

LIQUID SCINTILLATION COUNTERS AND THEIR APPLICATIONS TO PHYSICS*

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SCINTILLATION COUNTER CHARACTERISTICS

BEFORE going into the question of liquid scintillation counters and their applications to measurements in physics, we wish to review the properties of scintillators in general so as to give a better idea of the range of usefulness of the liquids. Table 1 gives some of the properties of the most commonly used scintillators. The figures listed under relative light output are only approximate, since there is quite a range of disagreement in the published values. We

TABLE 1

	Relative light output	Decay time (nμsec)	Typical applications
Anthracene	100	32	beta-particles
Plastic	50	2	counter telescopes
Liquid	70	2	large counters; neutron counters
NaI(Tl)	160	250	gamma-ray spectrometry
Xenon gas	80	10-100	fission particles
ZnS (Ag)	(200)	(100)	alpha-particles; slow neutrons

have taken as standard the best available anthracene crystals. Under 'plastic' we list the light output and decay time of the best commercially available plastic scintillators; the liquid scintillator is a solution of 8 g/l. of PBD† in xylene, from which the dissolved oxygen has been removed.

Let us first consider the organic scintillators. Two of their distinguishing characteristics are the short decay time of the emitted light, and their non-linearity. By the latter is meant the dependence of the light output not only on the total energy deposited in the scintillator, but also on the rate of energy deposition; as a result of this, the pulse from a 5 MeV alpha-particle is only about one-tenth as great as from an electron of the same energy.

The organic scintillator with the greatest light output is anthracene, and this is used when one is primarily concerned with energy resolution. Its

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†2-(4-biphenyl)-5-phenyl-1,3,4-oxadiazole.

disadvantages are large variations in quality between different crystals, its susceptibility to surface oxidation, and its relatively high vapor pressure. The latter makes it unsuitable for use in a vacuum, since sublimation will change the surface reflectivity, and hence the energy calibration. In addition, it suffers from the drawbacks common to most crystals: it is difficult to grow large, clear specimens, and its fragility makes it hard to handle. One can get away from these drawbacks by using plastics, which are the most generally useful organic scintillators for physical measurements. The liquids have slightly better properties than the plastics, but in small volumes they are less convenient to handle. We shall discuss their uses in the following sections.

The inorganic scintillators suffer much less from nonlinearity than do the organic ones. The best known one is NaI, which is used principally in gamma-ray spectrometers. Its advantages are its high light output and transparency to its own radiations, which make possible a good energy resolution. In addition, it contains iodine, which has a high photoelectric cross section for gamma-rays up to a few MeV. This results in a peak whose pulse height is proportional to the gamma-ray energy. In an organic scintillator, except at very low or very high energies, gamma-rays are detected only by the Compton effect: the result is a smear of pulse heights from zero up to a maximum value which depends on the gamma-ray energy. NaI is not much used for the detection of charged particles, because of its long decay time. In addition, it is deliquescent so that it must be protected from the air by some method such as canning.

Gas scintillators,¹ of which the best are xenon and xenon-helium mixtures, appear to be completely linear. This, and the possibility of making them very thin in terms of g/cm², make them useful for detecting heavily ionizing particles such as fission fragments. Their decay time seems to vary inversely with the pressure, and to lie in the range indicated in Table 1, for the pressures normally used.

ZnS is available only in the form of microcrystals, and is normally mixed with a binder and painted on a surface. It makes an alpha-particle detector which is relatively insensitive to beta- and gamma-rays. Since it is not used for lightly ionizing particles, we list its light output for alpha-particles, compared to the output from anthracene for electrons of the same energy. ZnS may be used in conjunction with boron as a neutron detector,² by the reaction $B^{10}(n,\alpha)Li^7$. The efficiency can be made about 30% for thermal neutrons, using normal boron; with enriched boron efficiencies of 80% or better should be possible. In addition to its high efficiency, such a counter has the advantages of speed and insensitivity to the gamma-rays that nearly always accompany neutrons. The decay time of the emitted light has been variously reported to have values from 0.1 to 10 μ sec. The reason for this wide range is that the decay is not even approximately exponential, but varies more closely at $1/t^{1.36}$; ⁽³⁾ about two-thirds of the light comes out in 0.1 μ sec, so this has been listed as the decay time.

The emission spectra of the various scintillators have not been listed. With appropriate wavelength shifters, they can all be made to lie near the maximum of the spectral response curves of the normally used photomultipliers.

In designing a scintillation counter, one would like to know the efficiency of the scintillator, or the total number of photons emitted for a given energy input. A number of measurements of this quantity have been made, and the published values range from 1.7 to 10% for the efficiency of anthracene excited by fast electrons. Taking a median value for this efficiency, one finds that in the best liquids one photon is emitted for every 100 eV of energy deposited by a particle at minimum ionization. By estimating the light collection efficiency, one can now arrive at the total number of photoelectrons corresponding to a given energy deposition in the detector.

APPLICATIONS OF LIQUID SCINTILLATORS

Liquid scintillators find the following applications in physics:

- Study of the scintillation process
- Fast timing experiments
- Solution of a source in the counter
- Neutron detectors
- Large counters.

We shall not discuss the first application here, but confine ourselves to the cases in which scintillation counters are used as tools in physical measurements.

Fast timing experiments

In timing experiments in which the limiting factor is the duration of light emission from the scintillator, Post⁴ has shown that the timing uncertainty, Δt , is inversely proportional to the maximum rate of emission of photons:

$$\frac{1}{\Delta t} \sim \frac{N}{\tau}$$

N being the light output and τ the decay time (the decay being assumed to be exponential). From Table 1, dividing the figures in the first column by those in the second, we get the following figures of merit:

liquid	35
plastic	25
anthracene	3

The organic crystals used in fast counters are stilbene, terphenyl and diphenyl acetylene. The average figure of merit for all of these crystals is about 10; but this seems to vary widely for different crystals, and for some stilbene crystals may be as high as 30. From these figures it would seem that the choice lies between the plastics and the liquids. Decay time measurements in the range of

2 μsec are somewhat uncertain, but the best liquid scintillator is probably superior to the best plastic. With the present state of electronics, this superiority is not very great and in designing a counting system one would have to balance it against the greater ease of handling a plastic.

With organic scintillation counters it is now possible to measure time intervals of the order of 10^{-10} – 10^{-11} sec, by taking a large number of measurements and matching the time delay curve to one obtained with the counters reversed. Since 10^{-11} sec is the time it takes light to travel 2mm in the scintillator, one is near the limit imposed by the size of the detector necessary to intercept a beam of reasonable dimensions.

Solution of a source

It is sometimes desirable to dissolve or suspend a source in a liquid scintillation counter, in order to get the maximum possible counting efficiency. The techniques for doing this are discussed fully in other papers presented at this conference, so we shall not deal further with this application.

Neutron detectors

The liquid scintillator offers the only known way of detecting neutrons above thermal energy with a high efficiency. The neutrons are detected by their interaction with cadmium or boron which is dissolved in the solution, the cadmium as the octoate and the boron as methyl borate. Neutron capture in cadmium gives an average of three or four gamma-rays, with a total energy of 9.1 MeV. A large scintillator is necessary to catch these gamma-rays efficiently, and the resulting pulse is big enough to allow one to discriminate against the high background that one gets in a counter of this type. Neutron capture in boron usually results in 2.3 MeV going to the decay products, and 0.5 MeV to a gamma-ray. Because of the nonlinearity of the scintillator, the alpha particle and Li^7 recoil nucleus together give a pulse equivalent only to 40 keV, based on the response to relativistic electrons. This makes it desirable to use boron in a small counter, with high light collection efficiency, in order to minimize the background.

For low energy neutrons, one may use a small boron-loaded counter. A counter of this type was found to have an efficiency greater than 95% for thermal neutrons, dropping to 50% at a neutron energy of 150 eV.⁵ The mean neutron capture time was 0.5 μsec at the concentrations used, and this was short enough so that the detector could be used to determine neutron energies by measurement of the time-of-flight between the source and the detector. Such a counter has a high efficiency and a fast response, and with pulsed neutron sources can be used to measure the neutron energy and discriminate against gamma-rays produced at the same time as the neutrons. It will not discriminate against gammas from other sources, and is only efficient for low-energy neutrons.

As the neutron energy increases, so does its range in the scintillator, and one needs a large counter to detect high energy neutrons efficiently. Such a counter is shown in Fig. 1.⁶ This was used to measure the distribution of fission neutron multiplicities, for which a high efficiency was essential. The detector is cylindrical, 70 cm long and 70 cm in diameter, and is viewed by 90 2 in. photo-

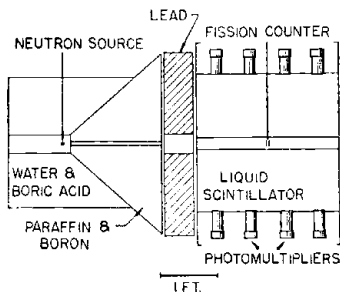


Fig. 1. Liquid scintillation counter for neutron detection.

multipliers. Along the axis is a hollow cylinder, in the center of which is placed the fission counter. Each pulse in the fission counter triggers an oscilloscope sweep, on which are recorded pulses in the detector. There is a prompt pulse due to gamma-rays and proton recoils from the neutrons. Then, as the neutrons are slowed down and captured by the cadmium, each capture gives another pulse. Delays are of the order of $10 \mu\text{sec}$, so that the pulses can be sorted out. Since the ranges of the capture gamma-rays are comparable to the radius of the cylinder, it is unlikely that all the available energy will be deposited in the scintillator. As a result the capture pulse height distribution is roughly uniform from zero to 9.1 MeV. In order to discriminate against the background, it was necessary to reject the smallest pulses, and this resulted in a 12% loss. Five per cent of the neutrons leaked out of the detector, and in 2% of the cases the delays were too long to be recorded on the scope, making the overall efficiency 81% for neutrons of a few MeV.

With a larger detector the efficiency could be higher. However, the efficiency cannot increase more than linearly with the dimensions, and the backgrounds will go up faster than this, so that one does not gain very rapidly.

Large detectors

Apart from financial considerations, the maximum size of a scintillation counter is determined by the transparency of the material to the radiation it emits. The peak of the emission spectrum of POPOP, the most popular wavelength shifter for the large detectors, is at 4200 \AA . At this wavelength the best solvents have optical mean free paths of about 5 m. A measurement of the mean free path of light of this wavelength in the scintillating solution gives a smaller value. However, light absorbed by the solutes will in general be re-radiated, so that this is not true absorption, but scattering; and the main

effect is to increase the distance the light has to travel in reaching the photomultipliers. If one wishes to collect a reasonable fraction of the light emitted in a scintillation and to collect it uniformly over the volume of the detector, the maximum detector size is limited by light absorption to 1 or 2 m.

Toluene and triethylbenzene are equally good solvents, and the choice between them is made by weighing the greater cost of TEB against the fire hazard and toxicity of toluene. Terphenyl is to be preferred as the primary solute, because it is about as good as any other, and much cheaper. POPOP has been widely used as the secondary solute, but there may be others which are equally good.*

The use of large scintillators to detect high energy neutrons was discussed in the last section. Another use that has been made of them is as shields for smaller counters. According to one theory of double beta decay, one expects that one nucleus may change to another by simultaneously emitting two electrons, instead of one electron and one neutrino, as in ordinary beta decay. In experiments to detect double beta decay, the predicted counting rates are so low that one must make every effort to reduce the backgrounds. In one such experiment,⁷ this was done by immersing the detector in the center of a cylindrical scintillation counter 90 cm high and 90 cm in diameter. This not only acted as a cosmic ray shield, but was also expected to reduce the gamma-ray background by detecting some of those gamma-rays which were scattered out of the central region, and which therefore would otherwise have given counts. The energy region of interest was around 4-5 MeV, and in this region it was found that the greatest part of the background was due to neutrons, produced either by the cosmic rays or by nearby neutron sources. These neutrons were captured in the iron of the detector walls, producing gamma-rays of high enough energy to give background counts up to 8 MeV.

A liquid scintillation counter has been constructed at Los Alamos to measure the strength of the interaction between cosmic ray μ -mesons and carbon nuclei. Since it was desired to minimize the background counting rate above 5 MeV, the counter, which is a cylinder 120 cm by 120 cm, was made of polyethylene. This does not reduce the background at lower energies, since the neutrons now capture in hydrogen, giving 2.2 MeV gamma-rays; but it is hoped that it will result in a substantial reduction of the high energy background. Polyethylene does not appear to be attacked chemically either by toluene or TEB; but it is permeable to toluene, so the counter is used with TEB.

A counter for the detection of the neutrino must necessarily be large, because the extremely small interaction cross section makes necessary a large number of target nuclei. The first Los Alamos neutrino counter was later modified to count neutrons, and is the one shown in Fig. 1. The second model, which is perhaps the most impressive scintillation counter built to date, is

*See the chapter on the chemistry of the counting solution.

described by F. Reines in another chapter of this report. Its size is near the limit imposed by light transmission in the solvent.

CALIBRATION

The calibration of a large scintillation counter is often best done by using the cosmic rays. The great majority of the cosmic ray counts are due to single particles at minimum ionization, passing through the detector, preferentially in the vertical direction. These produce a spectrum with a peak, the position of which gives the energy response of the system. Apparatus to measure time delays in the microsecond region may be tested by recording the decays of μ -mesons stopping in the detector. For a neutron source, one can use lead bricks, in which neutrons are generated in cosmic ray stars, and by the capture of μ -mesons. These techniques have been discussed previously.⁸

CONCLUSION

The examples which have been given of the use of liquid scintillation counters in physics do not cover the field, and were picked out mainly because they are the ones with which the author is most familiar. Liquid and plastic scintillation counters have several unique advantages over other types of detectors, the principal ones being high speed and sensitivity, coupled with energy discrimination. Their main drawback is their sensitivity to background radiations.

REFERENCES

- ¹ J. A. NORTHROP and R. A. NOBLES. *I.R.E. Transactions* NS-3, No. 4, 59 (1956); C. EGGLEER and C. M. HUDDLESTON. *ibid*, page 36.
- ² K. H. SUN, P. R. MALMBERG and F. A. PECIAK. *Nucleonics* 14, No. 7, 46 (1956).
- ³ H. PALEVSKY, H. R. MUETHER and A. STOLOVY. *Phys. Rev.* 93, 920 (1954).
- ⁴ R. F. POST. *Nucleonics* 10, No. 6, 56 (1952).
- ⁵ L. M. BOLLINGER and G. E. THOMAS. *Rev. Sci. Instrum.* 28, 489 (1957).
- ⁶ B. C. DIVEN, H. C. MARTIN, R. F. TASCHEK and J. TERRELL. *Phys. Rev.* 101, 1012 (1956).
- ⁷ C. L. COWAN, Jr., F. B. HARRISON, L. M. LANGER and F. REINES. *Nuovo Cim.* 3, 649 (1956).
- ⁸ F. B. HARRISON, C. L. COWAN, Jr. and F. REINES. *Nucleonics* 12, No. 3, 44 (1954).