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Roger Wallace October 18, 1962

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October 17, 1962

ABSTRACT

The proton-recoil and gamma-ray pulse-shape discriminating, He^{3} , He⁴, Li⁶, double pulsing, moderated BF₃, nuclear emulsion, threshold and semiconductor types of 4π neutron spectrometers are described. These nine types are compared as a function of neutron energy on the basis of absolute sensitivity and energy resolution. Recent developments in the pulseshape discriminating and semiconductor types are outlined. Experimental calibrations of several of the more promising types are included. The discrimination against gamma rays of each type is described. The application of these instruments to neutron field dosimetry measurements of biological significance is discussed .

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FOUR- π FAST-NEUTRON SPECTROMETERS FOR DETECTION AND DOSIMETRY

Roger Wallace

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I. INTRODUCTION

Four- π neutron spectrometers, as contrasted to the many other types of neutron spectrometers, have been developed relatively recently which depend on a prior knowledge of the neutron-source point or the beam line of the neutrons striking the spectrometer.¹ In the case of the time-of-flight proton-recoil, ranger, chopper, or crystal types of spectrometer, the source point and perhaps zero time of origin of the neutrons must be known in order to measure the flight time or to allow the measurement of a recoil angle and associated proton energy. If the point of origin of a neutron is not known, a new approach must be made to measuring neutron-energy spectra. It is conceivable that a collimator could be used to define the incident direction in order to make the resulting proton recoil angle meaningful. The use of such a collimator would largely defeat the purpose of such a measurement in that it would introduce a large mass around the detector and consequently distort the neutron spectrum .

To avoid this distortion other approaches can be made. If we analyze the statistical distribution of proton recoil energies resulting from the recoil of monoenergetic neutrons of a few MeV, we find that all energies from that of the primary neutron on down to zero are equally probable. This rectan_: gular distribution, shown in Fig. 1, is naturally somewhat distorted by the range -energy relation for these protons and other instrumental considerations. Nevertheless, in principle if one assumes that the relation between. the incident monoenergetic neutron energy and the resulting proton recoil spectrum

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is known for a variety of incident neutron energies, then the pulse-height spectrum produced by an unknown incident-neutron spectrum can be unfolded by a simple differentiation if the ideal rectangular shape is actually or assumed to be present.

If the modified non.rectangular shape is to be taken into consideration, then a matrix inversion process will allow the desired result to be achieved with considerably more effort and less precision. This latter process may introduce some practical but surmountable difficulties such as negative probability values, which are of course meaningless. These considerations are indicated in Fig. l for a delta-function neutron energy distribution and for three mixed delta functions. The extension of this basic idea to more complex spectra is the basis for several of the instruments that we will discuss. The actual relation between the proton recoil distribution and the neutron spectrum i s

$$
N_{n}(E)dE = -\frac{dN_{p}(E)}{dE} \frac{E dE}{4\pi n \sigma_{np}(E)}, \qquad (1)
$$

where $N_n(E)$ is the neutron spectrum, $N_p(E)$ is the proton spectrum (converted from pulse heights), n is the number of target protons per cm^{-2} , and $\sigma_{nn}(E)$ is the n-p elastic cross section.

This relation is the basis for the organic-scintillator, double-pulsing, nuclear-emulsion, and some semiconductor types of 4π spectrometers. The same relations corrected for the recoil of a heavier nucleus are the basis for the He⁴ type.

II. DESCRIPTION OF THE VARIOUS TYPES A. Organic Scintillators

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Probably the most promising development in the 4π neutron-spectrometer field in the past few years has been that of electronic circuits that can discriminate against gamma rays while transmitting proton recoil pulses with high efficiency.^{2, 3} This allows the high gamma-ray detection efficiency of the organic scintillators to be largely eliminated. Most neutron fields, especially those that approach the detector from all directions, are accompanied by a considerable number of gamma rays usually due to 2.2 MeV n-pcapture γ rays. Unfortunately many detectors for neutrons are also sensitive to gamma rays, especially if they also have an output pulse height which is proportional to the incident neutron energy. It is therefore a very significant

step forward to be able to discriminate against the gamma-ray-induced pulses in the case of the organic scintillators, which in turn are such ideal neutron detectors for the application of the statistical recoil unfolding technique described in the Introduction.

B. He⁴ Noble-Gas Scintillator

Helium-4 gas of sufficient purity will emit light directly when a charged particle passes through it.⁴ The recoil spectrum of the He⁴ ions will have the same rectangular shape as that of the protons shown in Fig. 1 except that the maximum energy possible for He⁴ is $4A / (1+A)^2 = 64\%$ of that of the neutron. This scaling down of the recoil energies is a kinetic effect due to the nonequality of the masses of the neutron and the He^4 . The He^4 is used under several atmospheres of pressure to limit the ranges of the recoils to the sensitive volume of the counter.

C. $\operatorname{Li}^{6}I(\text{Eu})$ Scintillator

Fast neutrons produce the Li⁶ + n \rightarrow He⁴ + T + Q = 4.78 MeV reaction in this scintillator. The He⁴ and T deliver $E_n + 4.78$ MeV energy to the phosphor. $5-8$ Although it is ideal for the detection of neutrons with energies of a few MeV, this reaction tends to mask pulses from low-energy neutrons of a few hundred kilovolts since they do not add much to the already large 4. 78-MeV Q value. Also, for neutrons above 10 MeV competing processes set in, making the pulses no longer uniquely associated with one neutron energy.

D. Double-Pulse Type

The double-pulse spectrometer is represented by two closely related types. $9-12$ The first consists of a liquid scintillator in which boron or cadmium is dissolved, and the other is a plastic scintillator surrounded by or near a thermal-neutron detector such as a BF^3 proportional counter. The principle of operation is that an incident neutron's energy is transferred almost entirely to a recoil proton, which is detected in those collisions in which the recoil neutron is left with only a small amount of energy. In these particular collisions the ,neutron is more likely to be captured by the thermal detector, after a time necessary to thermalize. This time is characteristically from 10 to 20 μ sec. When actuated by an initial pulse due to the proton, the associated electronic circuits start a delayed "gate." If a second pulse

in the thermal-neutron detector arrives 10 to 20 usec later, the original proton recoil pulse, which has been traveling along a delay line in the meantime, is admitted to the pulse-height analyzer. The delayed second pulse is not detected when the initial "proton" pulse was a spurious one due to gamma rays. Unfortunately in most neutron fields there are enough gamma rays to cause a fairly large number of accidental gamma-gamma coincidences, which appear to the electronic circuits as proton pulses. The γ -ray rejection by this technique is not as good as in the case of category A organic scintillators. Consequently, these double-pulsed spectrometers are mainly of historical interest, and they will not be mentioned further, except that they are indicated by the shaded regions on the final graphs of efficiency, energy, and energy resolution. Perhaps the high intrinsic efficiency of these spectrometers is misleading, because their vulnerability to accidental coincidences forces them to be used at somewhat lower counting rates than other types, thus cancelling to some extent their high efficiency.

E. He³ Counters

The reaction He³ + n \rightarrow T + p + 0.77 MeV is the basis for several recent developments of spectrometers which do not operate statistically as is the case in types -A and B, but in which each neutron is individually associated with a pulse that is uniquely related to its energy, as in types C
and D.¹³⁻¹⁶ The 770-keV energy release is very useful in making it possil The 770-keV energy release is very useful in making it possible to detect neutrons with energies less tpan 1 MeV, which is quite difficult in many other types of spectrometers. The He³ spectrometers usually take the form of proportional counters operating on He^3 with some other gas added to limit the proton ranges and improve counter performance. Anticoincidence counters have been added to the walls of some proportional counters to elim: imate those proton pulses that would otherwise escape into the nonsensitive volume and thus appear with too small a pulse height. $^{16,~17}$. At energies above 1 MeV an ambiguity is possible in that He^{3} recoils which have not been involved in a nuclear reaction but only in an elastic collision will compete with the He³ (n, p)T reaction. This ambiguity can be removed in the data reduction, 14 but it reduces the resolution of the spectrometer for higher energies.

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F. Moderated BF³

A BF³ counter, surrounded by various thicknesses of moderator which in turn is contained inside a Cd cover, will have a counting-rate variation with moderator thickness (see Fig. 2) which changes with the incident-neutron energy. $^{\bf l}$ These curves are plotted in Fig. 3 as counting rate vs energy for a constant moderator thickness. It is seen that they permit this detector to be used as a very crude spectrometer, with the exception of the 6-cm thickness which is almost energy-insensitive. Although the resolution of this system is very low, the sensitivity is fairly high, and the widespread availability of the $BF³$ detector makes this an important method in spite of its serious limitations.

G. Nuclear Emulsion

The length of proton recoil tracks produced in nuclear emulsion can be measured with a properly equipped microscope. This measurement yields a proton range spectrum, which can be converted to a proton energy spectrum which in turn can be inverted, by a process related to differentiation, to the incident-neutron energy spectrum. $18-22$ The extreme simplicity of the detector is perhaps misleading when the overall effort necessary to exploit this method is considered. Since this is a statistical process like A and B, a very large number of proton recoil tracks must be measured, each one individually, by a fairly well-trained operator in order to carry out the pseudo differentiation process with any precision. The data can be recorded and subsequently reduced by computer methods. This detector is recommended when the 4π spectrum is required simultaneously at many points in space, and electronic methods would be too costly to duplicate on this scale.

H. Threshold Detectors

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The use of U^{238} , P^{31} , S^{32} , Ag^{107} , I^{127} , C^{12} and Bi^{209} in reactions whose thresholds vary from 1.1 to about 50 MeV provides simple, inexpensive detectors.^{23, 24} The half lives of all reaction products except U^{238} and B_1^{209} are long enough to allow the sample to be taken to a counter for assay. In many applications the efficiency of the threshold detectors is too low, although several recent developments have allowed this to be increased in some special cases. In applications where neutrons are present for a very

short time with very high intensity as is the case near a nuclear explosion or during the burst of a critical assembly, are the only detectors that can be used. There is practically no possibility of saturation by high intensities. The spectral resolution, while crude, is better than that of type F .

I. Semiconductors

These are the most recent detectors to be developed, and in spite of their very low efficiency offer some very attractive features. $8, 24-27$ Their extremely small size and electronic simplicity are great advantages. Basically they detect charged particles such as protons, alphas, or fission products. Thus they can be used to. detect recoil protons from hydrogen or reaction products from a variety of nuclear reactions. 5

III. GAMMA-RAY DISCRIMINATION

Since, like neutrons y -rays are uncharged their simultaneous detection by any of these spectrometers distorts the neutron spectral measurement and produces a spectrum that is a combination of γ and neutron effects. It is .important that this be avoided, and therefore a discussion of the relative y. ray rejection of these spectrometer types. is in order

Although organic scintillators (type A) are quite sensitive to γ rays . the rejection ratios for well-designed pulse-shape discriminating circuits are of the order of one thousand or better. This is sufficiently high for practical purposes. unless they background is very intense relative to the neutron flux.

Type B He⁴ recoil counters are fairly insensitive to γ rays because of the low Z of He. Rejection ratios of twenty to one are obtained experimentally.

In type C, $Li^{6}I(Eu)$ crystals, there is good discrimination of the lowenergy y rays because of the high Q value of 4.78 MeV which is added to the $He⁴$ and T emitted in the basic neutron-detection reaction.

The double-pulsing type-D spectrometer is quite vulnerable to γ interference, especially at high counting rates.

Type E He³ spectrometers have excellent gamma discrimination, because of the low Z of the He and the 770-keV Q value of the He³ (n, p)T reaction. In addition when the counter is surrounded by an anticoincidence counter ring to reject escaped protons and ions produced in the counter walls, the γ counts, which are largely due to wall effects, are automatically eliminated except at the ends.

Moderated BF³ detectors (type F) are very insensitive to gamma rays. Again this: is due to the large Q value of the B^{10} (n, d) Li^7 reaction, as well as to the inherent insensitivity of a gas counter to γ rays.

Nuclear emulsion, treated as type G, responds to γ rays by producing random developed grains. Since these are rarely aligned in tracks. it is easy for the microscope scanner to eliminate γ contamination. If the background random grain density becomes high, however, it interferes with the measurement of the length of the proton recoil tracks themselves. On a dose basis, there is about a ten to one rejection of γ rays by emulsion.

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Threshold detectors (type H) are fairly immune to gamma interference, especially for low-energy γ rays. In the particular case of Bi^{209} fission, the γ sensitivity is small even for γ rays with several BeV energy.

In type I, semiconductors have such a small sensitive region and usually so little material around them that their γ response is small. In addition, . the low ionization density along electron tracks in the. thin sensitive region tends to reduce the γ pulse heights, permitting effective discrimination.

In general γ rejection is accomplished by pulse-height discrimination using electronic bias in types B , C , E , F , and I. In the special case of type A, the discrimination is on the basis of pulse shape rather than height. In ·typeD, it is on a time· identification basis, in-type G by visual means, and in type H on the basis of the small cross section for gamma-induced activities.

While y rejection is a major hurdle in neutron spectroscopy it has been solved fairly well in many of the types that are outlined here. In general γ discrimination breaks down in most types when the γ count rate becomes very high. This is usually the result of the piling up of many small γ pulses that arrive close to each other in time and produce an accumulative effect which cannot be distinguished from a real neutron-induced pulse. ·Even in the case of the type A pulse-shape discrimination method, the irregularly shaped γ accumulation pulses coming from γ pileup are not as easily rejected by the shape-sensitive circuits as are the single γ pulses for which the circuits were intended.

IV. SENSITIVITY AND RESOLUTION

Sensitivity and resolution are generally mutually exclusive qualities in spectrometers.^{29, 30} Only by comparison of these properties can a wise choice be made between the several types. Statistical fluctuations reduce the resolution of all types, and since this effect is fairly obvious, it will not be discussed in this brief resume.

Organic scintillators _(A) suffer from nonlinear pulse-height output as a function of the deposited energy. In addition there are losses of protons from the scintillator in small scintillators, and in large ones there may be multiple neutron collisions which make incorrect the spectral unfolding outlined in the Introduction.

Thus there is a correct size for the organic scintillators, intermediate between these two extremes, for maximum energy resolution for any particular incident-neutron energy. This ideal size is usually a few em on a side. In the case of a spectrum that extends over a wide energy range, it would probably be best to use several different sizes of scintillator. The efficiency of an organic scintillator 2 em on a side is usually greater than 0.1 over the energy range from 1 to 10 MeV. However, to actually detect 1-MeV neutrons is difficult, and the usual lower limit is 2.5 MeV. competing recoil pulses are produced by the carbon in the scintillator. Their pulse spectrum is superimposed on that of the H recoils. Fortunately this effect 1s small.

The $He⁴$ recoil detectors (B) have the same wall- and end-effect losses of resolution as the organic scintillators and the same type of limitations on the lower and the upper size of the sensitive volume. In some of these detectors, loss of resolution occurs because of collisions with Xe which has been added to limit the recoil ranges, thus producing a spurious pulse distribution which is superimposed on that of the $He⁴$. Energy resolution improves and sensitivity decreases as the pressure in the counter decreases.

The efficiency of a $Li^{6}I(Eu)$ crystal (C) a few centimeters on a side is equal to a few percent for neutrons of a few MeV energy. For the smaller crystal sizes, the resolution is limited by the escape of the He⁴ and the T products. In the largest sizes, light-collection efficiency might decrease. The main limitation on the resolution is, of course, the size of the pulse that you wish to detect relative to the Q value of 4.78 MeV for the Li⁶ (n, T)He⁴ reaction. Resolutions near 30% are practical in favorable energy ranges.

No comment on the double-pulse spectrometer (D) will be made here other than the inclusion of this type in the following figures.

The He³ counter (F) has a fairly low efficiency, since it is a gas-filled device. Pressures up to 10 atm are often used. Large diameter is desirable, but this requires higher voltages with consequent breakdown difficulties. The end losses also reduce resolution. Both the end losses and the side losses can be compensated for in principle by a proper treatment of the data. The anticoincidence schemes also reduce the efficiency with increasing energy, since more and more proton pulses actuate the anticircuit. At best, the efficiency is several orders of magnitude below that of types A, F, and G, and somewhat below B. Although the resolution of the He^3 counter depends on the usual statistical limitations and such characteristic proportionalcounter difficulties as recombination, due to the presence of electronegative gases such as O_2 or H_2O , the resolution also depends on special He 3 problems such as the possible presence of radioactive T and the additive Q value of 0.77 MeV for the He³ (n, p)T reaction. The resolution is thus limited at the low-energy end by the Q value and at the high-energy end by the presence of competing reactions. The useful energy range in which there is a reasonable resolution is more limited in the He³ spectrometer than in most of the other types, but the spectrometer's ability to work at energies somewhat below the limits of most of the other types is its main virtue. This makes it a device that should be thought of as complementary to one of the other types.

The efficiency of the moderated BF₃ counter (F) varies with moderator thickness, which is reasonably high however. This variation is not shown in the following figures other than as a shaded area because of the very low energy resolution.

The efficiency of emulsion (G) is moderately high, and the resolution is largely a matter of the statistics. It is however a formidable task to build up enough statistics to increase the resolution above a few percent.

The efficiencies of some of the threshold detectors (G) are shown as dots on Fig. 4. In general the efficiency is low, but the phoswich technique of interposing layers of the threshold material between layers of plastic scintillator considerably increases this efficiency. The crudeness of the resolution is second only to that of the moderated $BF₃$ counter because of the infrequent spacing of the threshold values and the shape of the activation cross sections as a function of energy above the thresholds. Threshold detectors are compared in Table I.

The efficiency of the semiconductor detectors (I) depends on the reaction probability of the material that is used *to* get charged particles which in turn are detected in the sensitive layer. Higher efficiencies can be obtained by using thicker layers, naturally at the cost of a reduction in the energy resolution. Efficiencies are of the order of 10^{-6} , and reaction-layer thicknesses are a fraction of a micron. This extremely low efficiency is somewhat offset by the high resolution and small size of the detector. About 3 eV of energy are required *to* produce an electron-hole pair in a semiconductor, compared *to* about 30 eV in a gas counter. This means that for equal energy deposition, the statistics are a factor of $\sqrt{10}$ better in the semiconductor.

V. CONCLUSIONS

The sensitivity and resolution are shown in Figs 4 and 5. The details of the production of these curves are given by $Kim.$ ¹ Reasonable assumptions .about maximum and minimum detector sizes, minimum pulse-height bias, resolving times, counter filling pressures, counter operating voltages, sensitive layer thicknesses in the semiconductors, and converter thicknesses have been made in each case. These curves express qualitative relations only.

A high value for the efficiency/ resolution ratio η/δ makes a desirable spectrometer. Unfortunately this ratio is not as meaningful as might be hoped, since the energy resolution varies by a small amount in comparison to the range of variation of the efficiency. The ideal spectrometer would be represented by a point in the upper right corner of Fig. 5. It is seen that with the exception of that for the semiconductor the curves for the actual spectrometers hug the ordinate, and few of them have better than 10% resolution.

. The conclusion which seems best supported by the se qualitative relations is that from 2.5 to 15 MeV the pulse-shape-discriminating organic scintillator--probably in several different sizes- -is *to* be preferred, while below 1 MeV, He³ is the only convenient choice. These two energy ranges can probably be fitted together by a slight downward extension of the organicscintillator lower limit, and the removal of the high-energy ambiguities of the ${\rm He}^3$ counter in the treatment of the data. Both of these instruments require a multichannel pulse -height analyzer and modest data-handling and computer facilities. At present, a single spectrometer which produces a direct readout

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and a few percent resolution of the energy distribution of neutrons approaching the detector from a $4-\pi$ direction seems to be beyond the scope of our technology.

The need for $a.4\pi$ neutron spectrometer was originally motivated by a desire to be able to multiply the resulting spectra by the RBE as a function of energy in order to secure the REM dose in biologically significant radiation fields. During the time that these spectrometer developments and calculations have been carried out, the 1 to 10 range of RBE values has apparently contracted somewhat, perhaps to as little as one to two. Thus the need for such a device has been somewhat reduced for the calculation of REM values. However, in the evaluation of the effectiveness of reactor and accelerator shieldins, such a spectrometer retains its practical value. It is hoped that the next few years will see the increased use of some of these relatively new tools.

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Table I. Comparison of various threshold detectors.

FIGURE CAPTIONS

- 1. The relation between a monoenergetic incident neutron and the resulting proton (or heavier A) recoil spectrum, and its extension to the measurement of an unknown neutron spectrum from its proton recoil spectrum.
- 2. The BF_3 counting rate as a function of paraffin thickness for various neutron sources, corrected to an isotopic flux distribution. The entire assembly was covered with Cd.
- 3. Average efficiency of a BF₃ proportional counter as a function of neutron energy for various paraffin thicknesses, corrected to an isotropic flux distribution. Averaging counting rates over the 4π solid angle, an error of up to 10% is present.
- 4. Efficiency vs energy of various counters at the conditions stated for comparison. The relative vertical positions of curves are not fixed but can be moved up or down vertically by several factors, by changing the conditions stated. The top curve does not imply the best counter. It merely states that it has a relatively high efficiency. The usefulness of a counter is determined by various factors such as energy resolution, gamma sensitivity, energy range, and time response as well as by efficiency.

-Curves (A) to (I) are drawn from the calculated reaction probabilities and represent the highest theoretically possible efficiency at the conditions stated. Downward arrows are drawn to indicate that the actual efficiency will be smaller by several factors. Curve (D) is the probable range of efficiency for a large $($ 30 \times 30 in.) Cd-loaded organic scintillator. Curve (F) shows an efficiency of the order of 10^{-2} in the energy range shown.

- 5. Qualitative relation between efficiency and energy resolution for various counters.
	- $\mathcal{A}(A)$ Plastic scintillation counter for $d = 1$ in. and $B = 0.1$ MeV.
	- (B) He⁴ scintillation counter for $d = 10$ cm and 5 atm.
	- (C) Li⁶I(Eu) scintillation counter for $d = 1$ cm.
	- (D) Cd-loaded liquid scintillator.
- (E) He³ proportional counter for 5 atm. and $d = 5$ cm. Energy resolution obtained by Batchelor et al., i.e., about 5% at 0.12 MeV and 12% at 1 MeV.
- (F) BF₃ counter with cover of 6 cm of paraffin.
- (G) Nuclear emulsion for $d = 600 \mu$ thickness.
- (1) Semiconductors. Drawn from the best energy resolution of 0.38% obtained by Dearnaley et al. (Ref. 26) and the efficiency range of 10^{-5} to 10^{-7} . The sideward arrow is drawn to indicate that usually a poorer energy resolution than the indicated value is obtained. Downward arrows are drawn to indicate that the actual efficiency will be smaller by several factors. Numbers on curves indicate neutron energy in MeV. The range of efficiency (η) of various counters is ofthe order of the seventh power, while the range of energy resolution (δ) of these counters is of the order of the third power. The η/δ vs energy does not give correct interpretation of the importance of the counter, since the small variation in the range of energy resolution is more important than the same variation in the range of efficiency.

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Fig. 4

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