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Measurement of fluence
at the D-line fast neutron facility
at iThemba LABS

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Abstract

Measurements of fluence were made for the neutron beams produced at the fast neutron beam facility of iThemba LABS, using an NE213 organic liquid scintillator detector and a $^{238}\text{U}$ fission ionisation chamber. Neutron beams were produced by irradiating a 6 mm natural lithium target with pulsed proton beams obtained from the $k = 200$ separated sector cyclotron. Three incident proton beam energies were used in this work, namely 65.99 MeV, 99.44 MeV and 203.33 MeV. From time-of-flight measurements with the NE213 scintillator, the spectral fluence of the neutron beams was obtained. Pulse shape discrimination was used to correct for gamma ray induced signals in the NE213 detector. Simultaneous measurements of the neutron beam flux were performed at 0° and 16°. The $^{238}\text{U}$ detector was used to obtain peak fluence measurements relative to the $^{238}\text{U}(n,f)$ cross section.
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My mother Ruth and brother Edwin, who have inspired me to reach further than I dared to dream was possible.

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Chapter 1

Introduction

The demand for well characterised quasi-monoenergetic neutron beams above 20 MeV that service a wide range of applications has grown in recent years [Me97] as illustrated by the following examples: the development of dosimeters for commercial airline crew [No06] who are now classified as occupationally exposed personnel [ICRP90]; spectrometry to determine the radiation damage on astronauts and equipment which suffer exposure to high energy neutrons during space flights [No06], and dosimetry for applications in shielding and radiation protection at high energy accelerators [Ta03].
Other uses for well characterised neutron beams include research in neutron therapy dosimetric methods, which require neutron energies up to 70 MeV [He09], the development of techniques for contraband detection [Bu04], testing the radiation hardness of electronic devices [Fr00] and calibrating detectors for use in nuclear physics experiments. Furthermore, developments of techniques for the transmutation of nuclear waste and the production of neutron beams for accelerator driven systems [Bl06] will require neutron beams that are characterised to within very small uncertainties [No07].

Numerous measurements of neutron fluence have been performed with neutron beams of energy less than 20 MeV [Ha79] and the techniques that are used for neutron spectrometry in this energy range are well established [Br02]. These techniques usually involve a combination of pulse height spectra unfolding, and where pulsed neutron sources are available, analysing time-of-flight spectra [Sc02]. However, comparatively fewer measurements of neutron spectra are available for energies above 20 MeV and the techniques for high energy neutron spectrometry are relatively less well established [Na02]. The reason for this is two-fold; first, truly monoenergetic neutron beams cannot be produced from bombarding light metal targets with protons. A continuum of lower energy neutrons will always contribute to the total neutron spectrum. The second reason is that the lack of detailed interaction cross section data for high energy neutrons with $^{12}$C, which is a major component of organic scintillator detectors, hampers the development of high energy neutron calibration fields.

The aim of this work was to measure the spectral fluence and the total peak fluence of the quasi-monoenergetic neutron beams that are produced at the fast neutron beam facility (D-line) at iThemba LABS. Nano-second pulsed neutron beams with energy between 25 MeV and 200 MeV are available at the neutron vault of the iThemba LABS D-Line. Time-of-flight measurements were carried out by increasing the time separation between proton bunches from 50 ns up to 500 ns using a beam pulse selector. Spectra were measured with a new neutron beam monitoring system that was recently installed at the facility. The detector system comprises an NE213 organic liquid scintillator, a $^{238}$U fission chamber and an NE102 plastic scintillator. The data yielded measurements of the spectral fluence as well as the total peak fluence of the neutron beams.
Chapter 2

Fast neutron detection

2.1 Introduction

Neutrons are uncharged, hence they are not affected by the Coulomb force. They do not produce direct ionization events and their cross section for interaction with matter is generally low [Kr88]. As a result, neutron detection is usually accomplished via the detection of secondary events produced by nuclear reactions, such as (n,p), (n,α), (n,γ) and (n,fission) or scattering against light nuclei which will become ionized and excited by energetic neutrons.
Neutrons of different energy are detected in many different ways. Common reaction mechanisms for both fast and slow neutrons are the (n,p) and the (n,α) reactions which lead to the direct detection of the charged products. The target elements of most neutron detectors are typically the isotopes of light nuclei that have a relatively large cross section for neutron capture.

2.2 Fast neutron scattering

The most important reaction for fast neutron (> 20 MeV) spectroscopy is elastic neutron scattering. In this process, an incident neutron transfers part of its kinetic energy to the scattering nucleus giving rise to a recoil nucleus. A common target for fast neutron scattering is the hydrogen nucleus [Kn00]. The cross section for neutron elastic scattering from hydrogen is relatively large and its energy dependence is known accurately across a wide energy range and to a high degree of accuracy [Ma68], as shown in figure 2.1. Fast neutrons scattering off hydrogen can transfer up to 100% of their kinetic energy in a single ‘head on’ collision.

![Figure 2.1 Elastic neutron scattering cross sections, in barns, for neutrons scattering on $^1$H, covering several orders of magnitude of the neutron energy range from about $10^{-3}$ eV to $10^7$ eV [Mu09].](image)
Typically, only a small fraction of incident kinetic energy can be transferred in collisions with heavy nuclei. The variation of the amount of energy that can be transferred from an incident neutron to a target is a function of the angle of scattering and can be described by kinematic equations. For a neutron of energy $E_o$, and of relative mass one, scattering elastically from a nucleus of relative mass $A$ (the mass number), and at a laboratory angle, $\varphi$, then the energy of the scattered neutron, $E(\varphi)$, can be shown to be [Co86],

$$E(\varphi) = E_o \frac{A^2 + 1 - 2\sin^2 \varphi + 2\cos \varphi \sqrt{(A^2 - \sin^2 \varphi)}}{(A + 1)^2}. \quad (2.1)$$

Maximum neutron energy loss occurs when the scattering angle is $180^\circ$, and is thus given by

$$\Delta E = E_o - E(180^\circ) \quad (2.2)$$

$$= E_o \frac{4A}{(A + 1)^2}. \quad (2.3)$$

Protons can be assumed to have the relative mass one, thus $A = 1$. Therefore, the maximum scatter angle is $90^\circ$ and hence for a proton target, equation (2.1) becomes

$$E(\varphi) = E_o \cos^2 \varphi, \quad (2.4)$$

which gives $E(\varphi) = 0$ for all $90^\circ \leq \varphi \leq 180^\circ$ and can be simplified to

$$E(\varphi) = E_o \cos^2 \varphi, \quad (2.5)$$

provided that $\varphi$ is restricted to the range $0^\circ - 90^\circ$. In the case of fast neutron detection, a proton is scattered with an angle $\theta$ to the initial neutron direction and the variation of the proton energy as a function of this angle is given by,

$$E_p(\theta) = E_o \cos^2 \varphi. \quad (2.6)$$

The recoil protons that result from fast neutron scattering are relatively easy to detect and this makes the (n,p) reaction ideal for fast neutron spectroscopy. Detection of the recoil protons can provide information on the energy of the incident neutrons. This is why organic scintillators, which are proton-rich hydrocarbons, serve as the basis for a wide variety of applications in fast neutron detection.
2.3 Scintillators

A scintillator is a type of material that absorbs radiation incident on it, which could be photons, neutrons, protons or other particles. It then converts the energy deposited in the detector material by ionization into a fast pulse of light (scintillation) through subsequent de-excitation mechanisms. This light pulse then enters a photomultiplier, via a light guide, where it is collected by a photocathode. The light falling on the photocathode produces electron pulses via the photoelectric effect. These electron pulses are amplified in the photomultiplier by a multi-stage sequence of focusing dynodes. Secondary emission and electron multiplication occurs at each dynode as the electron pulse is accelerated towards the anode (see figure 2.2). The electronic pulses can then be further amplified and processed by suitable electronics in order to give information about the incident radiation.

Different types of scintillation detectors are used in neutron spectroscopy [Br07] and the choice of detector usually depends on the type of neutron detection reaction that is employed. Factors such as the incident neutron beam energy, the time structure of the beam and the purpose of the investigation also determine the type of detector to be used. A thorough review of the different types of scintillator detectors, their properties and their applications in fast neutron spectroscopy is covered in works by Birks [Bi64], Harvey and Hill [Ha79] and Klein and Brooks [Kl06].

Since neutrons are generally detected indirectly after interacting with other charged particles, neutron detectors need to be fairly dense and opaque to neutrons. A good scintillator should therefore have a relatively high efficiency for converting the energy of recoil particles to fluorescent radiation. Scintillators should be transparent to their own radiation and the scintillation light spectrum should be sufficiently well matched to the photomultiplier response [Kl06].
Figure 2.2 Schematic diagram of a scintillator. Radiation incident on the detector interacts with the detector material leading to the production of photons by the scintillator. The photons strike a photo cathode producing electrons which are accelerated to the first dynode. Electron multiplication occurs in the photomultiplier along the dynode chain until the electron shower reaches the anode.
2.4 Organic scintillators

The most commonly used types of scintillation detectors in neutron spectroscopy are the organic scintillators [Br79]. Organic scintillators consist of aromatic hydrocarbon compounds that emit a flash of light or scintillation when exposed to radiation. In the case of neutron detection, the neutron is detected indirectly through the production of secondary charged particles, protons, alphas, gammas and other ions. These are released when the neutrons interact with the hydrogen and carbon atoms of the detector material. Therefore the detector provides the target nuclei for neutron interaction as well as the source of the scintillation.

The charged recoil particles transfer their energy to the detector material by exciting the valence electrons of the organic scintillator. The scintillator electrons are delocalized and occupy a π-molecular orbital structure within the organic molecules [Bi64]. These electron orbitals have an energy level structure of spin singlet and spin triplet states and each state has a fine structure of excited vibrational states. A diagram of these energy levels is shown in figure 2.3.

Ionization energy deposited in the detector leads to excitation of the higher singlet electron states, $S_1$, $S_2$ and $S_3$, which promptly decay without emitting any radiation, by a process of internal conversion to the $S_{10}$ state. This singlet excitation then radiatively decays to one of the vibrational states of the ground state, $S_{00}$ to $S_{03}$. This is the source of the prompt component of the scintillation light output described later in this chapter by equation (2.9). The transitions of the triplet excited states initially also occur through a process of internal conversion to the lowest triplet state $T_1$. The lifetime of the $T_1$ state is longer than that of the singlet state. It decays through a transition system with the $S_1$ state, known as the intercrossing system shown in figure 2.3, to the $S_0$ state. Some molecules that are in the $T_1$ state are thermally excited back to the $S_1$ state and proceed to decay through normal fluorescence. This transition constitutes the delayed or slow component of the scintillation light output which is referred to in the discussion on pulse shape discrimination in section 2.6.
Figure 2.3 The energy levels of an organic scintillator molecule’s $\pi$-electron orbital structure. The absorption arrows point up and the fluorescence arrows point down. The dashed arrow pointing towards $T_1$ shows the inter-system crossing. Adapted from [Kn00].
2.4.1 Liquid scintillators

Organic liquid scintillators typically comprise about 95% of a solvent such as toluene, xylene or dioxin, about 5% of a primary activator such as 2,5 diphenyl-oxazole (PPO) and about 0.5% of a wavelength shifter, such as 1,4-di(2-(5-phenyl-oxazolyl)) commonly known as POPOP. The ionizing particle interacts with the solvent and transfers its energy to this bulk material. The excitation energy of the solvent is transferred to the activator solute which produces almost all the scintillation. The wavelength shifter then absorbs the light produced by the primary scintillant and reradiates it at a longer wavelength to which the scintillator does not respond [Co86].

The most common liquid organic scintillator is the NE213 (equivalent brands are BC501A from BICRON and the EJ301 from Eljen Technology). The primary solvent is xylene, \( \text{(C}_6\text{H}_4(\text{CH}_3)_2) \) in which a small amount of naphthalene \( (\text{C}_{12}\text{H}_8) \) is dissolved. It also contains trace amounts of the wavelength shifter, POPOP [Nu]. Naphthalene reduces quenching effects in the detector, enhances the slow component of the light output and also improves the pulse shape discrimination capability of the detector [Bu69].

When used in fast neutron detection, liquid scintillators are used for counting neutrons as well as for spectrometry. This is achieved either by the use of the time-of-flight method, if the neutron beam is produced via a pulsed neutron source, or by unfolding the detector pulse height spectra to compare with calculated or simulated spectra [Sc02].

Spectrometry measurements are often performed in mixed neutron and gamma fields. Liquid organic scintillators such as the NE213 are well suited to these conditions as they can distinguish neutron induced events from gamma induced events in the detector. Separation of neutron pulses from gamma pulses is achieved via pulse shape discrimination [Kl02]. Liquid organic scintillators also have other advantages, such as high sensitivity to incident particle energy, a fast time response of approximately 2 ns and relatively short recovery times which lead to good time resolution. This makes them the preferred material for fast neutron spectrometry [Kl06]. The properties of some common organic scintillators are shown in table 2.1.
Table 2.1 Properties of plastic and liquid scintillators used for neutron detection [Cr91].

<table>
<thead>
<tr>
<th>Type</th>
<th>Name</th>
<th>Light Output (% of anthracene)*</th>
<th>Decay const(ns)</th>
<th>Wavelength of Max emission (nm)</th>
<th>H/C Atomic Ratio</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plastic</td>
<td>NE102A</td>
<td>65</td>
<td>2.4</td>
<td>423</td>
<td>1.104</td>
<td>General use</td>
</tr>
<tr>
<td></td>
<td>NE104</td>
<td>68</td>
<td>1.9</td>
<td>406</td>
<td>1.100</td>
<td>Fast timing</td>
</tr>
<tr>
<td></td>
<td>NE111A</td>
<td>55</td>
<td>1.6</td>
<td>370</td>
<td>1.103</td>
<td>Ultrafast timing</td>
</tr>
<tr>
<td>Liquid</td>
<td>NE211</td>
<td>78</td>
<td>2.6</td>
<td>425</td>
<td>1.248</td>
<td>General use</td>
</tr>
<tr>
<td></td>
<td>NE213</td>
<td>78</td>
<td>3.7</td>
<td>425</td>
<td>1.213</td>
<td>F. neutron (PSD)</td>
</tr>
<tr>
<td></td>
<td>BC501A</td>
<td>78</td>
<td>3.7</td>
<td>425</td>
<td>1.213</td>
<td>F. neutron (PSD)</td>
</tr>
</tbody>
</table>

*Anthracene, an organic crystal, is one of the oldest organic materials and has the highest scintillation efficiency (light output per unit energy) of any organic scintillator. The light output of other organic scintillators is usually expressed as a percentage of the light output of anthracene.

However, liquid scintillators are susceptible to solvent impurities, especially dissolved oxygen. Oxygen contamination leads to quenching of the triplet excitations that govern the slow component of the scintillator light decay. Significant distortions may occur in the detector pulse height response and loss of pulse shape discrimination capabilities may directly result if the scintillator is contaminated. It is imperative that the scintillator housing be airtight. Bubbling pure nitrogen through the scintillator removes oxygen from the scintillator cell [Kl06].

2.4.2 Plastic scintillators

Plastic scintillators are solutions of organic scintillators in a solid plastic solvent. They are also widely used in neutron spectroscopy, with the most common plastics being polyphenylbenzene, polystyrene and polyvinyltoluene. The common primary solutes are PBD (2-phenyl,5-(4-biphenylyl)-1,3,4-oxadiazole), p-Terphenyl and PBO [Le94]. A small proportion of POPOP is often added as a wavelength shifter.
A distinguishing feature of plastic scintillators is that they produce very fast signals, approximately 2 ns, with a very short decay time of 2 ns to 3 ns and high scintillation light output. Plastic scintillators such as the NE102A and the NE111A are therefore preferentially used as counters in fast neutron spectroscopy. They are also easily shaped and moulded, hence can be cut to any desired shape or size. Since they are relatively inexpensive, they are often chosen when large volume detectors are required [Kn00].

### 2.5 Scintillator response functions

The response of an organic scintillator refers to the pulse height spectrum output or light output of that scintillator for a given particle and energy. The lineshape of a detector is the response function measured for monoenergetic radiation of a single type and the response matrix refers to a set of lineshapes measured for a range of incident energies [Bu98]. The response function of organic scintillators to ionizing particles of different mass, charge and energy is generally non-linear and this is well understood across a fairly wide energy range [Le94]. A semi-empirical model was suggested by Birks [Bi64] to describe the non-linear response of organic scintillators to charged particles. The model explained this non-linearity as a result of quenching effects in the detector medium and it is governed by the specific energy loss (energy deposited per unit length) in the path of the ionizing particle, \( dE/dx \). The scintillation light output per unit path length, \( dL/dx \), is related to the specific energy loss of a given type of particle, \( dE/dx \), by:

\[
\frac{dL}{dx} = \frac{A dE}{dx} \left(1 + kB \frac{dE}{dx}\right),
\]

where \( A \) is the absolute detector efficiency and \( kB \) is the parameter relating the density of ionization centres to \( dE/dx \). Here, \( k \) is the fraction of ionization centres in the path of the particle that will lead to quenching and \( B \) is a proportionality constant referred to as the Birk’s parameter. The product \( kB \) is obtained by fitting the Birk’s formula to experimental data for a specific scintillator. The scintillator response depends on the energy, mass and charge of the ionizing particles. It is governed by the Bethe-Bloch relation and becomes more non-linear for more heavily ionizing particles as shown in figure 2.4.
Chou [Ch52] suggested a semi-empirical analysis which modified Birk’s formula for the relationship between the specific light output and the energy loss in a small element of a charged particle track by introducing an additional free parameter to Birk’s relation. The expression suggested by Chou was,

\[
\frac{dL}{dx} = \frac{A\frac{dE}{dx}}{1 + kB\frac{dE}{dx} + C\left(\frac{dE}{dx}\right)^2},
\]

(2.8)

where \(C\) is a constant. The Chou equation was considered a generalisation of Birk’s theory and it approaches Birk’s formula (equation 2.7) for small values of \(dE/dx\).

**Figure 2.4** The light response (relative light output) of a BC400 (NE102 equivalent) plastic scintillator to ionization due to different charged particles [Sa01].
The pulse height (PH) spectrum of an organic scintillator to incident radiation will depend on the size and geometry of the particular detector and the response characteristics of the photomultiplier. The energy and type of the incident radiation and the nature of the interactions induced by that particular radiation in the detector, as well as the cross sections of these interactions also affect the detector pulse height response [Bu98].

The response of an organic scintillator to incident neutrons is due to two main reactions; recoil protons resulting from n-p scattering and charged particles resulting from $^{12}\text{C}(n,x)$ reactions. Up to 20 MeV in neutron energy, n-p scattering events are the dominant light producing reactions in organic liquid scintillators and the spectral fluence of neutron beams can be obtained by unfolding the pulse height spectra. This is a relatively straightforward process as the n-p cross sections are well known and can be used to calculate the efficiency of the detector and hence the calculation of the neutron fluence can then be performed. The detector pulse height spectra can be compared with simulations of the response functions of the detector made using Monte Carlo calculations such as SCINFUL [Di88], NRESP [Di82] and CECIL [Ce79] codes.

Figure 2.5 shows the measured pulse height spectra from an NE213 organic liquid scintillator fitted with the calculated response functions. Up to about 20 MeV, the n-$^{12}\text{C}$ reaction cross sections can be accurately described by simulated response functions and good agreement is usually obtained between experimental and calculated responses [KI02]. With increasing incident neutron energy, the reactions of neutrons with the carbon component of organic scintillator detectors increasingly become the dominant light producing reaction. From energies exceeding 8 MeV, incoming neutrons can release up to three alpha particles from the breakup of $^{12}\text{C}$ [Kn00] via reactions such as $^{12}\text{C}(n,n')3\alpha$ and $^{12}\text{C}(n,\alpha)\text{Be}$. At higher energies, one or more combinations of protons, deuterons, tritons, $^3\text{He}$ ions, alphas, Li, Be, and B ions can appear in the reaction products. When neutron energies are above 90 MeV, the $^{12}\text{C}(n,x)$ reactions that occur in an organic scintillator can proceed via at least 65 reaction channels [Ke53] all of which include at least one charged ion in the product [Bu90]. At higher incident neutron energies, the contributions of n-p reactions to pulse height spectra are eclipsed by n-$^{12}\text{C}$ reactions.
Figure 2.5 Pulse height spectra, $L$, measured for (a) 2.52 MeV, (b) 7.93 MeV, (c) 15.0 MeV and (d) 18.98 MeV neutrons (histograms) and compared with response functions calculated with the NRESP-code (lines) [Kl02].

This makes simulating PH spectra more difficult for high energy neutrons as the cross sections of $^{12}$C(n,x) that are required to calculate the response matrix of the detector to n-C interactions are either not accurate enough [Bu02] or not available [Ti09], [Br04]. The upper pulse height regions represent the response of the detector to the maximum energy neutrons and hence are attributed to n-p scattering. These regions can be isolated and the light yield due to protons can be extracted and used for calculating the detector efficiency and the neutron fluence. However, this can only be done for neutron energies up to 70 MeV as the useful PH region diminishes with increasing neutron energy [No02b]. Figure 2.6 shows the PH spectrum of 46 MeV neutrons measured with an NE213 detector and fitted with a response calculated using SCINFUL. The PH region to the left of the dashed line includes contributions from n-C interactions and it shows the disagreement between the measured and calculated response functions.
Figure 2.6 The response of an NE213 scintillator to 46 MeV neutrons. The upper pulse height region \( h > 130 \), is associated with the detection of \( n-p \) scattering events only. It shows good agreement between the measured response functions (thick line) and the calculated response function (thin line). The region \( L < 130 \), includes the contribution of \( n-C \) interactions to the detector response [Br02].

A significant disadvantage of organic scintillators is that when proton recoils are produced by high energy neutrons, large volumes of the detector are required to limit the escape of recoil protons due to their long range in the scintillator. 100 MeV protons, for example, have a range of about 9 cm in a liquid scintillator [Ki06]. The phenomenon of escaping recoil protons is referred to as the wall-effect or edge-effect. It leads to a loss of neutron-gamma discrimination capability for the highest energy proton recoils [Na01]. This happens because the pulse shapes of the escaping particles become closer in appearance to the pulse shapes of Compton scattered electrons. The use of large detector dimensions to limit particle escapes, however, increases the probability of multiple neutron scattering events occurring within the detector. Hence, a compromise has to be found between the need for maximum detector size and minimising the chance of multiple scattering by a single neutron in the detector.
Several methods have been devised to mitigate the disadvantages of insufficient knowledge of the response matrix as well as finite detector size on scintillator response functions to high energy neutrons. Wall-effects have been successfully minimised by the use of higher density and inorganic scintillators such as CsI(Tl). Gunzert-Max et al. [Gu05] employed a BaF$_2$ scintillator to detect 198 MeV neutrons and it was found to have twice the detection efficiency of a similar sized NE213 detector. Buffler et al. used multiple slabs of NE213 scintillators, stacked back to back, to measure neutron energy spectra up to 150 MeV [Bu02]. This system was designed such that escape events could be vetoed and only events which stopped in one or both scintillators and deposited all their energy in the system were analyzed.

A compact liquid organic neutron spectrometer (CLONS) was designed by Brooks et al. [Br07], to measure neutron energy spectra up to 200 MeV. It used both the pulse height ($L$) and pulse shape ($S$) measurements from an NE213 detector to derive an $LS$-signature of the different charged particles in the detector. The $L$ and $S$ data from events due to particles stopping in and escaping from the detector was then unfolded as an alternative to pulse height spectrum unfolding. Figure 2.7 shows the pulse height ($L$) and pulse shape ($S$) spectrum of quasi-monoenergetic neutrons of energy 180-200 MeV. The regions corresponding to escaping ions ($e$) and non-escaping protons ($p$), deuterons ($d$) and alpha particles are indicated. $B1$–$B3$ are the boundary lines used in the derivation of the $LS$-response functions.

Nakao et al. [Na02] performed measurements of peak fluence on a quasi-monoenergetic neutron field at the RIKEN ring cyclotron facility in Japan with neutron energies from 66 MeV up to 206 MeV. Time-of-flight selected response functions were measured with a 12.7 cm diameter by 12.7 cm long NE213 scintillator. These were compared to responses that were calculated using the Monte Carlo code, CECIL, both including and excluding proton events due to wall-effects. The escaping protons were identified and isolated by their pulse shape, which is close to that of Compton scattered electrons. Reasonably close agreement was found between measured responses (with and without escaping protons) and calculated responses (also with and without escaping protons) for 74 MeV to 78 MeV neutrons, and for 205.8 MeV to 207.8 MeV neutrons, see figure 2.8.
Figure 2.7 The pulse height \( L \), and pulse shape \( S \), versus counts spectrum for quasi-monoenergetic neutrons of energy 180–200 MeV incident on a 5 cm x 5 cm NE213 organic liquid scintillator. The separation in the spectrum due to different ionizing particles is clear [Br07].
Figure 2.8 The measured response functions of a 12.7 cm long NE213 scintillator to neutrons are compared with and without escaping proton events, to the Monte Carlo calculated responses at $E_n = 74$-78 MeV (top) and at $E_n = 205.8$-207.8 MeV (bottom) [Na02].
2.6 Pulse shape discrimination

The scintillation light output, or yield curve, produced by organic scintillators in response to the passage of radiation is the sum of two exponential decays. These are, a prompt fluorescence which represents the “fast” component of the scintillation decay usually less than 5 ns long, and a “slow” longer-lived component, with a typical decay time of several hundred nanoseconds. This two-component exponential can thus be written as [Le94],

\[ N = A \exp\left(-\frac{t}{t_f}\right) + B \exp\left(-\frac{t}{t_s}\right), \]  

where \( N \) is the number of photons emitted (light output) at time \( t \), \( A \) and \( B \) are the relative magnitudes of the fast and slow light output respectively while \( t_f \) and \( t_s \) are the fast and slow time decay constants. The two decay curves described by this equation are shown in figure 2.9.

![Figure 2.9](image-url)  

**Figure 2.9** The scintillation light output of an organic detector resolved into its slow and fast components as a function of time. The solid line represents the total light decay curve. The time axis and light output axis are both in arbitrary units [Le94].
The majority of the light yield occurs in the prompt component of the scintillator light output and is affected by the ionization density along the path of the charged particle that excites the emission of light. However, the fraction of light that appears in the slow component is highly characteristic of the ionizing particle and is more strongly stimulated by a high ionization density. This is a result of the excitation of different fluorescence mechanisms in the scintillator material by particles of different ionizing power. Recoil protons, for example, which are generated by fast neutrons in the detector will be associated with the slow component of light decay, whereas electrons, which are near the minimum ionization density in the MeV range, will be associated with the fast component of the decay. Thus, it is possible to distinguish between different types of ionizing particles that might deposit the same amount of energy in the detector.

This is the basis of pulse shape discrimination [Co86], [Kn00], [Le94] which is used to discriminate between gamma ray induced pulses and neutron induced pulses when carrying out spectrometry measurements in a mixed neutron-gamma field. To further illustrate the difference in excitation properties of different ionizing particles, figure 2.10 shows how the decay of the scintillation light output of an organic crystal detector varies with the type of particle that is depositing energy in the detector material.

Different methods [Ow62] are employed to achieve neutron gamma discrimination when using organic liquid scintillators in mixed radiation fields. A variety of electronic circuits have been designed to carry out this function with the two most common being the zero crossover method [Pe79] and the charge integration (or constant fraction discrimination) method [Ad78].

The charge integration method [Pl76] was developed at AERE Harwell and is available as a commercial unit, the LINK 5010. The LINK’s operation involves two integrating circuits which operate in parallel and are coupled directly to the anode of the detector. One circuit integrates the signal for 25 ns and holds the result while the other integrates for 500 ns. These timings are especially suitable for the NE213 liquid organic scintillator. Pulse shape discrimination is obtained by comparing the two integrator outputs. A decision is made electronically by the LINK as to whether the pulse was induced by a neutron or a gamma ray.
Figure 2.10 The time dependence (ns) of scintillation pulses in the organic crystal, stilbene, (equal intensity at time zero) when excited by alpha particles, fast neutrons and gamma rays [Bo61].

The zero crossover method requires that the photomultiplier signal be passed through a bipolar shaping circuit such as a double delay line. The zero crossing of this pulse depends on the rise time and shape of the initial pulse, but is independent of its amplitude. A measure of the pulse shape is given by the time duration between the leading edge of the initial pulse and the zero crossing of the bipolar shaped pulse [Cr91]. A coincidence resolving time of 100 ns to 150 ns is usually necessary to distinguish the fast neutron induced signals from the gamma ray induced signals (see figure 2.11) [Co86].
Figure 2.11 An illustration of the zero crossover method of pulse shape discrimination showing bipolar pulse shapes and the time spread of the zero crossover signal [Co86].

### 2.7 The fission chamber detector

The specification of a reference neutron field requires that the measurement of the peak neutron flux be made relative to a known standard neutron interaction cross section [No04]. For neutron energies between 20 MeV and 200 MeV, the parallel plate fission chamber [Ga90] can be used for this purpose if the neutron fission cross section of the target fissile material is a known standard in the energy range. The fission chamber consists of anode plates, which are usually 5 or 6 platinum backed plates onto which layers of fissionable material have been deposited.
The deposition is done either by vacuum evaporation or by a painting technique. These plates are arranged alternately with tantalum collector plate electrodes inside an ionization chamber. For fast neutron detection, the fission chamber is placed in the path of a neutron beam and it can be used to determine the fluence of the beam. Neutrons enter the chamber and are detected when they induce fissions with the fissionable material on the plates. The fission fragments ionize the fill gas inside the chamber and the ionization events are processed with suitable electronics. The most common choice of fill gas is P-10 gas (90% volume argon, 10% volume methane) at atmospheric pressure and typical ionization chamber voltages of 100 – 200 V are applied, see figure 2.12.

**Figure 2.12** The fission chamber is operated within the ionization chamber region of the gaseous ionization detectors [Mu09].
The thin layer of fissile material, which is typically uranium, is in direct contact with the P-10 gas and when a fission event occurs, the two fission fragments travel in nearly opposite directions. The two fragments share about 160 MeV of energy but have a short range, in the case of uranium, the range is typically about 7 μm which is equivalent to a deposit of 13 mg/cm². Thus, fission fragments that are produced at a depth of more than 7 μm in the detector plate may not reach the gas and cause ionization. However, according to Crane and Baker [Cr91], most fission fragments exit at a grazing angle so their path length is longer than the minimum needed to escape. Figure 2.13 shows the construction of a basic parallel plate fission chamber.

The requirement to keep the coating thin in order to allow the fission fragments to enter the gas means that a typical fission chamber will contain a very low quantity of fissile material. It will thus have very low detection efficiency. For thermal neutrons, the intrinsic efficiency of a ²³⁵U fission chamber is typically 0.5% to 1% and for fast neutrons it is even lower [Cr91].

\[
\text{Intrinsic efficiency} = \frac{\text{number of events registered}}{\text{the number of events impinging on the detector}}
\]  

(2.10)

This is not necessarily a disadvantage as this means the fission chamber can ideally be used as a neutron beam monitor. It can be placed directly in the path of a neutron beam and it will ‘count’ neutrons without greatly affecting the absolute fluence of that beam.

The uniformity of the deposit thickness is crucial to fission chamber design. Traditionally, fissile materials were deposited onto plates using the painting technique, but chambers of this type exhibited a large inhomogeneity in deposit thickness which posed serious problems with the pulse height data quality. It was discovered that more homogeneous deposits could be achieved through vacuum evaporation. The variation in the thickness of fissile layers on the fission chamber plates also results in a loss of fission events below the alpha decay threshold. This in turn distorts the calculation of the fission fragment detection efficiency of the chamber and the evaluation of the spectral fluence of the neutron beam.
Figure 2.13 Basic fission chamber construction with 5 platinum backed fission foils and 6 tantalum collector plates [Ga90].

The common target nuclides used in the construction of fission chambers are $^{238}\text{U}$, $^{235}\text{U}$, $^{239}\text{Pu}$, $^{237}\text{Np}$, $^{209}\text{Bi}$ and nat$^{\text{Pb}}$. The choice of fission element is usually based upon the energy range of the experiments and the purpose of measurements. The $^{238}\text{U}$ fission chamber is ideal for fast neutron spectroscopy due to its sensitivity to fast neutrons and it has no cross section to slow and thermal neutrons. This is in contrast to other common fissile elements such as $^{235}\text{U}$, $^{239}\text{Pu}$ and $^{237}\text{Np}$ which are sensitive to thermal and low energy neutrons, especially below 20 MeV, as shown in figure 2.14. The basic chamber design that is used for measurements of neutron induced fission cross sections [No02a] is similar to the design used for measurements of neutron fluence and spectral fluence of quasi-monoenergetic neutron beams as well as the calibration of neutron dosimeters. $^{238}\text{U}$ fission chambers have been used together with NE213 detectors to measure experimentally the neutron fluence and spectral neutron fluence in numerous experiments [Sc99]. They are commonly used as fluence monitors and transmission detectors [No02b].
Figure 2.14 Fission cross sections (in barns) of some common target nuclides used in fission chambers as a function of neutron energy (in eV) [Kn00].

The fission reaction of neutrons on $^{238}\text{U}$ and $^{235}\text{U}$ has an extremely large $Q$-value (about 200 MeV). Therefore, detectors that are based on fission reactions can give output pulses that are orders of magnitudes higher than those induced from competing reactions or incident gamma rays. This gives the advantage that a very clean discrimination can be obtained between neutrons and gammas by setting a discriminator threshold in the measured pulse height data (see figure 2.15). The $^{238}\text{U}$ fission chamber is therefore highly insensitive to photons and low energy neutrons [Sc99].

The commonly used $^{238}\text{U}$ and $^{235}\text{U}$ fission chambers being naturally radioactive have an irreducible alpha induced pulse that gives an undesirable count rate independent of the neutron fission ionization. This alpha particle count rate dominates the lower pulse height regions of the uranium fission chamber pulse height spectrum.
Figure 2.15 The pulse height spectrum obtained from 5 MeV neutrons irradiating a $^{238}$U fission ionization chamber [Zh09]. The part of the spectrum that shows rapidly increasing counts below channel 400 is due to alpha particle induced ionization events in the detector.

Alpha particles have an average energy of approximately 5 MeV, so these alpha pulses can be discriminated against based on their pulse height. The setting of a pulse height threshold on the fission chamber spectrum to discriminate fission events from alpha particle events is easy to reproduce with high precision. Gayther [Ga90] suggested setting a pulse height threshold of $0.45P$, where $P$ is the pulse height value at the peak of the pulse height spectrum. This procedure is discussed in further detail in section 5.3.
2.8 Time-of-flight measurements

A neutron time-of-flight spectrum can be obtained from measurements of the neutrons beams that are produced via the irradiation of light targets such as $^7$Li, $^{10}$Be and $^{12}$C with pulsed proton beams. The corresponding spectral fluence distribution is obtained by converting the time-of-flight spectrum to an energy spectrum. The neutron energy spectrum is characterised by a quasi-monoenergetic high energy peak and a continuum of lower energy neutrons (see figure 2.16).

![Energy spectra of quasi-monoenergetic neutron beams generated from bombarding a 10 mm thick $^7$Li target with 70 MeV to 210 MeV protons at the RRC in Japan [Na02].](image)

*Figure 2.16 Energy spectra of quasi-monoenergetic neutron beams generated from bombarding a 10 mm thick $^7$Li target with 70 MeV to 210 MeV protons at the RRC in Japan [Na02].*
The high energy neutrons are more strongly forward peaked than the continuum neutrons which are relatively isotropic, thus making it necessary to perform fluence measurements at $0^\circ$ and wider emission angles. A subtraction method can then be employed to correct for the effect of the continuum fluence on the total neutron fluence in the measured response [No02b]. Quasi-monoenergetic neutrons can be obtained from the synthetic difference spectrum that is yielded by subtracting the wide angle spectrum from the $0^\circ$ spectrum [Si05]. Comparing the fluence of peak neutrons to the total neutron fluence including the low energy continuum will yield the relative spectral fluence of the beam.

**Figure 2.17** The relative spectral fluence ($\Phi_E/\Phi$) for the UCL beams (36.4 MeV, 48.5 MeV and 62.9 MeV protons) and the iThemba LABS beam (100 MeV protons on 5 mm thick Li). The NE213 measurements are shown by the thick lines and the UFC measurements are shown in the thin lines. The horizontal error bars indicate the widths of the high energy peaks as calculated from proton energy, kinematics and target thickness [No02b].
Nolte et al. [No02b] performed measurements of spectral fluence at the neutron beam facility of the Université Catholique de Louvain (UCL), Belgium, and at iThemba LABS, South Africa. The UCL measurements were performed on neutron beams produced from 36.4 MeV, 48.5 MeV and 62.9 MeV protons impinging on a 3 mm thick Li target. Figure 2.17 shows the relative spectral fluence measured with an NE213 detector and a $^{238}$U fission chamber. A low energy cut-off of about 3 MeV was found in the spectral fluence measured with the scintillator and the fission chamber. Below this energy, the spectral fluence was measured with the PTB Bonner sphere system [Wi02] and the results justify a constant extrapolation of the relative spectral fluence measurements down to lower energies.

2.9 Other neutron fields

The neutron fields that are obtained at accelerator and cyclotron facilities are often characterised by an inherent time structure. The neutrons are typically produced via the bombardment of a light metal target using nanosecond pulsed proton beams of variable width and current, as well as energy. This makes it possible for spectrometry to be performed on fast neutrons using time-of-flight and pulse height unfolding methods to select the quasi-monoenergetic component of the neutron beams [No04]. The neutron fields that are produced at facilities such as those at IRSN (France), PTB (Germany) and RIKEN (Japan) are referred to as reference fields. High energy neutron fields, such as those at iThemba LABS (South Africa), and UCL (Belgium) are well characterised quasi-monoenergetic neutron beams with well specified pulse selection and can be measured over long flight paths. They are essential for characterising the response functions of different types of neutron detectors for different applications [Bu90], [He09] for neutron energies of up to 200 MeV. Specifying these neutron fields requires measurement of the peak fluence and spectral fluence of the neutron beam [No04]. Neutron time-of-flight spectra can be obtained from measurements made using organic liquid scintillators and uranium fission chambers as these instruments have relatively fast response times and good time resolution [Kl06].

Organic liquid scintillators and fission chambers are used in various applications in neutron metrology. Measurements of fission cross sections of $^{235}$U, $^{238}$U, $^{209}$Bi and natPb were performed by Ralf Nolte et al. [No07] using quasi-monoenergetic neutron beams of up to 200 MeV and this
relied heavily on well characterised reference neutron beams. The results of these measurements went on to become part of the internationally accepted standard neutron fission cross sections of the relevant elements [NIST08]. Measurements of neutron flux and spectral fluence were conducted together with Monte Carlo calculations using FLUKA, to design improvements to the shielding of the target area of the iThemba LABS neutron beam facility [Ad10]. Taniguchi et al. [Ta03] employed liquid scintillators to measure high energy neutron spectra between 6 MeV and 800 MeV behind an iron and concrete lateral shielding of the Final Focus Test Beam (FFTB) at the Stanford Linear Accelerator Centre (SLAC). The measurements were performed behind thick concrete shields that were placed outside the FFTB housing. Measurements of this type are becoming increasingly important as more high energy accelerator facilities are built worldwide and the shielding of such facilities has to be developed [Th85].

The dosimetry for airline crew who regularly fly at commercial flight altitudes requires calibrating, testing and comparing different detectors such as tissue equivalent proportional counters (TEPC) using quasi-monoenergetic reference neutron fields [Mo08]. Neutron detectors have been installed on board long distance passenger jets to measure the flight altitude neutron flux that is produced by the interaction of galactic cosmic radiation with the atmosphere [ICRU10]. The data has been used to calculate the response functions of these detectors and compile standards that can be applied across different dosemeter systems in the aviation industry [Wi10].

Tippawan et al. [Ti09] used plastic scintillators as part of an eight piece, three-element telescope detector arrangement in the measurements of double differential cross sections for light ion production in carbon induced by 96 MeV neutrons. The scintillators were used as an active anti-coincidence collimator to discard the signals from particles that did not stop in the desired detector.
Chapter 3

The experiments

3.1 The iThemba LABS fast neutron beam facility

The experiments that made up the present work were conducted at the neutron beam facility at iThemba LABS, a separated sector cyclotron (SSC) in Faure, Cape Town. The experiments were performed under the project PR156: “Tests of neutron detectors for beam monitoring at the D-line”. Data was obtained over three consecutive weekends in August 2009 at three different proton beam energies: 65.99 MeV, 99.44 MeV and 203.33 MeV. The experimental area at the separated sector cyclotron facility at iThemba LABS that is dedicated to neutron work is called the D-line vault.
3.2 Neutron production

Pulsed proton beams accelerated from the separated-sector cyclotron at iThemba LABS were directed to the D-line vault where they irradiated a metallic natural lithium target, ($^{nat}$Li is composed of 92.5% $^7$Li) 6 mm thick and of density 0.534 g/cm$^3$. The time separation between successive proton beam pulses at 200 MeV was enlarged from 33 ns to 235 ns, by letting through one in seven pulses using an electrostatic deflector system.

Pulsed neutron beams were produced via the $^7$Li(p,n)$^7$Be reaction which has a $Q$-value of -1.664 MeV. The target was placed in a target holder, which is a water cooled ladder assembly that can hold up to four targets inside an evacuated scattering chamber. These targets can be moved in or out of the beam remotely. The first position in the target holder was occupied by a quartz ruby which was used to precisely focus the proton beam onto the target position before the $^7$Li target was placed in the beam. A closed circuit television camera installed inside the target area of the vault showed the ruby glowing brightly when irradiated by the proton beam, hence the beam could be precisely focused. The second position was left empty and when selected, background measurements could be carried out at the same conditions as the neutron measurements. Adjustments of the proton beam width were made from the control room to ensure that the halo of the beam did not irradiate any parts of the target holder assembly. The third position was occupied by the 6 mm lithium target and the fourth position was occupied by a $^{12}$C target. The $^{12}$C target was 8 mm thick graphite of density 2.266 g/cm$^3$.

The target area was separated from the detector area by iron and concrete shielding 3 m thick. Blocks of wax, 25 cm thick covered the iron wall inside the detector area to thermalise stray neutrons and the wax was covered by sheets of borated polyethylene, 4 cm thick (see figure 3.1). Multi-angle fan collimators of cross section area 4.75 cm x 4.75 cm made neutron beams available at angles of 0°, 4°, 8°, 12° and 16°. During this measurement campaign, neutrons were detected at 0° and 16° to the incident proton beam.

Protons that did not interact with the target as well as other charged reaction products were extracted from the neutron beam by deflection magnets. The magnets are designed and positioned to bend protons of up to 200 MeV by 15° away from the incident proton beam direction to a Faraday cup buried inside the collimator shielding [Mc93]. A 10 cm thick block of
graphite was positioned at the collimator entrance to remove protons and charged particles that remained in the beam and stopped them from reaching the detectors.

Figure 3.1 An overview of the main neutron vault (D-line vault) of the iThemba LABS neutron beam facility. The target area, where the target holder and bending magnets are located, is on the right of the diagram. The multi-angle collimator fan is in the middle and the detector area is on the left of the diagram. The positions of the detectors in the vault are indicated. For clarity, only the NE213 is shown in the 16° beam line, without the monitors. The graphite blocks (shaded black in the diagram) are placed at the collimator entrance, where the neutron beams enter the collimator from the target area.
3.3 NE213 organic liquid scintillator

The NE213 organic liquid scintillator detector was 5.08 cm in diameter and 10.16 cm long (see figure 3.2 and figure 3.3) and the scintillator cell was optically coupled to an XP2020 model photomultiplier tube by means of a light guide. This photomultiplier was a 12 dynode stage system operated at a high cathode voltage of -1750 V with the anode at earth potential for all the experiments with neutron beams.

The detector was gain stabilized using a built-in LED and a PIN diode inside the PMT and near the scintillator, combined with a feedback system. The system adjusted the light output of the LED and kept the gain changes to a minimum. Gain drifts in the photomultiplier are normally brought about by variations in the detector count rate, changes in temperature, and changing magnetic fields around the detector [Mo10].

![Diagram of NE213 detector](image)

Figure 3.2 A schematic diagram of the NE213 detector showing the dimensions of the scintillator and the position of the gain stabilizing LED and the light guide (LG) that optically couples the active part of the scintillator to the photomultiplier tube (PMT).
Figure 3.3 The NE213 organic liquid scintillator that was used in the experiments.

3.3.1 NE213 electronics

A fast anode signal was obtained from the photomultiplier base of the scintillator and this signal was split between a constant fraction discriminator (CFD) unit along a timing circuit and a pulse shape discriminator (PSD) in a different circuit. From the leading edge of the anode input, the CFD produced a logic pulse which was sent to start a time-to-amplitude converter (TAC) in the timing branch of the circuit. This TAC was stopped by a suitably delayed pulse obtained from the radio frequency (RF) cycle of the cyclotron. It thus produced a pulse whose amplitude was proportional to the time interval between the start and stop signal on the TAC and also carried the neutron beam time-of-flight information. The pulse was sent through a delay amplifier to an analogue-to-digital converter (ADC) for data acquisition of the time-of-flight parameter, $T$. The pulse shape, $S$, pulse height, $L$ and time-of-flight, $T$, events were recorded by a FAST ComTec multiparameter data acquisition system (MPA-3) for further offline analysis (see figure 3.4).
Figure 3.4 Schematic arrangements of the electronic modules of the NE213 detector assembly.
Table 3.1 The abbreviations used for the electronic modules of the NE213 detector assembly.

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Anode</td>
</tr>
<tr>
<td>ANTICO</td>
<td>Anti-coincidence</td>
</tr>
<tr>
<td>CFD</td>
<td>Constant Fraction Discriminator</td>
</tr>
<tr>
<td>CO</td>
<td>Coincidence</td>
</tr>
<tr>
<td>D</td>
<td>Dynode</td>
</tr>
<tr>
<td>DLA</td>
<td>Delay Line Amplifier</td>
</tr>
<tr>
<td>DISC</td>
<td>Discriminator</td>
</tr>
<tr>
<td>HV</td>
<td>High Voltage</td>
</tr>
<tr>
<td>PSD</td>
<td>Pulse Shape Discriminator</td>
</tr>
<tr>
<td>TAC</td>
<td>Time to Amplitude Converter</td>
</tr>
<tr>
<td>RF</td>
<td>Radio frequency</td>
</tr>
<tr>
<td>LED</td>
<td>Light Emitting Diode</td>
</tr>
<tr>
<td>L</td>
<td>Pulse Height parameter</td>
</tr>
<tr>
<td>S</td>
<td>Pulse Shape parameter</td>
</tr>
<tr>
<td>T</td>
<td>Time-of-Flight parameter</td>
</tr>
</tbody>
</table>

3.3.2 Pulse shape discrimination

The zero crossover method was used to distinguish light pulses produced from scattering neutrons and those produced by Compton scattered gamma rays in the detector. The anode signal was directed through a Comtec 2160A Pulse Shape Discriminator module (PSD), where it was integrated and differentiated in a preamplifier to produce a bipolar shaped pulse. The zero crossing of this pulse depended on the rise time and shape of the initial anode pulse. The signal was then fed to a high gain limiting amplifier and the zero-crossing points corresponding to gammas and neutrons were thus greatly enhanced. The CFD output from the timing pulse was used to start a TAC in the pulse shaping circuit, which was stopped by a signal from the PSD whenever the bipolar pulse of the neutron signal crossed the zero crossover point. The TAC logic output produced a pulse shape parameter, $S$, which was sent to a delay amplifier and then to an ADC for data acquisition.
The linear output of the scintillator was extracted from the 9th dynode. It was directed through a preamplifier and an Ortec 460 delay line amplifier (DLA) which produced the pulse height parameter, $L$. This output was split, with one branch being delayed and sent to an ADC while the other branch of the DLA output was sent through a discriminator and a logic unit. The logic unit branch, in coincidence with the TAC output from the pulse shaping branch and the timing branch, triggered an event gate that prompted the data acquisition system to accept an event.

### 3.4 Uranium fission chamber

The Institute for Reference Materials and Measurement, “IRMM”, parallel plate fission chamber (PPFC) detector was used for the experiments to measure relative fluence. The outer casing of the fission chamber was made from pressed steel and the windows were made of 0.15 mm thick tantalum. Inside this casing, there were six platinum backed electrode plates coated on both sides with $\text{U}_3\text{O}_8$ and each of these electrode plates had a sensitive diameter of 76 mm (see figure 3.5). These plates were arranged in alternating layers, 5 mm apart, with tantalum electrodes (or collector plates). These collected the charge produced by fission fragments ionizing the fill gas in the chamber. The chamber contained a total of 240.931 mg of fissile uranium-238 which was deposited on the plates by electro-spraying [No07c]. This mass of $^{238}\text{U}$ was specified with an estimated uncertainty of about 10% [Mo10].

During measurements, the chamber was suspended from an aluminium frame and was attached to the corners of this frame by means of thin wires. The reason for this arrangement was that when used with an ‘open’ neutron flux configuration, such as that employed at the PTB and iThemba LABS, the effects of neutrons scattering off the apparatus that held the detector and back into the chamber was minimized (see figure 3.6).
**Figure 3.5** An internal view of the “H21” uranium fission chamber. It is similar in construction to the IRMM fission chamber and the H22 fission chamber, but has five platinum electrodes coated with $^{238}$U that are separated by tantalum electrodes.

**Figure 3.6** The IRMM uranium fission chamber is shown suspended from its steel frame. During operation, the tubes circulate P-10 gas from the tank (shown on the right) throughout the chamber.
The fission chamber was not perfectly evacuated and this made it necessary to flush out the air inside it with P10 gas, which is a mixture of 90\% argon and 10\% methane, for at least 30 minutes prior to use and continuously during operation. Air contains water and other electronegative atoms and molecules that interfere with charge transfer to the fission products in the detector and affect the detector’s pulse height output.

The fission chamber had a time resolution of 4 ns and the efficiency of fission fragment detection was rated at 96\%. Some properties of the chamber are shown in table 3.2, where $A$ and $B$ refer to the two sides of each of the six coated plates.

**Table 3.2** Properties of the uranium coating of the IRMM parallel plate fission chamber.

<table>
<thead>
<tr>
<th>Plate num.</th>
<th>Coating thickness $A$ ($\mu g/cm^2$)</th>
<th>Coating thickness $B$ ($\mu g/cm^2$)</th>
<th>Mass of $^{238}\text{U}$ ($mg$) $A$</th>
<th>Mass of $^{238}\text{U}$ ($mg$) $B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>412</td>
<td>405</td>
<td>18.690</td>
<td>18.373</td>
</tr>
<tr>
<td>2</td>
<td>407</td>
<td>408</td>
<td>18.463</td>
<td>18.509</td>
</tr>
<tr>
<td>3</td>
<td>453</td>
<td>423</td>
<td>20.550</td>
<td>19.189</td>
</tr>
<tr>
<td>4</td>
<td>450</td>
<td>534</td>
<td>20.414</td>
<td>24.225</td>
</tr>
<tr>
<td>5</td>
<td>468</td>
<td>431</td>
<td>21.231</td>
<td>19.552</td>
</tr>
<tr>
<td>6</td>
<td>427</td>
<td>493</td>
<td>19.371</td>
<td>22.365</td>
</tr>
<tr>
<td></td>
<td>Total mass of $^{238}\text{U}$ ($mg$)</td>
<td></td>
<td>240.931</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total no. of $^{238}\text{U}$ atoms</td>
<td></td>
<td>6.095E+20</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Total no. of $^{238}\text{U}$ atoms (barn)</td>
<td></td>
<td>1.344E-05</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Activity (Bq)</td>
<td></td>
<td>2996</td>
<td></td>
</tr>
</tbody>
</table>
3.4.1 Fission chamber electronics

The output signal that was obtained from the fission chamber pre-amplifier was a typical unipolar signal, characterised by a sharp rising edge and a long decay of approximately 50 µs. This signal was then split between an Ortec 474 differentiating timing filter amplifier (TFA) and an integrating Ortec 571 amplifier.

The integrating amplifier typically produced a Gaussian shaped pulse, of approximately 1 µs FWHM that approximated the full signal at the input but added far less noise to it. This amplified signal was then split between a delay amplifier and a timing single channel amplifier (TSCA). The delay amplifier simply amplified the signal and sent it directly to an ADC (see figure 3.7) from where it would be recorded as a pulse height signal in the data acquisition. The branch that passed through the TSCA was then passed through a logic unit. This signal, in coincidence with a gate-and-delay (GDG) signal, acted as a switch to trigger the data acquisition to accept an event.

In the other branch, the TFA produced a fast negative output pulse, with characteristic steep edges that are good for timing applications. This pulse was passed to a constant fraction discriminator. The CFD was triggered to produce an output signal characteristic of the rise time of the input and this output pulse was used to provide the start signal to a TAC.

The TAC was stopped by the cyclotron RF signal and it generated a logic timing output pulse. This TAC output pulse went straight to the ADC from where it was recorded as a time-of-flight pulse, while another branch of the TAC logic output went to the GDG where it was used to trigger the data acquisition by a coincidence in the logic unit. The setting of the trigger threshold was done very carefully. It was set just high enough to exclude most, but not all of the alpha events. This reduced the dead-time in the system that would be caused by the alpha events, but still made it possible to see where the neutron fission events began.
Figure 3.7 A schematic representation of the arrangement of the electronics for the $^{238}\text{U}$ fission chamber detector system.
Table 3.3 The abbreviations of the electronics system for the fission chamber detector.

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Amp</td>
<td>Amplifier</td>
</tr>
<tr>
<td>DA</td>
<td>Delay Amplifier</td>
</tr>
<tr>
<td>CFD</td>
<td>Constant Fraction Discriminator</td>
</tr>
<tr>
<td>CO</td>
<td>Coincidence</td>
</tr>
<tr>
<td>FC</td>
<td>Fission Chamber</td>
</tr>
<tr>
<td>GDG</td>
<td>Gate and Delay Generator</td>
</tr>
<tr>
<td>HV</td>
<td>High Voltage</td>
</tr>
<tr>
<td>PSD</td>
<td>Pulse Shape Discriminator</td>
</tr>
<tr>
<td>TAC</td>
<td>Time to Amplitude Converter</td>
</tr>
<tr>
<td>TFA</td>
<td>Timing Filter Amplifier</td>
</tr>
<tr>
<td>TSCA</td>
<td>Timing Single Channel Analyser</td>
</tr>
<tr>
<td>L</td>
<td>Pulse Height parameter</td>
</tr>
<tr>
<td>T</td>
<td>Time-of-Flight parameter</td>
</tr>
</tbody>
</table>

3.5 Beam monitors

In both the 0° and 16° measurements, constant beam monitoring was done using a disc-shaped, 2 mm thick NE102A plastic scintillator monitor and a smaller $^{238}\text{U}$ fission chamber monitor from the PTB, called the H22.

The arrangement of the monitors and detectors used in the vault is shown in figure 3.1. Upon exiting the collimator, the neutron beam passed through the gain-stabilised NE102A detector. This plastic scintillator which was also optically coupled to an XP2020 photomultiplier tube was operated in transmission mode as a fluence rate monitor. Both the fast anode signal and the slow dynode signal were monitored, however, the fast signal was more useful for making comparisons with the other monitors. A pulse height threshold was applied to the NE102 to make it insensitive to gamma rays.
The beam then passed through the \( ^{238}\text{U} \) fission chamber monitor which was placed right next to the NE102A detector. This H22 fission chamber monitor (see figure 3.5) was of the same design as the IRMM fission chamber detector that was used for the measurement of the neutron fluence and was described in section 3.3. The main difference was that it had only 5 coated electrodes in total hence a smaller mass of \( ^{238}\text{U} \) (45.34 mg in total) than the IRMM fission chamber. It was easy to set up and operate, and like the IRMM fission chamber almost completely insensitive to photons and low-energy neutrons. As a result of the smaller mass, it had a lower capacitance than the IRMM fission chamber and consequently, faster response and shorter recovery times than the larger IRMM fission chamber.

The setting of a pulse height threshold required to discriminate fission events from alpha particle events was easy to reproduce with high precision on both fission chambers. However, the low neutron sensitivity of the H22 fission chamber prevented monitoring at low beam currents and over short measuring periods. The insufficient counts made the relative errors in the counting statistically larger. In this range, the monitoring was mainly accomplished by the NE102A detector. However, at the higher energies and high currents required for the fluence measurements with the IRMM fission chamber, the H22 monitor provided more reliable readings than the plastic scintillator, hence a correlation had to be established between the two monitor readings at intermediate energies.

The beam current obtained from the Faraday cup was also used as a monitor, although care had to be taken especially with measurements at low beam currents. This was mainly due to the effects of leakage currents and problems with the beam focusing at low currents, hence the ratios of the readings on the different monitors had to be observed carefully across the entire energy range at which measurements were made.

3.6 Measurements

3.6.1 Beam profile measurements

The spatial profile of the iThemba LABS neutron beam was well characterised in a previous experiment [No06] at 100 MeV and 200 MeV proton beam energies (see figure 3.8). In that
procedure, the beam profile was measured using a 2 mm thick Polymethyl methacrylate (PMMA) plate and $^7$LiF thermo-luminescent diodes (TLDs). The TLDs were inserted in the PMMA holder in two vertical and two horizontal rows and mounted such that the central crossing point of the horizontal and vertical rows was the position of the nominal beam centre. After neutron fluence measurements were performed, with the plates centered on the detector axis and in the path of the neutron beam, the plates were analyzed and the shape of the beam at the detector face was obtained.

The image plate uses photo stimulated luminescence (PSL). It consists of bariumfluorobromide doped with europium (BaFBr:Eu2+) in a plastic matrix. The PSL process is similar to thermo luminescence (TSL). Radiation releases electrons from the Eu2+ ions into the conduction band. These electrons get trapped in metastable states and are released later by irradiation with visible laser light in the reader. The recombination of the electrons with the Eu3+ ions yields red light which is proportional to the absorbed dose. The image plate is sensitive to the neutron and photon component of the beam. So the procedure used to obtain the neutron beam profile would be to take a picture with and without a neutron converter (Lucite sheet) in front of the image plate and produce a difference image which would then be neutrons only. However, the photon fraction should be small. The result of the measurements showed that at a distance of 8 m from the target, the neutron beam width at both energies was wider than the active area of the NE213 detector that was presented to the beam [No07b].

It was thus deduced that the detectors and monitors were all uniformly covered by the beam and these results could be directly applied to our measurements as the collimator openings and flight paths were unchanged for our experiments. See Appendix I for a complete description of the spatial measurements of the beam profile.
Figure 3.8 Beam profiles for the neutron beam measured using TLDs (solid line) and the image plate (dashed line). The distance from the Li target was 8 m for both measurements at the energies (a) 100 MeV and (b) 200 MeV [No06].
3.6.2 Neutron beam measurements

The data in table 3.4 shows the proton beam energies and the resulting maximum neutron beam energies at which the experiments were conducted. During measurements at high proton beam energies with the NE213 scintillator, typical beam currents of 0.5 nA were employed. The average energy loss experienced by protons in the lithium target, $\Delta E$, and the maximum energy of the resulting neutron beam, $E_{0,\text{calc}}$, (average energy of the peak neutrons) were calculated using the thickness of the target and the $^7\text{Li}(p,n)^7\text{Be}$ reaction kinematics as well as the reaction $Q$-value.

A feature of the $^{238}\text{U}$ fission chamber was that it had a low sensitivity to passing neutrons, thus high proton beam currents, typically 300 nA, were employed when measurements were made with the fission chamber in order to produce high neutron fluxes.

Table 3.4 The parameters used in the experiments with the 6 mm thick Li target and both the NE213 detector and the uranium fission chamber detector in the beam.

<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>$\Delta E$ (MeV)</th>
<th>$E_{0,\text{calc}}$ (MeV) at 0°</th>
<th>Beam Freq (MHz)</th>
<th>Beam Period (ns)</th>
<th>NE213 Detector</th>
<th>Uranium Fission Chamber</th>
</tr>
</thead>
<tbody>
<tr>
<td>65.99</td>
<td>2.674</td>
<td>62.99</td>
<td>16.373</td>
<td>427.5</td>
<td>5.0</td>
<td>9.158</td>
</tr>
<tr>
<td>99.44</td>
<td>1.933</td>
<td>96.81</td>
<td>19.664</td>
<td>355.98</td>
<td>5.0</td>
<td>9.158</td>
</tr>
<tr>
<td>203.33</td>
<td>1.163</td>
<td>201.08</td>
<td>26.000</td>
<td>269.20</td>
<td>0.5</td>
<td>8.000</td>
</tr>
</tbody>
</table>

The overlap energy of slow neutrons arriving at the detector face, simultaneously with fast neutrons of the successive pulse, was found to be between 3 MeV to 5 MeV at an average distance of 8 m for the three beam energies. Thus a threshold for timeframe overlap was set up in the detector system at 4 MeV for the measurements.
Chapter 4

Results: Spectral neutron fluence

4.1 The neutron time-of-flight spectrum

The time-of-flight spectrum of the iThemba LABS neutron beams shows the energy distribution of neutrons obtained from the proton bombardment of a metallic $^7$Li target. The spectrum at 0° is characterised by a sharp peak, which consists of the highest energy neutrons and a broad Maxwellian-like continuum that extends down to lower energies (see figure 4.1). To the right of the main peak is a low energy gamma ($\gamma$) ray peak. These gamma rays arise from the de-excitation of the $^7$Li + p system. Photons of energy 0.47 MeV are produced from the first excited state of $^7$Li which is excited by $^7$Li(p,p$'$). A time independent gamma ray background is also
present in the neutron spectrum. These $\gamma$-rays are produced further down the proton beam line before the target and are mostly less than 4 MeV [No11]. The neutrons in the high energy peak are produced from transitions to the ground state and the first excited state of the target nucleus, $^7$Be ($E_x = 0.429$ MeV) which is stable against particle emission. The lower energy tail is made up partly of neutrons that arise from higher energy excitations and neutrons that are produced via many-body breakup reactions, as well as multiple scattering inside the target. Neutrons that are slowed down by interactions with the collimator, the detector housing [No04] and scattering within the detector also contribute to the low energy continuum.

![Time-of-flight spectrum of neutrons](image)

**Figure 4.1** The time-of-flight spectrum of neutrons produced from 65.99 MeV protons irradiating a 6 mm thick lithium target and detected at 0°. The time-of-flight channel scale was calibrated to a time (ns) scale and a non-linear energy scale (MeV) above the graph.
Additionally, when protons irradiate the more massive atoms of the target holder, they cause reactions which give rise to slower neutrons. These will also appear in the lower energy continuum of the time-of-flight spectrum.

The width of the main neutron peak in figure 4.1 is due in part to incident protons losing energy in the lithium target, as a result of the target thickness, before undergoing a (p,n) reaction. These energy losses are continuous over the entire range from zero up to a maximum determined by the incident proton energy and the thickness of the target [Au72]. The resulting spread in the energy of the incident protons reacting with $^7$Li atoms leads to a corresponding spread in the energy of the neutrons emitted from the target. Three body breakup reactions in $^7$Li as well as a small contribution from the unresolved first excited state of $^7$Be and higher order excitations, also contribute to the broadening of the high energy peak.

The time resolution of the entire detector system also contributes measurably to the width of the main neutron peak. It adds an uncertainty to the time-of-flight measurement of the order of the full width at half maximum of the gamma ray peak.

### 4.2 Time calibration

A calibration of the time-of-flight in figure 4.1 yielded the time scale, $T$ (ns), and a neutron energy scale, $E_n$ (MeV), shown above the spectrum. Both scales run in an opposite direction to the time-of-flight channel scale. The position of the gamma peak at 30.53 ns is crucial to calibrating the time scale and energy scale. Essentially, the gamma rays are produced from (p,n) reactions in the target. They are detected in the liquid scintillator at a time represented by the time-of-flight ADC channel, $T_\gamma$, determined by the velocity of light, $c$, and the distance, $d$, between the centre of the detector and the centre of the target. The exact time when gamma rays are emitted from the target corresponds to a ‘time zero’ position on the time-of-flight channel, $T_o$, which is indicated in figure 4.1. This channel corresponds to the position of zero time-of-flight and for the measurements performed with 65.9 MeV protons, $T_o$ was calculated to be at channel number = 1048 using the equation,

$$T_o = T_\gamma + (d / c) F,$$  (4.1)
where $T_\gamma = 972$ channel, $F = 2.51 \pm 0.01$ channels/ns is the time calibration factor obtained from the TAC (see figure 4.2), $d = 9.1586$ m is the neutron flight distance and $c = 0.30$ m/ns.

![Figure 4.2](image)

**Figure 4.2** The TAC was run in a self-stopped mode through a calibrated delay box. This produced sharp peaks over the full range of the TAC, separated by 20 ns, evenly spaced over the TAC range every 50 channels. This yielded a time calibration factor of 2.5 channels/ns.

The channel number of the high energy neutron peak, $T_n = 825$, was read off the spectrum. The average speed, $v$, of peak neutrons, from the time-of-flight, was calculated to be

$$v = \frac{d \cdot F}{(T_o - T_n)} = 0.104 \text{ m/ns.}$$  \hspace{1cm} (4.2)

This gives a value of $\beta = v/c = 0.347$ for neutrons produced by 65.99 MeV protons. Thus, the average energy of the peak neutrons incident on the NE213 detector, $E_n$, was calculated from the time-of-flight spectrum using the neutron rest mass, $m_n = 939.565$ MeV and the relativistic equation,
\[ E_n = m_n(\gamma - 1), \quad (4.3) \]

\[ = m_n \left( \frac{1}{\sqrt{1 - \beta^2}} - 1 \right) = 939.565 \left( \frac{1}{\sqrt{1 - 0.347^2}} - 1 \right), \]

\[ = 62.24 \text{ MeV}. \]

The average peak energies for the neutrons produced from 99.44 MeV and 203.33 MeV protons were calculated in a similar way to be \( E_n = 95.74 \text{ MeV} \) and \( E_n = 197.29 \text{ MeV} \) respectively.

The gamma ray peak in figure 4.1 was fitted with a standard Gaussian function (see figure 4.3). The full width at half maximum (FWHM) of this Gaussian fit of the gamma peak was measured to be 1.8 ns. This gives an indication of the timing resolution of the detector system.

\[ \Delta T_\gamma = 1.8 \text{ns} \]

**Figure 4.3** A Gaussian fit (bold line) of the \( \gamma \)-peak in the time-of-flight spectrum of the neutron beam produced by 65.99 MeV protons on \(^7\text{Li}\) gives an indication of the detector time resolution.
4.3 Neutron-gamma discrimination

The NE213 detector data was processed to separate the gamma ray events from neutron events in the spectra by the use of pulse-shape discrimination (PSD) described in chapter 3. From a plot of the pulse height versus pulse shape parameters, the values of $L$ and $S$ were chosen such that events due to gamma rays were separated from neutron events. The software separation is shown in figure 4.4. After removing the $\gamma$-ray events from the NE213 data, an analysis of the neutron-only spectra was performed.

![Figure 4.4](image.png)

*Figure 4.4* Event density spectrum of the pulse height parameter, $L$, versus pulse shape parameter, $S$, of neutrons produced by 65.99 MeV protons. The dashed line shows the separation of events that are due to interactions of neutrons with the detector material from events that are due to gamma rays.
Figure 4.5 An orthographic projection of the L versus S spectrum of figure 4.4 showing counts (vertical) versus pulse height, L, and pulse shape, S, for 62.24 MeV neutrons and the associated gamma ray peak. The ‘valley’ between the peaks indicates the position of the locus that separates the gamma ray data from neutron density data.
The time-of-flight parameter $T$, versus the pulse height parameter $L$, spectrum for events due to neutrons only is shown in perspective in figure 4.6. Projecting this spectrum onto the $T$ axis gives the neutron only time-of-flight spectrum (see figure 4.7) and projecting onto the $L$ axis gives the NE213 detector pulse height spectrum in figure 4.13.

Figure 4.6 A perspective plot of counts (vertical) versus pulse height parameter, $L$, and time-of-flight parameter, $T$, of the neutrons produced by the interaction of 65.99 MeV protons with the $^7$Li target. The detector was at 0° to the incident proton beam direction. Gamma rays have been removed from the spectrum.
4.4 Measurements of spectral fluence

The results of the neutron beam spectral fluence measurements are presented in the neutron-only time-of-flight spectra. These measurements were done at 0° and 16° and at the three incident proton beam energies: 65.99 MeV, 99.44 MeV and 203.33 MeV with a Li target. Figure 4.7 shows the time-of-flight channel (T), versus the normalised detector counts for the 0° measurements at these three energies. Figure 4.8 shows the time-of-flight (T) channel versus normalised detector counts measured at 16° for the three proton beam energies.

The measurements at 0° show a sharper high energy neutron peak than the 16° spectra measurements. Resonance structures appear in the continuum of the lower time-of-flight channels for all spectra in both the 0° measurements and the 16° degree measurements. The small structures to the right of the main peak in the $E_p = 65.99$ MeV and $E_p = 203.33$ MeV measurement spectra of figures 4.8 and 4.10 are due to breakthrough in the PSD.

The target material was changed to 8 mm thick graphite and a corresponding neutron time-of-flight spectra was recorded (see figure 4.9) at 0° and at 16° (see figure 4.10). Neutrons were produced from the irradiation of the graphite target with protons via the reaction $^{12}\text{C}(p,n)^{12}\text{N}$ which has a reaction $Q$-value of -13.120 MeV. The main feature of this neutron beam spectrum is that the high energy neutrons are less strongly peaked in the time-of-flight spectrum than they are for neutrons produced from the $^7\text{Li}$ target at the same incident proton energy.
Figure 4.7 Neutron only time-of-flight (T) spectra for (a) 65.99 MeV, (b) 99.44 MeV and (c) 203.33 MeV protons bombarding 6 mm thick $^7$Li, measured at 0° to the incident proton beam direction. The TAC settings were changed during the week (between measurement weekends) hence the time-of-flight channel scale is arbitrary.
Figure 4.8 Time-of-flight (T) spectra for (a) 65.99 MeV, (b) 99.44 MeV and (c) 203.33 MeV protons irradiating 6 mm thick $^7$Li, measured at 16° to the incident proton beam direction.
Figure 4.9 The time-of-flight spectra of neutrons produced from an 8 mm thick $^{12}$C target irradiated by (a) 65.99 MeV, (b) 99.44 MeV and (c) 203.33 MeV protons, measured at 0° to the incident proton beam direction.
Figure 4.10 The time-of-flight spectra of neutrons produced from 8 mm thick $^{12}$C target irradiated by (a) 65.99 MeV, (b) 99.44 MeV and (c) 203.33 MeV protons, measured at 16° to the incident proton beam direction.
4.5 NE213 pulse height response

The response function of an NE213 detector for a particular type of radiation depends on the type and energy spectrum of the radiation, as well as the shape and geometry of the detector [Bu90]. A pulse height calibration of the NE213 detector as well as measurements of its response functions were performed using radioisotopic sources of γ-rays and the neutron beam. This was done to find the detector pulse height threshold and experimental gain, as well as to obtain a measurement of the true zero of the MPA-3 pulse height parameter, \( L \). The γ-calibration sets up a scale for the pulse height spectra which can be reproduced on any ADC [Ti92].

Several γ-ray sources were used for the calibration. These were: \(^{137}\text{Cs}\) which beta decays to an excited state of \(^{137}\text{Ba}\), and consequently decays to its ground state via the emission of a 0.661 MeV γ-ray. \(^{22}\text{Na}\) predominantly decays by β⁺ emission to the 1.275 MeV excited state of \(^{22}\text{Ne}\). In AmBe, the reaction \(^{9}\text{Be}(\alpha, n)\)\(^{12}\text{C}\) proceeds partially via the first excited state of \(^{12}\text{C}\) which then decays by the emission of a 4.44 MeV photon. The pulse height spectra of these γ-rays, as well as a background spectrum were measured with the NE213 detector under the same experimental conditions as the neutron measurements were performed, with the gain set at 1mV/channel for the γ-ray measurements.

The main features of the NE213 detector response function to the 4.44 MeV γ-ray spectrum of AmBe are the Compton peak, the Compton edge and the pair production double escape peak (see figure 4.11). The Compton peak and edge were used as calibration points with the pulse height channel of the Compton peak being associated with 95% of the calculated maximum Compton electron energy [Ma78]. The energy of the Compton edge, \( E_c \), was calculated using [Kn00]

\[
E_c = \frac{2E_\gamma^2}{2E_\gamma + m_e c^2}, \quad (4.3)
\]

where \( m_e c^2 = 0.511 \text{ MeV} \) is the electron rest mass energy. \( E_c \) was found to be 4.198 MeV for AmBe, 1.062 MeV for \(^{22}\text{Na}\) and 0.477 MeV for \(^{137}\text{Cs}\) γ-rays. The double escape peak of AmBe arises from the pair interaction of the 4.44 MeV γ-rays on the \(^{12}\text{C}\) atoms of the NE213 detector where two annihilation γ-rays were produced. These promptly escaped from the detector resulting in the 3.42 MeV double escape peak.
Figure 4.11 Pulse height response of the NE213 detector to photons from (a) $^{137}$Cs, (b) $^{22}$Na, (c) AmBe and (d) the background radiation. The Compton edge of $^{137}$Cs ($E_\gamma = 0.662$ MeV), $^{22}$Na ($E_\gamma = 1.275$ MeV) and AmBe ($E_\gamma = 4.44$ MeV) as well as the double escape peak of the 4.44 MeV $\gamma$-ray ($E_\gamma = 3.42$ MeV) pulse height spectra were used as calibration points.
The AmBe double escape peak provides an unambiguous calibration reference as it is insensitive to both multiple scattering and the detector pulse height resolution [Bu98]. The pulse height channel of the AmBe double escape peak and the $E_c$ of $^{22}$Na, $^{137}$Cs and AmBe were used as reference points for a calibration of the detector pulse height scale. The pulse height channel of the Compton edge was read off at half the height of the Compton peak of the corresponding γ-ray spectrum [Kn72] which is a good approximation for relatively small detector sizes [Di82b]. When these reference points were plotted against the corresponding energy, they showed a linear relationship between the electron energy and the pulse height scale (see figure 4.12) and thus a calibration (linearity factor) was obtained in the form of a linear equation.

**Figure 4.12** Pulse height channel scale dependence on electron energy. The reference points were obtained from the Compton edge of $^{22}$Na, $^{137}$Cs and AmBe γ-ray spectra and the double escape peak of AmBe. These spectra were measured under the same conditions as the neutron beam measurements with 203.33 MeV protons. The amplifier voltage gain was 1 mV/channel.
Figure 4.13 Pulse height (L) spectra for the (a) $E_p = 65.99$ MeV, (b) $E_p = 99.44$ MeV and (c) $E_p = 203.33$ MeV measurements at 0° with the NE213 liquid scintillator.
The PH spectra of the $E_p = 203.33$ MeV measurement (figure 4.13(c)) looks different compared to the lower energy spectra because the 200 MeV recoil protons do not stop in the detector. The PH scale in figure 4.13 was recalibrated to an electron energy scale (figure 4.14) with units of MeVee (MeV electron equivalent). The MeVee is a conventional unit that is taken to correspond to the energy deposited by a 1 MeV electron in the detector [Mo95]. This recalibration was done using the linearity factor obtained from $\gamma$-ray pulse height parameters in figure 4.12. These $\gamma$-ray measurements were performed under the same conditions as the $E_p = 203.33$ MeV neutron fluence measurement. The recalibration made it possible to express the response of the scintillator to different charged particles, such as protons, in terms of the equivalent electron energy that would produce the same pulse height response. The conversion of the PH scale was made taking into consideration that the gain during the course of the neutron measurements was 4mV/channel.

**Figure 4.14** Pulse height spectrum from the $E_p = 203.33$ MeV measurement calibrated to a MeVee scale. The recalibration was done using values obtained from the linear fit in figure 4.12.
4.6 Relative spectral fluence

The neutron time-of-flight spectra were re-binned to neutron energy spectra for the three incident proton beam energies. The energy spectra were corrected for NE213 detector efficiency. The efficiency of the NE213 detector was previously calculated using the Monte Carlo code SCINFUL [No11] for a 2.5 MeVee threshold. This was done using the best evaluations available for the cross sections for n-p elastic scattering and all n-$^{12}$C reactions. Figure 4.15 shows this efficiency as a function of energy.

![Figure 4.15](image-url)  
*Figure 4.15 The NE213 detector efficiency calculated as a function of incident neutron energy. The efficiency was calculated using the Monte Carlo code SCINFUL for a 2.5 MeVee threshold.*
All neutron energy spectra were divided by this efficiency after the PH thresholds were set to 2.5 MeVee. Both the $0^\circ$ and $16^\circ$ spectra were then normalised to the same monitor counts (see table 4.1 for the monitor values labelled $M$). This made it possible to compare the spectral distribution of the neutron beam that was measured at $0^\circ$, to the neutron beam that was measured at $16^\circ$, for the same beam energy. The $16^\circ$ spectra were further adjusted to match the $0^\circ$ spectra by dividing them by a factor of 1.2 (65.99 MeV), 1.2 (99.44 MeV) and 1.1 (203.33 MeV) in order to match them in the region from 0 MeV to 30 MeV and avoid negative values of fluence. The neutron energy spectra for the $E_p = 65.99$ MeV measurement are shown in figure 4.16.

![Figure 4.16](image)

Figure 4.16 Relative spectral fluence measured at 9.158 m at $0^\circ$ (bold line) and 8.000 m at $16^\circ$ for neutrons produced by 65.99 MeV protons impinging on a 6 mm thick $^7$Li target. The $16^\circ$ spectrum has been normalised by a factor of 1.2. The peak neutron fluence makes up a small fraction of the total neutron fluence in the $16^\circ$ spectrum.
Figure 4.17 Relative spectral fluence measured at 9.158 m at 0° and 8.000 m at 16° for neutrons produced from 99.44 MeV protons irradiating a 6 mm thick ⁷Li target. Peak neutrons make a much smaller contribution to the total fluence in the 16° spectrum than continuum neutrons.

It was observed that the contribution of high-energy neutrons to the 16° spectra decreases with increasing incident proton energy, whereas the ratio of the fluence of high energy neutrons to the fluence of continuum neutrons shows a marginal increase for the 0° measurement with increasing energy (see table 4.1). At $E_p = 65.99$ MeV, the peak to continuum ratio is much larger for the 0° spectrum than it is for the 16° spectrum. The high-energy peak of the 16° spectrum is relatively small when compared to its low-energy continuum component. For the $E_p = 99.44$ MeV measurements (figure 4.17), the ratio of peak to continuum neutrons stays relatively large for the 0° spectrum, but gets even smaller for the 16° spectrum.
Figure 4.18 Relative spectral fluence measured at 8.000 m at 0° and 8.000 m at 16° for neutrons produced by 203.33 MeV protons irradiating a 6 mm thick $^7$Li target. The high energy peak almost completely disappears from the 16° spectrum.

The high energy peak almost completely disappears from the 16° spectrum measured at $E_p = 203.33$ MeV (see figure 4.18) but a strong high-energy neutron peak can still be observed at 0°.

The highest energy neutrons arise from nuclear transitions to the unresolved ground state and first excited states of the product $^7$Be in the $^7$Li(p,n)$^7$Be reaction. These neutrons are strongly forward peaked whereas the continuum neutrons are more isotropic, hence the continuum flux is less affected by varying the angle at which a neutron beam measurement is made. This is shown in the difference between the neutron flux in the high-energy peak of the 0° measurement and the 16° measurement (see table 4.1). The 16° spectra are more strongly influenced by the low energy continuum of the neutron beam and yield interesting insight into the reaction kinematics.
A difference spectrum was obtained by subtracting the normalised 16° spectra from the normalised 0° spectra of the neutron beam. This made it possible to isolate the high-energy neutrons of the main peak and thus obtain a nearly monoenergetic or quasi-monoenergetic neutron beam. These 0° - 16° difference spectra measurements are shown in figure 4.19.

**Table 4.1** The parameters of the spectral fluence distribution of the iThemba LABS neutron beam.

<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>$\Delta E_{\text{calc}}$ (MeV)</th>
<th>$E_{0,\text{calc}}$ (MeV)</th>
<th>$F$ (ch/ns)</th>
<th>$T_n$ (ch)</th>
<th>$E_1$ (MeV)</th>
<th>$N_0/N$</th>
<th>$N_{0/16}^\circ/N$</th>
<th>$N_{0/16}^{16\circ}$</th>
<th>$M^* (10^7)$</th>
<th>$M^* (10^7)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>65.99</td>
<td>2.674</td>
<td>62.99</td>
<td>2.51</td>
<td>829</td>
<td>62.24</td>
<td>0.58</td>
<td>0.42</td>
<td>3.14</td>
<td>2.15</td>
<td></td>
</tr>
<tr>
<td>99.44</td>
<td>1.933</td>
<td>96.81</td>
<td>2.51</td>
<td>754</td>
<td>95.74</td>
<td>0.64</td>
<td>0.35</td>
<td>3.68</td>
<td>1.73</td>
<td></td>
</tr>
<tr>
<td>203.33</td>
<td>1.163</td>
<td>201.08</td>
<td>2.52</td>
<td>535</td>
<td>197.29</td>
<td>0.68</td>
<td>0.20</td>
<td>4.88</td>
<td>2.49</td>
<td></td>
</tr>
</tbody>
</table>

$E_p$ is proton energy, $E_{0,\text{calc}}$ is the mean energy of the high energy neutrons calculated from $\Delta E_{\text{calc}}$, the proton energy loss in the target and the $Q$-value. $E_1$ is the mean energy as calculated from the time-of-flight. $F$ is the time calibration factor, $T_n$ is the time-of-flight channel number of the neutron peak. $N_0/N$ is the ratio of the fluence of peak neutrons to continuum neutrons obtained from integrals of the neutron energy spectra. $M$ is the number of monitor counts.

A correction was made for low energy neutrons that registered counts on the neutron monitors, but were subsequently vetoed by the NE213 detector because of the low energy threshold. Having recalibrated the pulse height scale to a MeVee scale, the threshold was calculated in units of electron energy, and subsequently the correction was used in the calculation of neutron densities by integrating neutron energy spectra.

The width of the high energy neutron peaks in figure 4.19 is dominated by the detector resolution. It becomes increasingly wider at higher energies than what the proton energy loss in the target, $\Delta E_{\text{calc}}$, would suggest. This happens because the energy resolution becomes poorer at higher energies, due to shorter neutron flight times as well as limited time resolution [Gu05].
Figure 4.19 The $0^\circ$ - $16^\circ$ difference spectra measurements performed with the neutron beam energies; $E_n = 62.24$ MeV, $E_n = 95.74$ MeV, $E_n = 197.29$ MeV. The spectra are shown offset by 0.005 on the vertical axis for clarity.
Chapter 5

Results: Peak neutron fluence

Measurements performed with the Institute for Reference Materials and Measurements (IRMM) $^{238}$U fission ionization chamber detector allowed for a calculation of the peak neutron fluence, $\Phi_o$ (neutrons/cm²), using the equation [No07b]

$$\Phi_o = \prod_i k_i \frac{N_{f,exp}}{\sigma N_u},$$  \hspace{1cm} (5.1)

where $N_{f,exp}$ represents the measured fission counts from the detector, $\prod k_i$ are the various correction factors, $N_u$ is the total number of fissile $^{238}$U atoms on the fission chamber plates and $\sigma$ is the $^{238}$U fission cross section.
5.1 Number of $^{238}\text{U}$ atoms in the fission chamber

The total mass of uranium in the IRMM fission chamber was measured to be 0.2409 g and specified to within 10% uncertainty. This amount of uranium was deposited on six tantalum plates, back-to-back, by means of electronic spraying in the form of highly purified uranium oxide ($\text{U}_3\text{O}_8$) with an approximate density of 8.3 g/cm$^3$ [No09]. The total number, $N$, of fissile uranium atoms contained in the $\text{U}_3\text{O}_8$ is

$$N = \frac{0.2409 \text{ g} \times (6.0221415 \times 10^{23} \text{ atoms/mol})}{238.0507826 \text{ g/mol}} \tag{5.2}$$

$$= 6.0950 \times 10^{20} \text{ atoms of uranium.}$$

The relative abundance of $^{238}\text{U}$ in natural uranium is 0.999825 (see Appendix II), therefore the number, $N_u$, of $^{238}\text{U}$ atoms is

$$N_u = 0.999825 \times (6.0950 \times 10^{20}) \tag{5.3}$$

$$= 6.0939 \times 10^{20} \text{ atoms.}$$

The standard unit of scattering cross section is the barn ($10^{-24} \text{ cm}^2$), thus $N_u$ can be expressed in terms of the total atomic area available for neutron fission

$$N_u = (6.0939 \times 10^{20}) \times (10^{-24} \text{ cm}^2/\text{barn}) \tag{5.4}$$

$$= 0.00060939 \text{ (cm}^2/\text{barn}).$$

5.2 $^{238}\text{U}$ Neutron fission cross section

The $^{238}\text{U}$ fission cross sections ($\sigma$) that were used in the calculation of $\Phi$ (see equation 5.1) were obtained from the International Nuclear Data Committee (INDC) Nuclear Data Standards [Ca97]. The $^{238}\text{U}(n,f)$ cross section exhibits a minimum incident energy threshold near 1 MeV which eliminates the response to thermal neutrons and $\gamma$-rays. The $^{238}\text{U}$ cross section is relatively
large and the variation of the cross section with neutron energy is fairly smooth (see figure 5.1) which allows for neutron fluence measurements to be made.

Figure 5.1 The fission cross section (barn) for $^{238}U$ as a function of the incident neutron energy [IAEA-NDS]
5.3 Fission chamber spectra

Each event recorded with the $^{238}\text{U}$ fission chamber detector yielded pulse height and time-of-flight data. Figure 5.2 shows an orthographic plot of fission events as a function of pulse height parameter $L$, and time-of-flight parameter $T$, from 62.24 MeV neutrons measured by the fission chamber. The large peak in the spectrum observed at higher time-of-flight channels corresponds to the detection of the maximum energy neutrons. Projecting this spectrum onto the pulse height ($L$) axis yields the pulse height spectrum shown in figure 5.3 (a).

The fission chamber pulse height spectra (figure 5.3) exhibit a large contribution of counts from $\alpha$-particle events at the lower pulse height channels. These $\alpha$-particles are a result of the natural radioactivity of uranium and can be observed as the large peak in the perspective plot of figure 5.2. A hardware threshold was applied in the electronics of the detector which excluded most, but not all, of the $\alpha$-particles just above zero pulse height [No07]. By limiting the alpha events, this threshold helped to reduce dead time and pile up in the detector electronics. The shape of the fission fragments pulse height spectrum obtained from the fission chamber is relatively stable and is independent of the fluence rate [Mo10].

The $\alpha$-particles from $^{238}\text{U}$ have average energies of 5 MeV whereas two fission fragments can share up to 160 MeV [Cr91]. Based on this difference, a pulse height threshold based on the $\alpha$-particle energy was set in the detector software to veto the alpha counts and other light reaction products [Ga90] from fission events. The position of this threshold in the pulse height spectrum is indicated by the vertical dashed line in figure 5.3.

The time-of-flight spectra measured with the fission chamber (figure 5.4) are due to neutron fission events, as well as the time independent $\alpha$-particle events. The black spectra in figure 5.4 show the time-of-flight information of neutron events and include the $\alpha$-particle events. After the $\alpha$-particle discriminator was applied in the detector software, the neutron-only time-of-flight spectra (red spectra) were obtained. The position of the discriminator determines the ratio of alpha counts that were suppressed in the time-of-flight spectra.

The uranium fission chamber time-of-flight spectrum also consists of a high energy peak and a lower energy continuum. The separation of the high energy peak from the low energy continuum
was not very clear in the measured spectrum due to the time resolution of the fission chamber. A correction was made to compensate for the overlap of these components, by fitting a Gaussian function to the peak and continuum components of the measured time-of-flight spectrum. Adjustments were then made to the convolution in order to reproduce, as accurately as possible, the time-of-flight spectrum that was measured by the fission chamber. The width of the Gaussian fit was then adjusted to reproduce the shape of the high-energy peak in the measured time-of-flight peak. Figure 5.5 shows the neutron time-of-flight spectra fitted with a standard Gaussian distribution between vertical dashed lines. The measured fission counts, $N_{f,exp}$, were obtained by taking the integral under the Gaussian fit and between the dashed lines.

**Figure 5.2** An orthographic plot of the fission chamber counts (vertical) versus the pulse height parameter, $L$, and the time-of-flight parameter, $T$, for 62.24 MeV energy neutrons measured with the IRMM fission chamber at a distance 9.291 m from the centre of the target at 0°. Projecting this plot onto the $L$-axis yields the PH spectrum shown in figure 5.3(a). Projecting it onto the $T$-axis yields the time-of-flight spectrum in figure 5.4(a).
Figure 5.3 Fission chamber pulse height (L) spectra for neutrons of energy (a) 62.24 MeV, (b) 95.74 MeV and (c) 197.29 MeV measured at 0°. The dashed line in each pane indicates the position of the pulse height threshold which was set to exclude alpha particle events.
Figure 5.4 Fission chamber time-of-flight spectra obtained at neutron energies (a) 62.24 MeV, (b) 95.74 MeV and (c) 197.29 MeV measured at 0°. The black spectra consist of neutron fission events and α-particle events. The red spectra show the time-of-flight structure after the threshold was applied in the software and the dashed lines indicate where $N_{f,exp}$ was calculated.
Figure 5.5 The neutron-only time-of-flight spectra (red histogram) at (a) 62.24 MeV, (b) 95.74 MeV and (c) 197.29 MeV measured at 0°. The high energy neutron peaks were fitted with a standard Gaussian function. The number of fission counts due to peak neutrons, $N_{f,exp}$, was calculated by integrating the fission counts under the Gaussian fit between the dashed lines.
5.4 Correction factors

Correction factors were calculated in order to relate the measured value of fission counts, \( N_{f,\text{exp}} \), to the value that would be obtained under reference conditions, \( N_f \) [No07c].

\[
N_f = \prod k_i N_{f,\text{exp}} . \tag{5.5}
\]

The overall correction factor for each neutron beam measurement, \( k_{\text{total}} \), was calculated from the product of the partial correction factors and folded into the fluence calculation. A description of each of the partial correction factors, \( k_i \), that are used to determine \( N_f \), how they arise and how they are calculated, is described in this section.

5.4.1 Correction for the fission fragment detection efficiency, \( k_1 \)

The inefficiency of the fission chamber in fission fragment detection is primarily a result of the incomplete linear momentum transfer that characterizes \(^{238}\text{U}(n,f)\) nuclear interactions. The anisotropy of fission fragments which results from angular momentum transfer [Ca74] during these interactions compounds this inefficiency.

The efficiency of the IRMM fission chamber in the neutron energy region above 20 MeV had not been calculated prior to the fluence measurements at iThemba LABS. Instead, the results of efficiency calculations that were performed previously on the PTB fission chamber, the H21, were used [Mo10]. Comparisons showed that the IRMM fission chamber had a neutron detection efficiency approximately 10% higher than the H21 detector [Mo08] at a neutron beam energy of 15 MeV. Comparisons of pulse height measurements performed in the energy region below 20 MeV showed that the IRMM results and the H21 results were in agreement to within 1% [Mo10]. The fission chamber efficiency was determined as a function of incident neutron energy (see figure 5.6). A correction factor, \( k_1 \), for the efficiency, \( \varepsilon \), of the IRMM fission chamber was calculated:

\[
k_1 = \frac{1}{\varepsilon} . \tag{5.6}
\]
Figure 5.6 The efficiency, $\varepsilon$, of the IRMM fission chamber as a function of incident neutron energy.
Figure 5.7 The correction factor, $k_2$, for neutron absorption, multiplication and attenuation in the fission chamber as a function of incident neutron energy.
5.4.2 Correction for neutron absorption by the chamber structure, $k_2$

A correction was determined for the absorption of neutrons by the fission chamber apparatus, their subsequent multiplication due to the production of secondary neutrons via $(n,xn)$ reactions as well as attenuation within the chamber. The correction factor, $k_2$, was calculated using the Monte Carlo code MCNPX. The correction makes an adjustment for the peak neutron fluence, $\Phi_o$, that would exist at the position of each electrode, had the chamber structure not been present. MCNPX was used to calculate the spectral fluence, $\Phi_o$, in the chamber averaged over the area of the fissile layers on the six electrodes from the uncollided fluence $\Phi$ (at the chamber entrance). The correction factor, $k_2$, was plotted as a function of neutron energy in figure 5.7.

5.4.3 Correction for the applied threshold, $k_3$

A software threshold was applied in the detector pulse height parameter to discriminate $\alpha$-particle events from neutron fission events. Inevitably, alpha events extend to higher pulse height channels above the threshold. These counts decrease asymptotically to zero. A correction was made for these alpha counts by approximating the alpha counts by a linear decrease with increasing pulse height channel (the red line in figure 5.8). Integrating the counts under this extension yields a value, $N_\alpha$, for these ‘extra’ $\alpha$-particle counts.

Neutron fission events still occur at low pulse height values, below the alpha discriminator (hardware) threshold down to zero pulse height. By taking a flat extrapolation of the fission pulse height spectrum to zero pulse height (the horizontal green line in figure 5.8), it was possible to calculate a correction for these ‘lost’ events, $N_{Th}$.

The correction factor for these effects was calculated by adding the neutron fission counts lost due to the PH threshold, $N_{Th}$, and subtracting the alpha counts that occur above the threshold, $N_\alpha$, from the experimentally measured fission counts, $N_{f,exp}$:

$$k_3 = (N_{Th} - N_\alpha + N_{f,exp}) / N_{f,exp}. \quad (5.7)$$
**Figure 5.8** Pulse height channel ($L$) spectra for 62.24 MeV neutrons at $0^\circ$. The vertical dashes indicate the $\alpha$-particle PH threshold. The green line indicates the horizontal extrapolation for fission counts from the threshold to zero PH and the red line indicates the approximation for alpha counts that are detected above the PH discriminator threshold.

### 5.4.4 Correction for dead-time, $k_4$

Dead-time exists in all types of detectors and is due to the finite time required by the system to process an event in the detector and produce an output pulse [Le94]. During this time, the detector may not be sensitive to other events. The combination of digital and analogue components in the fission chamber data acquisition equipment results in a dead-time correction for the system given by:
\[ k_4 = k_{DAQ} k_{analogue} \]  

(5.8)

where \( k_{DAQ} \) is the dead-time coefficient for the digital component and \( k_{analogue} \) is the dead-time coefficient for the analogue component of the detector system. A value for \( k_4 = 1.001 \) was estimated for the measurements at \( E_n = 62.24 \text{ MeV} \) and \( E_n = 95.74 \text{ MeV} \).

### 5.4.5 Correction for the reference distance, \( k_5 \)

The fission chamber was positioned at a different distance, \( d \), from the target (see tables 5.1 to 5.3) for each measurement as the same detectors had to be moved between the 0° and 16° positions between measurements. However, the neutron fluence had to be specified at a particular reference distance, \( d_{ref} = 8 \text{ m} \). Thus, a correction, \( k_5 \), was calculated to adjust for the difference in flight path between the experimental distance and the reference distance:

\[ k_5 = \left( \frac{d}{d_{ref}} \right)^2. \]  

(5.9)

### 5.4.6 Correction for neutron beam attenuation, \( k_6 \)

The neutron beam flux attenuates along the flight path from the target to the detector. A correction was made for the attenuation of the neutron beam in air from the target position to the adjusted reference position of the fission chamber. The attenuation coefficient in air, \( \Sigma_{air} \), which accounted for the attenuation of neutron flux between the experiment distance, \( d \) and reference distance, \( d_{ref} \) [No07c], was plotted as a function of neutron energy in figure 5.9 and the correction factor, \( k_6 \), was calculated as:

\[ k_6 = \exp (-\Sigma_{air} (d_{ref} - d)). \]  

(5.10)
Figure 5.9 The attenuation coefficient in air of the neutron beam, $\Sigma_{\text{air}}$, as a function of incident neutron energy.
5.4.7 Correction for the neutron beam profile, \( k_7 \)

Due to the inhomogeneous nature of the iThemba LABS neutron beam profile at the detector face, the mean neutron fluence averaged over the detector face, \( \Phi_{avg} \), is different from the actual neutron fluence at the centre of the detector, \( \Phi \). Essentially, the beam deviates from a rectangular profile about the centre in both the vertical and horizontal planes and becomes irregular. A Monte Carlo code BPR.EXE was used specifically for the rectangular neutron beam profile at iThemba LABS [No07b] and a correction factor for the beam profile was calculated (see Appendix I):

\[
k_7 = \frac{\Phi}{\Phi_{avg}}.
\]  

(5.11)

5.4.8 Correction for the time resolution of the fission chamber, \( k_8 \)

As a result of its relatively poor time resolution, the fission chamber detector can not sufficiently resolve the fast neutron peak and the slow continuum in the time-of-flight spectra. This means a continuum component is present in the analysis of fission events induced by the peak neutrons. A correction for this was calculated by simulating the time-of-flight response of the FC using a high resolution instrument, such as an NE213 detector and the \(^{238}\text{U} \) reference cross sections. The peak and continuum components of the simulated spectrum, which could then be sufficiently resolved, are fitted with standard Gaussian distributions. These were then adjusted to reproduce the shape of the Gaussian fit of the peak and continuum components in the measured time-of-flight spectrum. In this way, a correction for the detector time resolution was found:

\[
k_8 = 1.
\]  

(5.12)

5.4.9 Total correction

The overall correction factor was obtained from the product of the partial correction factors \( k_i \) to \( k_8 \) (see tables 5.1 to 5.3) using

\[
k_{total} = \prod k_i.
\]  

(5.13)
5.5 Uncertainty in measured quantities

The process of measuring and calculating every variable in equation (5.1) inherently introduces an uncertainty that is associated with each measured variable. Consequently, $\Phi_o$ will also have an uncertainty associated with it. For the purposes of this work, the treatment of measurement uncertainties, as well as their propagation, was done in accordance with the Joint Committee for Guides in Metrology (JCGM) Guide to the Expression of Uncertainty in Measurement (GUM), [GUM08]. This classification makes possible the determination of a transferrable and consistent standard uncertainty associated with each measurement that was performed during the experiments [Ki02].

According to the GUM method, uncertainties are classified as either Type A or Type B uncertainties depending on how the measurement is performed. Type A uncertainties arise from the statistical analysis of repeated observations with dispersion, and are obtained from a probability density function (PDF) that is derived from the observed frequency distribution. Type B uncertainties arise when methods other than statistical analysis are applied to a series of observations [GUM], [Ka07]. The uncertainty associated with the measurement of $N_{f,exp}$ was treated as a Type A evaluation and calculated as the square root of the measurand. The Type B uncertainties were treated either as a triangular PDF, or a rectangular PDF. The choice of PDF depended on the information that was available about the instrument. A triangular PDF gives a standard uncertainty $u(x)$ of the form:

$$u(x) = \frac{1}{2} \frac{(x_{right} - x_{left})}{\sqrt{6}},$$

(5.14)

whereas a rectangular probability function gives a standard uncertainty $u(x)$ of the form

$$u(x) = \frac{1}{2} \frac{(x_{right} - x_{left})}{\sqrt{3}},$$

(5.15)

Where the $x_{right}$ and $x_{left}$ are the lower and upper limits of the measurand for the PDF in use.

The standard uncertainties associated with the variables of equation (5.1), including the correction factors, are listed in table 5.1, table 5.2 and table 5.3.
Table 5.1 The standard uncertainties obtained for the measured parameters at $E_{n} = 62.24$ MeV.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>$x_i$</th>
<th>$u(x_i)$</th>
<th>Uncertainty Evaluation</th>
<th>PDF</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_{f,exp}$ (counts)</td>
<td>7610</td>
<td>87</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$N_{n} \times 10^{-4}$ (cm$^2$/b)</td>
<td>6.09</td>
<td>0.61</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$\sigma$ (barn)</td>
<td>1.576</td>
<td>0.027</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$d$ (m)</td>
<td>9.291</td>
<td>0.001</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_1$</td>
<td>1.058</td>
<td>0.016</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_2$</td>
<td>0.989</td>
<td>0.005</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_3$</td>
<td>1.079</td>
<td>0.006</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$k_4$</td>
<td>1.001</td>
<td>0.001</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_5$</td>
<td>1.349</td>
<td>0.002</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$k_6$</td>
<td>1.004</td>
<td>0.004</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_7$</td>
<td>1.037</td>
<td>0.010</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_8$</td>
<td>1.000</td>
<td>0.045</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_{total}$</td>
<td>1.587</td>
<td>0.050</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 5.2 The standard uncertainties obtained for the measured parameters at $E_n = 95.74$ MeV.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>$x_i$</th>
<th>$u(x_i)$</th>
<th>Uncertainty Evaluation</th>
<th>PDF</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_{f,exp}$ (counts)</td>
<td>5708</td>
<td>75</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$N_n * 10^{-4}$ (cm$^2$/b)</td>
<td>6.09</td>
<td>0.61</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$\sigma$ (barn)</td>
<td>1.402</td>
<td>0.040</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$d$ (m)</td>
<td>8.842</td>
<td>0.001</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_1$</td>
<td>1.058</td>
<td>0.016</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_2$</td>
<td>0.989</td>
<td>0.005</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_3$</td>
<td>1.079</td>
<td>0.006</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$k_4$</td>
<td>1.001</td>
<td>0.001</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_5$</td>
<td>1.222</td>
<td>0.002</td>
<td>Type A</td>
<td>Gaussian</td>
</tr>
<tr>
<td>$k_6$</td>
<td>1.002</td>
<td>0.003</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_7$</td>
<td>1.037</td>
<td>0.010</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_8$</td>
<td>1.000</td>
<td>0.045</td>
<td>Type B</td>
<td>triangular</td>
</tr>
<tr>
<td>$k_{total}$</td>
<td>1.435</td>
<td>0.050</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The peak neutron fluence, $\Phi_0$, was calculated from equation (5.1) at the three neutron beam energies $E_n = 62.24$ MeV, $E_n = 95.74$ MeV and $E_n = 197.29$ MeV. The combined standard uncertainty for the peak neutron fluence, $u(\Phi_0)$, was calculated using the equation [GUM08]:

$$
u(\Phi_0) = \frac{1}{\Phi_0} \sqrt{\sum_i \left( \frac{u(x_i)}{x_i} \right)^2 } .$$

The values of $\Phi_0$ and their associated uncertainty are shown in Table 5.4. For large beam currents, such as those employed in the fission chamber measurements, the uncertainty of the counting procedure corresponds to the statistical uncertainty of the fission chamber reading and is typically in the range 1% to 2%. For small beam currents, an uncertainty of 2% was estimated by analyzing the influence of the irradiation conditions (e.g. varying the beam current and focusing) on the readings of the two beam monitors [Sc99].
Table 5.4 The peak fluence of the iThemba LABS neutron beam and other parameters obtained during the measurements of neutron fluence.

<table>
<thead>
<tr>
<th>$E_n$ (MeV)</th>
<th>$\sigma$ (barn)</th>
<th>$N_{f,exp}$</th>
<th>$k_{total}$</th>
<th>$t$ (s)</th>
<th>$d$ (m)</th>
<th>$\Phi_o \times 10^{+6}$ (n/cm$^2$)</th>
<th>$Q$ (nC)</th>
<th>$\Phi_o/Q$ (n/cm$^2$/nC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>62.24</td>
<td>1.576 ± 0.027</td>
<td>7610 ± 87</td>
<td>1.592 ± 0.050</td>
<td>6940</td>
<td>9.291</td>
<td>12.58 ± 0.65</td>
<td>2079338</td>
<td>6.05 ± 0.31</td>
</tr>
<tr>
<td>95.74</td>
<td>1.402 ± 0.040</td>
<td>5708 ± 75</td>
<td>1.435 ± 0.050</td>
<td>5565</td>
<td>8.842</td>
<td>11.68 ± 0.98</td>
<td>2070921</td>
<td>5.64 ± 0.47</td>
</tr>
<tr>
<td>197.29</td>
<td>1.314 ± 0.040</td>
<td>2548 ± 50</td>
<td>1.515 ± 0.041</td>
<td>13918</td>
<td>8.837</td>
<td>4.82 ± 0.53</td>
<td>627604</td>
<td>7.68 ± 0.84</td>
</tr>
</tbody>
</table>

In the table, $\sigma$ is the neutron fission cross section of $^{238}$U, $t$, is the duration of the neutron fluence measurement and $N_{f,exp}$, represents the measured number of fission counts obtained from integrating the Gaussian fit to the experimental time-of-flight spectrum peak. The peak neutron fluence integrated over the total measurement time, $\Phi_o$ (neutrons/cm$^2$), was divided by the total charge measured by the beam charge monitor, $Q$ (nC). This allowed for the fluence to be calculated as a ratio of the total beam charge, $\Phi_o/Q$ (neutrons/cm$^2$/nC).

The measurement characteristics of the IRMM fission chamber were made traceable to an international reference standard. This was done by conducting simultaneous calibration measurements with the PTB fission chamber, the H21, which is an international standard reference device [Mo08]. The composition of uranium in the IRMM fission chamber was certified as a uranium isotopic standard (see Appendix II).
Chapter 6

Conclusion

6.1 Spectral fluence

Measurements of the neutron fluence were carried out at the D-line fast neutron facility at iThemba LABS. Time-of-flight spectra were obtained using the high resolution NE213 scintillator and these were re-binned to energy spectra. Pulse shape discrimination was used to separate neutron events from $\gamma$-ray events.

The spectral distribution of the iThemba LABS neutron beam was found to consist of a strongly forward-peaked high energy component and an isotropic continuum of lower energy neutrons.
The peak neutrons were produced from transitions to the ground state and first excited state of the target \((E_x = 0.429 \text{ MeV})\) in the neutron production reaction \(^7\text{Li}(p,n)^7\text{Be}\) while continuum neutrons were produced from break-up reactions, higher order excitations, as well as neutrons that had interactions with the collimator [No02b].

The ratio of the neutron fluence in the high energy peak to fluence in the continuum was found to increase with increasing neutron beam energy for 0° spectra. However, the converse was found to be true for neutron fluence measurements performed at 16°, as the ratio of the peak fluence to the continuum fluence in the wide angle measurements diminished at higher neutron energies.

By performing difference measurements with the normalised 0° and 16° spectra, it was possible to exclude the low energy fluence in the continuum from the total neutron fluence. In this way, spectra of quasi-monoenergetic neutron beams were obtained at 0°.

### 6.2 Relative fluence

The peak fluence of the iThemba LABS neutron beam was calculated relative to the recommended \(^{238}\text{U}(n,f)\) scattering cross section [Ca97] using the IRMM \(^{238}\text{U}\) fission chamber. Time-of-flight and pulse height measurements were measured with the fission chamber at both 0° and 16° to the neutron beam direction. Alpha fission events were excluded from the spectra by the use of a threshold based on the energy of the fission fragments.

The number of fission counts that could be attributed to peak neutrons, \(N_{f,exp}\), was obtained by integrating under a Gaussian fit of the high energy neutron peak in the time-of-flight spectra. The peak fluence of the neutron beam was calculated for each measurement energy using equation (5.1). In order to account for the deviations from reference conditions, several corrections were made and folded into the calculation of the relative fluence.
6.3 Future work

A wide scope of further developments could arise out of this work. These include comparisons of the measured spectra, with Monte Carlo simulations. The measurement of neutron fluence at the iThemba LABS D-line makes it possible to characterise different neutron detectors, in a variety of arrangements.

The neutron beam can be used to develop fission chamber detectors based on other fissile materials, besides uranium, with a view to improving the time resolution of fission chambers. More precise measurements of neutron fission cross sections of $^{235}\text{U}$ and $^{238}\text{U}$ can be carried out in the energy range covered by the iThemba LABS neutron beam.

Also, further investigations are required into the correlations between the uncertainties in the total fluence and the various measured quantities such as fission chamber efficiency, total number of fissile atoms in the fission chamber presented to the beam and correction for continuum overlap with peak neutrons [No07].
References


[Ch52] C. N. Chou, Phys. Rev. 87 (1952) 904


[Ha79] J. A. Harvey, N. W. Hill, Nuc. Instr. and Meth. 162 (1979) 507


[Ma68] J.B. Marion and F.C. Young, Nuclear Reaction Analysis, North Holland, Amsterdam, (1968)


[Sa01] Saint-Gobain Crystals and Detectors, “Scintillation Products” Information Sheet, (2001), Ohio, USA


Appendix I

Beam profile measurements

Determination of the spatial beam profile

The spatial beam profile at iThemba LABS was determined using image plates mounted behind a 2 mm Polymethyl methacrylate (PMMA) plate. In addition, thermo-luminescent diodes (TLDs) were inserted in a PMMA holder in two vertical and two horizontal rows. The TLDs were covered by a 2 mm PMMA layer and mounted behind the image, such that the central crossing point of the horizontal and vertical rows was the position of the nominal beam centre. The distance of the image plates and the $^7$LiF TLDs to the Li-target was about 6.69 m and 6.63 m for the 100 MeV and 200 MeV runs, respectively.

The spatial beam profile is parameterized by the following model:

$$\frac{\Phi(x, y)}{\Phi(0,0)} = p_x(x) \cdot p_y(y)$$

(a)
with

\[ p_i(z) = \frac{1 + a_iz + b_iz^2}{(1 + \exp(-(z+1)/w_i))(1 + \exp((z+1)/w_i))} \quad \text{with} \quad i = x, y \quad (b) \]

and

\[ z = \frac{x-x_0}{\delta_x} \quad \text{or} \quad z = \frac{y-y_0}{\delta_y} \quad (c) \]

where \( x \) and \( y \) are the horizontal and vertical coordinates of a right-handed coordinate system with the \( z \)-axis pointing to the Li-target, i.e. against the neutron beam direction. It has to be noted that the image plates and the TLDs integrate over the spectral distribution. It was therefore assumed that the energy dependence of the spatial profile as well as the contribution of \( \gamma \)-rays produced in the target either by direct reactions or by activation can be neglected. The slope of the profiles (shown in figure 3.8), in particular for 200 MeV, could have been caused by half-shadow effects due to non-perpendicular or non-central incidence of the proton beam on the Li target.

Table A1 shows the parameters of the beam profiles for the reference distance of 8 m. The parameters were determined by fitting the parameterization (b) to the TLD data and to corresponding one-dimensional intensity distributions produced by placing cuts on the image plate data. In both cases, the background was determined outside the beam area and subtracted.
Table A1 Parameters for the spatial beam profile at the reference distance of 8 m. The offsets \((x_0, y_0)\) of the position of the centroid of the neutron beam from the nominal beam centre could only be determined using the TLDs.

<table>
<thead>
<tr>
<th></th>
<th>TLD 100 MeV</th>
<th>image plate 100 MeV</th>
<th>TLD 200 MeV</th>
<th>image plate 200 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a_x)</td>
<td>-0.0265</td>
<td>0.00084</td>
<td>0.0258</td>
<td>-0.0365</td>
</tr>
<tr>
<td>(b_x)</td>
<td>-0.157</td>
<td>-0.204</td>
<td>-0.146</td>
<td>-0.149</td>
</tr>
<tr>
<td>(\delta_x) (cm)</td>
<td>4.853</td>
<td>4.850</td>
<td>4.911</td>
<td>4.905</td>
</tr>
<tr>
<td>(w_x)</td>
<td>0.0342</td>
<td>0.0210</td>
<td>0.0388</td>
<td>0.0188</td>
</tr>
<tr>
<td>(x_0) (cm)</td>
<td>0.041</td>
<td></td>
<td>-0.243</td>
<td></td>
</tr>
<tr>
<td>(a_y)</td>
<td>0.0831</td>
<td>0.0524</td>
<td>0.0659</td>
<td>0.162</td>
</tr>
<tr>
<td>(b_y)</td>
<td>-0.151</td>
<td>-0.197</td>
<td>-0.180</td>
<td>-0.203</td>
</tr>
<tr>
<td>(\delta_y) (cm)</td>
<td>4.859</td>
<td>4.863</td>
<td>4.899</td>
<td>4.906</td>
</tr>
<tr>
<td>(w_y)</td>
<td>0.0320</td>
<td>0.0202</td>
<td>0.0339</td>
<td>0.0286</td>
</tr>
<tr>
<td>(y_0) (cm)</td>
<td>0.038</td>
<td></td>
<td>0.277</td>
<td></td>
</tr>
</tbody>
</table>
Appendix II

Uranium certificates

$^{238}\text{U}$ certificate of analysis and mass certificates

The uranium fission chamber detector that was used in this work was obtained from the Institute for Reference Materials and Measurements (IRMM). The composition of uranium that was used in the construction of this device has been analysed by the National Bureau of Standards of the United States of America. A certificate of analysis was issued by the Office of Standard Reference Materials, which confirmed this composition of uranium to be a uranium isotopic standard. A certificate for the uranium mass was also provided.
Certificate of Analysis

STANDARD REFERENCE MATERIAL U-0002

Uranium Isotopic Standard

\[
\begin{array}{cccc}
234U & 235U & 236U & 238U \\
\text{Atom Percent} & 0.00016 & 0.01755 & <0.00001 & 99.9823  \\
\pm & .00001 & \pm .00005 & \cdots & \pm .00001  \\
\text{Weight Percent} & .00016 & .01733 & < .00001 & 99.9825
\end{array}
\]

The material consists of highly purified oxide, U3O8. The atomic weight of the material is calculated to be 238.0503 using the nuclidic masses 234.0409, 235.0439, and 238.0508.

The value for 235U is calculated from measurements made on samples spiked with high purity 233U to approximate the 235U concentration, the ratio 233U to 235U was measured on a triple-filament equipped thermal ionization mass spectrometer with d-c amplifier circuits. Ratio determinations were corrected for mass discrimination by measurements made under similar conditions on SRM U-509.

The value for 234U is calculated from measurements made on samples spiked with high purity 233U, the ratio 233U to 234U was measured on a two stage mass spectrometer using a pulse counting technique.

The limits indicated for the isotopic compositions are at least as large as the 95 percent confidence level for a single determination, and include terms for the inhomogeneities of the material as well as analytical error.

Mass spectrometry measurements at NBS were made by E. L. Garner and L. J. Moore using solutions prepared by L. A. Machlan.

The overall direction and coordination of the technical measurements leading to certification were performed under the chairmanship of W. R. Shields.

The technical and support aspects in the preparation, certification and issuance of this standard reference material were coordinated through the Office of Standard Reference Materials by J. L. Hague.

Washington, D.C. 20234
July 30, 1970

J. Paul Cali, Acting Chief
Office of Standard Reference Materials
INFORMATION SHEET

SP.88007

1. Description of the samples
Six double-sided \( \frac{238}{234} \) U deposits (\( \equiv 76 \text{ mm} \)) on Ta backings.

2. Preparation method
Suspension spraying.

3. Chemical analyses
Probe 685.

4. Isotopic analyses
NBS 0002 (see annex).

5. Metrological information (*)

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>U Content (ug/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>413</td>
</tr>
<tr>
<td>2</td>
<td>407</td>
</tr>
<tr>
<td>3</td>
<td>453</td>
</tr>
<tr>
<td>4</td>
<td>490</td>
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<tr>
<td>5</td>
<td>468</td>
</tr>
<tr>
<td>6</td>
<td>422</td>
</tr>
</tbody>
</table>

\[ T = \frac{441.6}{100} \times 3 \times 10^{-3} \times U \text{ (cm²)} \]

6. Additional information
The samples were mounted in your sample holder and transported to the Van de Graaff on 6.5.88.

R. Eykens
Sample Preparation

(*) Determined by weighing (uncertainty: ± 10 %)
<table>
<thead>
<tr>
<th>Measurement</th>
<th>Baseline</th>
<th>Number of items</th>
<th>Material description</th>
<th>Weight of element</th>
<th>Unit</th>
<th>Weight of fissile material</th>
<th>Unit</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>T</td>
<td>BC00685H</td>
<td>6</td>
<td>SSPP</td>
<td>0.00024</td>
<td>K</td>
<td>A</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**REMARKS:**

**PRO FORMA Nr:** R08/24

**DATE OF DISPATCH:** 03/07/03

**NAME AND SIGNATURE FOR DISPATCH:** Andreas Fessler

**DATE OF ACCOUNTANCY:** 03/07/03

**EUPM, B-2440 GEEL, BELGIUM:**

(EURATOM) Regulation no 3227/75 – Annex II