the four counting channels (fast neutrons (i.e. proton recoil followed by capture in \(^{6}\text{Li}\)), epithermal neutrons (i.e. capture in \(^{6}\text{Li}\)), thermal neutrons (i.e. capture in
Gd), and gamma-rays), plus the event file containing the digitised pulse data for the fast neutrons detected.

In trial runs it was noticed that there was a slight reduction in the recorded count rate when the LGB Pulse Digitiser was operated in the control room and connected to the detector via the installed cable looms in the accelerator hall. Also, on examination of the event files, there was found to be an increase in the proportion of mis-shaped pulses in the file presumably due to electronic noise or signal reflection in the very long cable. For these reasons it was decided to operate with the LGB Pulse Digitiser and data logging computer in the accelerator hall close to the detector. Data collection was started before leaving the hall, which results in some background counts accruing before and after the neutron irradiation. This background was very small, however, and closed circuit television cameras in the hall were used to monitor the digitiser LCD display and record the count information at the beginning and end of the neutron irradiation.

6.2.1 LGB Results (Lithium-6 Version)

The fluence delivered to the LGB $^6\text{Li}$ version on each of the irradiations, with and without shadow cone, is shown in Table 6.3. Dividing the number of counts in each of the four counting channels by the neutron fluence delivered gives the detector response per unit fluence. The counting channel results are displayed in Figures 6.3 to 6.9.
Table 6.3 Details of the fluence delivered during each of the irradiations for the $^6$Li version of the LGB spectrometer. Data taken from NPL Calibration Certificate.

The response of all counting channels for the irradiations without the shadow cone is shown in Figure 6.3, while that for irradiations with the shadow cone is shown in Figure 6.4. Results of the shadow cone irradiations must be normalised to fluence delivered before they can be subtracted from the non-shadowed run to remove the effect of room-scattered neutrons. This process leaves just the counts from neutrons arriving directly from the target. The 'direct' irradiation results are shown in Figure 6.5 for all counting channels. Results for non-shadowed, shadowed and direct irradiation are also plotted separately for each counting channel to indicate the relative fluence contribution from direct and room scattered neutrons. Figure 6.6 shows the fluence response of the fast neutron channel, Figure 6.7 shows the gamma ray channel, Figure 6.8 shows the epithermal neutron channel, and Figure 6.9 shows the thermal neutron channel.
Figure 6.3 Non-shadowed irradiation results for the LGB ($^6$Li version) counting channels showing the fluence response.

Figure 6.4 Shadow cone irradiation results for the LGB ($^6$Li version) counting channels showing the fluence response.
Figure 6.5 Direct irradiation results for the LGB (\(^6\)Li version) counting channels showing the fluence response.

Figure 6.6 Fluence response for the LGB (\(^6\)Li version) fast neutron counting channel.
Figure 6.7 Fluence responses for the LGB ($^6$Li version) gamma ray counting channel.

Figure 6.8 Fluence responses for the LGB ($^6$Li version) epithermal neutron counting channel.
Figure 6.9 Fluence responses for the LGB (6 Li version) thermal neutron counting channel.

The event file data is presented in its raw format is similar to a multi-channel analyser display, the channels being integrated pulse area and the vertical scale indicating the number of pulses counted in that channel. Each capture-gated event consists of two pulses, a proton recoil pulse and a 6 Li capture pulse (or 10B capture in the alternative spectrometer). Both pulses are displayed in this pulse area distribution, with the large area 6 Li capture pulses appearing as a broad peak on the right, and the proton recoil pulses mainly to the left of this.

The offline analysis of the capture-gated event file is now performed by a program called EVENTHANDLER that can read the event file format and selectively output data for analysis on a spreadsheet.
EVENTHANDLER is used to separate the proton recoil pulses from the $^6$Li capture pulses, then the pulses can be sorted by the spreadsheet into the chosen pulse area binning scheme. In the capture-gated detection principle, the proton recoil pulse area distribution should closely resemble the incident neutron energy distribution. Figures 10 through to 15 show the proton recoil pulse area distributions with a 20 bin structure.

It can be seen that the pulse area distribution for the 1.2 MeV irradiation that the majority of counts occur in a single bin at the low end of the scale (in the pulse area 100 – 200 bin). The fact that no counts were recorded in the preceding bin (pulse area 0 – 100) would seem to indicate that a minimum pulse area of 100 exists for a pulse to be recorded as a fast neutron induced recoil proton.

Figure 6.10 Proton recoil pulse area distribution for 144 keV mono-energetic neutrons in LGB ($^6$Li version).
Figure 6.11 Proton recoil pulse area distribution for 250 keV mono-energetic neutrons in LGB ($^6$Li version).

Figure 6.12 Proton recoil pulse area distribution for 565 keV mono-energetic neutrons in LGB ($^6$Li version).
Figure 6.13 Proton recoil pulse area distribution for 1.2 MeV mono-energetic neutrons in LGB (Li version).

Figure 6.14 Proton recoil pulse area distribution for 2.5 MeV mono-energetic neutrons in LGB (Li version).
6.2.2 LGB (Boron-10 Version)

The results for the version of LGB employing capture in $^{10}$B showed that the pulse digitiser settings are optimised for the $^6$Li version. Pulse area distributions for the $^{10}$B version at neutron irradiation energies of 2.5 MeV and 5.0 MeV are shown in Figures 6.16 and 6.17 respectively. No distribution could be plotted for the 1.2 MeV neutron irradiation as there were only 2 fast neutron events were recorded in 22 minutes of irradiation.

Figure 6.15 Proton recoil pulse area distribution for 5.0 MeV mono-energetic neutrons in LGB ($^6$Li version).
Figure 6.16 Proton recoil pulse area distribution for 2.5 MeV mono-energetic neutrons in LGB (10B version).

Figure 6.17 Proton recoil pulse area distribution for 5.0 MeV mono-energetic neutrons in LGB (10B version).
6.3 Discussion of LGB results

As with all organic scintillators, the light output in the LGB plastic from neutron induced recoil protons below 1 MeV cannot be distinguished from the light output of gamma-ray induced Compton electrons. Therefore, no capture-gated events (i.e. fast neutron counts) were expected for those measurements where the neutron energy was below 1 MeV because the pulse shape discriminator stage in the pulse digitiser should reject them.

Initially these unexpected counts were attributed to the long cables used when operating the digitiser and data logging computer in the control room. However, the effect persisted after these electronics were moved into the accelerator hall.

From the counting channel data, it is noted that the gamma-ray channel and thermal neutron channel both display a similar response over the three lower energies, 144, 250 and 365 keV. In the case of the thermal neutron channel, the imposition of the shadow cone shows that almost all the counts at these three energies were coming direct from the neutron producing target. Of course, thermal neutrons are expected to come from room return and hence to be largely independent of the original neutron energy. The epithermal neutron counting channel shows no such trend in the lower energies, and a much higher proportion of the counts are shown to be from room return in the shadow cone measurement.

The lower three neutron energies are all produced from the same LiF target using the \(^{7}\text{Li}(p,n)\) reaction. Other nuclear reactions are possible with the fluoride in the target such as \(^{19}\text{F}(p,\alpha)^{16}\text{O}\) accompanied by the emission of a 6.129 MeV gamma-ray. These high energy gammas are likely to be misinterpreted by the digitiser as coming from neutron captures in gadolinium inside the LGB detector, leading to an increase in the thermal
neutron count rate as previously noted. It is suggested here that high energy gamma-rays from the LiF target could also explain the fast neutron counts appearing for the three energies that are below the supposed 1 MeV threshold for the pulse shape discriminator.

6.4 N-Probe Monoenergetic Irradiation Results
The opportunity was taken to test the performance of the Bubble Technology N-Probe in mono-energetic fields and investigate the accuracy of its fluence and dose equivalent information. In addition, the N-Probe results would serve to confirm the neutron energy spectrum at the measurement position. NPL does not perform any neutron spectrometry itself during mono-energetic irradiations.

The N-Probe was mounted on a horizontal platform with the centre of the aluminium cap covering the NE-213 scintillation detector in line and level with the neutron producing target. This results in the 3He detector seeing a slightly lower neutron fluence since it is further from the target. Control of the N-Probe remotely from the control room is not possible. The instrument was instead set to acquire data before the accelerator hall was secured. The number of counts accumulating in the short time before the beam-flap is removed and the irradiation begun will be zero in most cases. When the beam energy is high, as it is for the 5 MeV irradiation, there is some production of neutrons in the tantalum beam-flap due to reactions with deuterium atoms that accumulate in the metal. Even then the effect of these on the N-Probe spectrum would be negligible due to the large distance from the beam-flap to the measurement position.
The contribution to the measured spectrum from scattered neutrons was assessed using the shadow cone technique.

Figure 6.18 shows the incident neutron spectrum unfolded by N-Probe on irradiations without the shadow cone for 1.2, 2.5 and 5.0 MeV. (Negative fluence values in some energy groups caused problems in determining the neutron spectrum after shadow cone correction.) N-Probe spectra show the expected monoenergetic peak in the correct energy group for each irradiation, together with some low energy scattered neutrons.

Correction for room-scattered neutrons could be applied to the total fluence in the measured spectrum by subtracting the reading for the shadow cone irradiation from that for the irradiation with no shadow cone. The result should be the reading for only the directly incident neutrons, and this can be compared with the NPL data on delivered fluence and dose equivalent. Results of this procedure are presented in Table 6.4. It can be seen that N-Probe over-estimates the fluence at all energies except 565 keV where it agrees very closely to the NPL value. Not long after these irradiations took place the N-Probe was recalled to BTI for a component replacement. It was returned in July 2002 with new versions of the DOSPEC software that resides on the Microspec analyser and the NVIEW software used for analysing the data on PC.
Figure 6.18. Neutron spectra unfolded by N-Probe from monoenergetic irradiations at a) 1.2 MeV, b) 2.5 MeV, and c) 5.0 MeV. Low energy neutrons present are from room scatter.
<table>
<thead>
<tr>
<th>Neutron Energy (MeV)</th>
<th>Neutron Fluence (cm(^{-2}))</th>
<th>Neutron Ambient Dose Equivalent (H^*(10)) ((\mu)Sv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NPL</td>
<td>N-Probe</td>
</tr>
<tr>
<td>5.0</td>
<td>1.35x10^6 ± 7.8%</td>
<td>2.09x10^6</td>
</tr>
<tr>
<td>2.5</td>
<td>4.72x10^5 ± 7.8%</td>
<td>8.89x10^5</td>
</tr>
<tr>
<td>1.2</td>
<td>4.02x10^5 ± 8.9%</td>
<td>5.58x10^5</td>
</tr>
<tr>
<td>0.565</td>
<td>1.11x10^6 ± 6.2%</td>
<td>1.09x10^6</td>
</tr>
<tr>
<td>0.250</td>
<td>5.45x10^5 ± 6.4%</td>
<td>1.50x10^6</td>
</tr>
<tr>
<td>0.144</td>
<td>1.32x10^6 ± 6.2%</td>
<td>3.12x10^6</td>
</tr>
</tbody>
</table>

Table 6.4. N-Probe fluence and dose readings from the unfolded neutron spectrum compared to NPL data. In almost all cases the N-Probe significantly overestimates the fluence, while dose is overestimated at high energies and underestimated at low energies.

Re-analysing the NPL data using the updated software showed considerable differences to fluence and dose estimations. A slight improvement in agreement with NPL data was seen at 2.5 and 5.0 MeV, but at 1.2 MeV the overestimate got worse. Previously, the N-Probe was in very good agreement with NPL data at 565 keV, but the new software gives an underestimate. At 144 and 250 keV there was a large overestimate with the old software, but this has now become an underestimate. In fact, the fluence reading is so low that the simple shadow cone subtraction method used fails at these two energies. (The shadow cone spectrum has a higher fluence reading than the non-shadowed spectrum.) Response function data used by the programs is unchanged, while there are only slight differences in the calibration data. The suspicion is that changes have been made to the working of the unfolding program, particularly with regard to the \(^3\)He counter. Figure 6.19 shows the N-Probe fluence
readings relative to the NPL conventional true value at each energy for the old and new software.

![Graph showing N-Probe fluence readings for the six irradiation energies relative to the fluence NPL fluence data comparing the old and new versions of the unfolding software.]

**Figure 6.19.** N-Probe fluence readings for the six irradiation energies relative to the fluence NPL fluence data comparing the old and new versions of the unfolding software.

### 6.5 Thermal Neutron Irradiations

To assess the thermal response of the lithium gadolinium borate spectrometer (LGB), the NPL Thermal Neutron Facility, commonly known as the ‘thermal pile’ was selected. Here thermal neutrons are produced by the moderation of fast neutrons, created by the reaction of a deuteron beam on beryllium targets, in a large graphite block. Very small items (< a few cm in size) can be placed in a cavity near the centre of the block. Larger items, such as the LGB, are irradiated in a ‘thermal column’ that extracts a beam of thermal neutrons from within the pile.

The Thermal Neutron Facility is housed in the same room as the low-scatter facility and runs on a separate beamline from the NPL Van de Graaff accelerator that passes...
through the entire length of the graphite moderator block. Fast neutrons are produced by a 2.8 MeV deuterion beam striking two beryllium targets, which are semi-circular in shape and placed 160 cm apart, equidistant from the centre of the block. A servo control system equalises the fraction of the deuterion beam incident on each target. A semi-circular tantalum plate fixed vertically in the beamline at the entrance to the graphite block acts as a ‘beam-dump’ and forms part of the thermal pile control mechanism.

Three ionisation chambers are located within the moderator block, one underneath each of the beryllium targets and another below the central cavity. Signals from these ionisation chambers are used to control the fraction of the deuterion beam striking the targets and the beam-dump, and hence control the neutron production in the pile, via a servo-system acting upon the vertical and horizontal steering plates in the beamline. The ionisation chambers below the targets control the vertical deflection of the beam to give approximately equal neutron outputs from each target. The ionisation chamber below the central cavity controls the horizontal deflection of the beam and thus the distribution of the beam between the targets and the tantalum plate. The desired thermal neutron flux output can be obtained by presetting the servo-system horizontal beam deflection.

Of course, the neutron field produced will not be purely thermal in energy, there will be an epithermal component from primary neutrons that have not been sufficiently moderated. The size of this epithermal component can vary because in reality there is more than one neutron producing reaction happening.

Primary neutrons are, of course, produced by the d-Be reaction in the beryllium targets, but there is also a small contribution from d-D reactions, and from the action
of the deuteron beam on contaminants deposited on the targets and beam-dump. Therefore, the ratio of the thermal component to the epithermal component depends on the deuteron beam conditions and the cleanliness of the vacuum system, targets and tantalum plate.

The thermal column used for the irradiation of the LGB, consists of a stainless steel cylinder (cross-sectional area 1000 cm\(^2\)) with cadmium-lined sides sunk into the graphite above one of the neutron-producing targets. The thermal neutron fluence in the column is monitored by a \(^{235}\text{U}\) fission chamber positioned in the graphite between the bottom of the column and the beryllium target. The column length can be adjusted in half-metre steps from 1 m to 3 m, and the monitor can be calibrated in terms of the fluence at any position in the column by use of gold foil activation.

**Figure 6.20** Construction of the NPL thermal pile showing the location of the beryllium targets fission chambers within the graphite block, and the position of the thermal column.
The thermal column set at the 1 m reference height. The effective centres of the LGB detectors were about 20 mm above the reference height. Three measurements were made for each instrument, one essentially "free-in-air", one with a 1 mm thick cadmium plate covering the end of the thermal column, and another with the cadmium plate again in place and a cylindrical enclosure of cadmium covering the entire instrument.

### 6.5.1 LGB Thermal Results

Table 6.5 shows the sub-cadmium fluence delivered for each irradiation and the fluence response of both versions of LGB in each of the four counting channels. The fluence value given by NPL is the Westcott sub-cadmium fluence free-in-air. The uncertainty on the gold foil fluence measurement is 1.3% at 95% confidence level. Further uncertainties, including uncertainty in the calibration of the NPL BF₃ detector used for monitoring and the effective temperature of the thermal neutron distribution, lead to an expanded uncertainty of 2.8%.

When the cadmium plate is positioned over the thermal neutron beam the effect on the four counting channels is not entirely what would be expected. The fast neutron count is reduced, perhaps because of fast neutrons being scattered away from the detector, but more likely indicating the effect of random coincidences (captures of thermal neutrons in $^6$Li) triggering capture-gated counting. The epithermal neutron count is reduced as expected, again showing we can have confidence in this counting channel, and perhaps supporting the random coincidence explanation of the fast neutron count. Both the thermal neutron and gamma ray count are increased significantly. A large number of prompt gamma rays are expected from neutron capture in the cadmium, which emits a series of gamma rays totalling 9 MeV on average. It is suggested here
that the thermal neutron counter is again being confused by these gamma rays, and mistaking them for the characteristic signal for neutron capture in gadolinium.

Placing the cadmium tophat over the detector (with the cadmium plate still in place) means the detector is completely surrounded with cadmium, and is therefore more completely shielded against thermal neutrons. However, the count rate for all counting channels increases slightly. It is proposed that in-scattering by the cadmium tophat of neutrons that would otherwise not have interacted with the detector is the cause of these increases. This would be the case if the neutron beam from the thermal column was very well collimated.

<table>
<thead>
<tr>
<th>LGB version</th>
<th>Irradiation details</th>
<th>Sub-cadmium Fluence (x10⁵ cm⁻²)</th>
<th>'Fast' Channel</th>
<th>'Epithermal' Channel</th>
<th>'Thermal' Channel</th>
<th>'Gamma' Channel</th>
</tr>
</thead>
<tbody>
<tr>
<td>⁶Li</td>
<td>Free-in-air</td>
<td>7.171</td>
<td>7.18E-02 ± 2.83%</td>
<td>2.62E-02 ± 2.81%</td>
<td>4.06E-02 ± 2.86%</td>
<td>1.67E+00 ± 2.80%</td>
</tr>
<tr>
<td></td>
<td>Free-in-air</td>
<td>3.720</td>
<td>8.67E-02 ± 2.85%</td>
<td>6.76E-01 ± 2.81%</td>
<td>9.64E-02 ± 2.85%</td>
<td>4.20E+00 ± 2.80%</td>
</tr>
<tr>
<td></td>
<td>Cd plate</td>
<td>5.555</td>
<td>3.17E-02 ± 2.94%</td>
<td>1.88E-01 ± 2.82%</td>
<td>1.40E-01 ± 2.83%</td>
<td>7.33E+00 ± 2.8%</td>
</tr>
<tr>
<td></td>
<td>Cd plate + cover</td>
<td>3.854</td>
<td>3.28E-02 ± 2.90%</td>
<td>2.00E-01 ± 2.82%</td>
<td>1.44E-01 ± 2.82%</td>
<td>7.44E+00 ± 2.80%</td>
</tr>
<tr>
<td>⁹Li</td>
<td>Free-in-air</td>
<td>4.314</td>
<td>9.11E-03 ± 3.22%</td>
<td>7.00E-02 ± 2.86%</td>
<td>1.22E-01 ± 2.83%</td>
<td>6.33E+00 ± 2.80%</td>
</tr>
<tr>
<td></td>
<td>Cd plate</td>
<td>4.428</td>
<td>3.86E-03 ± 3.70%</td>
<td>2.22E-02 ± 2.98%</td>
<td>1.49E-01 ± 2.83%</td>
<td>7.11E+00 ± 2.80%</td>
</tr>
<tr>
<td></td>
<td>Cd plate + cover</td>
<td>5.815</td>
<td>4.73E-03 ± 3.39%</td>
<td>2.34E-02 ± 2.93%</td>
<td>1.58E-01 ± 2.82%</td>
<td>7.36E+00 ± 2.80%</td>
</tr>
</tbody>
</table>

Table 6.5 Fluence responses measured on the NPL Thermal Column for the ⁶Li and ⁹B versions of LGB.


6.6 Conclusions

The monoenergetic responses for the LGB spectrometer show that the energy resolution becomes poorer with increasing neutron energy. This is due to incomplete thermalisation of the incident fast neutrons in the detector volume, the non-linearity of the proton light output function in the plastic scintillator, and the increasing importance of elastic and inelastic scattering from carbon. This poor energy resolution, and the complicated shape at higher energies, prevents the straightforward conversion from measured pulse area distribution to incident neutron spectrum and hence the ambient dose equivalent. This would require the use of an unfolding algorithm using a detailed response function set for many more energies than presented here.

It is also clear that the ‘thermal’ neutron counting channel, since it works by detecting the characteristic gamma ray signal from neutron capture in gadolinium, can be triggered by high energy gamma rays in the environment. This is suspected to be the case on the 144, 250 and 565 keV irradiations, where 6-7 MeV gamma rays were produced from the lithium fluoride target. It is suspected that this problem explains the unusual results in the thermal irradiations too due to prompt gamma rays from neutron capture in the cadmium shield. The waveform digitiser prevented any detailed study of this problem as the gamma ray pulses are not made available for examination.
Chapter 7

Measurements in Working Environments

7.1 Low Flux Reactor, Petten

The Low Flux Reactor (LFR) situated at NRG Petten in the Netherlands is a 30 kW graphite and light water moderated ARGONAUT reactor following the design originating from the Argonne National Laboratory in the United States. It has a single slab annular core situated between two concentric aluminium cylinders. The inner cylinder is filled with graphite and functions as the inner neutron reflector. The outer cylinder is surrounded by a graphite block that acts as the outer neutron reflector. Biological shielding consists of a thick layer of concrete enclosing the entire structure. One wall of the shield has a cavity containing a large graphite block called the thermal column, which produces an almost pure beam of thermal neutrons. The facility also has a large irradiation chamber in the wall opposite the thermal column, accessed by the removal of a section of the biological shield that is mounted on a trolley. This chamber is used for animal trials into boron neutron capture therapy (BNCT). The shielding on the other two walls is penetrated by beam tubes that gives access to fast and thermal neutrons. On the working platform on top of the reactor is a neutron radiography rig. Figure 7.1 shows a plan diagram of the reactor construction.

Experiments were performed at the LFR to test the functioning of LGB in reactor neutron spectra. A further objective of the work was to determine if the thermal counting channel of LGB would be affected by high energy gamma-rays that would be encountered in the reactor environment. Nitrogen-16 gas is produced in the reactor
coolant water via the reaction $^{16}\text{O}(n,p)^{16}\text{N}$. Radioactive decay of nitrogen-16 leads to the emission of gamma-rays at 6.13 MeV and 7.12 MeV with a half-life of 7.13 seconds.

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**Figure 7.1.** A diagram of the 30 kW LFR reactor at Petten showing the construction details.

To determine the effect of these nitrogen-16 gamma rays, a series of instruments were assembled on the access hatch directly above the reactor water dump tank, where the coolant water is discharged when the reactor is scrammed (shutdown). In this position the neutron fluence is very low, being outside the biological shielding, but there is a higher gamma ray fluence caused in part by nitrogen-16 in the pipes circulating the coolant. The instruments used were the $^6\text{Li}$ version of LGB, a Mk7 NRM neutron dosimeter and a 2" x 2" NaI(Tl) gamma ray spectrometer. The NaI(Tl) detector will only detect gamma rays, and the Mk7 NRM will only detect neutrons. The LGB is of course sensitive to both radiations. The 'epithermal' ($^6\text{Li}$ capture) channel should only
be sensitive to neutrons, and the ‘gamma’ channel only to gamma rays. The ‘thermal’ (gadolinium capture) channel is expected to be sensitive to both neutrons and high energy gamma rays. Pulses from the Mk7 NRM were counted using a counter/scaler box. An energy calibration had been carried out for the NaI(Tl) detector that allowed a region of interest to be set on the multichannel analyser where the nitrogen-16 peak would lie. This energy calibration could only be very approximate due to the lack of any long-lived radioisotope sources of sufficient energy.

The reactor was operated at a steady power for some time to allow the build up of nitrogen-16 in the coolant. A 60 second measurement was then made with each of the three instruments to establish the “steady” count rate. The reactor was then scrammed, sending all the coolant water to the dump tank in just a few seconds. At the instant of the scram, the three instruments were again set to take a measurement over 60 seconds. This is the “scram” count rate. Since 60 seconds is over 8 half-lives for the nitrogen-16 it should have largely decayed in this time. This procedure was repeated for three different reactor powers, 1, 5 and 10 kW. Results are presented in Table 7.1 below. Ratios of the scram count rate to the steady count rate are included in the table to highlight the nitrogen-16 effect more clearly.
### Table 7.1. Results of $^{16}$N investigation at LFR Petten for three reactor powers. Detector counts shown are for a 60 second measurement.

<table>
<thead>
<tr>
<th>Reactor Power</th>
<th>1 kW</th>
<th>5 kW</th>
<th>10 kW</th>
</tr>
</thead>
<tbody>
<tr>
<td>Detector</td>
<td>Steady</td>
<td>Scram</td>
<td>Ratio Scram/Steady</td>
</tr>
<tr>
<td><strong>Mk7 NRM</strong></td>
<td>17±4</td>
<td>23±5</td>
<td>1.35±0.32</td>
</tr>
<tr>
<td><strong>LGB thermal</strong></td>
<td>104±10</td>
<td>373±19</td>
<td>3.59±0.11</td>
</tr>
<tr>
<td><strong>LGB epithermal</strong></td>
<td>105±10</td>
<td>85±9</td>
<td>0.81±0.14</td>
</tr>
<tr>
<td><strong>LGB gamma</strong></td>
<td>8404±92</td>
<td>19030±138</td>
<td>2.26±0.01</td>
</tr>
<tr>
<td><strong>NaI(Tl)</strong></td>
<td>13692±117</td>
<td>31122±176</td>
<td>2.27±0.01</td>
</tr>
</tbody>
</table>

Starting with those instruments that only detect neutrons, as should be expected, the Mk7 NRM recorded very few neutron counts either at steady power or the period after the scram. Fewer neutron counts were expected after the scram, so the scram:steady ratio should be less than unity. This is true for 5 kW and 10 kW, but not at 1 kW due to poor counting statistics. The LGB ‘epithermal’ channel shows similar behaviour, but is more sensitive and so achieves a scram:steady ratio of less than unity for all three powers.

For the gamma ray only detectors, i.e. NaI(Tl) and LGB ‘gamma’ channel, the ratio should be greater than unity due to the enormous increase in nitrogen-16 gamma rays from the coolant water in the dump tank. This is true for all reactor powers for both these detectors. The ratio remains very stable for the LGB ‘gamma’ channel, which registers gamma rays depositing less than 2 MeV of energy in the detector, increasing
on average by a factor of 2.33. For the NaI(Tl) detector the ratio shows quite a large variation between different powers, but without a clear trend. It should be noted that the counts in the nominal position of the nitrogen-16 peak were taken as the NaI(Tl) reading.

The LGB ‘thermal’ channel ratio shows an increase of a factor of three or more in the counts post scram. This proves that high energy gamma rays in the environment are easily mistaken for thermal neutron events by the LGB data processing system.

7.2 Realistic Neutron Calibration Fields

7.2.1 Introduction

It has been suggested by many authors that the effect of dissimilarity between calibration field and workplace field on dosemeter response could be tackled by developing calibration fields that are more representative of the workplace field, a realistic neutron calibration field (RNCF). This would eliminate the need for field dependent correction factors to be applied to the dosemeter reading. Of course, this does not obviate the requirement for spectrometry to be performed in the workplace, as construction of a suitable RNCF relies on knowledge of the neutron energy spectrum.

7.2.2 Review of RNCFs

Many laboratories have experimented with RNCFs, using different techniques to simulate neutron fields that are of particular interest to them. The simplest approach possible has been taken by PTB in Germany, where the spectrum produced by the ISO-recommended radioisotope sources in an existing calibration facility has been investigated [Kluge, 1997]. In the standard calibration procedure for a neutron instrument, the contribution to a measurement from neutrons scattered from the walls
of the room is removed by means of a second measurement performed with a shadow cone imposed between the source and the detector, leaving just the direct contribution. Due to the modest dimensions of the room, and the thickness of the concrete walls, the scattered contribution consists mainly of low energy neutrons with an almost isotropic directional distribution. Different fields can be produced by the use of $^{241}$Am/Be or $^{252}$Cf sources of different strengths, or $^{252}$Cf with a D$_2$O moderator sphere. The later would normally be used with a cadmium cover to reduce the number of thermal neutrons emitted, but this can be removed to enhance the thermal component if desired. Further more, the realistic field can consist of the ‘scattered’ and ‘direct’ neutrons, or, by use of the shadow cone, just the scattered neutrons. Measurements have been made with Bonner spheres and calculations performed with MCNP for all the possible combinations of the variable parameters. The total fluence rate can be determined with a relative standard uncertainty of about 4%, and the ambient dose equivalent to around 5%.

Researchers in Canada have investigated the feasibility of producing fields representative of those around heavy water moderated CANDU reactors using moderator assemblies with either a $^{252}$Cf source or a sealed-tube neutron generator utilising the $^3$H(d,n) reaction [Nunes, 1997]. Monte Carlo simulations with MCNP were compared with spectra measured with Bonner spheres at various positions around CANDU power reactors [Nunes, 1996a] [Nunes 1996b].

GSF in Germany have constructed a facility based around neutrons produced by the Be(d,n) reaction by a Van de Graaff accelerator [Schraube, 1997]. Using a thick
target and a deuteron energy of 2.8 MeV, the average neutron energy along the axis of
the beam is 2 MeV, which is comparable to the energy of fission neutrons. The
neutron producing target is inside a 130 cm x 170 cm x 116 cm cavity with concrete
walls 25 cm thick. Further moderating materials can be introduced inside the cavity,
either on the walls or in front of the target, to modify the field characteristics, e.g. a 30
cm D$_2$O sphere in front of the target and 5 cm of iron on the walls. The field produced
is intended to simulate fields around commercial PWRs and MOX fuel transport
containers. Again, simulations were performed using MCNP, and here the problem of
describing the source term for accelerator produced neutrons is encountered. In the
model the source was approximated by 2 MeV monoenergetic neutrons isotropically
emitted from a disc 10 mm in diameter.

PTB also have their own version of Van de Graaff accelerator based RNCF, using the
$^7$Li(p,n) reaction. The neutron producing target is mounted on the end of a beamline at
the centre of a large experimental hall with very low room return. Around the target
are concentric spheres, first of polyethylene with a radius of 4.7 cm, and then of
graphite with a radius of 14 cm. The exact shape of the spectrum varies as a function
of angle with respect to the direction of the incident proton beam. Necessarily, the
usual procedures of neutron instrument calibration still hold with a shadow cone
measurement being needed to determine the direct contribution from the source
assembly. The final construction was the result of extensive Monte Carlo calculations
with a modified version of MCNP capable of describing neutron production in a thick
lithium target. Despite this there is still some discrepancy between calculated and
measured (Bonner sphere) results, probably due to certain simplifications used in the
source term such as neglecting energy or angular straggling. The discrepancy becomes larger with increasing angle.

Another accelerator driven RNCF has been constructed at Cadarache in France [Chartier, 1992], but taking a different approach to that described above. Instead of producing an extended neutron field around a central source, the aim was to produce a collimated beam into which instruments could be placed. Neutrons with a mean energy of 14.6 MeV originate from the $^3\text{H}(d,n)$ reaction driven by a 150 kV SAMES accelerator. The tritium target is surrounded by a $^{238}\text{U}$ hemisphere that converts the 14.6 MeV neutrons into fission neutrons. An iron shell 15 cm thick covers this to reduce the number of remaining 14.6 MeV neutrons by the $(n,2n)$ reaction. This setup sits inside a cylindrical scattering chamber with polyethylene walls into which additional moderating materials can be placed. This facility has been established for some time and can be considered well characterised. Measurements of the neutron field produced have been made, including a large-scale comparison exercise [Thomas, 1997], using proton recoil counters, Bonner spheres, tissue equivalent counters, area survey instruments, and personal dosemeters and show that the facility is capable of reproducing neutron fields found near power reactors and fuel transport containers [Posny, 1992].

Here in the UK, NPL have decided to create a RNCF representative of gas-cooled nuclear power stations. Initial Monte Carlo modelling results, accompanied by comparisons with workplace spectra and predictions of dosemeter response from SPKTBIB, are shown in NPL Report CIRM 27 [Thomas, 1999]. Designs based on
radioisotopic and accelerator-produced neutrons were considered. The design chosen
use the $^7\text{Li}(p,n)$ reaction with a 40 cm diameter D$_2$O moderator sphere as this gave the
best fit to the available gas-cooled reactor spectra and the accelerator based approach
gives the greatest control over the field produced. Trials of this service are expected to
begin soon.

Growing interest in the subject of RNCFs has led to the creation of ISO standard
12789 dealing with their production, characterisation and use [ISO, 2000].

7.2.3 Experiments at NEPTUNE

NEPTUNE is a zero-power reactor operated by Rolls-Royce and Associates plc at
their site in Derby, UK. The primary function of the reactor is to test fuel and core
designs for the Naval Nuclear Propulsion Programme.

An investigation [Palmer, 2002] was in progress to assess whether the reactor could
be used to provide a spectrum representative of a naval PWR. Such a facility, when
properly characterised, would provide a useful tool for researching dosemeter and
spectrometer performance. This work provided an opportunity to test the LGB
spectrometer in an unknown spectrum and compare the results with those from other
instruments and calculations.

Following the guidance found in the ISO standard, a Monte Carlo simulation of the
NEPTUNE facility was created with the MCBEND neutron transport code. A two part
model was built, the first part to predict the neutron leakage spectra from the reactor
tank (the source term), and the second to simulate neutron transport in the reactor hall.
Three positions of interest had been selected for the study:

1. Working platform; over a concrete-walled storage area, level with the height of the top of the reactor tank,
2. Top gantry; on a gantry several meters above position 1,
3. Radiography room; a small room in the corner of the reactor hall with lead-lined walls.

Figure 7.2. Plan of the NEPTUNE reactor hall showing the key features and the location of the measurement positions.

Work to characterise the neutron field at these locations has been performed using the standard Mk7 NRM area survey meter, a Duosphere instrument consisting of a pair of polythene spheres (5” and 8” diameter) containing a $^3$He counter (type SP9) on loan from BAe Systems Ltd. An estimation of spectral hardness could be made from the ratios of count rates in the 5” and 8” sphere using knowledge of their energy response contained in the SPKTBIB database. Each instrument was equipped with an electronic counter to record the number of pulses emitted, with the BAe spheres being capable of logging data accumulated over set time intervals. For these experiments the interval
was set a 10 seconds in order to provide a detailed record of the reactor operation. An approximate measurement of the gamma ray dose was also made using a Nuclear Enterprises PDR2 gamma monitor.

Figure 7.3 shows the Bonner sphere count rate data with time for one of the reactor runs. The different stages of operation can clearly be seen in this plot. Initially almost no neutrons are present, just an occasional count recorded from the small Am/Be sources in the installed reactor instrumentation. A $^{252}$Cf start-up source is then raised into position and the spheres begin to count at a steady rate until the count rate again returns to zero when water being pumped into the reactor tank covers the source. Neutrons begin to be detected again when the reactor becomes source-free critical and then the power is increased for the period of the measurement. These features appear again but in reverse after reactor shut down at the end of the measurement.

![Figure 7.3. Count rates for the 5" and 8" Bonner spheres positioned in the radiography room position.](image)
The Bubble Technologies N-Probe was used to measure the spectrum in the same locations at the same time as the moderator sphere measurements. These simultaneously acquired spectra are therefore affected by the proximity of the moderator spheres, but this was unavoidable because of time limitations and the nature of operations at NEPTUNE. It would not be possible to control the N-Probe during the run. It was therefore set to begin acquiring a spectrum before the reactor start-up sequence had begun. This had the drawback that neutrons from the $^{252}$Cf start-up source would be detected during the reactor's approach to power. Also, more seriously, the spectrum acquisition time would have to be altered before running the unfolding program, to account for most of the neutrons being detected in a small fraction of the overall measuring time.

### 7.2.4 Duosphere Results

To assist in understanding the meaning of the ratio value, a set of neutron spectra thought to be representative of the fields found on board submarine plant were chosen from the SPKTBIB catalogue. The expected reading for a 5” and an 8” Bonner sphere (using data for NPL $^3$He set) and a Leake type survey instrument were then calculated by folding the selected spectra with the appropriate response function. The expected ratio of the two spheres in each field could then be found.

The mean count rate for the set of Duospheres and the Mk7 NRM are presented in Table 7.2 below, together with the ratio of the 5” sphere reading to the 8” sphere reading.
Table 7.2 Mean count rate data for the Duosphere and Mk7 NRM in the three measurement positions and the 5"/8" ratios.

<table>
<thead>
<tr>
<th>Position</th>
<th>Instrument</th>
<th>Mean counts per second</th>
<th>5&quot;/8&quot; Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radiography Room 5&quot; sphere</td>
<td>49.285</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8&quot; sphere</td>
<td>29.082</td>
<td>1.695</td>
<td></td>
</tr>
<tr>
<td>Mk7 NRM</td>
<td>1.895</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Working Platform 5&quot; sphere</td>
<td>76.617</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8&quot; sphere</td>
<td>33.629</td>
<td>2.278</td>
<td></td>
</tr>
<tr>
<td>Mk7 NRM</td>
<td>3.404</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Top Gantry 5&quot; sphere</td>
<td>88.927</td>
<td></td>
<td></td>
</tr>
<tr>
<td>8&quot; sphere</td>
<td>59.159</td>
<td>1.503</td>
<td></td>
</tr>
<tr>
<td>Mk7 NRM</td>
<td>7.024</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The 5"/8" ratio can be compared to the figure calculated by SPKTBIB for the set of representative spectra selected by Palmer. Details of the dosimetric quantities for these spectra and the 5"/8" ratio are presented in Table 7.3. From the Duosphere measurement data the representative spectra that most closely matches that measurement position can be chosen.
Table 7.3. Representative spectra taken from SPKTBIB.

<table>
<thead>
<tr>
<th>Description</th>
<th>Index No.</th>
<th>Mean Energy (keV)</th>
<th>Mean ( w_R )</th>
<th>Mean ( H^*(10) ) (pSv cm(^2))</th>
<th>SPKTBIB 5''8'' Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR3 location 3, within containment</td>
<td>72</td>
<td>85.5</td>
<td>7.63</td>
<td>-48.55</td>
<td>2.51</td>
</tr>
<tr>
<td>PWR location 81 From IAEA318</td>
<td>80</td>
<td>142</td>
<td>8.34</td>
<td>61.6</td>
<td>2.23</td>
</tr>
<tr>
<td>Gosgen PWR, position 2</td>
<td>338</td>
<td>35.1</td>
<td>7.29</td>
<td>32.5</td>
<td>2.68</td>
</tr>
<tr>
<td>Caorso BWR inside reactor cavity</td>
<td>339</td>
<td>282</td>
<td>13.9</td>
<td>141</td>
<td>1.66</td>
</tr>
<tr>
<td>Czech PWR circulation pump room</td>
<td>382</td>
<td>52.5</td>
<td>7.45</td>
<td>40.2</td>
<td>2.44</td>
</tr>
<tr>
<td>Czech PWR reactor hall</td>
<td>389</td>
<td>305</td>
<td>13.4</td>
<td>141</td>
<td>1.59</td>
</tr>
<tr>
<td>Swiss BWR reactor level stairwell</td>
<td>470</td>
<td>30.7</td>
<td>6.78</td>
<td>27.4</td>
<td>2.66</td>
</tr>
<tr>
<td>Swiss BWR 16m level</td>
<td>472</td>
<td>53.6</td>
<td>7.53</td>
<td>36.7</td>
<td>2.47</td>
</tr>
<tr>
<td>Swiss BWR airlock into containment</td>
<td>474</td>
<td>139</td>
<td>10.8</td>
<td>80.7</td>
<td>2.01</td>
</tr>
</tbody>
</table>

The Radiography Room ratio of 1.695 is most closely matched by spectrum number 339 (the Caorso BWR inside the reactor cavity). For the Working Platform ratio of 2.278 the closest match would be spectrum number 80 (PWR Location 81 from IAEA318). The Top Gantry position has a ratio of 1.503, which is closest to spectrum number 389 (Czech PWR reactor hall).

7.2.5 N-Probe Spectra

The N-Probe spectra collected inside the NEPTUNE reactor hall at each of the measurement positions are shown below. With the data corrections for the actual irradiation time as a fraction of the total measurement time the unfolding proceeded without any problems. Figure 7.4 shows the N-Probe spectra for the Radiography
Room position. A repeat measurement was possible for this position and the two spectra are in reasonable agreement, indicating that the unfolding result is stable.

Figure 7.5 is the N-Probe spectrum for the Working Platform position. The neutron fluence is higher in this position, but the shape of the spectrum here does not differ significantly from the Radiography Room.

Finally, Figure 7.6 shows the N-Probe spectrum for the Top Gantry position. Here the spectrum shows an increased number of intermediate energy (in the range 10 keV to 500 keV) neutrons compared to the other two positions. For all three measurement positions, it can be said that the spectrum is quite soft, that is the majority of the fluence is of slow neutrons. None of the spectra display a prominent fission peak.
Figure 7.5 N-Probe spectrum for the NEPTUNE working platform position.

Figure 7.6 N-Probe spectrum for the NEPTUNE top gantry position.
7.2.6 Characterisation of NEPTUNE Conclusions

This work represents the beginning of a complete characterisation of the NEPTUNE test reactor hall. The BAe Systems Duosphere technique of assessing the spectral hardness using the ratio of readings from a 5” and 8” sphere appears to be quite successful. It would be desirable in future to obtain measurement data for a complete set of Bonner sphere diameters in the positions of interest within the reactor hall.

Despite the difficulty of having no control over data acquisition during the measurement the N-Probe behaved quite well and produced believable spectra for all three measurement positions. These confirmed the fluence spectrum to be fairly soft throughout the hall.

The Radiography Room would appear to offer the best position to site a fully characterised RNCF as it has a softer spectrum than the Working Platform and Top Gantry. It is conveniently situated on the ground floor, and has lead lined walls that will reduce the unwanted gamma dose.
Chapter 8

Conclusions

The importance of neutron spectrometry in the provision of accurate neutron dosimetry in the workplace has been emphasised in this work. Neutron spectrometry has not been practised routinely before due to the expense of neutron spectrometers, and the long measurement times and complex data analysis (unfolding) required. This also necessitates the use of specialist staff.

In the UK Naval Nuclear Propulsion Programme, neutron spectrometry has been carried out using the TNS. The TNS measurement results presented here show that performance is adequate. Deficiencies have been identified with the TNS detector response functions and spectrum unfolding software. However, the major obstacles to the wider use of the TNS are the overall size and weight of the system, and the extremely long data acquisition time necessary to achieve any detail in the intermediate neutron energy range from 50 keV to 1 MeV caused by the poor efficiency of the hydrogen-filled proportional counters.

It has been demonstrated that LGB has the properties to act as a high-sensitivity spectrometer for fast neutrons in high gamma-ray backgrounds. LGB also has several properties that would overcome the obstacles to routine spectrometry. The digital electronics fit in a very compact box, together with the HV power supply and can run off batteries, making the system man portable. The use of capture-gating, that is the combination of pulse shape discrimination on the proton recoil signals with the requirement for a subsequent neutron capture pulse, the rejection of gamma-ray
induced events should be very high. The LGB capability to measure the thermal and epithermal neutron fluence simultaneously to the fast means it could replace the NE213 and the two BF3 tubes in a future TNS replacement. The gamma ray counting channel could function as a gamma-ray dosimeter.

However, at only 2” diameter and 1.5” thickness, the LGB detector is not large enough to thermalise many neutrons before they escape from the detector volume, which leads to quite poor detection efficiency, and strong energy dependency in the measured efficiency as shown in the NPL irradiation results.

The near complete thermalisation of the incident fast neutron is also required in a capture-gated spectrometer if the measured recoil proton distribution is to resemble the original neutron spectrum. If neutrons are being captured with a considerable amount of their incident energy remaining, the result will be a significant degradation of the spectrometer resolution. This was the case with the monoenergetic irradiations of LGB.

The response of LGB is more complicated than for other loaded scintillators because it is doped with an inorganic material in particle form containing more than one isotope having a large neutron capture cross-section. For a lithium doped liquid scintillator it is easily assumed that the dopant is uniformly dispersed throughout the detector volume and, more importantly, all the scintillation light is produced in the organic liquid. In LGB, the initial fast neutron signal is scintillation light caused by recoil protons in the plastic scintillator. The subsequent delayed capture event occurs in a particle of the lithium gadolinium borate inorganic scintillator. It is assumed the range of the capture reaction products is small compared to the size of the crystal particle. The LGB spectrometer is therefore a composite of two separate scintillators, each having it own light output function. It will sometimes be the case that a recoil
proton passes through one of the inorganic particles and hence produces a different light signal to a neutron of the same energy that only travelled through the plastic. Similarly, sometimes the product of a capture reaction near to the surface of a crystal particle may travel into the plastic. The inevitable effect of this is a worsening of the resolution of both fast and capture signals.

The total amount of light from recoil protons produced by the slowing down of a fast neutron does not determine the original energy of the neutron unambiguously due to the nonlinearity of the light output function for organic scintillators. A neutron with energy of a few MeV loses 90% of its energy during the first 10 ns in the scintillator, which is of the same order as the characteristic time of the scintillation light. Therefore, it is impossible for the PMT to distinguish between light contributions from individual recoil protons.

Further developments of the digital pulse capture electronics would be necessary to progress this work any further. The lack of control over the pulse digitiser parameters, for example the threshold for neutron/gamma discrimination or the length of the delayed capture pulse window, made interpretation of certain results difficult. The probable interference to fast neutron counting from high energy gamma-rays from the lithium fluoride target at NPL could not be verified or eliminated because of this lack of control.

Even with greater control over data acquisition it is hard to see how the ‘thermal’ neutron counting channel (internal gamma ray signal from captures in gadolinium) could be made to work reliably. As demonstrated at the LFR, Petten this channel is very sensitive to high energy external gamma rays, such as from nitrogen-16. There will always be high energy gamma rays like this present in the environment around nuclear reactors, and there does not seem to be a way of discriminating against them.
If the use of unfolding methods to obtain the neutron energy spectrum is to be avoided, it is suggested that the size of the LGB detector be increased considerably. Monte Carlo studies could be used to determine the optimum size and composition of the spectrometer.

Continuing development with the present size spectrometer would make unfolding methods necessary. It would become important to have the capability to calculate detailed response functions and detection efficiencies for the LGB in capture-gated mode to provide the input data for spectrum unfolding. This could be achieved through the use of a bespoke Monte Carlo code that would track the charged particle reaction products in the two scintillators and calculate the quantity of light emitted.
References


[Danyluk, 2001] P Danyluk, DRaStaC, private communication, 2001


