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A High Resolution Neutron Spectrometry System for the 50 to 1500 keV Energy Region

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Abstract:

A high resolution spectrometry system, based on spherical SP2 hydrogen recoil proportional counters, has been successfully developed and tested. It has been carefully calibrated, and its response functions determined using monoenergetic neutrons measurements combined with calculations. The system covers the 50 to 1500 keV energy range where the dose equivalent per unit fluence varies markedly with neutron energy. Combined with the NE-213 scintillator system, it gives NPL a high resolution spectrometry capability for neutrons between 50 keV and 20 MeV, which is the region where the majority of the dose equivalent occurs in many workplace fields, and the high resolution capability complements the existing Bonner sphere system which covers the complete thermal to 20 MeV region, although with lower energy resolution.

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1. INTRODUCTION

Over recent years, it has increasingly been recognised that in order to improve estimates of neutron doses, there is a need to perform neutron spectrometry measurements in workplace fields where radiation workers are exposed to neutron doses. The need for such measurements stems from the fact that presently available neutron dosemeters, both area survey instruments and personal dosemeters, have inadequate dose equivalent responses as a function of energy. This deficiency in the performance of the dosemeters means that their readings can be significantly in error in fields with spectra which are different in shape from that of the neutron field used to calibrate the device. The calibration fields presently available are based on radionuclide sources, and are predominantly high-energy fields. Workplace fields, in contrast, often contain very significant numbers of low and intermediate energy neutrons, and systematic errors in dosemeter readings are to be expected. Without spectral information it is difficult to even begin to estimate the extent of these errors.

Spectrometry instruments have been developed at NPL over a number of years for use both in workplace environments and in the laboratory. The first of these was a Bonner sphere set(1-3). The main advantage of this system is that it covers the full range of neutron energies relevant to radiation protection. It does, however, have the disadvantage of poor energy resolution. Since the dose equivalent per unit fluence is a factor of about 30 times higher for high energy neutrons than for low and intermediate energy neutrons, a significant fraction of the dose in many fields occurs in the upper energy region. Also, the dose equivalent per unit fluence varies markedly with neutron energy between about 10 keV and 1 MeV so that the calculated dose equivalent can depend strongly on the exact shape of spectrum in this region. There is thus a much greater need for high resolution measurements in the upper energy region than elsewhere, and for this reason efforts have been made to develop high-resolution spectrometers to cover this range to augment the Bonner sphere data. An NE-213 scintillator system(4) was developed to cover the range above about 1 MeV, but this still left the important energy region just below 1 MeV. This report describes the selection, development, and characterisation of an appropriate high resolution spectrometry system to cover this region.

The desired characteristics of neutron spectrometers, particularly ones which will be used in workplace fields, are:

- coverage of a wide range of energies,
- an isotropic response,
- good energy resolution,
- an ability to discriminate between neutrons and photons,
- an ability to work reliably in the presence of high thermal and epi-thermal neutron fluences,
- reliability, ease of use, ease of transport,
- an ability to work under harsh environmental conditions, e.g. high noise and temperatures,
- the possibility of providing the results rapidly.

The first stage in this work was to investigate and identify a suitable high resolution instrument, i.e. one exhibiting most of the above characteristics, capable of measuring neutron spectra over the 50 keV to 1.5 MeV range. From earlier work it was realised that interference from the presence of high fluences of thermal and epithermal neutrons might well be a significant problem. The instrument chosen, and the reasons for the choice are described in the next section, while the development and characterisation of the instrument are described in the subsequent sections.

2. IDENTIFICATION OF A SUITABLE INSTRUMENT

2.1 Initial considerations

The requirement identified above is for a high resolution spectrometer to cover the energy range 50 keV to 1.5 MeV. The term "high resolution" needs to be quantified in some way. In deciding on a figure for the desired resolution some account must to be taken of what is physically achievable, and thus a realistic target value for the resolution cannot be finalised until all the instrumental options have been considered. Some broad guidelines as to what would be acceptable can, however, be stated.

The question of what constitutes adequate resolution is nearly impossible to answer. It depends on the spectral shape in the region of interest, and on the final dose equivalent accuracy required. In the majority of rather well-moderated spectra of interest very pronounced structures, e.g. pronounced peaks, are unlikely in the 50 keV to 1.5 MeV region. The accuracy needs for radiation protection are not particularly high. Certainly resolution of 30% or better would be a considerable improvement on anything achievable with Bonner spheres.

To be able to measure to a lower limit of 50 keV, the resolution at that energy must be better than 50 keV. However, an energy resolution of 50 keV at 1.5 MeV represents a percentage resolution of about 3%, which is probably much better than can be achieved. The resolution of the NE213 system, used in the region above 1 MeV, is about 30% at 1 MeV, although it improves at higher energies. On the basis of these considerations, the resolution to aim for with the new instrument should be in the of 10 to 30% range. It may be possible to do better at the upper end of this region, whereas it may not be possible to achieve anything as good as 30% at the lower end of the range.

The first step in identifying a suitable instrument was to investigate which spectrometers are available, and which ones other establishments have used for this energy range in the past. There is evidence of two types of device having been used for measurements of workplace spectra in Europe and the USA.

(a) Devices based on the detection of hydrogen recoil events (with the possible use of ^{4}He recoil devices to measure higher neutron energies).

(b) Devices based on the use of the ^{3}He(n,p)T reaction.

Before considering options (a) and (b) in more detail, all other instruments which can operate over the energy range of interest should be reviewed in case the use of any other type of device is feasible.

A Bonner sphere system of course covers the energy range of interest, but not with the required resolution. Since the response functions of Bonner spheres exhibit very broad maxima, it is very doubtful that any modifications, e.g. the use of more, or different material, spheres(5), will provide improvements in resolution for this system over the energy region from 50 keV to 1.5 MeV.

Another possibility is the use of threshold reactions to augment the Bonner sphere data. These, however, require fluence rates well above those relevant to neutron protection dosimetry level, and also do not provide the required resolution.

Semiconductor sandwich detectors based on either lithium or ^{3}He can be built and made to operate over the energy range of interest, but the devices are too complicated for field use and are also very inefficient as demonstrated by Marsh et al.(6).

A device which can detect neutrons over the relevant energy range is the lithium glass scintillator. There is no record of these devices being used as field instruments, and their poor light output and non-linear response would create severe problems(7).

Other devices which might be considered are Bubble, or Superheated Drop detectors (8-15). Both Apfel Entreprises (16) and Bubble Technology Industries(17) produce systems with spectrometric capabilities, but once again the attainable resolution is poor at present, and is unlikely ever to be good. Twenty years after their invention, Bubble detectors remain a challenging topic for research and development although they may yet provide the solution to the problems of finding a suitable dosemeter for practical applications.

We appear to be left with devices using either hydrogen (+ ^{4}He possibly) recoil, or the ^{3}He(n,p)T reaction. These detection mechanisms can be incorporated in different types of instruments, e.g. proportional counters or ion chambers, and these can be either spherical or cylindrical.

2.2 Devices Based on the ^{3}He(n,p)T Reaction.

Use of the ^{3}He(n,p)T reaction has certain attractions(18,19). The main one is that the pulse output spectrum from a spectrometer based on this reaction reflects the neutron spectrum more closely than the output from say a hydrogen recoil based device. This makes spectrum unfolding less prone to problems and uncertainty. In fact, until elastic scattering effects begin to be evident at about 1 MeV, the pulse output spectrum very closely resembles the neutron spectrum except for wall effects in the counter. A Q-value for the reaction of 764 keV means that even thermal neutrons have an energy of 764 keV in the pulse output spectrum so that discrimination against γ-radiation is relatively easy, except where there is significant γ-pile-up. Devices based on this reaction can definitely exhibit the required resolution, and do cover the relevant energy range(18,20).

Excluding ^{3}He sandwich devices, two types of ^{3}He based spectrometers are commercially available: ion chambers and proportional counters.

2.2.1 Ion Chambers

A number of papers (21-25) have been published on the ion chamber device of the Shalev and Cuttler design (20) available from 'Common Sense' (26). NPL acquired in 1993, as part of an extra-mural research project with the University of Birmingham(18), a Fast Neutron Spectrometer (FNS), based on a ^{3}He(n,p) "grid-ed" ionisation chamber of the Shalev-Cuttler type. On of its main strength remains its high energy resolution which can be upset by micro-phonics and mechanical vibrations. Two attempts have been made to use this spectrometer in simulated workplace environments without success as severe problems with pile-up made any analysis of the data impossible because pile-up of 764 keV events completely swamped everything. Not only is the 'double' pile-up at twice 764 keV very prominent, the pile-up extends to 3 x 764 keV, and possibly even higher. This was despite the fact that the instrument was completely enveloped in a cadmium sleeve of at least 1 mm thickness. The 764 keV events were thus not due to detection of incident thermal neutron, but were due to epi-cadmium neutrons detected either directly or after thermalisation within the counter.

In view of this problem it is very doubtful that this instrument could ever be useful for field measurements except perhaps in situations where there are very few neutrons below about 1 keV. It may be possible to reduce the effects of pile-up, and there are several options which might be tried:

(a) Use of boron or samarium rather than, or in addition to, cadmium to cut out more epi-thermal neutrons. This is probably not a complete solution and the spectrometer gets increasingly bulky and the neutron field is distorted by the additional absorbing sleeves.

(b) Use of pulse rise-time discrimination may be possible. The use of commercial pile-up rejection units could be considered, but this introduces problems of knowing which events the unit accepts or rejects with consequent problems of the stability of the efficiency calibration under different pile-up situations.

(c) "Stripping" off of the pile-up from the spectrum using a measured "pure" pile-up spectrum. This, if it were possible, would be very prone to uncertainties due to the fact that one is performing a subtraction process with two similar-sized large numbers.

In conclusion, there appears to be very little prospect of a working field spectrometer based on this instrument being successfully developed. It remains a very useful tool for laboratory spectrometry, such as for the measurement of neutron source spectra.

2.2.2 Proportional Counters

Endres et al.(27) in a general report on neutron dosimetry at nuclear plants published in 1981 reported measurements using a 2.5 cm (1") diameter (presumably cylindrical) counter. To reduce neutron pile-up they used a cadmium cover, and both pile-up rejection and pulse shape analysis. With this system they made measurements inside reactor containment and the report states that the spectrometer "seems to be operating properly".

A later report, from the same laboratory, in 1984, by Brackenbush et al.(28) concentrates on the characteristics of the ^{3}He spectrometer. It provides a great deal of relevant information about ^{3}He proportional counters and their use. While extolling the virtues of this spectrometer, the report, particularly in its recommendations section, repeatedly sounds warnings about the problems of pile-up. It says, for example, "pulse pile up problems can easily lead to errors in determining the spectrum between 20 keV and 700 keV".

They recommend the use of 1 to 3 mm of samarium, and say that a 1 cm thick boron carbide shield could also be used, but warn of the problems of scatter if these absorbers are present. They recommend keeping the ^{3}He pressure reasonably low at 2 to 4 atmospheres to reduce pile-up, (c.f. the 10 atmospheres and larger volume of the FNS(18,20,26) mentioned above), but this requires very long measurement times, e.g. overnight even in fields of 0.5 mSv h^{-1}. A technique of using a precision pulser is recommended to estimate pile-up, even after all the techniques to minimise it have been employed. They conclude that the spectrometer can be used in workplace environments, but note that the ^{3}He spectrometer indicates a "much larger contribution to dose equivalent than calculated by the multisphere spectrometer". The difference appears to be about 35%.

A third report, by Brackenbush et al.(29), expands a little on what is said in the 1984 report. Developments to the spectrometer, mainly in terms of the electronics, are described, and also some measurements in a plutonium processing area. Very much the same message is given, i.e., that it is a useful instrument for workplace spectrometry, but there is not a great deal of hard evidence.

In conclusion, ^{3}He proportional counter spectrometers appear to be an option, but in view of the problems with the FNS-1 and the comments about pile-up in the reports from the USA, great care should be taken before embarking on a lengthy development programme based on such an instrument.

2.3 Devices Based on Hydrogen or Helium (^{4}He) Recoil Detection

Proportional counters based on the detection of either hydrogen or helium recoils have been shown to work in the types of environment where field measurements need to be made. A set of such counters (usually 2 or 3) with different pressures, is required to cover the full energy range.

The first spherical hydrogen recoil proportional counter was suggested and developed by Benjamin, Kemshall and Redfearn (30,31) in 1964. The aim was to develop a counter having a response function which was independent of the direction of the incident neutrons. This was achieved by using a spherical cathode shell and a thin central anode wire and ensuring that the gas multiplication is reasonably constant within the "ionisation volume" by producing a uniform electric field within the counter. The counters have since been extensively researched, their response functions are now well understood and they have been widely used in neutron spectrometry (32-41). They have recently been successfully used in two intercomparison exercises: at the Silene reactor in Valduc (42) and at the Cadarache Moderator Assembly (43), both in France.

The transportable neutron spectrometer (TNS) from AEA Winfrith (UK) contains, amongst other instruments, hydrogen recoil counters. The device has been used successfully in environments where γ doses and thermal fluences are high(44). However, since the environments in which the measurements were made were 'military' it is hard to obtain any detailed information. The ROSPEC(45), produced by Ing in Canada, also uses hydrogen recoil counters, and has been used fairly extensively to measure 'field' spectra. Again, some of these are at military installations, but there is evidence that the device works. In both these systems the hydrogen recoil counter are clustered together in the "head" of the instrument and are all used at the same time. The degree of interference between these counters and the moderation produced in the field are not clear. Unfortunately, the TNS is no longer available, and the ROSPEC is very expensive.

For ^{4}He-filled proportional counters, one of the most recent studies of their use as neutron spectrometers, in the 144 keV to 14 MeV energy range, was carried out by Weyrauch et al.(46) who give an extensive bibliography of publications relevant to these counters.

2.4 Conclusion

In view of the above, a set of hydrogen recoil counters appears to be the best choice of instrument for measurements in the 50 keV to 1 MeV region. The most commonly used recoil counters are the spherical SP2 counters (Figure 1) made originally by AEA Technology, Winfrith. These are, however, no longer available from Winfrith, but it has, fortunately, been possible to acquire second hand SP2 counters from the National Radiological Protection Board (NRPB) and from the University of Birmingham.

The reminder of this report describes the development and calibration of a high resolution neutron spectrometry system based on 1, 3 and 9 atm hydrogen recoil counters of the SP2 type. In the SI system these pressures are about 0.1, 0.3 and 0.9 MPa; however, the values in atmospheres tend to be used as a matter of convenience. The three counters are referred to as SP2-1, SP2-3 and SP2-9 respectively throughout this work. Together they cover the important energy range (50 keV to 1.5 MeV) where the fluence to dose equivalent conversion factors are known to vary substantially. With the completion of this work, and in conjunction with the NE-213 and Bonner sphere counters, NPL possesses a means of measuring neutron spectra from thermal energies to 20 MeV with high energy resolution in the neutron region above about 50 keV.

3. THE HYDROGEN RECOIL BASED SPECTROMETRY SYSTEM

3.1 Introduction

The SP2 hydrogen recoil counters to be characterised, were obtained unfortunately, with very little information about their filling parameters. Initial tests were therefore carried out using radioactive sources to confirm that the counters were working and viable. This was followed by extensive measurements with monoenergetic neutron produced by the NPL 3.5 MV Van de Graaff accelerator using the ^{7}Li(p,n) and T(p,n) reactions. These measurements were used in conjunction with calculations of the response function and subsequent unfolded spectra to determine the characteristics of the counters, e.g., the filling pressure, the gas composition and the resolution parameters, etc., and hence their response functions. In this work, the response functions of the hydrogen recoil counters were calculated using a FORTRAN program, RESP, which is part of the SPHERE(47) computer programme package developed at PTB. RESP is based on the important work of Weise et al.(38), and takes account, not only of the wall effects, i.e., truncation of the recoiling hydrogen tracks by the counter walls, but also the variations in the gas amplification along the counter wire due to the non-uniformity of the electric field. The unfolding is carried out using three different programs. They are called GRAVEL, MIEKE and SPECAN and were developed by Matzke at PTB as part of the "so-called" HEPRO(48,49) computer programme package of unfolding codes. GRAVEL is a modified SAND-II program whereas MIEKE and SPECAN are Monte Carlo unfolding codes.

3.2 The Operating Principles of Hydrogen Recoil Counters

The SP2 counters consist essentially of a spherical stainless steel shell, about 4 cm diameter, acting as a cathode, and a central tungsten anode wire. For the 3 and 10 atm counters, the filling is about 90% hydrogen, the reminding 10% being methane which acts as a quenching gas, together with traces of ^{3}He used for energy calibration. The 1 atm counter filling is 100

% hydrogen, and it has about a 1 mm long deposit ^{239}Pu, an α-particle emitter, on its anode wire to act as an internal energy calibrator.

When a neutron is incident on such a hydrogen recoil counter, various processes can occur. These are listed below and illustrated in Figure 2.

- The incident neutron, if scattered by a hydrogen nucleus, produces a proton recoil.
- The electrons produced along the proton track drift towards the anode wire along an electric field line.
- Near the wire, the electron gains enough energy for the gas atoms to be ionised.
- The ionisation electrons can themselves produce further electrons and thus an avalanche (gas amplification) is produced.
- The ions are collected at the anode and a signal is produced.
- The signal is proportional to the amount of ionisation and thus to the energy of the recoiling proton (not of the incident neutron).
- A range of proton energies is obtained for each neutron energy depending on the angle between the neutron and the proton.

3.3 Advantages of Hydrogen Recoil Counters

The counters use the (n,p) scattering cross-section, which:

- is well known and changes monotonically with energy,
- is isotropic for neutron energies less than 10 MeV,
- can, because of the near equality of neutron and proton masses, be described by simple scattering theory so that response calculations are relatively straightforward,
- has a reasonable large absolute value so that the counters have a useable efficiency although measurements of several hours are still required in radiation protection applications.

Other attributes are

- high energy resolution,
- an isotropic response because the SP2 counters are spherical,
- an ability to work in high thermal and epithermal fields,
- they are tried and tested counters and the expertise in their use is available,
- they cover the 50-1500 keV range where fluence to dose equivalent factors vary rapidly with energy.

3.4 Disadvantages

Some of the disadvantages associated with these counters are:

- since the counters have a thin anode wire, they are highly micro-phonic; this problem may only be solved by enclosing counters in a firm, as thick as tolerable, metal box.
- three counters have to be used in succession and thus longer measuring times are required.

3.5 Dynamic Range

The lower energy limit is set by γ-ray induced pulses, and by the deterioration in resolution (a 1 keV proton yields only 30 ionisations), whereas the upper limit is set by wall effect distortions, which depend on the size of the counter and the stopping power of the filling gases since the proton range increases with increasing energy. A set of counters with different gas filling parameters (mixture content and pressure) and/or γ-ray discrimination techniques, must be used to cover the range from 50 keV to 1500 keV.

3.6 The Response Functions of Hydrogen Recoil Counters

3.6.1 The Ideal Response Function and Distortion Effects

For neutron energies below about 2 MeV, and for a spherical hydrogen recoil counter with 0.1 to 1 MPa pressure, the neutron mean free path is about 1 meter which is very much larger than the diameter of the counter. Thus, the counter is almost transparent to neutrons, the predominant reaction process being at most a single elastic scattering event of the incident neutrons by protons.

Since the scattering cross-section is isotropic in the centre of mass frame, it follows(7) that for an incident monoenergetic neutron fluence of energy E, the expected proton recoil energy distribution, $P(E)$, would ideally have the well known characteristic rectangular shape of Figure 3.

Provided all proton recoils are stopped within the counter volume, and that the ionisation is proportional to the energy E_p of the protons, the proton recoil spectrum $P(E)$ can be related to the neutron energy spectrum $\phi(E)$ by the equation(50).

$$\phi(E) = -\frac{E}{NV\sigma(E)}\frac{dP(E)}{dE}$$

where $\phi(E)$ is the fluence per unit energy at energy E, N is the number of protons per unit volume in the gas, V is the gas volume and σ is the hydrogen cross section. With this notation $P(E)\,dE$ would represent the number of recoil protons caused by a fluence $\phi(E)\,dE$ of neutrons which have an energy between E and $E+dE$.

Thus the neutron flux $\phi(E)$ would, in principle be obtained by simply differentiating $P(E)$, if it could only be measured.

This measurement is complicated by the distortion effects which result from the fact that, although W-values are independent of proton energy above a few keV(34,51), the number of ions collected at the anode does not give a measure of the proton recoil spectrum because :

- all recoil protons do not lose their entire energy within the counter before hitting the wall: wall distortion effects
- gas amplification is not constant over the entire volume as the electric field strength drops at the ends of the anode wire: gas amplification distortion effects

3.6.. The Real Response Function

The above effects distort the ideal response function. Allowing for these effects, the real response function can be written(38) as :

$$R(E) = \int d^3r \int d\Omega \int d(E') \,.\, T(E'.E' - E, r, \Omega)$$

where :

r is the n - p interaction point,
Ω is solid angle into which the proton is scattered,
E is the proton energy when produced,
E' is the proton energy when recorded,
and T is the transition function written as:

$$T(E', E' - E, r, \Omega) = \int dE'' V(E'.E' - E, r, \Omega)\ \ W(E'.E' - E'', r, \Omega)$$

where V is the gas amplification and W the wall effect amplitude, and $\int R(E)dE = 1$.
The wall and gas amplifications effects are calculated, and corrected for, following the methods outlined by Weise et al.(38), and the real response functions are obtained. In essence the Townsend theory for gas amplification is used(52) and the method of "image" electric charges is applied to the calculation of electric fields, which has the shape shown in Figure 4.

From the path length probability functions and the data of proton range as a function of energy, the wall effect $W(E'.E' - E'', r, \Omega)$ is calculated analytically following Snidow and Warren(53). The reader is directed to the paper by Weise et al.(38) for a full discussion of these various aspects of the response functions which are calculated in this work by the program RESP of the SHERE package.

The real response to a monoenergetic neutron field departs from the ideal rectangular shape not only due to the various distortion effects mentioned above, but also due to resolution, noise and γ-ray contamination effects as well. A "real" pulse height distribution spectrum, measured with the 3 atm counter for an incident energy of 565 keV is shown in Figure 5, and is compared to a spectrum calculated without any resolution broadening.

3.7 The Electronics System

A schematic diagram of the electronics system is shown in Figure 6. It is essentially a simple, single parameter arrangement, based on commercially available units: an Ortec 213 charge sensitive preamplifier, an Ortec 571 spectroscopy amplifier and a Canberra ADC which is part of the FAST dual-parameter data acquisition system used at NPL. The same electronics modules are used for all counters except that the 3.7 kV power is supplied to the 10 atm counter by a 'home made' unit capable of providing voltages greater than 3 kV without breakdown

4. ENERGY CALIBRATION

4.1 Internal Reference Sources

The 1 atm (SP2-1) counter has a small ^{239}Pu alpha particle emitter on the centre of its anode wire. The mean energy of the measured pulse height distribution of the alpha peak, although somewhat broad, corresponds to a fixed energy deposition in the counter. This is unaffected by the incident neutron field and is independent of changes in the gain or electronic amplification. It has a further advantage that the peak sits above the operating range (50-250 keV) of the counter and thus can be subtracted as background. The energy of the alpha particles is such that their range is much greater than the counter radius and only a small fraction (about 380 keV) of the energy is deposited in the counter. Although the measured Full Width Half Maximum (FWHM) of the peak is used to indicate whether the counter is operating well, it does not represent the intrinsic resolution of the counter. A pulse height distribution spectrum of the internal source, i.e., with no other source of irradiation, is shown in Figure 7. Some disadvantages of using internal calibration sources are discussed by Knauf and Wittstock(33).

The 3 and 9 atmosphere counters, which operate in the energy range 250 to 700 keV, and 600 to 1500 keV respectively, contain, in addition to hydrogen and methane, a trace of ^{3}He. Neutrons interact with the ^{3}He via the ^{3}He(n,p)T reaction which has a cross section with an essentially 1/v energy dependence where v is the neutron velocity. Since the cross-section for this reaction increases rapidly as the neutron energy decreases, the majority of reactions occur with low energy (≤1eV) neutrons. An energy equal to the reaction Q value (764 keV), is then deposited within the counter, with three quarters of the energy taken by the protons and the rest by the tritons. The reaction cross for thermal neutrons is in fact so large that contamination of the proton spectrum may occur in the presence of high thermal fluences, and must be subtracted before unfolding. This is possible since the thermal response of both counters have been determined at NPL using the thermal pile facility. In fact, the correction needs to be applied only for the 9 atmosphere counter since the thermal peak energy is beyond the range of operation of the 3 atm counter. The effective Q value of the reaction for these counters, i.e., the energy at which the thermal peak appears, is found to be about 780 to790 keV, not 764 keV. This is consistent with the findings of Brearly(34) who suggested that this might be due to the difference in the specific ionization of the protons and tritons involved.

To produce an intense source of thermal neutrons for testing the counters, a block, with dimensions, 30*20*20 cm, was constructed of high-density polyethylene. Two holes separated by about 8 cm of polyethylene house the SP2 counter and an ^{241}Am-Be source in an X3 capsule, respectively. The source and detector distance is reproducible within a couple of mm. This arrangement is used for instance to test and check that the counters are operating satisfactory, to set-up and adjust the electronics, and amplifier gains before and after measurements by looking at the peak channel position and its FWHM, which is similar to the intrinsic resolution of the counters for protons. Figure 8 shows a spectrum recorded by the 3 atm (SF2-3) counter within this "thermal block" arrangement.

4.2 Energy Calibration using Monoenergetic Neutrons

An energy calibration for the system was obtained using monoenergetic neutrons of well known energy, and the back bias of the system was determined by use of an Ortec 448 pulse generator. The channel number corresponding to the incident neutron energy is set (within ± one channel) equal to the channel number at which the pulse height is equal to one half the pulse height corresponding to the "knee" of the pulse height spectrum, as indicated in Figure 5. The energy calibrations obtained from the monoenergetic measurements carried out on separate occasions are shown, without any correction for shifts in the overall gain, in Figure 9 (a) to (c) for the three counters respectively; they suggest that the beam energy calibration and the detection system are stable and reasonably reproducible.

5. MEASUREMENT OF THE RESPONSE FUNCTIONS

5.1 Introduction

The response functions of the three SP2 counters were measured using monoenergetic neutrons generated by the NPL 3.5 MeV Van de Graaff accelerator. The ^{7}Li(p,n)^{7}Be and T(p,n)^{3}He reactions were used to cover the 50 to 1500 keV neutron energy range of interest. The experimental set-up is shown in Figure 10. The corrections for room scatter were performed using the shadow cone technique (54-55). The total neutron fluence to which the counters were subjected was determined using our calibrated long counters (54). Measurements and calculations were combined using a two complementary methods to determine the response function of the counters.

In the first method, use is made of the fact that the distributions calculated from the product of the integrated neutron fluence measured by the long counter and the resolution broadened response function should superimpose the measured multi-channel spectrum if the response function is correct. This follows from the matrix equation $Z = R\phi$ where Z is the measured pulse height distribution, R is the response function matrix and ϕ is the incident neutron spectrum. The response function for a monoenergetic spectrum, calculated using the programs RESP and GAUS from the PTB SPHERE package were accordingly optimised by adjusting the counter parameters such as the gas mixture, pressure, resolution parameters, etc. Counter parameters, and thus response functions, were considered optimum when calculations and measurements agreed well at a few monoenergetic neutron energies.

The resolution broadening of the response function, carried out by the program GAUS, assumes that the FWHM, as a function of energy E is given by:

$$FWHM = \sqrt{a + bE + cE^2}$$

where a,b and c are constants which are obtained by comparing measurement to calculation, as described above.

The form of the equation is based on the assumptions that:

- The constant parameter, a, accounts for noise contributions.

- The linear parameter, b, accounts for statistical variations due to fluctuations in the number of primary ion pairs and in the gas multiplication.
- The quadratic term, c, accounts for various effects such as those due to differences between the real field and the model field along the wire, impurities and instrumental effects e.g. instability in amplifier gains and high voltage. The non-uniformity of the field along the anode wire is taken into account by the model filed in the response function calculations.

The broadening due to target thickness was calculated and stripped off in quadrature. Once the first method, the quickest, has been applied and the counter parameters have been tuned, the full response matrix was calculated using RESP and was resolution broadened using GAUS program. The monoenergetic spectra were then unfolded and the resulting fluence was compared to the fluence measured with the long counter (method 2). To be able to correct for any downscatter neutrons, the response matrices were calculated for neutron energies beyond the dynamic range of each counter.

5.2 Calibration of the 1 atm. SP2 Counter (SP2-1)

The 1 atmosphere counter, which covers the energy range of about 50 to 250 keV, contains 100% hydrogen, and has a small ^{239}Pu α particle emitter source on its anode wire acting as an internal energy reference (see section 4.1). The counter was calibrated using monoenergetic neutrons of energy 70, 144 and 250 keV. Details of the electronics settings and of the measurements are summarised in the Appendix. A plot of channel number versus energy, as measured with a precision pulse generator, is shown in Figure 11, demonstrating the excellent linearity of the acquisition system. A pulse height distribution spectrum measured with the SP2-1 counter for 144 keV incident neutron energy is shown in Figure 12. It is compared to the calculated spectrum which results from the calculated resolution broadened response function times the total neutron fluence measured by the long counter (method 1 in section 5.1). The optimum FWHM parameters obtained as described above are given in Table 1 and a plot of the FWHM function against neutron energy is shown in Figure 13.

Table 1: Parameters used to calculate the response function of the 1 atm SP2 counter

Hydrogen content (%)	100
Methane content (%)	0
^{3}Helium content (%)	0
Filling pressure (MPa)	0.106
Operational voltage (V)	1300
Temperature at filling (°C)	25.5
Resolution parameters	A=(16.66 ± 0.21) B= (0.0910 ± 0.0039) C= (0.00502 ± 0.00001)

Examples of monoenergetic neutron spectra unfolded using the programs GRAVEL , MIEKE and SPECAN are shown in Figures 14 (a) to (c). The integral of the unfolded neutron spectra are compared to the fluence measured by the long counter (method 2 in section 5.1) to further test the response functions. The fluences measured with the long counter and with the SP2 counter agreed remarkably well for all three monoenergies as shown in Table 2. The percentage uncertainties quoted are statistical only and assume that the response functions are exactly known. It must also be noted that the uncertainties given by GRAVEL which is mainly used to prepare for MIEKE and SPECAN runs, does not incorporate a robust variance calculation algorithm.

Table 2: Neutron fluence measured by the 1 atm SP2 counter and unfolded by GRAVEL, MIEKE and SPECAN compared to long counter measurements. All fluences have been divided by 10^6 for convenience, and the uncertainties quoted are percentage values.

Energy	PLC fluence	GRAVEL	GRA/PLC %	MIEKE	MIE/PLC %	SPECAN	SPE/PLC %
70 keV	5.394 ± 2.0%	5.253 ± 1.4%	-2.60	5.400 ± 1.1%	0.10	5.353 ± 1.2%	-0.78
144 keV	6.693 ± 2.0%	6.639 ± 3.4%	-0.80	6.730 ± 1.8%	0.56	6.688± 0.48%	-0.07
250 keV	5.668 ± 2.0%	5.666 ±12.1%	-0.03	5.710 ±0.57%	0.74	5.687 ± 0.53%	0.33

Various parameters of the response functions were altered and methods 1 and 2 (section 5.1) were reiterated until the best possible agreement was obtained for all energies and for the same response function parameters. At the end of the calibration exercise, the parameters to be used for the 1 atm SP2 hydrogen recoil counter were obtained and are also given in Table 1 above.

5.3 Calibration of the 3 atm. SP2 Counter (SP2-3)

The 3 atmosphere counter (SP2-3) which covers the energy range of about 150 to 700 keV, contains hydrogen (about 90%), methane and a trace of ^{3}He acting as an internal energy reference (see section 4.1). The calibration exercise is similar for all counters except for the neutron energies. This counter has been calibrated using monoenergetic neutrons of energy 144, 250 and 565 keV. Details of electronics set-up and of the measurements are summarised in Appendix, and an example of a pulse height distribution spectrum measured with the SP2-3 counter for a 565 keV monoenergetic incident neutron field is shown in Figure 15. It is compared to the calculated spectrum which results from the calculated response function times the total neutron fluence measured by the long counter (method 1 in section 5.1). The optimum FWHM parameters obtained by comparing measurements to calculations are given in Table 3 and a plot of the FWHM against energy is shown in Figure16.

Table 3: Response function parameters of the 3 atm SP2 counter

Hydrogen content (%)	90
Methane content (%)	10
^{3}Helium content (%)	traces
Filling pressure (MPa)	0.3016
Operational voltage (V)	2400
Temperature at filling (°C)	22
Resolution parameters	A=(66.83 ± 0.24) B= (0.296 ± 0.002) C= (0.0050 ± 0.0001)

Examples of unfolded 565 keV monoenergetic neutron spectra are shown in Figures 17 (a) to (c). The integral of the unfolded neutron spectra are compared to the fluence measured by the long counter (method 2 in section 5.1) to further test the response functions. Very good agreement was obtained, as shown in Table 4, for the 144 and 565 keV measurements. The agreement is not so good at 250 keV, due to the fact that the target used was slightly damaged during the measurement. The percentage uncertainties quoted are statistical only and assume that the response functions are exactly known. Similarly to the 1 atm counter, various parameters of the response functions were altered and methods 1 and 2 (section 5.1) were reiterated until the best possible agreement was are obtained for all energies and for the same response function parameters. The optimum parameters obtained for the 3 atm SP2 hydrogen recoil counter are given in Table 3.

Table 4: Neutron fluence measured by the 3 atm SP2 counter and unfolded by GRAVEL, MIEKE and SPECAN and compared to long counter measurements. All fluences have been divided by 10^6 for convenience, and the uncertainties quoted are percentage values.

Energy	PLC fluence	GRAVEL	GRA/PLC %	MIEKE	MIE/PLC %	SPECAN	SPE/PLC %
144 keV	2.894±2.0%	2.887 ±3.9%	-0.24	2.929±0.57%	0.90	2.889±1.1%	-0.20
250 keV	2.711±2.0%	2.574±2.9%	-5.00	2.618±0.66%	-3.40	2.624±0.56%	-3.22
565 keV	11.296±2.0%	11.305±6.7%	0.06	11.335±0.33%	0.35	11.297±0.25%	0.01

5.4 Calibration of the 9 atm. SP2 Counter (SP2-9)

The 10 atmosphere counter (SP2-9), which is intended to cover the energy range of about 500 to 1500 keV, contains hydrogen (about 90%), methane and a trace of ^{3}He acting as an internal energy reference (see section 4.1). The calibration exercise is similar for the other counters except for the neutron energies. This counter has been calibrated using monoenergetic neutrons of energy 250, 565, 1200 and 1500 keV. Details of electronics set-up are summarised in the Appendix, and examples of pulse height distribution spectra measured with the SP2-9 counter for 250, 1200 and 1500 keV incident neutron energy are shown in Figures 18 (a) to (c). These are compared to the calculated spectrum which results from the calculated response function times the total neutron fluence measured by the long counter (method 1 in section 5.1). The optimum FWHM parameters obtained by comparing measurements to calculations are given in Table 5 and a plot of the FWHM against neutron energy is shown in Figure 19.

Table 5: Response function parameters of the 9 atm SP2 counter

Hydrogen content (%)	90
Methane content (%)	10
^{3}Helium content (%)	traces
Filling pressure (MPa)	0.9
Operational voltage (V)	3700
Temperature at filling (°C)	22
Resolution parameters	A= (917.8 ± 130.6) B= (5.04 ± 0.81) C= (0.009 ± 0.001)

Examples of unfolded monoenergetic neutron are shown in Figures 20 (a) to (c). The integral of the unfolded neutron spectra are compared to the fluence measured by the long counter (method 2 in section 5.1) to test the response functions. As shown in Table 6, the agreement is good except perhaps at 250 keV, which is just below the dynamic range of the 9 atm counter.

Table 6: Neutron fluence measured by the 9 atm SP2 counter and unfolded by GRAVEL, MIEKE and-SPECAN and compared to long counter measurements. All fluences have been divided by 10^6 for convenience, and the uncertainties quoted are percentage values.

Energy	FLC fluence	GRAVEL	GRA/FLC %	MIEKE	MIE/FLC %	SPECAN	SPE/FLC %
250 keV	1.459±2.0%	1.638 ±2.3%	12.30	1.516±2.3%	3.90	1.494±2.0%	2.40
565 keV	3.759±2.0%	3.648±3.2%	-2.90	3.709±0.52%	-1.30	3.660±1.3%	-2.66
1200 keV	1.886±2.0%	1.884±1.0%	-0.12	1.903±1.4%	0.85	1.892±7.1%	0.31
1500 keV	6.412±2.0%	6.421 ±8.0%	0.14	6.471 ±0.77%	0.92	6.420±0.54%	0.13

The percentage uncertainties quoted are statistical only and assume that the response functions are exactly known. Similarly to the other counters, various parameters of the response functions were altered and methods 1 and 2 (section 5.1) were reiterated until the best possible agreement were obtained for all energies and for the same response function parameters. The optimum parameters to be used for the 9 atm SP2 hydrogen recoil counter were obtained and are given in Table 5 .

5.5 Response to Thermal Neutrons

The response of the 3 and 9 atmosphere SP2 counters to thermal neutrons was determined by irradiating the counters with the calibrated NPL thermal pile assembly. The difference method was used: this consists of measuring the response with and without a 1 mm thick cadmium sleeve covering the SP2 counters in order to estimate and correct for the response of the counter to fast neutrons in the predominantly thermal beam. The response to thermal neutrons is obtained by subtracting the two spectra. An example is shown in Figure 21, for the 3 atmosphere counter.

To correct a measured spectrum for the thermal response of the counter, the number of counts in the full (p+t) peak is calculated after subtraction of the continuum of the measured spectrum. The thermal neutron response is then subtracted from the measured spectrum after normalisation to give the same number of counts in the (p+t) peak(33).

6. TEST MEASUREMENTS

6.1 The ^{241}Am-Li Source Measurement

A test measurement has been carried out using an ^{241}Am-Li(α,n) radionuclide neutron source in the NPL low-scatter Main Bay. The experimental arrangement is shown in Figure 22. Source and counters were positioned 8 cm from each other and measurement were made for 3 to 5 days in succession for each counter separately. The ^{241}Am-Li spectrum maximum energy is within the dynamic range of the 9 atm (SP2-9) counter and thus was directly unfolded. The spectra of the 1 and 3 atm counters, however, are contaminated by the so called "down scattering", i.e., pulses from neutrons with incident energy greater than the operating range of the counter. Thus the procedure used is the following:

1. The higher energy pulse height distribution measured with the 9 atm counter (SP2-9) is unfolded which results in a neutron spectrum (labeled AMLIC) covering the energy range from about 600 to 1500 keV - Segment 1.

2. The latter spectrum is folded with the response function of the 3 atm counter (SP2-3) to calculate the contribution to the pulse height distribution, measured by SP2-3, of neutrons with energy beyond its dynamic range. The folding is done using either a special option within RESP of the SPHERE package or the program FALT of the HEPRO package.

3. The "downscatter" is subtracted.

4. Unfolding is carried out to yield the neutron spectrum in the energy range of about 250 to 600 keV, corrected for down scattering (labeled AMLIB) - Segment 2.

5. The procedure is repeated for the 1 atm counter to yield a neutron spectrum (AMLIA) in the energy range 50 to 250 keV that is corrected for downscattering - Segment 3.

Normalisation factors were of course applied to account for the duration of measurements which were different from one counter to the other. Figure 23 shows the full AMLI spectrum when all three segments are added together.

7. SUMMARY AND FUTURE DEVELOPMENTS

The SP2 hydrogen recoil counters have been calibrated and characterised and their response functions accurately determined. Thus, a high resolution spectrometry system has been developed to cover the 50-1500 keV range which gives NPL a high energy resolution spectrometry capability for 50 keV to 20 MeV neutrons. Future developments will involve better integration of the system with the NE213 and Bonner sphere systems (which can be used for thermal to 20 MeV neutrons although with lower resolution) and possibly extending the lower energy limit down to few tens of keV using γ-discrimination techniques.

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Appendix: The electronics settings for the three SP2 counters

1 atm SP2 Counter

Amplifier		ADC		Power Supply		Pulser		Alpha peak	
Coarse gain Fine gain Shaping time	50 6 6μs	LLD PeakDetect Gain/Range Offset Mode	0.2 Auto 256,256 0 PHA Anti-Coin	Voltage (V)	1300	Dials Norm. Peak Channel	0-3-0-00 8 237	Channel FWHM (keV)	196 45

3 atm. SP2 Counter

Amplifier		ADC		Power Supply		Pulser		Thermal peak	
Coarse gain Fine gain Shaping time	20 5 6μs	LLD PeakDetect Gain/Range Offset Mode	0.2 Auto 256,256 0 PHA Anti-Coin	Voltage (V)	2400	Dials Norm. Peak Channel	0-4-0-00 10 245	Channel FWHM (keV)	116 105

9 atm SP2 Counter

Amplifier		ADC		Power Supply		Pulser		Thermal peak	
Coarse gain Fine gain Shaping time	20 10.44 6μs	LLD PeakDetect Gain/Range Offset Mode	0.5 Auto 256,256 0 PHA Anti-Coin	Voltage (V)	3700	Dials Norm. Peak Channel	0-9-0-00 8 241	Channel FWHM (keV)	212 58

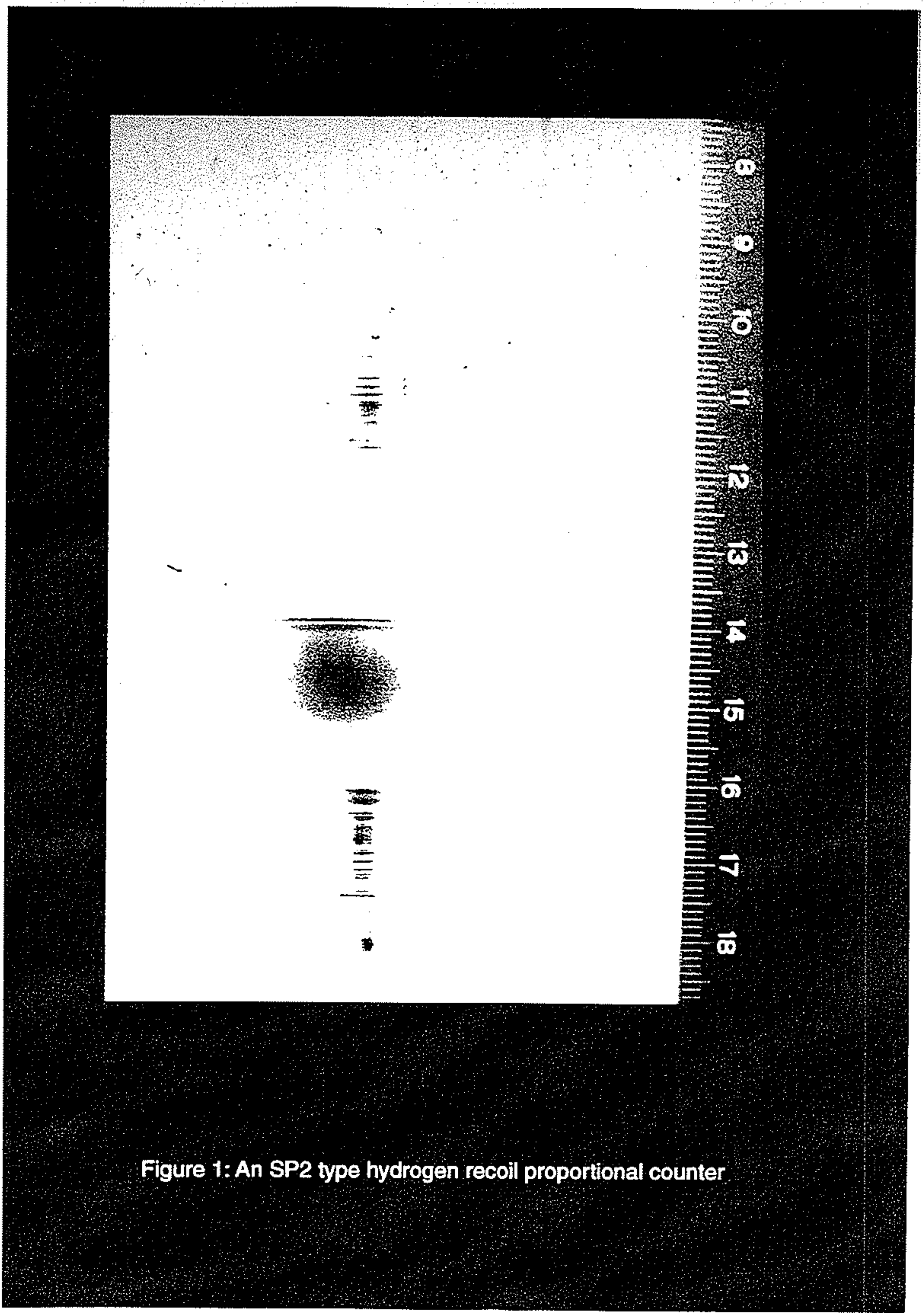


Figure 1: An SP2 type hydrogen recoil proportional counter

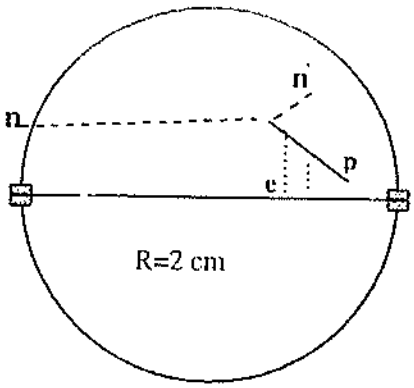
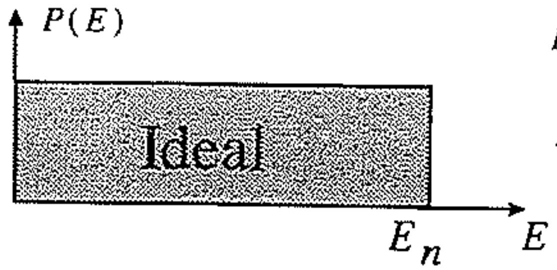


Figure 2: Schematic diagram for a spherical hydrogen recoil counter and the processes which occur in it when a neutron scatters off a hydrogen nucleus.



$$P(E) = \frac{1}{E_n} \text{ if } E \leq E_n$$

$$P(E) = 0 \text{ if } E < E_n$$

Figure 3: The ideal response function of a recoil counter for monoenergetic neutrons.

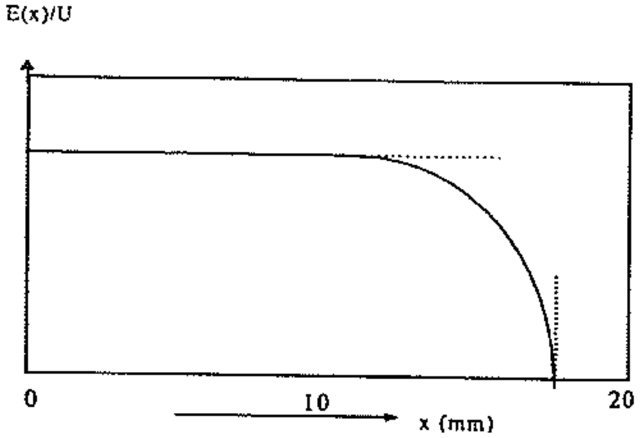


Figure 4: The electric filed strength E(x) on the anode wire surface divided by the voltage U between anode wire and cathode sphere; x is the distance from the equatorial plane of the counter. From Weise et al [38].

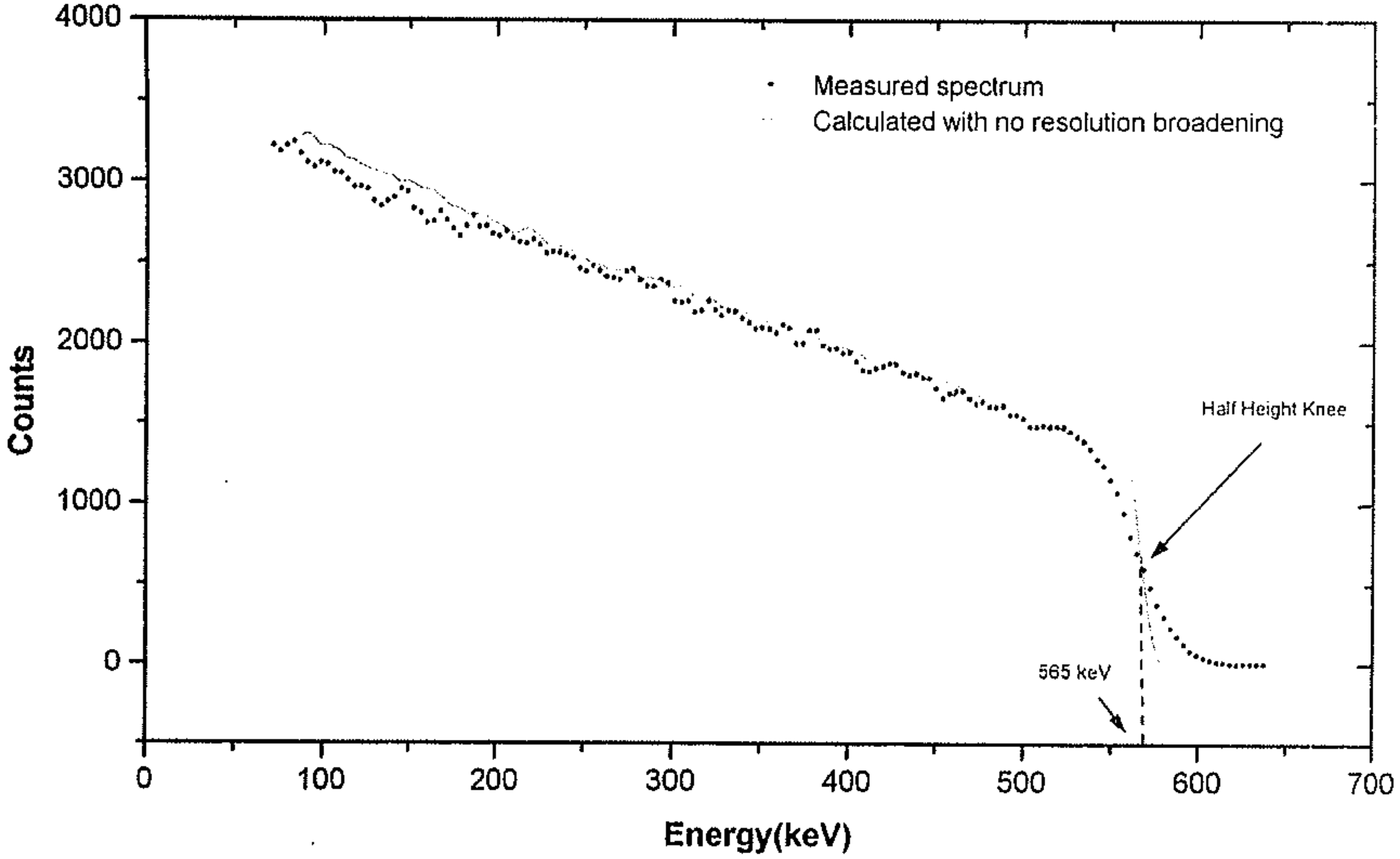


Figure 5: Proton recoil spectrum for a 565 keV monoenergetic fluence measured with the 3 atm SP2 counter and compared to a calculation with no resolution broadening.

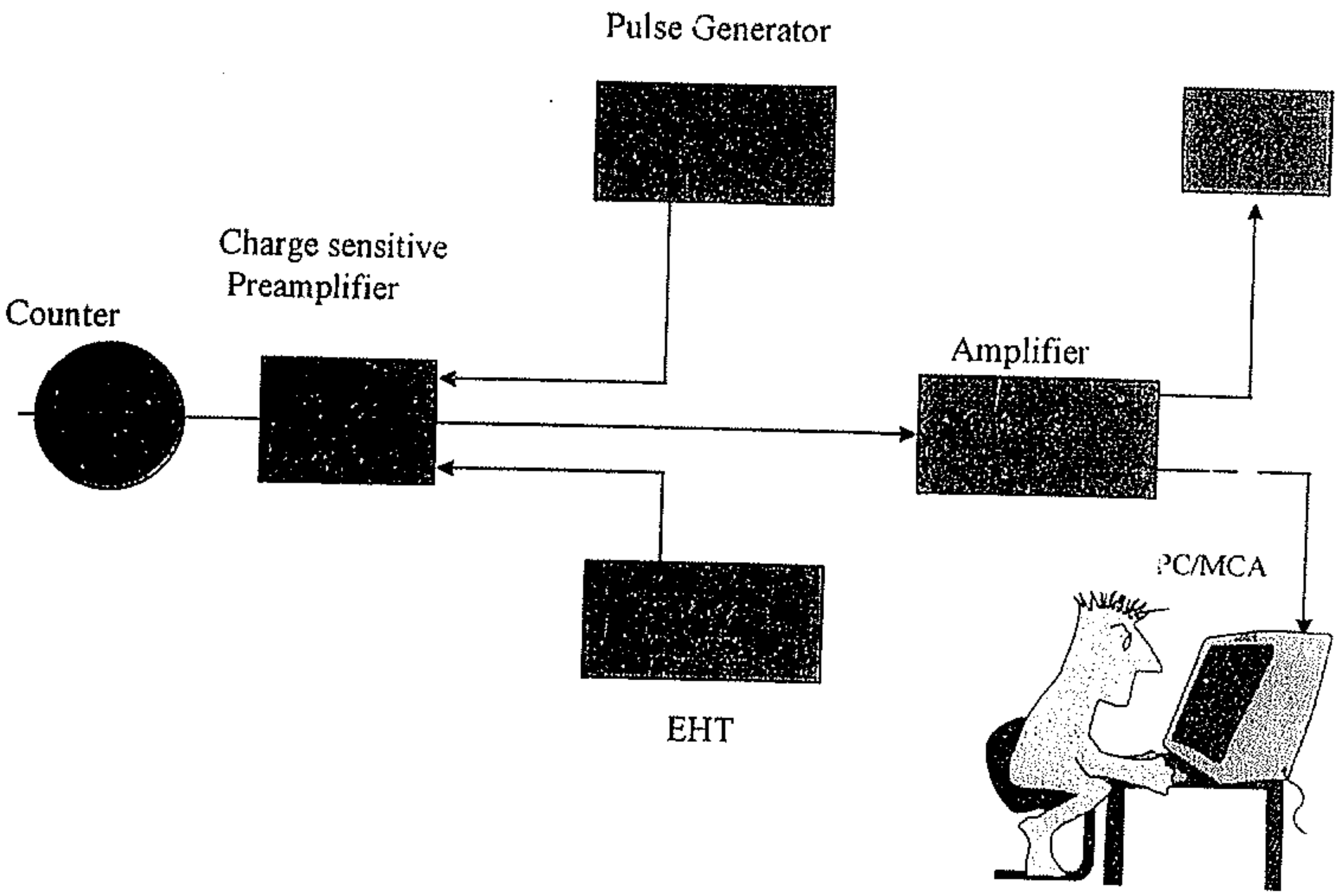


Figure 6: Schematic diagram of the electronic and data acquisition setup

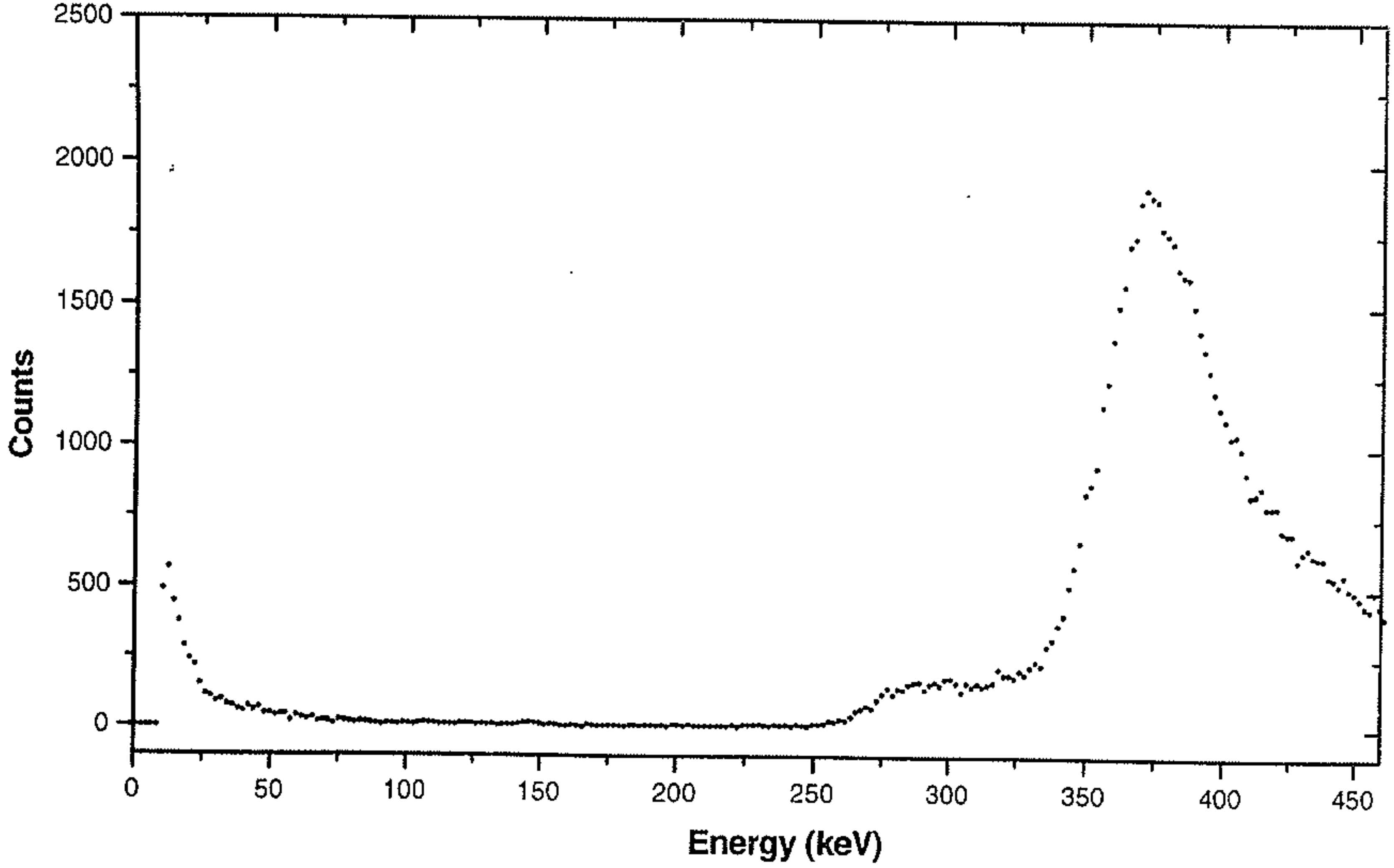


Figure 7: Spectrum of the ^{239}Pu internal source measured overnight with the 1 atm. SP2 counter

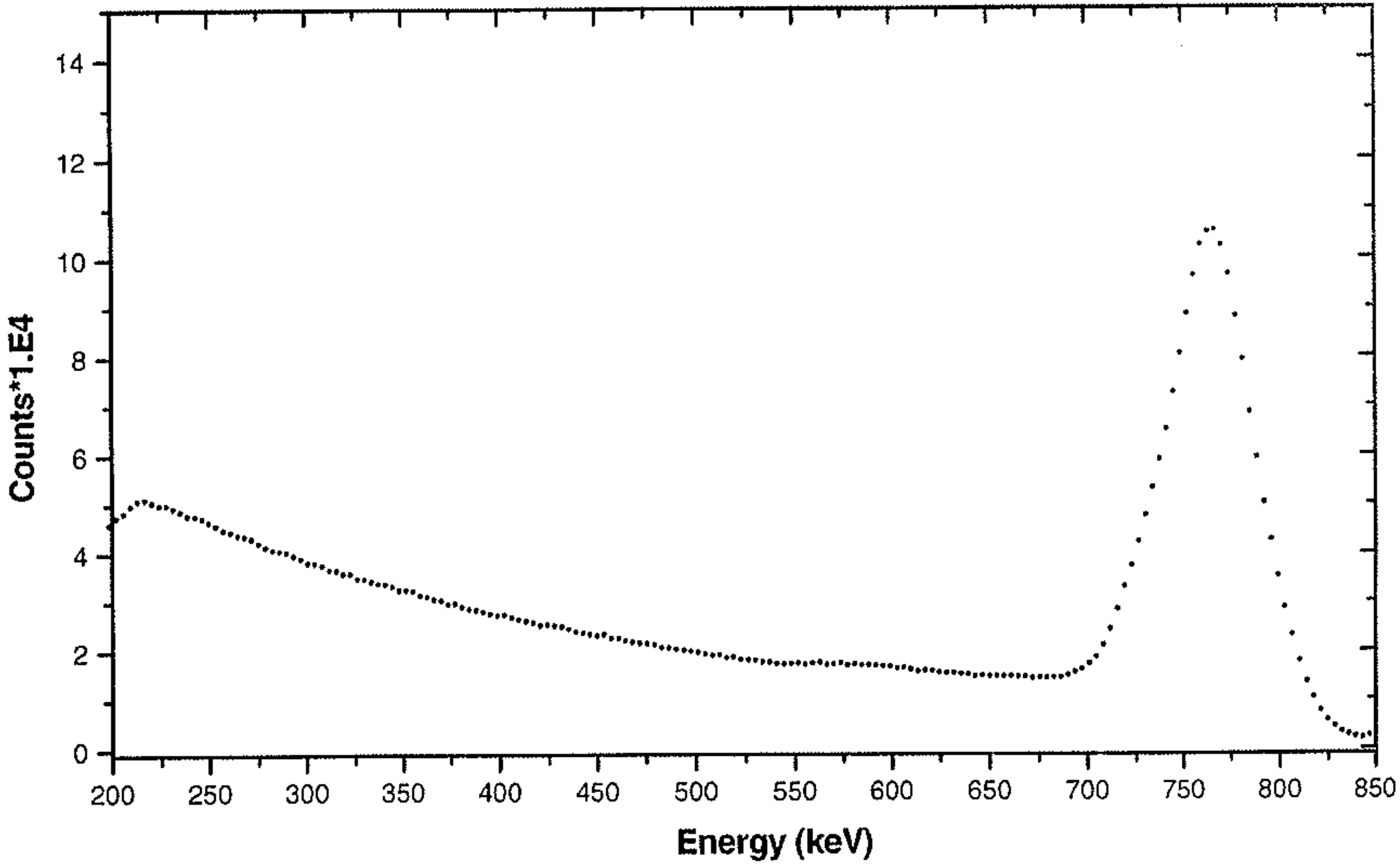


Figure 8: Spectrum of thermalised neutrons from an ^{241}Am-Be source measured with the 3 atm. SP2 counter

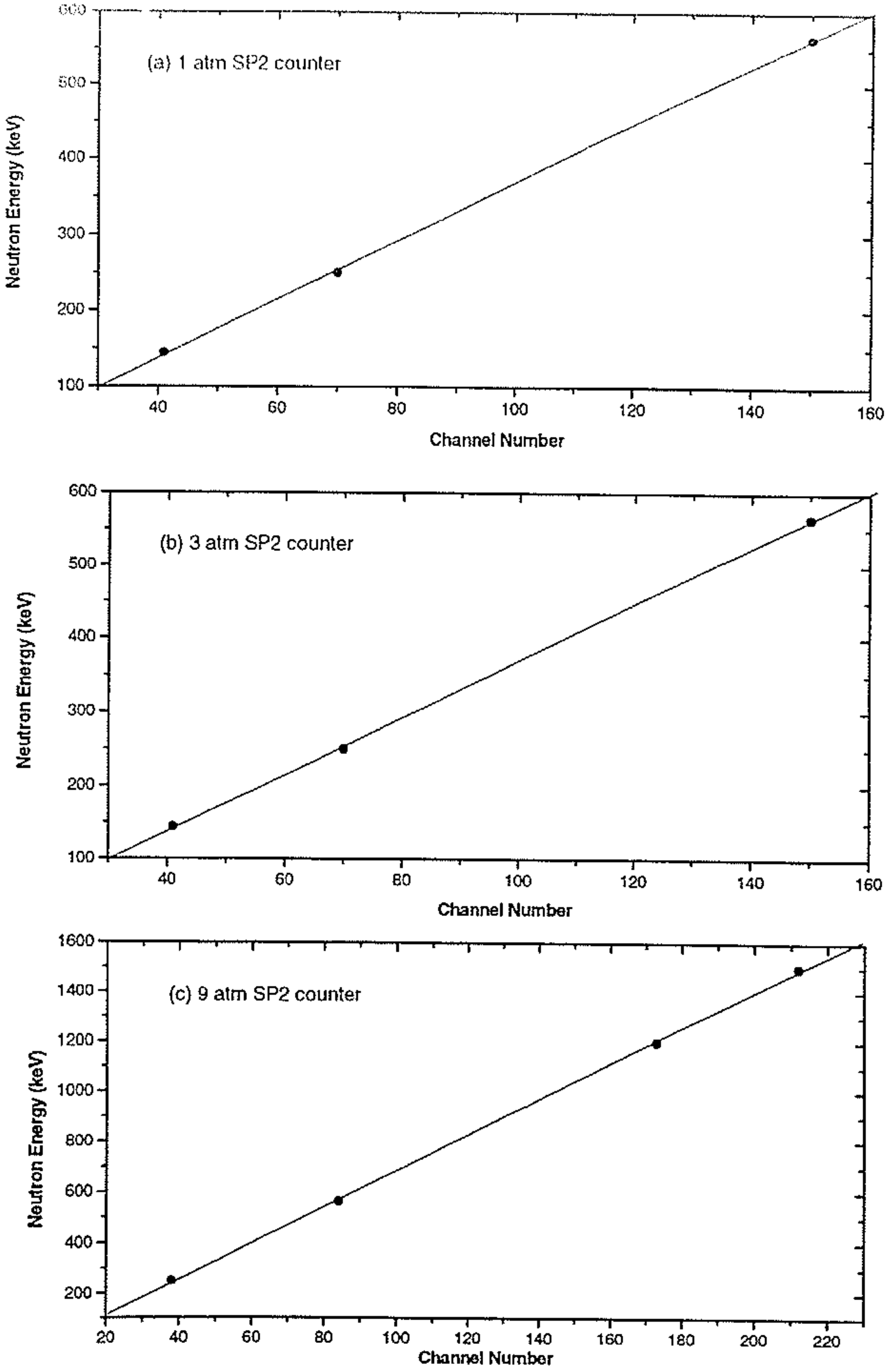


Figure 9: Energy calibration of SP2 counter from monoenergetic measurements made on separate days.

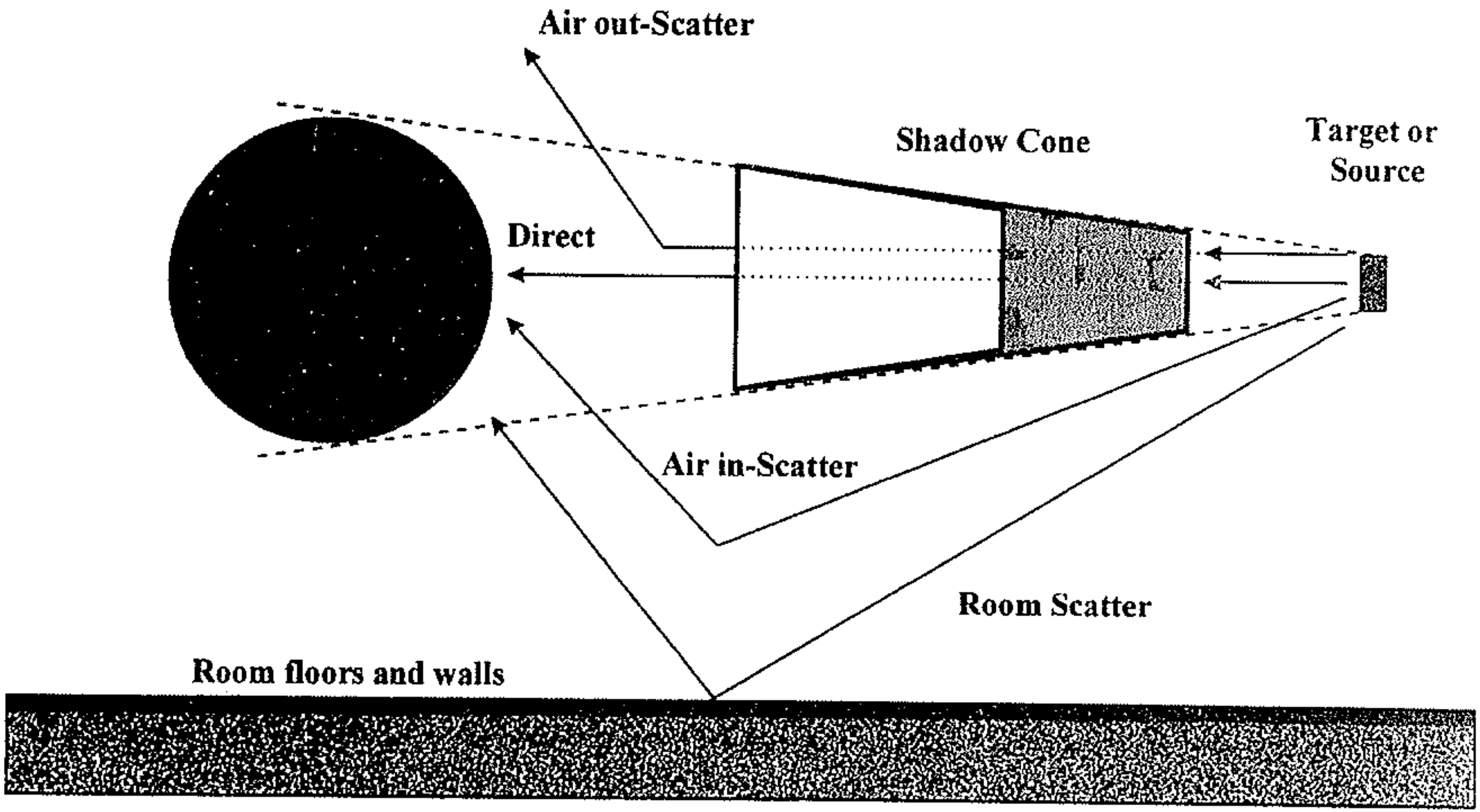


Figure 10: Experimental arrangement and scatter components (not to scale)

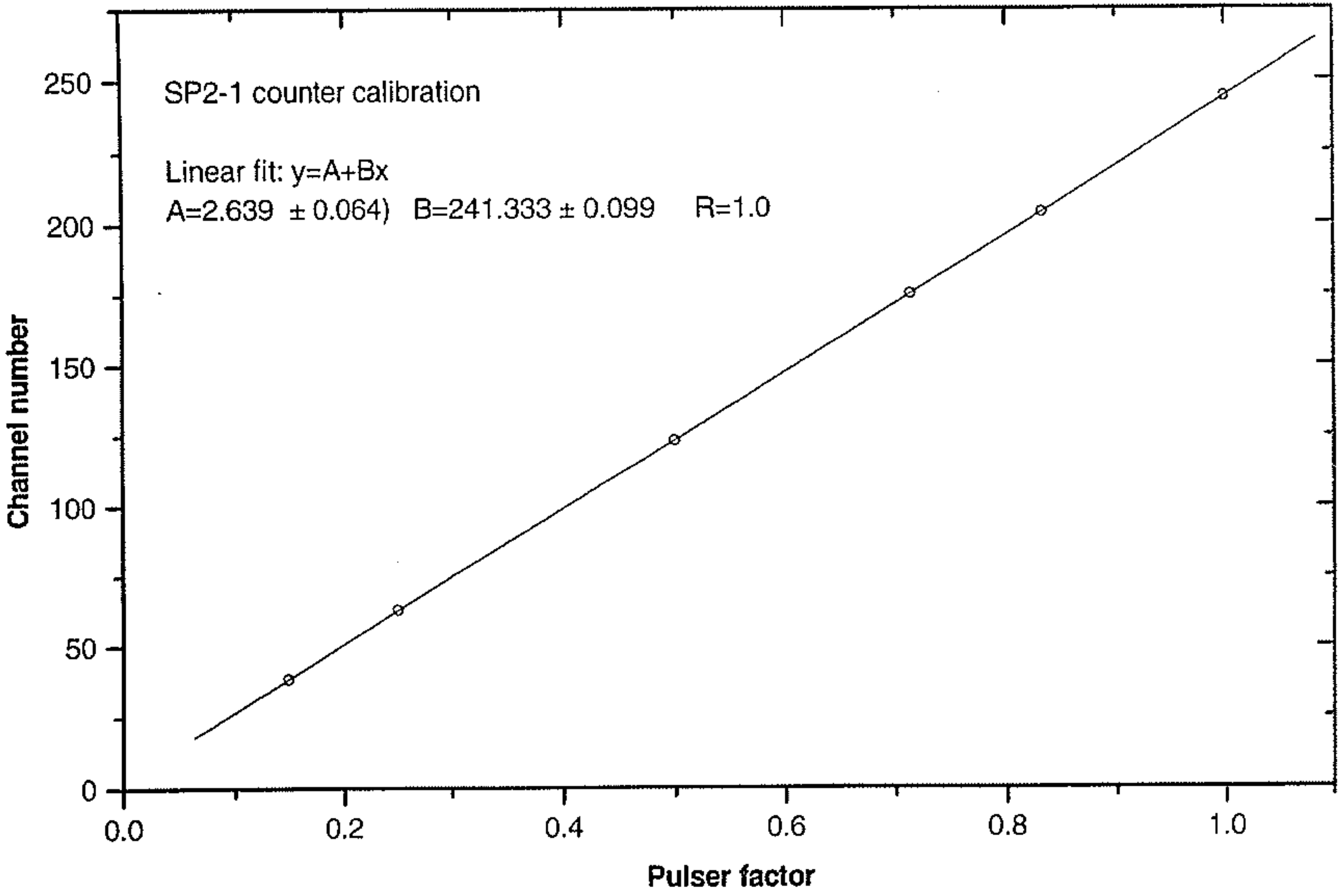


Figure 11: Pulser calibration for the 1 atm SP2 counter

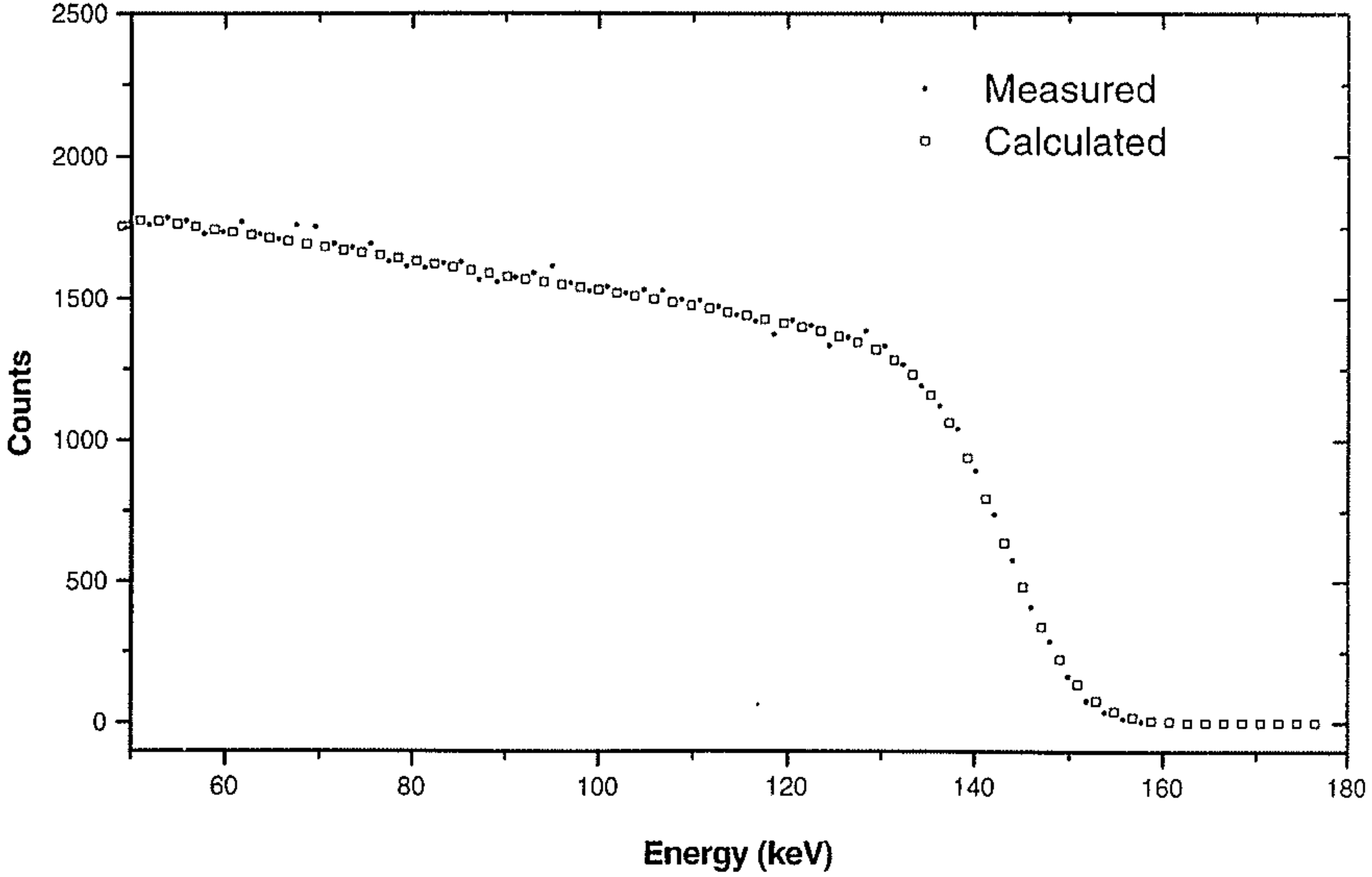


Figure 12: A 144 keV monoenergetic spectrum measured with the 1 atm SP2 counter and compared to calculation

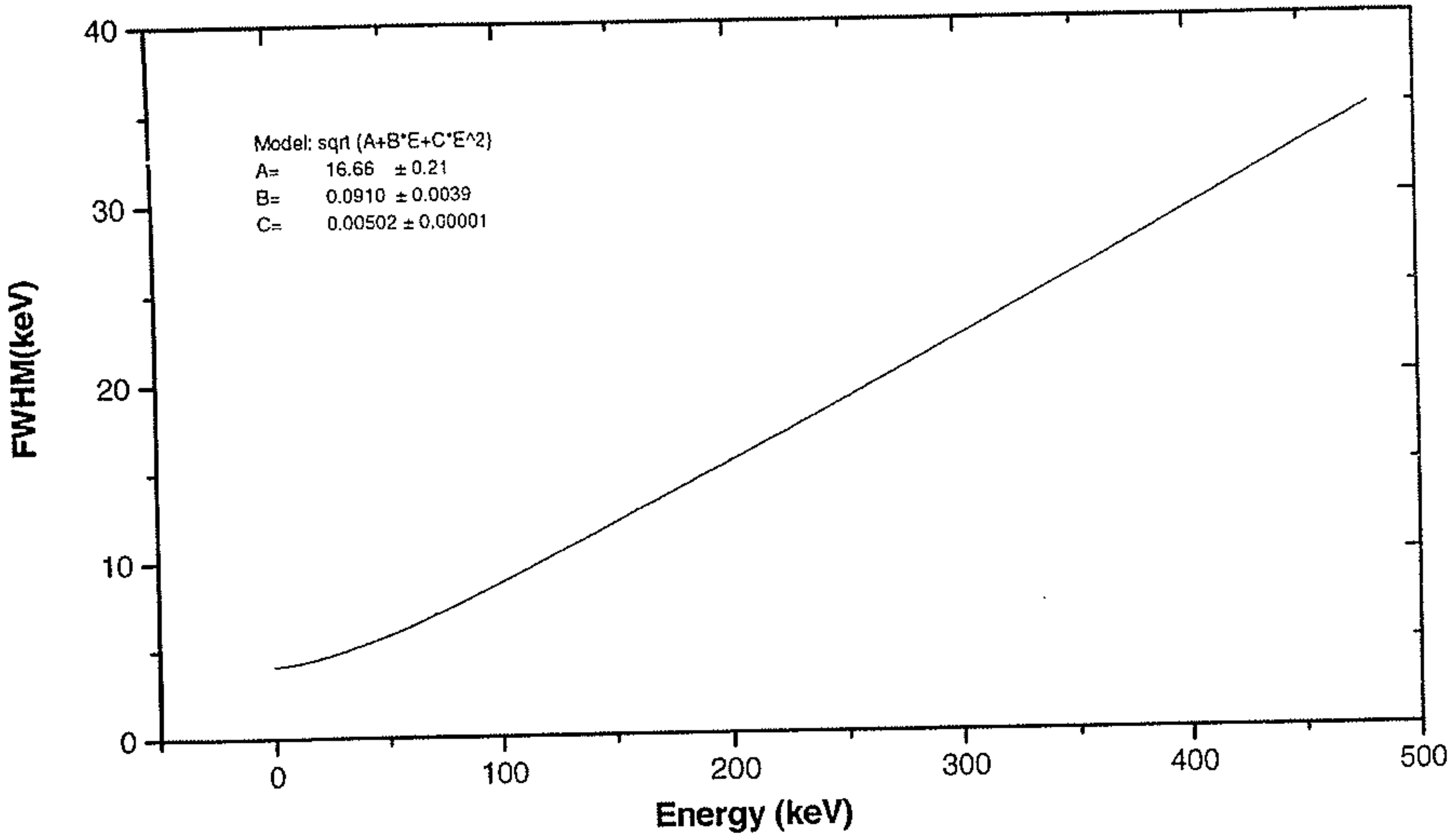


Figure 13: FWHM of the resolution function for the 1 atm SP2 counter as a function of energy

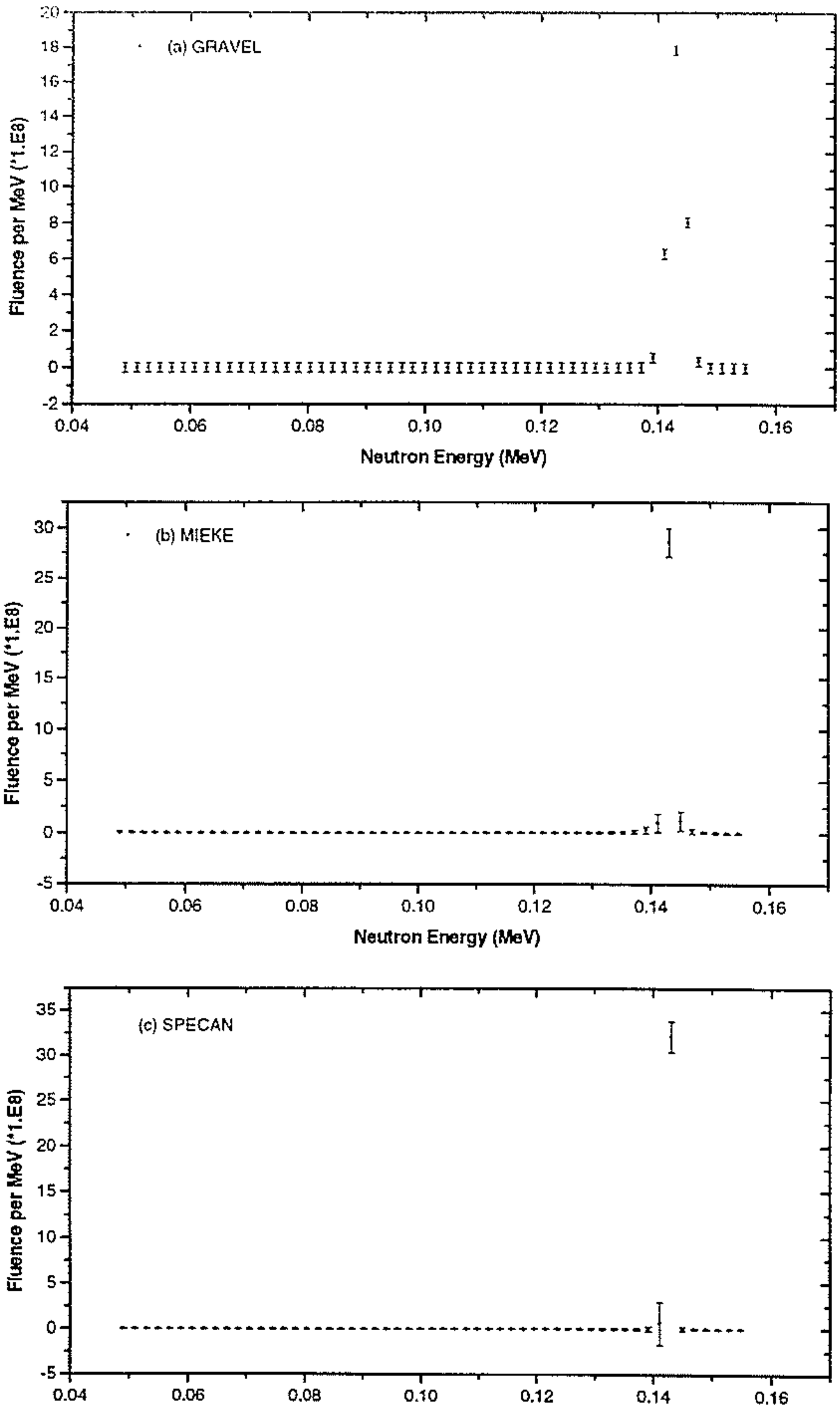


Figure 14: The 144 keV monoenergetic spectrum measured with the 1 atm SP2 counter and unfolded using: (a) GRAVEL, (b) MIEKE and (c) SPECAN

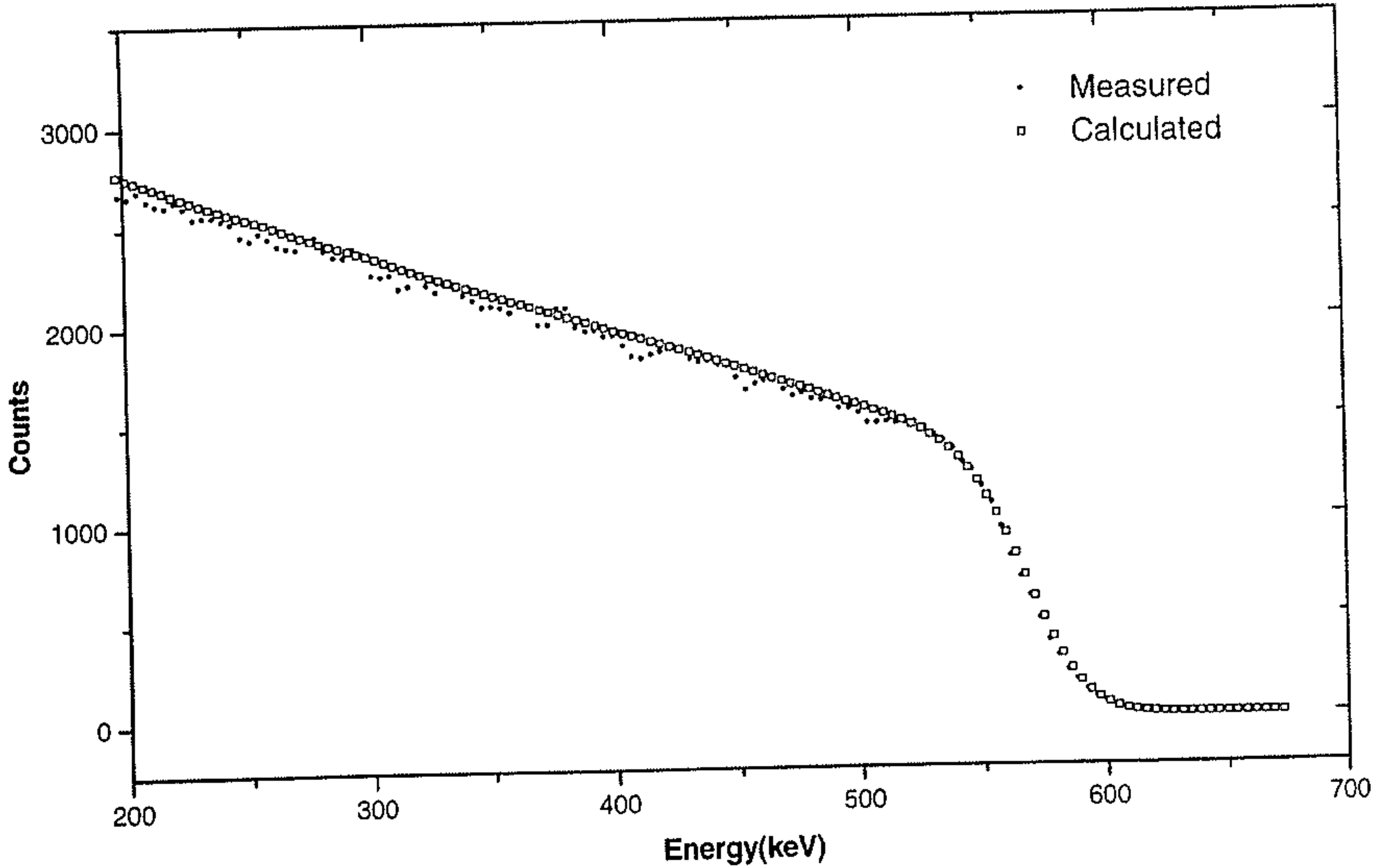


Figure 15: A 565 keV monoenergetic spectrum measured with the 3 atm SP2 counter and compared to calculation.

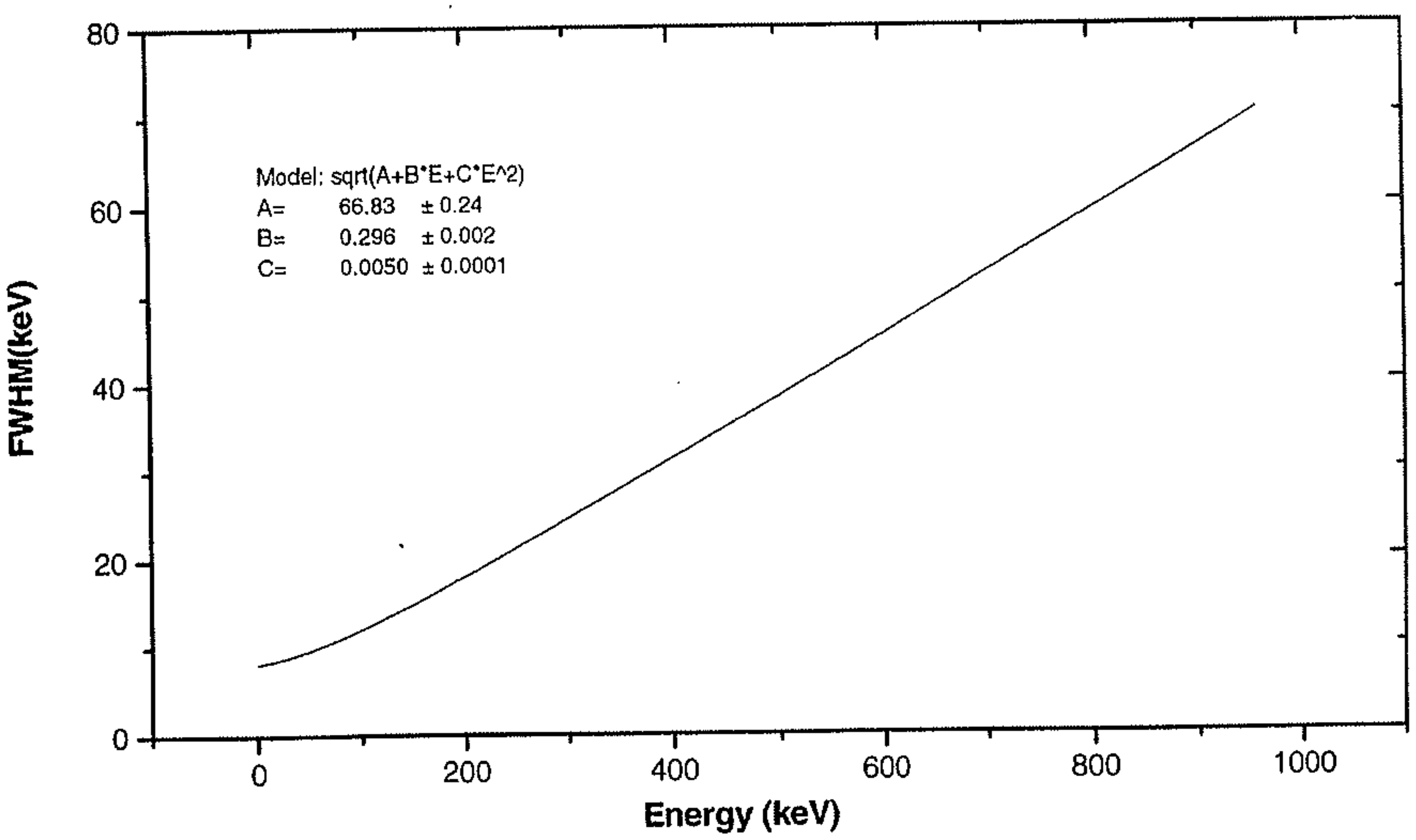


Figure 16: FWHM of the resolution function for the 3 atm SP2 counter as a function of energy

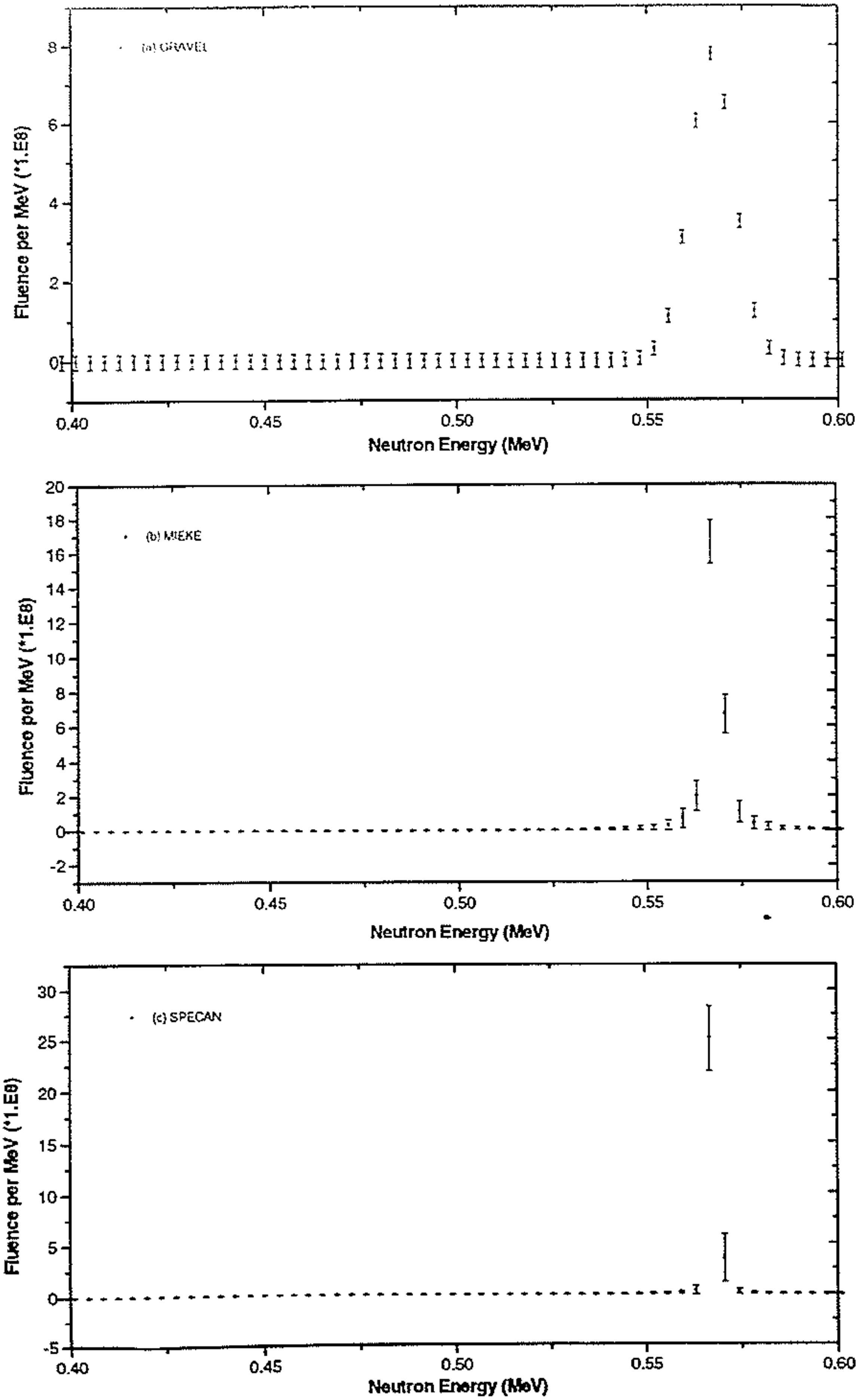


Figure 17: The 565 keV monoenergetic spectrum measured with the 3 atm SP2 counter and unfolded using: (a) GRAVEL, (b) MIEKE and (c) SPECAN

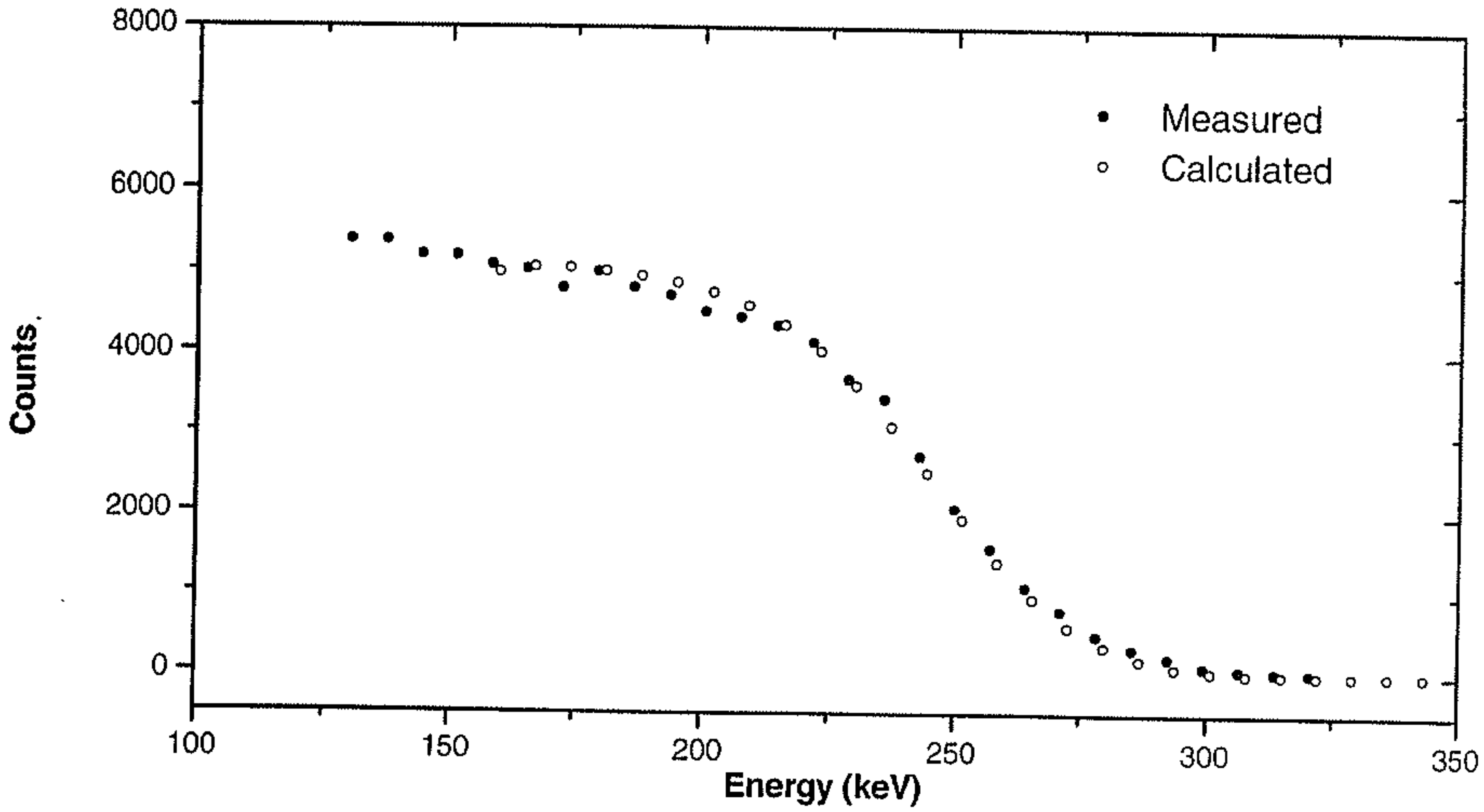


Figure 18 (a): A 250 keV monoenergetic spectrum measured with the 9 atm SP2 counter and compared to calculation. It shows that a spectrum as low in energy as 250 keV can be reasonably measured with the counter.

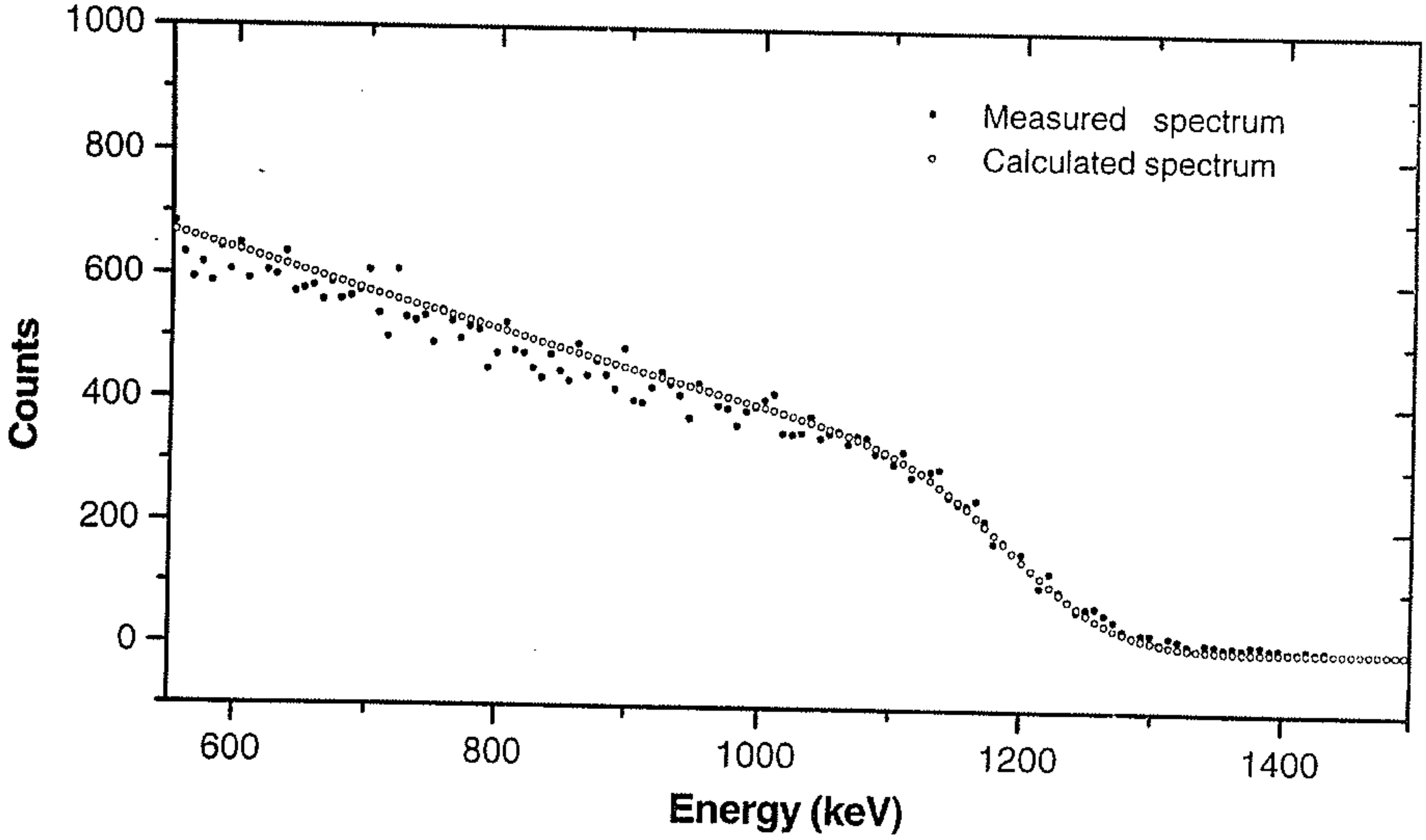


Figure 18 (b): A 1.2 MeV monoenergetic spectrum measured with the 9 atm SP2 counter and compared to calculation.

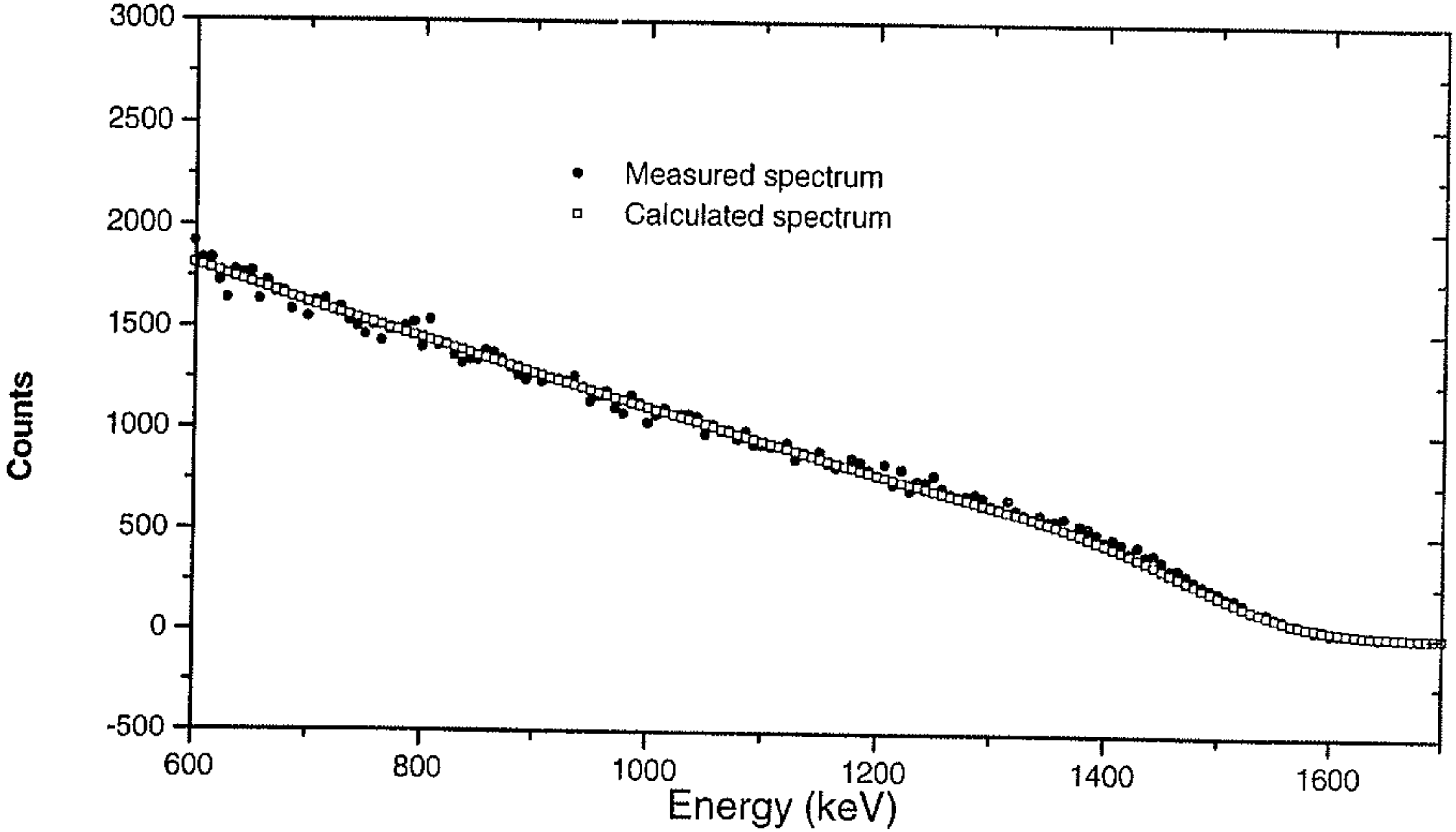


Figure 18 (c): A 1.5 MeV monoenergetic spectrum measured with the 9 atm SP2 counter and compared to calculation.

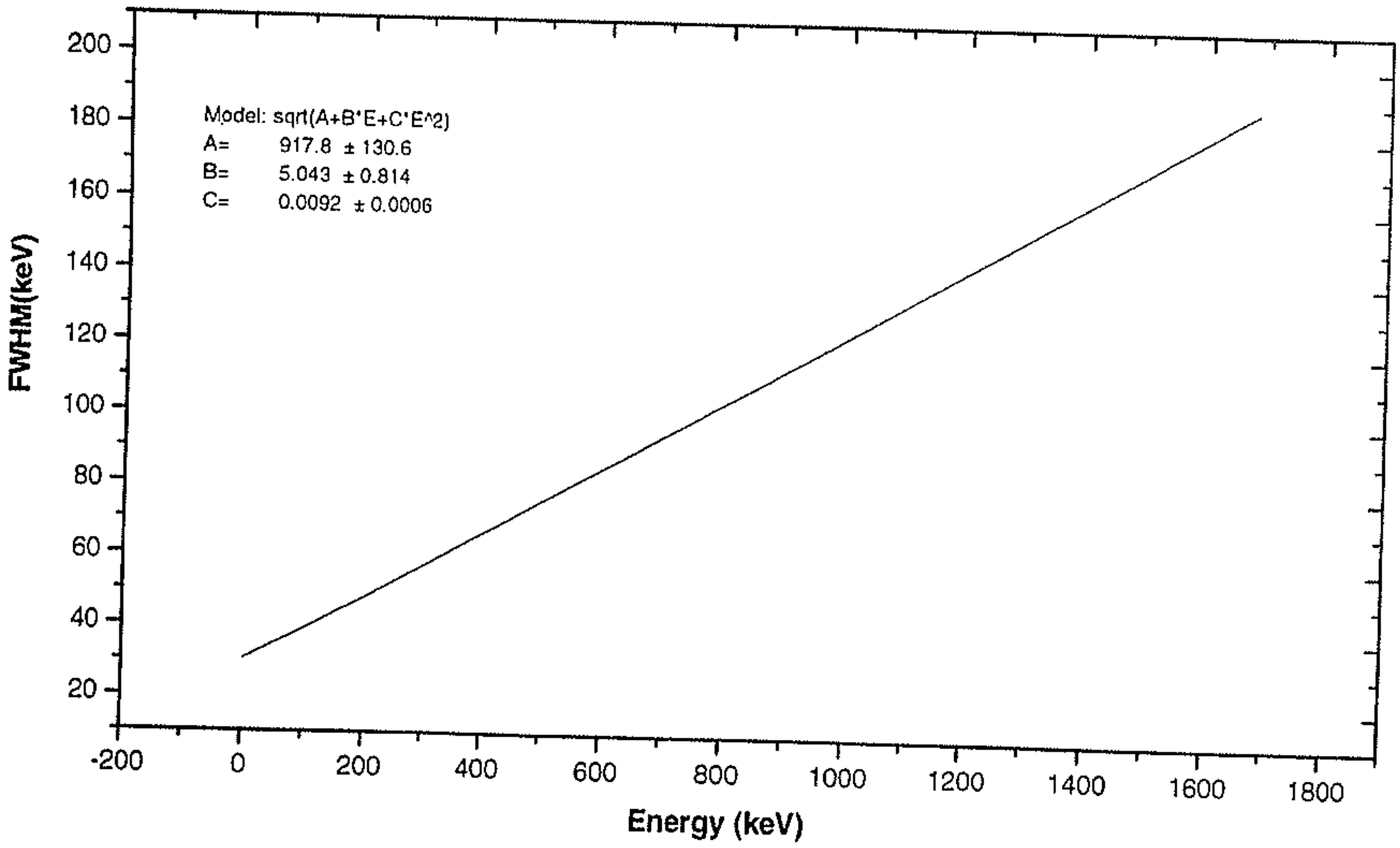


Figure 19: FWHM of the resolution function for the 9 atm SP2 counter as a function of energy

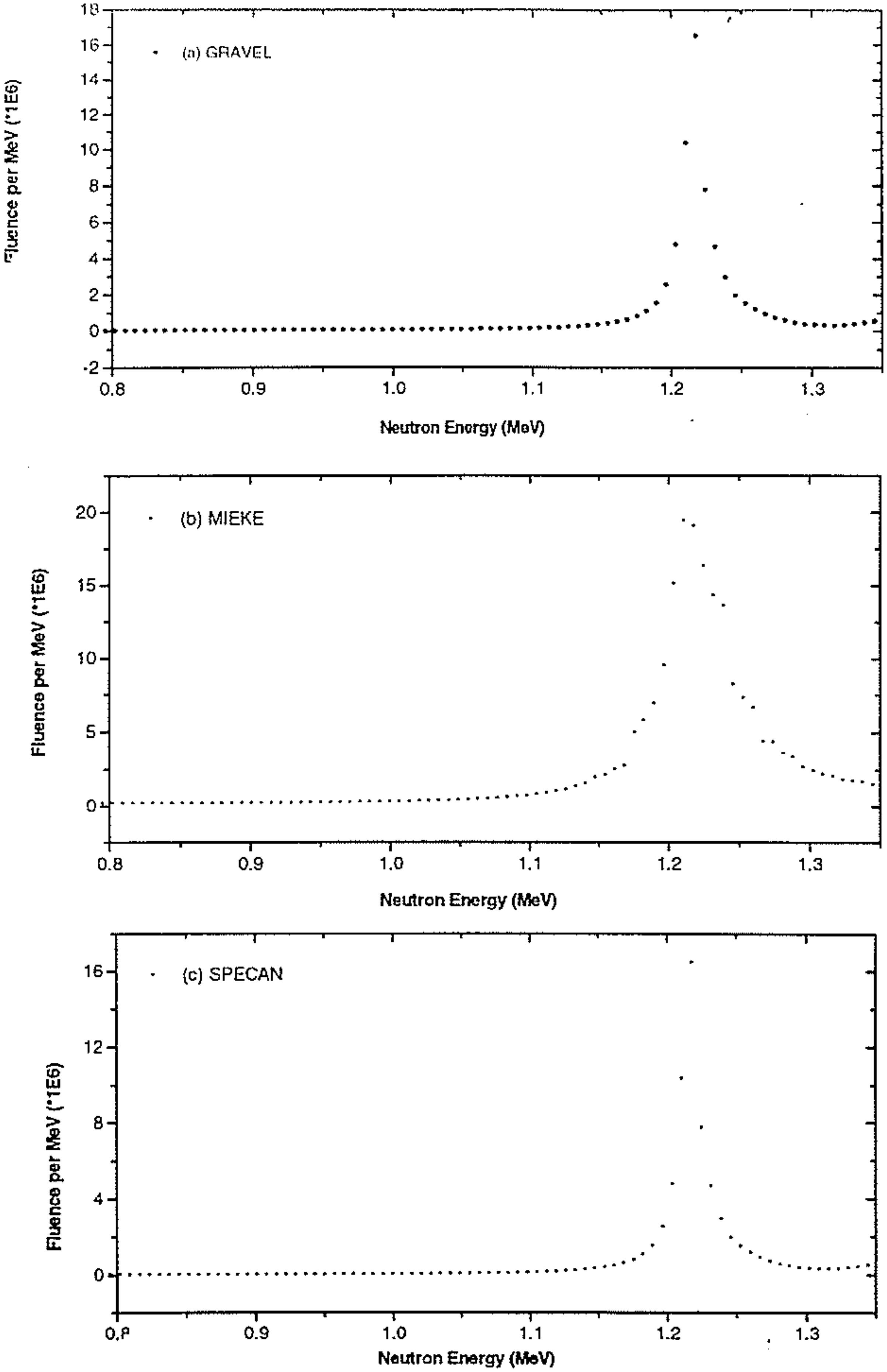


Figure 20: The 1.2 MeV monoenergetic spectrum measured with the 9 atm SP2 counter and unfolded using: (a) GRAVEL, (b) MIEKE and (c) SPECAN

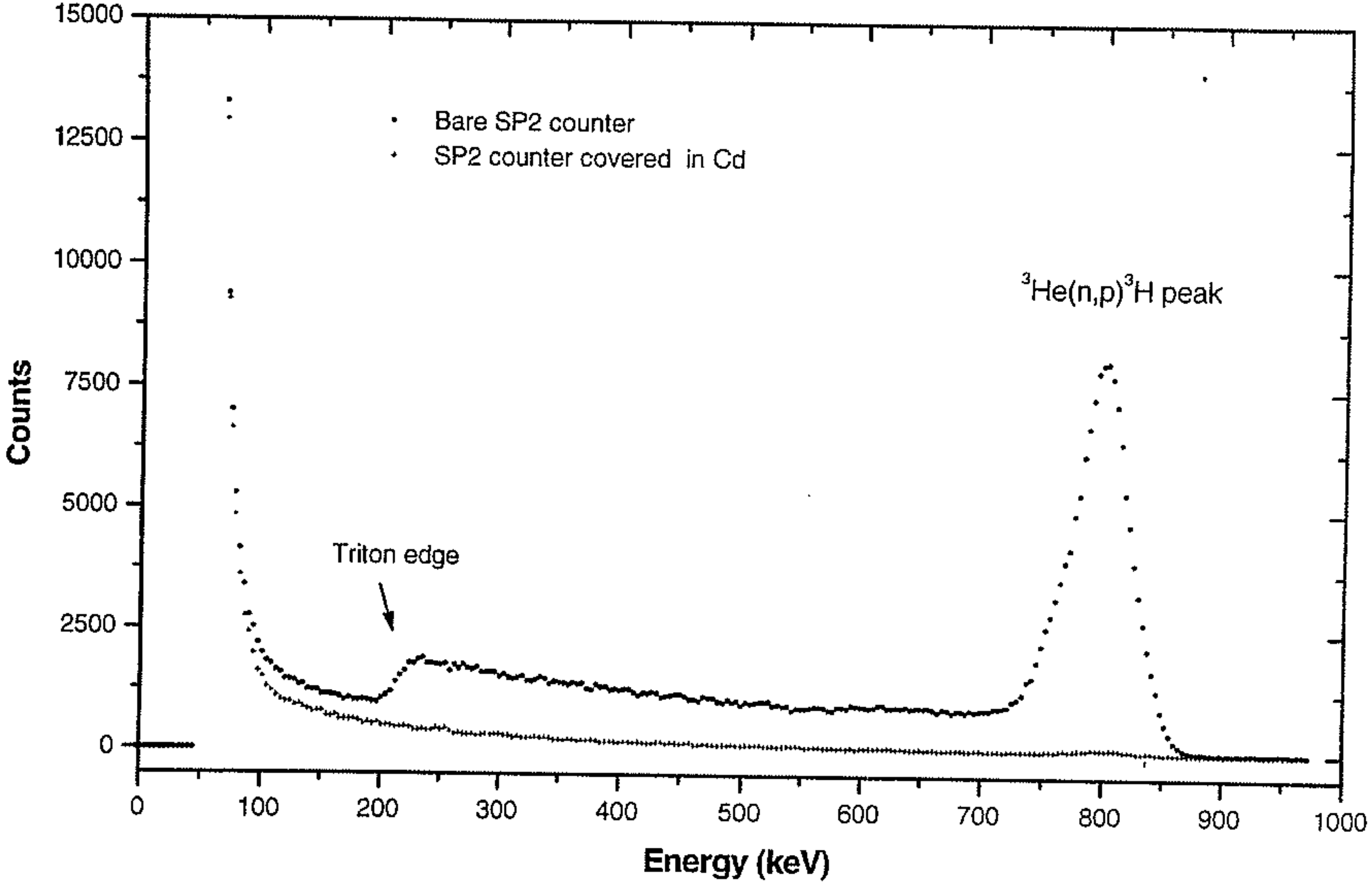


Figure 21: Response of the 3 atm counter to thermal neutrons with and without Cd