

A COMPARISON BETWEEN LARGE VOLUME LIQUID AND PLASTIC SCINTILLATORS

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INTRODUCTION

WE have been concerned in Leeds since 1949 with the measurement of gamma-radiation emitted from living human subjects. Our first apparatus^{1,2} was based on large high-pressure ionization chambers in a surface laboratory; it enabled most γ -emitting isotopes to be assayed at levels well below the maximum permissible body burdens. In a 2 hr run, the natural gamma-ray emission from uncontaminated persons could be measured with a probable error of ± 15 – 20% . Generally, these 'natural γ -rays' are due almost entirely to the presence of the K^{40} isotope in body potassium and some 300–500 photons of 1.46 MeV energy are emitted per second from adults.

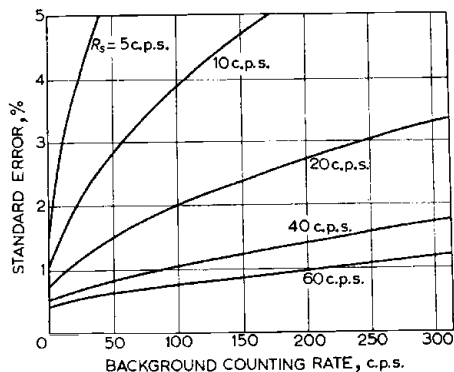


Fig. 1. Illustrating the connection between standard error, the response-rate (R_s) from the radiation source and background rate. Curves calculated for 15 min observation of source and 1 hr background.

The need for a much more sensitive apparatus became apparent and our clinical colleagues pointed out the usefulness of an apparatus which would allow the total potassium content of a patient to be measured swiftly and accurately. A standard error of about $\pm 2\%$ is considered desirable, to be obtained from a measurement of not more than 30 min duration. In Fig. 1,

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the connection is illustrated between counting statistical error, the response rate from the subject, and background rate; these curves are based upon a 15 min subject observation and a 1 hr background. Above a response rate of about 20 counts/sec, the statistical error is fairly independent of the background rate (providing this latter is constant), but at lower response rates there is a marked dependence of error on background, and sensitivity is greatly aided by a rigorous reduction in background.

The high-pressure ion-chamber apparatus suffered from another limitation, that is, its inability to provide information about the quality of the radiation emitted. Since the nature of radioactive contamination is not always known, it was felt that a new and more sensitive apparatus should at the same time be capable of providing spectrometric information. When MILLER and MARINELLI⁸ discovered Cs¹³⁷ to be present in human subjects, the need for spectrometric information was further emphasized. About 200 photons/sec of 0.66 MeV are emitted from contemporary (1957) Cs¹³⁷ burdens, as compared with the 300–500 photons/sec of 1.46 MeV from body potassium.

In 1952, our sensitivity criteria narrowed the choice of technique which could be exploited and the only practical solution appeared to lie in the use of large volume organic scintillators.

To allow the maximum flexibility it was decided to adopt a unit form of construction since this has the following advantages—(a) the number and position of units can be altered to accommodate a seated or supine subject; (b) by examining the output from each unit separately, a crude localization can be effected; (c) a spare unit held in reserve eases maintenance difficulties; (d) development work is facilitated.

Design of test unit (Fig. 2)

This was designed originally to accommodate a liquid although it later proved equally adaptable to housing an oval block of plastic scintillator. The oval scintillator tank and lid are aluminium castings enclosing a volume approximately $20 \times 10 \times 6$ in. A gasket, consisting of an asbestos core with a Teflon envelope to give toluene resistance, provided the liquid seal between lid and tank. Cold setting, toluene resistant, Shell Epikote Resin 815 with Curing Agent T was used to seal the two glass windows, the gas stopcocks, the filling orifice and the copper bellows. The gas stopcocks were fitted to enable liquid scintillators to be freed from oxygen and its quenching effects by flushing with nitrogen. To avoid the possibility of gas bubbles trapped at the windows affecting optical coupling, the unit was completely filled with liquid. Volume changes were accommodated by the copper bellows.

With the plastic scintillator, the two 5 in. diameter photomultipliers were optically coupled directly to its surface using a silicone oil (Midland Silicones 710). When liquids were used it was however necessary to insert a $\frac{3}{4}$ in.

Lucite light guide between the viewing window and the photomultiplier; this enabled the photomultipliers to be removed by sliding them sideways clear of the ridges on the aluminium casting. The light guides, which were provided with aluminium foil reflectors on their cylindrical surface, introduced a 9% reduction in pulse height.

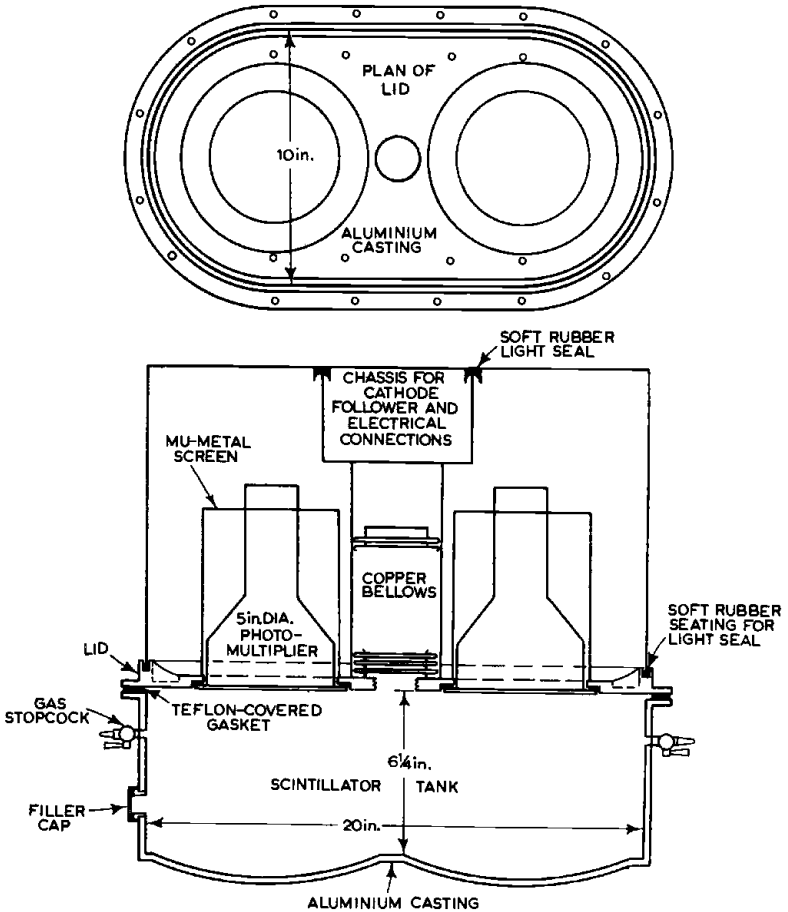


Fig. 2. Scintillation test unit. Top: plan view of lid (aluminium casting). Below: simplified section through unit showing scintillator tank and accessories, light cover and photomultipliers.

The output from the two paralleled 5 in. diameter Du Mont Type 6364 photomultipliers was fed via a cathode follower and a modified FRANCIS, BELL, HARRIS⁴ amplifier to a Sunvic pulse amplitude analyzer operated on its sixty channel range.

During the design and construction of these units, Nuclear Enterprises Ltd. produced a lead-loaded liquid scintillator containing 5% of lead by weight and also made available large plastic scintillators at a cost competitive with conventional liquid scintillators. It was decided to test both of these in addition to a conventional liquid scintillator in our units and a plastic block was commissioned to fit inside the casting.

Energy resolution considerations

In comparing the performances of the three scintillators, the chief interest lay in their spectral responses. Providing that pulse height is not too dependent upon the position of the scintillation event relative to the photomultiplier apertures, then the chief contribution to pulse height spread will be statistical fluctuations in the number of electrons released from the photocathodes. Since the available photocathode area is usually restricted by considerations of simplicity and economy, the yield of photoelectrons for a given event will be governed by the product of (a) the efficiency of the conversion of initial electron energy to technical fluorescent photons and (b) the fraction of these photons intercepted by photocathode surface. A measure of the technical efficiency of a scintillator is given by comparing the pulse height obtained with small volumes of the scintillator with similar small volumes of anthracene under conditions where reflectance and transmittance (see later) are both close to unity. According to Nuclear Enterprises, the pulse height for small volumes of NE 101 plastic scintillator is 50% of that obtained from anthracene, and for NE 314 lead-loaded scintillator the corresponding figure is 25%; deoxygenated solutions of PPO and POPOP in toluene should give a pulse height not less than 70% of that from anthracene (see e.g. PRINGLE *et al.*⁴).

When the fraction, p , of the total wall area covered by photocathode is small, the average fraction, F , of fluorescent photons falling on photocathode area is given by the following approximate formula:

$$F \simeq \frac{p}{1 - rt}$$

Where $r = (1 - p)r^1$, r^1 being the average reflectance of walls to technical photons,

t = average fraction of technical photons transmitted through the scintillator between successive reflections.

When using the plastic scintillator, the white painted walls of the casting were smoked with magnesium oxide to give a reflectance of 0.98 in the wavelength region of interest. A large number of toluene-resistant white paints were tested with an EEL reflectometer using the blue filter which gave a spectrum with peak intensity at about 4300 Å. Surfaces were tested dry and when wetted with toluene. The highest reflectance value we were able to

obtain was 0.88 (for both wet and dry surfaces) with rutile titanium dioxide pigment in a Group 37 enamel supplied by International Paints Ltd. This high value was obtained on an aluminium test panel, but on the pitted aluminium casting a rather lower average reflectance of 0.83 was found. It is not

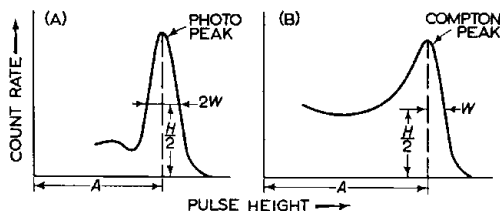


Fig. 3. Types of monochromatic γ -ray spectra obtained with (a) NaI (TI) crystal, showing photo-peak and (b) organic scintillator showing Compton peak.

possible to measure t directly in a scintillator due to scattering by absorption and re-emission processes. Mean free paths of at least 5 m may be obtained in pure toluene⁶ but this value will be reduced by the presence of primary and secondary scintillators. The lead-loaded solution had a slightly brownish appearance, and in our large test units this undoubtedly was responsible for t values much less than unity.

Experimental results

All response curves were obtained with the units inside a steel room with 9 in. thick walls and a 12 in. floor.

The figure of merit we have used to compare the spectral performance of organic scintillators is the 'half-resolution'. We define 'half-resolution'

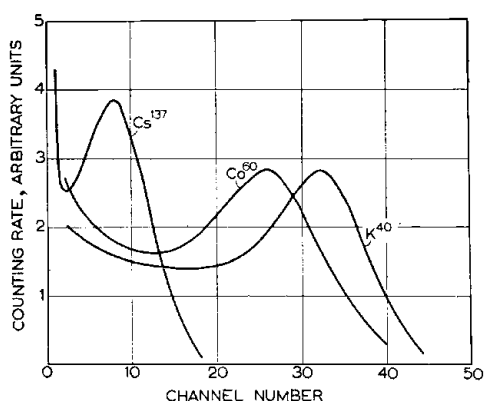


Fig. 4. Spectral response curves obtained with oxygen-free liquid scintillator, (3.7 g 2,5-diphenyloxazole (PPO) and 0.1 g 1,5-di-2-(5-phenyloxazolyl)-benzene (POPOP) per litre toluene) in $20 \times 10 \times 6$ in. test unit.

TABLE 1

Radiation source and photon energy in MeV	Plastic scintillator			Liquid scintillator		
	Pulse height of peak (arbitrary units)	Expected peak* position	Half-resolution %	Pulse height of peak (arbitrary units)	Expected peak* position	Half-resolution %
Cs ¹³⁷ , 0.66	9.7	10.1	26	12.0	12.5	33
Co ⁶⁰ , 1.17, 1.33	21.3	21.9 (approx.)	26 (combined peak)	26.6	27.2	24 (combined peak)
K ⁴⁰ , 1.46	26.1	26.1 (refce.)	14.5	32.4	32.4 (refce.)	16
Na ²² { 1.28 0.51				29.2 8.4	28.0 8.6	18.5 43

*Calculated on the assumption that the ratio of the pulse height of peaks corresponds to the ratio of the maximum energy of the Compton electrons. The K⁴⁰ peak was used as reference.

(see Fig. 3) as $100 W/A\%$ where W is the 'half-width' on the high energy side of the asymmetric Compton peak at half peak height, and A is the pulse height of the peak.

Spectral response curves obtained with the conventional liquid scintillator (3.7g PPO and 0.1g POPOP per l. of toluene, deoxygenated) are shown in Fig. 4. Removal of oxygen by flushing with nitrogen produced a 34% increase in pulse height.

Similar curves for the NE 101 plastic scintillator are given in Fig. 5. Results are compared in Table 1. It will be seen that the plastic gives the slightly better performance, but in neither case are the Co^{60} twin Compton peaks resolved.

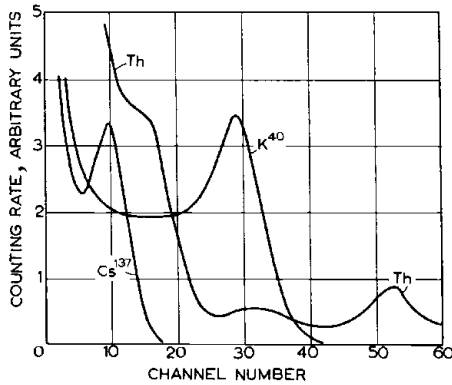


Fig. 5. Spectral response curves obtained with $20 \times 10 \times 6$ in. plastic scintillator (Nuclear Enterprises (G.B.) Ltd. NE 101) in test unit.

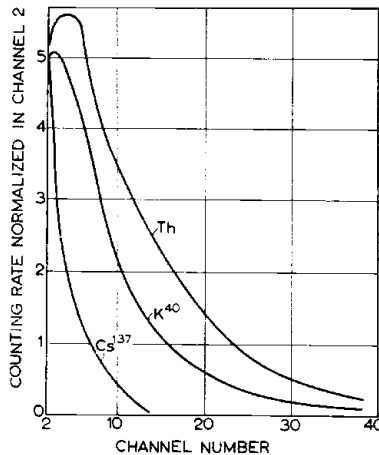


Fig. 6. Spectral response curves for oxygen-free lead loaded liquid scintillator (Nuclear Enterprises (G.B.) Ltd., NE 314) in $20 \times 10 \times 6$ in. test unit. Counting rates for Cs^{137} , K^{40} and 'old' thorium are normalized in channel 2.

The disappointing curves obtained with the deoxygenated NE 314 lead-loaded scintillator are illustrated in Fig. 6. The counting-rate for the different isotopes has been normalized in channel 2. No Compton or photo-peaks were obtained and the pulse height was of the order of one thirtieth of that obtained from the conventional liquid and the plastic scintillator. Unless the scintillator was poisoned, it seems that the chief cause of this large decrease was light absorption produced by the coloured lead compound. Oxygen removal produced an increase in pulse height of approximately 15%.

Application to body γ -ray monitors

As a result of these comparisons, it was decided to make plastic scintillators the basis of a sensitive body γ -ray monitor, relying on a NaI (TI)

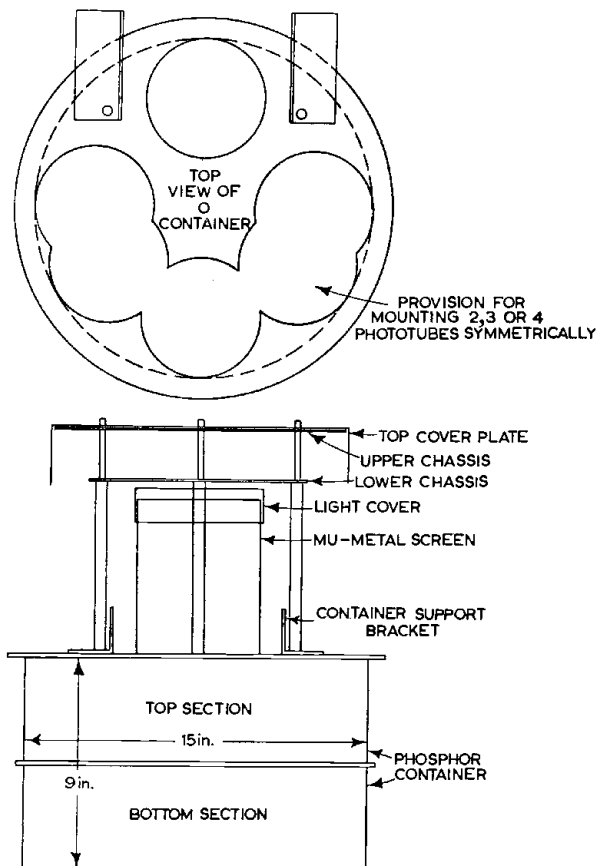


Fig. 7. View of scintillator unit to accommodate a plastic cylinder 15 in. dia. \times 9 in. The top section is designed for symmetrical mounting of two, three or four 5 in. diameter photomultipliers. Blanking disks are fitted to cover the apertures not in use and to provide a reflecting surface opposite the scintillator. Construction is in thin sheet steel.

crystal for supplementary spectral information. Three NE 102 plastic scintillators, 15 in. diameter and 9 in. thick have been prepared and these can be viewed by two, three or four 5 in. diameter photomultipliers arranged symmetrically (see Fig. 7) or one 16 in. diameter Du Mont photomultiplier, type K1328. So far experiments have been carried out with three and four, 5 in. diameter photomultipliers. The 'half-resolution' for the K^{40} Compton peak using three photomultipliers was 18.5% and with four was 15%. These results are slightly inferior to those anticipated on the basis of equal coefficients of transmittance in NE 101 and 102 scintillators; the manufacturer's claim was assumed that the light output of NE102 is 12–13% greater than that from NE101, although in our context this claim will apply only to small scintillator volumes.

Both the oval, $20 \times 10 \times 6$ in. plastic scintillator unit and a cylindrical, 15 in. diameter \times 9 in. unit with four photomultipliers, were tested for response to body potassium by observing that portion of the K^{40} spectrum above the Cs^{137} contribution. Units were placed about 3 in. above the thighs and 1 in. from the chest of the sitting subject. The potassium response for males with the oval unit was about 7 counts/sec above a background rate of 12.2 counts/sec; with the cylindrical unit, the typical male response was 12 counts/sec above a background of 20 counts/sec. Under these conditions, either unit would yield a counting statistics standard error for the potassium response of about $\pm 2\%$ for a 15 min subject observation and a 1 hr background. In the final assembly it is intended to use three or four units disposed in such a way that variations in specific response with the size and shape of subject are minimized.

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