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Neutron spectrometry—historical review and present status

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Abstract

Methods of neutron field spectrometry, other than those depending on the use of pulsed neutron sources, are surveyed. Neutron spectrometers are compared with particular reference to characteristics such as energy resolution, useful energy range, neutron detection efficiency and response functions.

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Keywords: Neutron spectrometry; Neutron spectrometer; Response function; Response matrix; Unfolding

1. Introduction

Neutron spectrometry is as old as the famous experiments [1–3] in which, to quote Chadwick [4], "the neutron revealed itself to us". The discovery of the neutron rested on measurements of neutron energy made by scattering neutrons on hydrogen [2] or nitrogen [3], and measuring the energies of the resulting recoiling nuclei. Neutron spectrometry has contributed much to the development of nuclear physics since 1932 and has also become an important tool in several other fields, notably nuclear technology, fusion plasma diagnostics, radiotherapy and radiation protection.

Methods of neutron spectrometry can be classified into seven groups based on the principle used to sense or measure neutron energy: (1) methods in which the neutron is scattered and the energy of a recoiling nucleus is measured, as in the discovery of the neutron; (2) methods based on measurements of the energies of charged particles released in neutron-induced nuclear reactions; (3) methods in which the neutron velocity is measured; (4) threshold methods, in which a minimum neutron energy is indicated by the appearance of a neutron-induced effect such as radioactivity, a specific gamma-ray energy or a phase transition; (5) methods in which the neutron energy distribution is determined by unfolding a set of readings of detectors (or detector geometries) which differ in the energy-dependence of their response to neutrons; (6) methods based on neutron diffraction; and (7) methods in which the time-distribution of the slowing down of a short burst of high-energy neutrons in a suitable medium is measured.

One of the most widely exploited methods of neutron spectrometry is the pulsed-beam time-of-flight technique [5], belonging to group 3 above. We will not discuss this particular technique, however, because this Workshop specifically excludes pulsed-beam methods. Groups 6 and 7 are omitted for similar reasons, hence this survey is

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confined to spectrometers belonging to groups 1–5, but excluding methods in group 3 that depend on the use of a pulsed neutron source.

2. Neutron spectrometry 1932–2000

Three phases can be identified in the evolution of neutron spectrometry to date: 1932–1959, 1960–1979, and 1980–2000, respectively. The first and second phases are documented in review articles collected in the well-known volumes edited by Marion and Fowler [6] and Bromley [7]. One of the goals of this Workshop is to review progress during the third phase. In this brief survey we attempt to identify some highlights in this progress.

Most neutron spectrometers used today are based on methods that were introduced before 1960. Recoil spectrometers in use by 1960 included ionization chambers and proportional counters [8], cloud chambers [9], nuclear emulsions [10], organic scintillators [11], recoil telescopes [12,13] and double-pulse (capture-gated) neutron spectrometers [14]. Neutron spectrometers based on measuring energies of charged reaction products included the ^{3}He-proportional counter spectrometer [15] and the ^{6}LiI(Eu) scintillation crystal [14,16]. Time-of-flight methods [17] and threshold radioactivation methods [18] were also prominent during this period.

Notable developments during the second phase, 1960–1979, were the introduction of the Bonner sphere method [19] and the advances made in neutron spectrometry techniques based on gaseous ionization detectors [20] and scintillation detectors [21]. An associated important development was the advent of computer unfolding methods for determining neutron spectra from few and multichannel measurements [21–24]. Other notable developments during this period were the first applications of semiconductor detectors to neutron spectrometry [25] and the introduction of the superheated drop detector [26].

The third (post-1979) phase also brought considerable technological progress but will probably be remembered more for the impact of computers on neutron spectrometry. In particular, neutron spectrometry has improved as a result of the comprehensive methods of calculation that are now available, firstly to generate the response functions or to calculate the neutron detection efficiency of the detector system, e.g. by Monte Carlo simulation, and secondly to unfold (deconvolute) the spectral neutron fluence from the spectrometer readings. The response matrix, which summarizes the detector response as a function of incident neutron energy and other variables, e.g. angle of incidence, provides the basis for this unfolding process. To date, response matrices are mostly based on calculated responses or evaluated excitation functions which must be validated by experimental measurements. One of the most important measures of the quality of a neutron spectrometer is the degree to which its response matrix is known and understood. The computer codes that have been developed for these operations, though vital to neutron spectrometry, are too numerous for us to list or discuss in this survey. When appropriate they will be referred to by name only.

3. Neutron spectrometry—the present status

3.1. Spectrometry of recoil nuclei

Recoil spectrometers are either detectors in which recoils at all angles to the incident neutron direction are accepted for measurement, or recoil telescopes, in which recoils at a particular angle (preferably 0°) are selected for analysis. The spectrometer response function (pulse height spectrum resulting from bombardment with monoenergetic neutrons) is typically a broad continuum for the first category and a narrower group in the case of a telescope.

Fig. 1 shows measured and calculated response functions of a proton recoil proportional counter detector spectrometer for monoenergetic neutrons of energy 565 keV [27]. The two response functions are in good agreement and show the expected form of a continuum that extends from zero pulse height to a well-defined upper limit. The accuracy with which this limit is defined provides a measure of the pulse height resolution of the spectrometer.

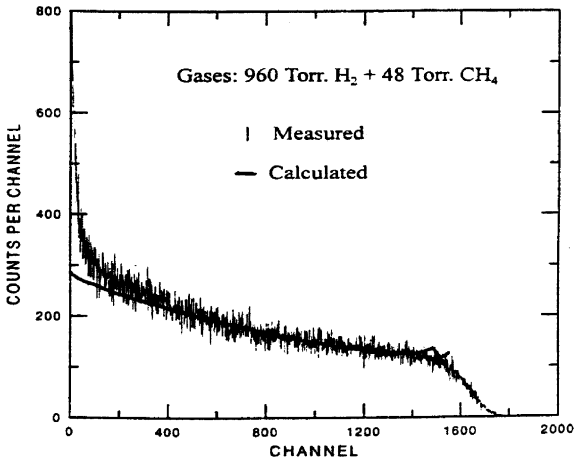

Fig. 1. Measured and calculated pulse height distributions for 0.565 MeV neutrons incident on a cylindrical proportional counter (active volume 38 mm diameter × 178 mm). From Ref. [27].

Recoil detector spectrometers based on proportional counters are widely used in the energy range 50 keV to a few MeV [28–30]. Pulse height resolution varies chiefly as the square root of the recoil energy deposited and is typically about 10% (FWHM) for 1 MeV recoil ions.

For neutron energies above about 5 MeV proton escape (wall effect) limits the usefulness of proton recoil proportional counters. Although ^{4}He recoil devices can extend the energy range upwards (to about 15 MeV), organic scintillators are usually the preferred detector medium for spectrometers at higher energies [21,31], in particular stilbene crystals and liquid scintillators, due to their property that neutron and photon events can be separated by pulse-shape analysis. Many examples of measured and calculated response functions for organic scintillators, at neutron energies up to about 15 MeV, are available in the literature [21,31–35]. The response functions of these detectors are dominated by n–p elastic scattering in this energy range. Since the n–p cross-section is well-known, response matrices and detection efficiencies can be computed accurately and neutron spectra can be reliably unfolded from measured pulse height spectra. It is particularly interesting to note that the neutron energy resolution obtained after unfolding can be 4–5 times better (narrower) than the corresponding pulse height resolution [31,36].

Charged particles produced by neutron interactions with the carbon nuclei in the scintillator make significant contributions to the response functions of organic scintillators at incident neutron energies above about 8 MeV, but are adequately accounted for in simulated response functions for energies up to about 15 MeV [31]. For higher incident energies [37–40] it becomes increasingly difficult to simulate these contributions because the cross-section data that are required are either not available or not accurate enough. Fig. 2 shows, for example, response functions of a liquid scintillator measured and calculated for 46 MeV neutrons [39]. Components due to charged products of n–C interactions contribute to this response function in the region to the left of the dashed line ($h < 130$), where the disagreement between calculated and measured responses is very apparent. However, agreement is good in the upper region of the response function ($h > 130$) which is due to n–p elastic scattering only. The simulated response function can therefore be used to determine the neutron detection efficiency, based on the n–p cross-section, for the threshold $h = 130$. An appropriate set of measured

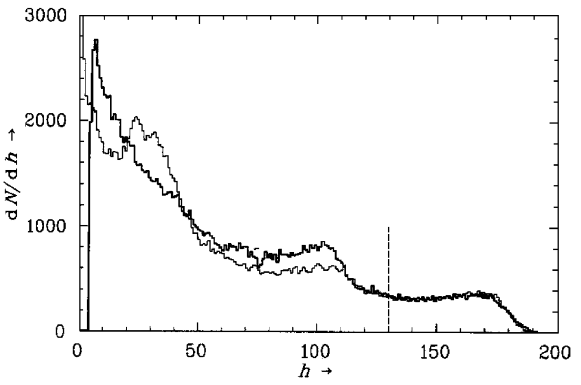

Fig. 2. Number of counts per pulse height interval, dN/dh, as a function of the pulse height, h, measured with a NE213 scintillation spectrometer (thick line) and calculated using the code SCINFUL (thin line) for 46 MeV neutrons. The incident neutron beam was parallel to the axis of the scintillator (51 mm diameter × 102 mm). From Ref. [39].

response functions normalized in this way can then be used to build a reliable response matrix for unfolding purposes [41] or to determine the energy dependence of the neutron detection efficiency for the pulse height threshold actually applied. This method is satisfactory for neutron energies up to about 70 MeV [37] but is seriously limited at higher energies, due to the rapid increase of proton range (in the scintillator) with energy. Correct consideration of the response of recoils not fully stopped in the scintillator is another difficulty because the n/γ-discrimination is also affected. New approaches for counteracting this problem and extending the range of scintillation spectrometers beyond 150 MeV are presented in contributions to this Workshop [42,43].

Recoil telescope spectrometers strive to achieve a simple response function, ideally a single sharp peak at a pulse height uniquely related to the neutron energy. A wide variety of designs have been used to do this. Some recent examples based on combinations of two or more detectors are reported in Refs. [39,44,45]. Another design [46] uses narrow rods of scintillator to form the telescope. Recoil telescopes can provide simple response functions but the price of doing so is usually a very low neutron detection efficiency, typically $<0.01\%$. Fig. 3 shows spectra of neutrons from the ^{7}Li(p,n)^{7}Be reaction measured using a telescope in which an annular radiator (polyethylene) was used [45] in order to increase detection efficiency without sacrificing energy resolution. The spectra shown were measured for incident protons of energy 43 and 68 MeV and are compared with measurements made for the same neutron beams, using the pulsed-beam time-of-flight method. Another example of the use of annular geometry, at lower energy (14 MeV), is presented in a contribution to this Workshop [47].

Another approach that is used to achieve a simple response function is the capture-gated neutron spectrometer [14,48–52]. This is a recoil detector spectrometer, usually a liquid or plastic scintillator of volume 0.5–3 l, which selects events in which neutrons transfer all of their energy by elastic and inelastic scattering within the scintillator. A delayed coincidence between the summed pulse height signal and the subsequent (0.2–50 μs)

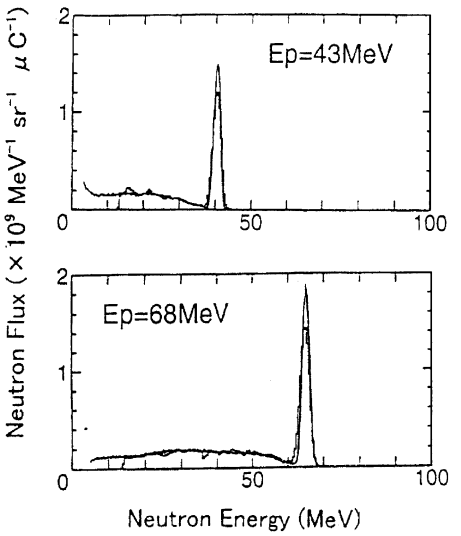

Fig. 3. Neutron spectra from the ^{7}Li(p,n)^{7}Be reaction for incident proton energies of 43 and 68 MeV, measured using a proton recoil telescope with annular radiator (histograms) and by the pulsed beam neutron time-of-flight method (lines). From Ref. [45].

signal due to capture of the neutron after moderation to a low energy (<10 eV) is required. Neutron capture is detected by doping the organic scintillator (liquid or plastic) with ^{10}B or ^{6}Li, or by incorporating a separate low-energy neutron detector in the system. The neutron energy is obtained from the summed pulse height signals which in general generate a broad distribution (FWHM $\sim 50\%$) due to the non-linear pulse height response of the scintillator. However, detection efficiencies of about 1–10% are achievable. New types of capture-gated spectrometers which achieve better pulse height resolution and high suppression of external backgrounds are described in contributions to this Workshop [53,54].

Some recoil spectrometers that might be regarded as exotic or unusual may also be noted with interest. A scintillation spectrometer based on neutron–deuteron scattering in a deuterated anthracene crystal has achieved a pulse height resolution of 4% (FWHM) for 20 MeV neutrons [55]. A spectrometer based on observation of the recoil broadening of internal conversion lines from the decay of certain states that are excited by inelastic neutron scattering in a germanium

semiconductor detector has been described [56]. A scintillation tracking detector system, SONTRAC, based on orthogonal layers of tightly packed plastic scintillator fibres and incorporating image intensifiers has been developed [57] to record recoil proton tracks for the measurement of neutron spectra in the energy range 20–250 MeV.

3.2. Methods using nuclear reaction products

The nuclear reactions ^{3}He(n,p)^{3}H, ^{6}Li(n,α)^{3}H, ^{10}B(n,α)^{7}Li, ^{12}C(n,α)^{9}Be and ^{28}Si(n,α)^{25}Mg are the main candidates for use in neutron spectrometers in which the neutron energy is determined by measuring the energies of charged reaction products. The ^{3}He(n,p)^{3}H reaction ($Q_{gs} = 0.764$ MeV) is a leading method for neutron spectrometry in the energy range 50 keV to about 5 MeV. Several types of ^{3}He-spectrometer are used, for example proportional counters [58,59], gridded ionization chambers [60–62] and sandwich spectrometers [63–65]. Pulse height resolutions of 15–40 keV (FWHM) for neutron energies <2.5 MeV have been reported [61,62] using ^{3}He-filled gridded ionization chambers and about 50 keV for the entire energy range from 100 keV to 15 MeV using ^{3}He-sandwich spectrometers [64,65].

Fig. 4a (points) shows a response function measured using a ^{3}He proportional counter, for monoenergetic neutrons of energy 2.413 MeV [59]. The peaks at high and low energy (3.17 and 0.76 MeV, channels 947 and 226), are due to ^{3}He(n,p)^{3}H reactions produced by incident neutrons and thermal neutron background, respectively. The continuum stretching from low energy to 1.6 MeV (channel 500) is due to ^{3}He recoils from ^{3}He(n,n)^{3}He elastic scattering. The response functions of ^{3}He-spectrometers can be simulated accurately [58,59,61], as shown in this example, hence reliable neutron energy spectra can be obtained by unfolding pulse height spectra measured by these instruments.

The ^{10}B(n,α)^{7}Li reaction is not as well suited for spectrometry because it populates both the ground state and first excited state (0.48 MeV) of ^{7}Li with similar probability. The ^{6}Li(n,α)^{3}H reaction has been used in scintillation spectrometers [16] and sandwich detectors [66]. However, the use of

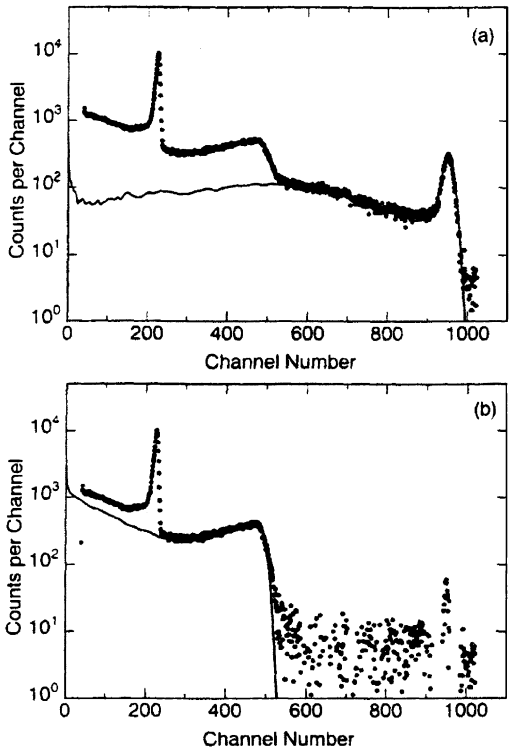

Fig. 4. (a) Measured response function (points) of a ^{3}He-proportional counter (sensitive volume 25 mm diameter × 151 mm) to 2.413 MeV neutrons and calculated response function (line) for the component due to the ^{3}He(n,p)^{3}H reaction only. (b) Difference (points) between the data and calculation shown in (a) and calculated response function (line) for recoils from ^{3}He(n,n)^{3}He elastic scattering. Calculations were made using the code NRESPG. From Ref. [59].

scintillation spectrometers has been limited by factors such as their sensitivity to gamma rays, non-linear pulse height response and contributions to the response function arising from neutron interactions with other constituents of the scintillator.

Diamond and silicon semiconductor crystals are convenient media for use as neutron spectrometers based on the reactions ^{12}C(n,α)^{9}Be$_{gs}$ [67–69] ($Q = -5.70$ MeV) and ^{28}Si(n,α)^{25}Mg$_{gs}$ [70] ($Q = -1.65$ MeV), particularly for applications in plasma diagnostics. The ground state transitions are prominent in the energy spectra for both reactions, for incident neutron energies below about 20 MeV, and a pulse height resolution of about 2% (FWHM) can be attained.

3.3. Neutron time-of-flight method

In this method neutron energy is determined by measuring the neutron flight time over a known distance. Unless the neutron source is pulsed, detectors are required to register both the start and termination of the neutron flight. Two methods are used to do this. In the first method the neutron is scattered in a start detector, for example an organic scintillator, and the time of flight to a second detector at a known distance and the angle is measured. In the second method the start signal is provided by an associated particle or quantum that is emitted from the neutron source at the same time as the neutron. The associated particle time-of-flight method has been employed, for example, to measure the spectrum of neutrons from the spontaneous fission of ^{252}Cf [71,72] or to characterize neutron time-of-flight detectors [72,73]. The scatter detector method has been used in studies of neutron scattering and to measure neutrons emitted from nuclear reactions and nuclear fusion [40,74–77]. Time-of-flight and other techniques used to measure neutron spectra from the JET tokamak are discussed and compared in contributions to this Workshop [77,78].

3.4. Threshold methods

Radioactivation methods are used extensively for measuring neutron fluence [18,79,80]. Some of these methods are based on endoergic neutron-induced nuclear reactions ($Q<0$). The observation of a radioactive product or daughter resulting from such a reaction indicates that the neutron energy must exceed the threshold for the reaction. Thus the energy spectrum of a neutron field may be determined [79] by comparing measurements of several different neutron-induced activities (with different threshold energies), made by irradiating an appropriate set of target foils in the field. The response matrix is then based on the evaluated excitation functions of the reactions employed [80].

Another type of threshold method is based on measurements of gamma rays or internal conversion electrons excited by neutron inelastic scattering in a germanium semiconductor crystal or in a suitably selected scattering material surrounding such a crystal [81]. The excellent energy resolution of this detector makes it possible to identify and measure many different gamma lines that are excited by $(n,n'\gamma)$ reactions. Neutron energy sensitivity is provided by the energy thresholds for populating different excited states.

A comparatively recent arrival in the field of neutron threshold spectrometers is the superheated drop (bubble) detector [26,82–85]. The detection process in these systems is well understood [86–88] and is characterized by a threshold energy (related to the critical radius for bubble formation) which depends on the liquid used, the energy of the neutron and the temperature and pressure of the liquid. A neutron spectrum can be measured by recording the bubble production rate as a function of temperature and then unfolding these measurements, using an appropriate response matrix [82], to obtain the spectrum. A measurement of the energy spectrum of neutrons from the spontaneous fission of ^{252}Cf, made using a prototype Bubble Interactive Neutron Spectrometer (BINS) [82], is shown in Fig. 5a. Developments in this method of neutron spectrometry are reviewed by d'Errico et al. at this Workshop [89].

3.5. Methods using multisphere systems

The prototype multisphere system was the neutron spectrometer introduced by Bramblett, Ewing and Bonner in 1960 [19], which subsequently became known as the Bonner Sphere Spectrometer (BSS). The original BSS comprised a small thermal neutron detector (cylindrical ^{6}LiI(Eu) crystal, 4 mm diameter $\times$ 4 mm), positioned at the centre of a polyethylene sphere. Five spheres of different diameters between 2 and 12 inch were used to produce five detector systems (geometries) with distinctively different response functions, as shown in Fig. 6. The BSS can determine neutron spectra over a wide energy range (see Fig. 6) but with relatively poor energy resolution. A neutron spectrum is determined by measuring the detector count rate for each of the 5 geometries and then

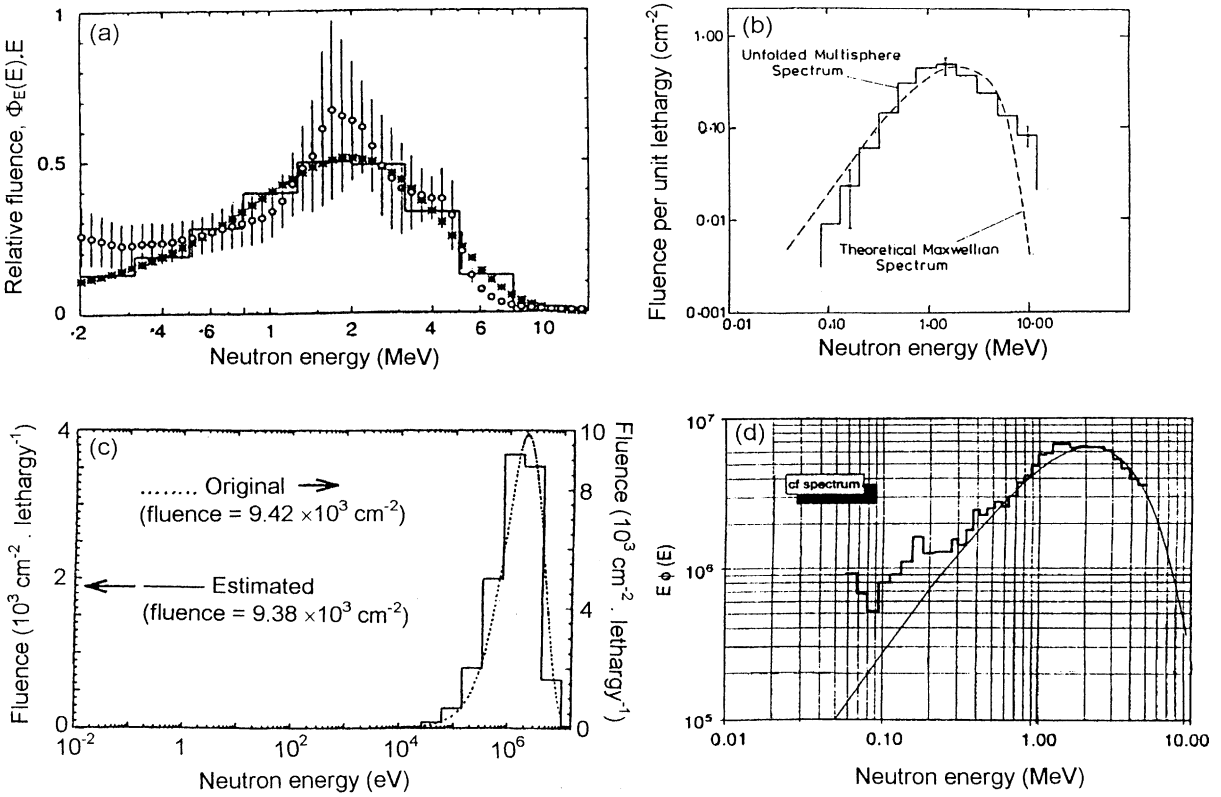

Fig. 5. Energy spectrum of neutrons from ^{252}Cf spontaneous fission, measured using four different neutron spectrometers: (a) the BINS spectrometer [82]; (b) a BSS [90]; (c) the spectrometer of Toyakawa et al. [96]; and (d) a ROSPEC spectrometer [29]. In (a) the histogram shows the reference spectrum of neutrons from spontaneous fission of ^{252}Cf and the points show results obtained using two different unfolding procedures. In (b)–(d) the histograms show the unfolded spectra and the curves show reference spectra. The enhancement in the histogram at low energies in (d) is attributed to room-scattered background [29]. Figures are taken from the references quoted.

unfolding these measurements using the response matrix represented by Fig. 6. A measurement of the neutron spectrum from spontaneous fission of ^{252}Cf, made using a modified BSS [90], is shown in Fig. 5b.

Bonner sphere spectrometers are widely used in radiation protection applications. Developments in this important branch of neutron spectrometry are critically reviewed at this Workshop by Thomas and Alevra [91]. Among the many contributions to the Workshop in this field we draw particular attention to those describing methods whereby BSS are being adapted to measure spectra up to neutron energies in the GeV range [92–95].

An interesting approach among new designs that have evolved from the BSS is to use a single block of moderator containing either several extended, position-sensitive thermal neutron detectors [96], or a number of small thermal neutron detectors [97] mounted at different positions. The position of neutron detection then replaces the sphere diameter as a variable in the response matrix and all measurements are made simultaneously, without interruption for geometry changes. The geometry of these systems also endows them with a rough direction-sensing capability. A measurement of the neutron spectrum from spontaneous fission of ^{252}Cf, made using the spectrometer of Toyakawa et al. [96], is shown in Fig. 5c.

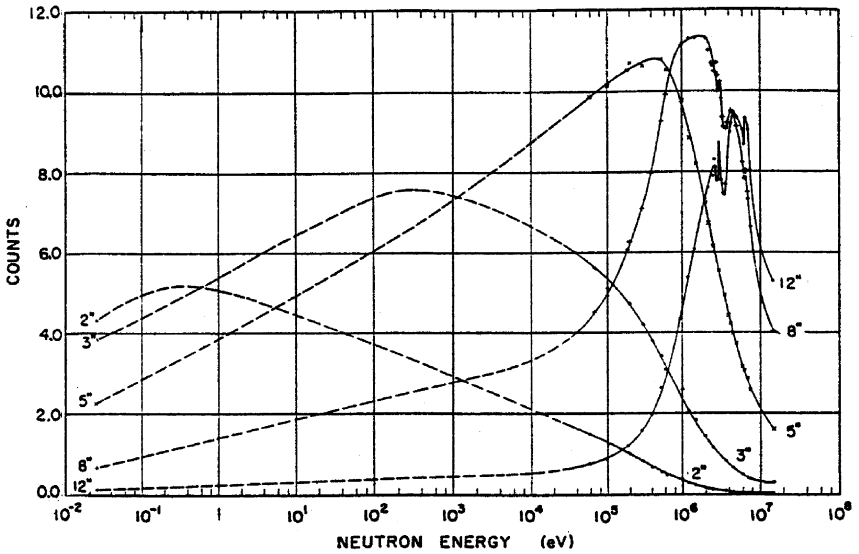

Fig. 6. Response functions of the original Bonner sphere spectrometer, showing the count rate of the ^{6}LiI(Eu) scintillation crystal detector (4 mm diam. × 4 mm) as a function of neutron energy for five different polyethylene sphere diameters between 2 and 12 inch. From Ref. [19].

3.6. Neutron spectrometer combinations

In the case that neutron spectra with a wide energy range have to be investigated with much better energy resolution than achievable with multisphere systems, e.g. to investigate shielding or to test transport calculations in some detail, two or more spectrometers of different types may be combined [98]. A set of proportional counters filled with hydrogenous gases at different pressures, may cover the energy range between 20 keV and some MeV, e.g. the commercially available ROSPEC system [29]. A measurement of the energy spectrum of neutrons from the spontaneous fission of ^{252}Cf, made using this system, is shown in Fig. 5d. Additional ^{3}He- or $^{10}BF_3$-filled counters, bare and Cd-covered, extend the energy range down to thermal energies and an additional ^{4}He-recoil counter may be used to cover the energy range between 1 and 15 MeV [99]. Alternatively, if a liquid scintillator is added [100], the upper energy limit may be increased substantially, to 20 MeV or even 100 MeV, depending on the size of the (stacked) detector system used. In addition, the photon component of the field can simultaneously be investigated [31].

4. Summary

Table 1 presents a comparison of characteristics of 11 different neutron spectrometers selected from the types discussed in Section 3. Further information about these and other types of neutron spectrometers may be obtained from the literature quoted in this section and from general references on radiation detection techniques [101]. The references quoted in Table 1 are those from which the data shown in the table were obtained. Dimensions and other details of the spectrometers referred to in the table can be found in these references. Typical characteristics (at a specific incident neutron energy) are listed for spectrometers 1–8. Spectrometers 9–11 are inherently different from 1–8 in the way that they function and perform. Spectrometers 1–8 all consist of a single detector (or system of detectors) which captures all the data needed to determine the neutron energy distribution in a single measurement. Spectrometers 9–11, in contrast, invoke multiple measurements in their functioning, either through the use of a number of different detectors (9) or by making several measurements under different conditions or geometries (10 and 11).

Table 1
Neutron spectrometer characteristics

Spectrometer				Typical characteristics for		
No.	Type	Ref.	Energy range (MeV)	Energy (MeV)	Resolution (FWHM)	Detection efficiency
1	Recoil proportional counter	[27]	0.05–5	1	10%[a]	3%
2	Organic scintillator	[31]	2–150	8	4%[a]	20%
3	Recoil proton telescope	[45]	1–250	60	4%[a]	$<0.05\%$
4	Capture-gated	[49]	1–20	5	50%[a]	1%
5	^{3}He gridded ionization chamber	[61]	0.05–10	1	2%[a]	0.3%
6	^{3}He-semiconductor sandwich	[64]	0.1–20	1	50 keV[a]	0.1%
7	Diamond semiconductor	[68]	8–20	14	1%[b]	1%
8	Time-of-flight	[74]	1–15	2.5	5%[c]	$0.05\,cm^{-2}$
9	Foil radioactivation	[79]	0.2–20	—	—	—
10	Superheated drop (bubble)	[82]	0.1–20	—	—	—
11	Multisphere	[91]	10^{-8}–200	—	—	—

[a] Pulse height resolution.
[b] Energy resolution.
[c] Time-of-flight resolution.

Spectrometers 1–8 can be designed or tuned to achieve relatively good neutron energy resolution. The energy resolution achieved by spectrometers 9–11 is much poorer and is sensitive to the number of measurements made and the procedures used in unfolding neutron spectra from these measurements. Typical characteristics such as those shown for spectrometers 1–8 in Table 1 cannot be meaningfully given for spectrometers 9–11.

Applications of neutron spectrometry can be grouped into two broad categories; those in which energy resolution is the primary requirement and those in which it is more important that the spectrometer should be simple to operate and capable of operating over a wide energy range. In nuclear physics and plasma diagnostics energy resolution is usually a high priority, hence spectrometers of types 1–8 in Table 1 (discussed in Sections 3.1 and 3.3) are likely to be favoured. The current interest in neutron spectrometry for plasma diagnostics has stimulated useful comparisons [62,74,77,78] of these types of spectrometers. The improved energy resolution (relative to pulse height resolution) attainable through spectrum unfolding [31,36] could also enhance neutron spectrometry in nuclear physics and plasma diagnostics.

Applications of neutron spectrometry in the fields of nuclear technology, radiation protection and radiotherapy are normally less demanding in regard to energy resolution but require the spectrometer to be simple to operate and that neutron fluence be measured accurately over a wide energy range. BSS systems [91], also in combination with other spectrometers discussed in Section 3, can meet these requirements.

Two challenges facing the development of neutron spectrometry in these fields can be highlighted. The first is the need to extend neutron spectrometry into the GeV region [92], a need which is important to safety in air travel and in work environments close to high energy accelerators. The second is to provide reliable methods of neutron spectrometry (besides the pulsed beam time-of-flight method [17]) over the energy range 1 eV to 100 keV [96,102]. This energy region has become more important recently, in view of current interest in the possibility of boron neutron capture therapy [103].

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