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Large area liquid scintillation and Čerenkov counters

J. C. BARTON,* C. F. BARNABY,† B. M. JASANI† and C. W. THOMPSON*

*Northern Polytechnic, London, N.7
†Department of Clinical Research, Medical Research Council, University College Hospital Medical School, London

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A counter of 1 m² area and 10 cm deep is described which uses medicinal paraffin as the scintillator solvent. Although only one-third as efficient as other liquid scintillators it is cheap and has a much higher flash point. Using two 5 in. photomultipliers the counter had a resolution of 62% for the cosmic ray peak. For comparison purposes a Čerenkov counter of similar area was studied. Using a wavelength shifter, which improved the performance by a factor of five, this counter has a comparable resolution but needs to be very much deeper than the scintillation counter.

1. Introduction
This paper discusses the performance of counters of area 1 m² or more, which are required for experiments in medical, cosmic-ray and high-energy physics. The large scintillation counters of which details have been published have used either plastic phosphor (Clark 1960, Suga et al. 1961) or p-terphenyl dissolved in toluene (Green 1958). The plastic phosphor is convenient in use but its initial cost is very high and its life uncertain (Barnaby and Barton 1962), whilst the liquids are rather less expensive but have low flash points and create a considerable fire hazard. We have therefore attempted to construct a cheap liquid scintillator using a less inflammable material. Several workers have reported the use of large non-directional liquid Čerenkov counters but the details given, except for the kerosene filled counter of Porter and Sherwood (1956), are inadequate to provide a useful comparison with the performance of the scintillation counter. A similar counter has therefore been constructed and tested and, in particular, the effect of water purity and the use of a wavelength shifter have been studied.

2. Choice of liquid for scintillation counter
It is generally agreed that the fire hazard from xylene or toluene scintillators is so high that their use in large counters must be avoided. Various alternatives have been suggested such as tri-ethyl benzene or deca-hydronaphthalene (available as decalin, Cowan 1961). These liquids provide scintillators of comparable efficiencies and higher flash points but they are expensive, require careful purification and, deca-hydronaphthalene in particular, deteriorate rapidly in the presence of oxygen. We have therefore experimented with a series of other liquids which are cheap, safe and of high transparency, in an attempt to find a more suitable one. It has been found that ordinary medicinal paraffin (liquid paraffin, B.P.) is a possible solvent. Its performance with p-terphenyl and POPOP is shown in figure 1, normalized to the performance of a plastic scintillator (NE 102) measured under similar conditions. It was found that the paraffin would not dissolve more than 1 g l⁻¹ of the p-terphenyl.

Assuming that the plastic phosphor and the other liquid scintillators all have an efficiency of about 200 ev per photon (Currie et al. 1961), we conclude that the maximum efficiency of the paraffin solution is about one-third; i.e. 600 ev per photon. However, the paraffin is much cheaper, and safer than the other liquids, as shown in table 1.

![Figure 1. Variation of the pulse height for single cosmic-ray particles with concentration of p-terphenyl in liquid paraffin (POPOP concentration always 1% of terphenyl concentration). Cell size 10 cm in diameter, 5 cm deep.](image)

Table 1. Comparison of cost and safety of various scintillators

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Cost per litre</th>
<th>Flash point (*c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xylene</td>
<td>£ 1 5 0</td>
<td>17</td>
</tr>
<tr>
<td>Toluene</td>
<td>1 10 0</td>
<td>5</td>
</tr>
<tr>
<td>Tri-ethyl benzene</td>
<td>4 0 0</td>
<td>58</td>
</tr>
<tr>
<td>Decalin*</td>
<td>1 0 0</td>
<td>58</td>
</tr>
<tr>
<td>Medicinal paraffin</td>
<td>0 2 6</td>
<td>229</td>
</tr>
<tr>
<td>Plastic Ne 102</td>
<td>10 0 0</td>
<td>(0†)</td>
</tr>
</tbody>
</table>

* Decalin requires further purification; note also that the recommended primary solute for decalin is PPO, which is twelve times more expensive than the terphenyl used with the other solvents.
† Plastic scintillator is available more cheaply in some other countries.
Another important advantage of medicinal paraffin is that it is relatively inert and has no deleterious effect on Perspex. The samples of medicinal paraffin obtained from Boots Ltd. were found to be 10% more efficient than those from the Esso Petroleum Co. Ltd.

3. Design and construction of counter

We have discussed previously (Barnaby and Barton 1960) the relative advantages of different optical arrangements for scintillation counters. It was shown that the optical efficiency is much better if the photomultipliers are in optical contact with one side of the scintillator and that light guides are necessary to reduce non-uniformity in the response. A rectangular shape was therefore chosen with one photomultiplier at each end (figure 2).

![Figure 2. Diagram of the liquid scintillation counter.](image)

The liquid was contained in a 3-in. Perspex tank, 138 cm \times 74 \text{cm} \times 12 \text{cm}, provided with two short ‘light guide’ extensions, 21 cm long, also made of Perspex. This tank was mounted in a light-tight wooden box on a series of rubber studs to minimize frustration of the total internal reflection. The Perspex tank was viewed by two 5-in. photomultipliers, E.M.I. type 9583 B, mounted one at each end in optical contact. The tank was filled with medicinal paraffin containing 0.8 g 1\text{-}l^{-1} p-terphenyl and 0.008 g 1\text{-}l^{-1} POPOP to a depth of 12 cm; the end sections were filled with pure paraffin, and depended for their effect on total internal reflection.

4. Performance of counter

The measurements required for the proper assessment of a counter of this sort are (i) the uniformity of response over the area of the counter and (ii) the resolution of the counter.

The counter was operated by equalizing the gain of the photomultipliers, adding their outputs in a simple mixing circuit and analysing the amplified output with a single-channel analyser. The uniformity of the counter was measured by operating a cosmic-ray telescope, consisting of two pairs of Geiger counters, each counter 36 cm \times 3.7 cm, in coincidence with the counter. In this way the \(\mu\)-meson peak of the cosmic radiation was obtained using only a small part of the counter. By comparing the pulse heights corresponding to the peaks of the curves the variation in response was obtained. The variation along the length of the counter is shown in figure 3; the variation across the width was everywhere small. It is seen that rather longer light guides would lead to a much more uniform response; these were not provided because it was mechanically inconvenient to increase the overall size. However, even the short light guides are better than none as without them the non-uniformity would be worse. Preliminary experiments with an optical model had predicted rather better response so it is believed that absorption of the scintillation light in the liquid is partly responsible for the non-uniformity.

The resolution is normally obtained by measuring the width of the \(\mu\)-meson peak at half amplitude of the differential curve for the cosmic-ray background in the whole counter. Because of the asymmetry of the curve, due to more complex cosmic-ray events, we have in fact used twice the distance from the energy at half amplitude on the low energy side to the energy at the peak.

In order to eliminate pulses from sources such as photomultiplier noise and Čerenkov radiation in the light guides, the two photomultipliers were operated in coincidence and the coincidence circuit output used to gate the differential analyser. In this way the resolution was found to be 62% (figure 4).

![Figure 3. Results of the uniformity experiment (using cosmic-ray telescope).](image)

![Figure 4. Integral and differential pulse height distributions of cosmic-ray background in the liquid scintillation counter with the photomultipliers in coincidence.](image)

A, integral pulse height distribution; B, differential distribution. Channel width 4 V.

Several factors contribute to this resolution: (i) variations in the directions and energies of cosmic rays entering the counter;
(ii) the Landau contribution, due to the statistical variation in the energy absorbed from the $\mu$-mesons by the liquid phosphor, of about 20%;

(iii) the variation in the proportion of light collected from different parts of the phosphor;

(iv) the statistical spread in the number of photoelectrons produced by the photomultipliers.

It is important to try to estimate the number of photoelectrons which result from the traversal of the counter by a cosmic-ray particle. This can be done in two different ways, though both are very rough. Firstly, if the width of the peak were due solely to the statistical spread in the number of photoelectrons, then it is possible to calculate that the most probable number liberated by each cosmic ray would be about 20. Making reasonable allowances for the other effects we estimate that this number should be increased to between 30 and 40. Secondly, since the most probable energy-loss of $\mu$-mesons in the phosphor was 18.7 MeV the number of photons produced was $18.7 \text{MeV}/600 \text{ev} = 3.1 \times 10^4$ photons. Roughly half of the emitted light is lost into the four escape cones through the sides of the counter. Of the remainder we can estimate that about a half makes such long paths and so many reflections that it is also lost. Hence only one-quarter of the emitted photons reach either end of the counter, of which areas the photocathodes cover one-tenth. The total number of photoelectrons produced should therefore be $3.1 \times 10^4 \times 0.25 \times 0.1 \times 0.1 = 80$ or 80 photoelectrons, where it is assumed that the effective quantum efficiency of the photocathodes is 10%. Figure 3 shows that the response of the central part of the counter is little more than half that of the extreme ends so that, allowing for additional loss in the light guides, the two estimates appear consistent.

Experiments were carried out using other depths of liquid. With only 8 cm of liquid the width of the cosmic ray peak increased to about 90% and with still smaller depths the performance fell off very quickly.

The possibility of replacing the liquid filled light-guide with a hollow reflecting guide was also examined. This was simulated by emptying the end sections and wrapping them with ordinary aluminium foil. The resolution again deteriorated from 62 to 80%, but the simplicity of the suggested arrangement may make it useful. Also it has the advantage that there are no longer any Čerenkov pulses produced by mesons traversing the light guides, which in some cases are comparable to the true scintillation pulses.

5. Comparative experiments with large Čerenkov counter

In view of the smaller efficiency of the liquid scintillation counter using paraffin it was thought that a comparison with a Čerenkov counter of the same area would be useful. A non-directional Čerenkov counter was constructed by lining a wooden box with white Darvic (I.C.I. Ltd.). The sensitive area of the counter was 100 cm x 100 cm and the depth could be varied up to 100 cm. The sensitive volume was viewed by a single 5 in. photomultiplier placed at the centre of the top of the counter. The arrangement was such that the photomultiplier and the top Darvic lined face of the box could both be immersed in the water. It was found that ordinary London tap water gave a response so poor as to be useless in practice. Water was therefore passed through ion-exchange columns and its purity assessed by conductivity measurements. In this way it was found that a significant improvement was obtained for purities up to distilled water standard (≈7 µmhos cm$^{-1}$) but beyond that the improvement was negligible. On the other hand mixing the sodium salt of 2-naphthylamine-6:8-disulphonic acid (sometimes known as amino-G acid) in distilled water at a concentration of 0.1 g l$^{-1}$, to act as a wavelength shifter and to make the light emission isotropic (Heiberg and Marshall 1956), gave an improvement in response of a factor of about 5. Higher concentrations of up to 0.2 g l$^{-1}$ gave no further improvement in response. Using a depth of 100 cm and the wavelength shifter dissolved in distilled water at a concentration of 0.1 g l$^{-1}$ the uniformity was measured by moving a Geiger counter telescope, operated in coincidence with the counter, over the counter area. The variation in response, measured by the position of the $\mu$-meson peak in the differential pulse height distribution of the cosmic-ray background, was not more than ±12%. The resolution of the $\mu$-meson peak in the whole counter was 50% if the curve was measured only on the high energy side of the cosmic-ray peak. The resolution is not well defined for a counter of this shape as those non-vertical particles which make short path lengths in the counter cause a long plateau below the peak, as seen in figure 5.

![Figure 5. Integral and differential pulse height distributions of cosmic-ray background in the Čerenkov counter of 100 cm depth.](image)

$A$, integral pulse height distribution; $B$, differential distribution. Channel width 1 v.

The results at smaller depths are shown in table 2. It is seen that the performance is moderately good for 50 cm but becomes very poor for 25 cm.

<p>| Table 2. Performance of Čerenkov counters of various depths |
|-------------------------------|--------------|-----------------|-----------------|</p>
<table>
<thead>
<tr>
<th>Liquid</th>
<th>Depth of liquid (cm)</th>
<th>Resolution (%)</th>
<th>Non-uniformity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>De-mineralized water</td>
<td>50</td>
<td>$\pm 25$</td>
<td>$\sim 10$</td>
</tr>
<tr>
<td>De-mineralized water +</td>
<td>25</td>
<td>183</td>
<td>$\pm 40$</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>95</td>
<td>$\pm 25$</td>
</tr>
<tr>
<td>amino acid</td>
<td>75</td>
<td>60</td>
<td>$\pm 25$</td>
</tr>
<tr>
<td></td>
<td>100</td>
<td>49</td>
<td>$\pm 12$</td>
</tr>
</tbody>
</table>

$N = \text{No. of photoelectrons per $\mu$-meson traversal.}$

In order to provide a direct comparison between the light output of the liquid scintillator and of the Čerenkov liquid, the solution containing the wavelength shifter was used to fill the Perspex box previously used for the scintillator. With other conditions the same the solution was found to give one-third as much response as the scintillator.
6. Discussion of results

The performance of both the scintillation counter and the Čerenkov counter described above are comparable with, or better than, others which have been reported so that a detailed comparison will not be made. The improvement on using a wavelength shifter in the Čerenkov counter is more striking than was found by Heiberg and Marshall (1956) or Saito and Suga (1959) but it is not surprising as these authors were studying much smaller counters and used a different geometric arrangement.

It is seen that the performance of either type of counter is adequate for many experiments in cosmic-ray and high energy physics, since both counters give clear meson peaks in their differential pulse height distributions. The deeper Čerenkov counters show a large proportion of pulses due to oblique particles traversing the counter with short track lengths, which may be a nuisance in some applications, although the 50 cm deep counter would appear adequate for most purposes. The shape of the scintillation counter is certainly the more convenient as two or more could be placed directly one above the other; an obvious application is in the construction of 'transition chambers' of the type described by Tanaka (1961). For each counter the total cost is not more than double that of the photomultipliers required. This makes the scintillation counter more expensive although the use of two photomultipliers does enable noise pulses to be eliminated by means of coincident operation.

The long-term stability of these counters is not yet well determined. The paraffin scintillation counter has been operated for several months and has not deteriorated noticeably (less than 10%); the same solution has been used for three separate fillings of the counter—more efficient liquid scintillators would certainly suffer from such treatment. Also bubbling nitrogen through the scintillator did not lead to any appreciable improvement. It seems possible that the more viscous liquid paraffin is inherently less likely to dissolve oxygen. We have at present no experimental evidence on the stability of the Čerenkov solution. Saito and Suga (1959) have reported a significant deterioration in the presence of oxygen but when the solution was kept away from light and air its life was much longer.

In their present form neither of these counters is suitable as a γ-ray counter as the number of photoelectrons is only 2 per mega electron volt for the scintillation counter and 0.5 per mega electron volt for the 50 cm Čerenkov counter. For the scintillation counter about ten times the photocathode area would be necessary to provide reliable detection of γ-rays of 500 kev. Another possibility is to mix a proportion of a more efficient solvent, such as tri-ethyl benzene, with the paraffin. Preliminary experiments on a smaller counter suggest that almost a two-fold improvement in efficiency can be obtained with only 10% of tri-ethyl benzene.

The design of still larger counters will depend critically on the transparency of the liquid used. Although spectrophotometer measurements show that the absorption length in pure paraffin for light of 4000 Å is several metres, the study of non-uniformity (figure 3) suggests a much smaller value for the solution.

A secondary solute with an emission maximum at a wavelength greater than that of POPP may be necessary for larger counters. In any case it seems doubtful whether the cost per square metre will be much less than for the counters described, so it may be simpler to run a number of these counters in parallel.

Acknowledgments

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References


