FIRST PART OF ROUND-TABLE ON 'CHEMISTRY OF THE COUNTING SAMPLE'

F. NEWTON HAYES

Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

THE liquid scintillation counter contains a light detecting system made up of reflectors, photomultipliers and an electronic assembly. These were covered during the session on instrumentation.

That which for the moment is called the counting sample is the chemical system in the liquid scintillation counter which produces light. It may range in complexity from an unadulterated liquid scintillator to one so loaded as to contain the source of exciting radiation and even an agent which gives the whole system a gel structure.

This round-table may be of most value to those who do internal sample counting where the liquid scintillator is continuously and variably adulterated. But, for this discussion to explore the complex, it must adequately develop the simple systems and thereby prove of use to those who use external sample counters.

TABLE 1

Components of the Liquid Scintillator

Solvent (S)	
if $S=S_1+S_2+S_3$ . . . . , $S_1>S_2>S_3$ in concentration	
Solute (σ)	Primary solute (σ_1)
	Secondary solute (σ_2)
	Neutron capture solute (σ_n)
	Gamma conversion solute (σ_γ)

TERMINOLOGY

The system of terminology used at Los Alamos, which applies to liquid scintillators and their uses in counting, is shown in the first two figures.

Table 1 lists the components of the liquid scintillator which is considered to be a combination of solvents and solutes necessary to provide proper scintillating action. The term solvent is used in the broadest sense to include both mechanistic and solubility functions. If designations are desired for a multi-component solvent system, primary and secondary, etc. are used in the order of solvent abundance. The solutes are called primary and secondary when two are used, on a mechanistic basis from the order in energy transfer. To qualify as a solute, the substance must be an emitter in the energy transfer sequence whose emission spectrum[1] is of long enough wavelength to give a moderate response with an S-11 photocathode.

Solutes which improve the response for certain uncharged particles might be called the neutron capture solute or the gamma conversion solute, examples of these will be given later.

The term phosphor has been applied to both the liquid scintillator and the scintillation solute. This is disliked because of any possibility of implication of similarity between liquid scintillation and phosphorescence.

The term fluor in place of liquid scintillator seems inappropriate since fluorescence does not require a solvent transfer mechanism such as found in liquid scintillators. The term fluor may, however, be used in place of scintillation solute.

Descriptive terms, such as spectrum shifter and wavelength shifter, have been applied to the secondary solute but should be restricted to the grade of second order synonyms; in other words, they should be used in a parenthetical sense.

Table 2 shows some terms for internal-sample counting. The gelling agent is that material added to a liquid scintillator to produce a gel scintillator. The radioactive sample which is dissolved in a liquid scintillator is called the sample solute. The insoluble radioactive sample which is suspended in a gel

TABLE 2

Nomenclature for Internal-sample Counting

Liquid scintillator + gelling agent → gel scintillator		
Sample solute		Suspended sample
	Counting sample	
	Internal standard	

scintillator is called the suspended sample. The system which results from either of these last two additions is the counting sample.

When the count rate from a counting sample must be translated into a disintegration rate and this must be carried out within the sample, that further material which is added is called the internal standard.

A METHOD FOR SCINTILLATOR COMPARISONS

The term relative pulse height will occasionally be referred to in this discussion. It is used for scintillator comparison. The heart of an apparatus for determining pulse heights is shown in Fig. 1.

A Cs^{137} source is deposited on Mylar film, which is attached to a hemispherical reflector whose concave surface is coated with evaporated alumin-

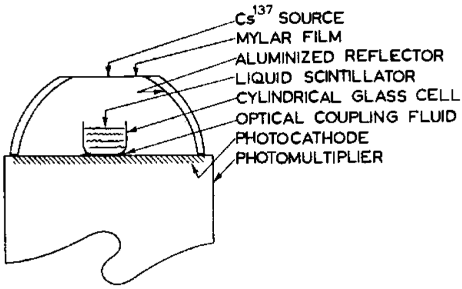

FIG. 1. Optical system for pulse-height determination.

um. Energy from the source excites the liquid scintillator which is contained in a cylindrical glass cell. For different special applications, the cell may be made of quartz, Pyrex or any other of a great variety of glasses. An optical coupling fluid is between the bottom of the cell and the top of the envelope of the erect flat-face photomultiplier. The pulse height is obtained by analyzing the discriminator voltage level resulting from the resolved line spectrum of the internal conversion electron from the excited Ba^{137} daughter of Cs^{137}.

LIQUID SCINTILLATION SOLVENTS

The best liquid scintillation solvents[2, 3] are alkylbenzenes. Below are shown the most notorious examples of these. Toluene is liked because it is efficient, cheap, transparent and available. Phenyl*cyclo*hexane and xylene are sometimes used instead of toluene. The most understandable argument for the use of these is local preference, since pure samples of all the solvents in this figure give essentially the same pulse height. Both xylene and triethylbenzene are commercially offered as mixtures of positional isomers. The most abundant isomer in each of these cases is shown.

Triethylbenzene is favored in certain installations where fire hazard is of great concern. It was used both as scintillation solvent and hydrogen source in the experiment on the identification of the free neutrino.[4]

p-Cymene has a specialized interest in that it would be a poor choice as a solvent for low-level counting with an added C^{14} sample solute. Commercially available *p*-cymene contains natural C^{14} at the level of about 13 disintegrations/min g of cymene. This has proved useful in a study[5] of contemporary natural C^{14}, since *p*-cymene can be synthesized from many natural oils derived from plants and trees with wide geographic distribution. In a 90 ml volume yielding 50% C^{14} counting efficiency, a 'live' *p*-cymene sample will give about 600 counts/min. The corresponding synthetic 'dead' *p*-cymene background is about 60 counts/min.

Some useful binary solvents are shown below. The emphasis in the upper set is to make a better chemical solvent out of a good scintillation solvent, and the emphasis in the lower set is to make a better scintillation solvent out of a good chemical solvent.

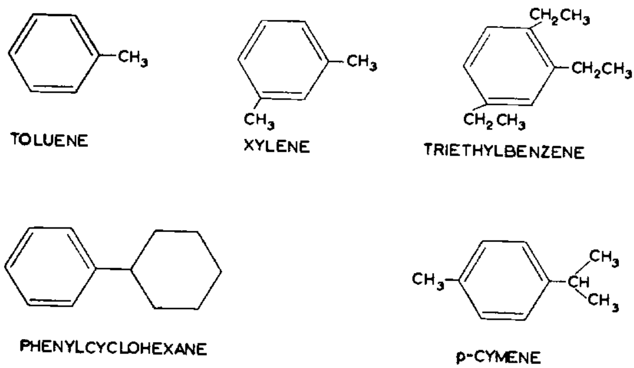

Fig. A

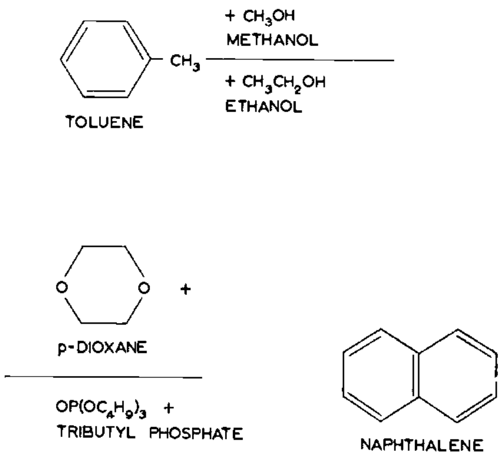

Fig. B

Toluene and methanol were used in the first part of the Los Alamos neutrino experiment[6] to allow dissolution of the neutron capture solute, cadmium propionate. One of the first systems for tritium water counting was toluene and ethanol.[7]

The *p*-dioxane and naphthalene combination was pioneered by KALLMANN[8] and is now used in the preferred method for counting with water systems.[9] Tributyl phosphate and naphthalene are useful after an extraction procedure wherein tributyl phosphate extracts a radioactive material from an aqueous solution.

In any liquid scintillator system, purity of the components is a very important consideration, because of possible quenching or light absorption due to contaminants. The solvent, which is by far the most abundant component in a liquid scintillator, should receive special attention with regard to procurement and maintenance of requisite purity.

REFERENCES

1 D. G. OTT, F. N. HAYES, E. HANSBURY and V. N. KERR. *J. Amer. Chem. Soc.* **79**, 5448 (1957).

2 H. KALLMANN and M. FURST. *Nucleonics* **8** (3), 32 (1951).

3 F. N. HAYES, B. S. ROGERS, and P. C. SANDERS. *Nucleonics* **13** (1), 46 (1955).

4 C. L. COWAN, Jr., F. REINES, F. B. HARRISON, H. W. KRUSE and A. D. MCGUIRE. *Science* **124**, 103 (1956).

5 F. N. HAYES, E. C. ANDERSON and J. R. ARNOLD. Paper P/68 *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy*, **14**, 188, United Nations, New York, (1956).

6 C. L. COWAN, Jr., F. REINES, F. B. HARRISON, E. C. ANDERSON and F. N. HAYES. *Phys. Rev.* **90**, 493 (1953).

7 F. N. HAYES and R. G. GOULD. *Science* **117**, 480 (1953).

8 H. KALLMANN, M. FURST, and F. H. BROWN. *Nucleonics* **13** (4), 58 (1955).

9 W. H. LANGHAM, W. J. EVERSOLE, F. N. HAYES and T. T. TRUJILLO. *J. Lab. Clin. Med.* **47**, 819 (1956).