SECTION I

MODERN CONCEPTS IN LIQUID SCINTILLATION COUNTING

Chapter 1

Applied Liquid Scintillation Counting

D.L. Horrocks

Beckman Instruments, Inc., Fullerton, Ca 92634, U.S.A.

INTRODUCTION

The scintillation process in solutions is comprised of several different steps. The ultimate result of the scintillation process is the conversion of part of the energy from some nuclear transformation into a number of photons. The number of photons is related to the energy deposited in the solution and the type of particle which imparts that energy. The excited scintillator molecules which produce the observed fluorescence determine the energy (wavelength) distribution of the emitted photons. The number of photons is also dependent upon such factors as the amount of scintillator solute present in the solution, the presence of certain impurities (quenching), the optical transmission properties (colour quenching) and scintillation efficiency of the total system.

Composition of a typical scintillator solution

Most liquid scintillator solutions are composed of three main parts; the solvent(s), the solute(s) and the sample. In special applications there may be more than one solvent for such purposes as increasing the solubility of the sample or increasing the energy transfer probability. Also it is often necessary to use more than one scintillator solute in order to shift the wavelength spectrum of the photons to more efficiently match the spectral response characteristics of the photon measuring device (multiplier phototube) or to reduce the effects of a high extinction coefficient for a given wavelength distribution by one of the components in the solution.

To understand the role of the solvent(s) and the solute(s) in the scintillation processes it is necessary to study the solutions without foreign substances present, such as the sample. These types of studies can be done using radioactive sources external to the solution for the production of excited molecules. Another type of external source is an ultra-violet radiation source.

The solvent plays at least two very important roles in the scintillation process. The solvent molecules accept a part of the energy from the exciting source and, in so doing, excited solvent molecules and ionised solvent molecules are produced. Upon recombination of solvent ions with electrons some of the solvent molecules are left in an excited state. The solvent molecules also act as an efficient means of transferring excitation energy through the solution to the fluor molecules. This solvent-solvent transfer process is very efficient. The solvent molecules themselves are very poor scintillators for several reasons, some of which are as follows:

(a) Solvent molecules have low probabilities for the emission of photons, i.e. a low quantum efficiency.
(b) The energy (wavelength) distribution for the photons emitted by the solvent molecules is of a range (2000-3000 Å) where most multiplier phototubes have reduced sensitivity.
(c) The probability of re-absorption of emitted photons by the solvent itself (self-absorption) is high due in part to the high solvent concentration.
(d) The life of the excited solvent molecule in the fluorescing state is relatively long (about 30 ns) which renders it very vulnerable to competitive processes which do not lead to fluorescence, i.e. quenching or energy transfer.

The role of the solute(s) in this process is to trap the excitation energy of the solvent molecules and efficiently release part of that energy in the form of photon energy. Whereas solvent molecules have low fluorescence efficiencies (0.07 to 0.40), the commonly used solute molecules have fluorescence efficiencies near 0.90. The probability of solute self-absorption is greatly reduced by the low solute concentration in the scintillation solution. The probability of competitive processes, such as quenching, are greatly reduced by the short decay time of these solute excited molecules, usually between 1 and 2 ns.

At the relatively low concentration of 3 to 10 g/litre, the solute molecules are capable of trapping 100% of the solvent excitation energy, i.e. the energy transfer is 100% efficient. Before the excited solute molecule emits a photon there is an intramolecular de-excitation by a non-radiative process by which a small part of the excess energy is lost in the form of vibrational energy of the molecule. All fluorescence occurs between the zero vibrational level of the first excited singlet energy level of the solute molecule and any of several vibrational energy levels of the ground state of the molecule. Figure 1 shows some of the many processes which can occur during the scintillation process.

Excitation processes

As an ionising particle (electron, alpha particle, proton, etc.) passes through a scintillator solution, slows down and is finally stopped, many different events occur. Figure 2 depicts a highly simplified representation of some of the more important events. A more detailed description of these events is given by Laustriat.[1] The largest part of the particle energy is expended in the form of kinetic (thermal) energy of the solution molecules and does not lead to the production of excited solvent molecules. Along the particle track ions, excited molecules, free radicals and secondary electrons are among the different products formed. The secondary electrons will produce their own track of events, which have been called 'spurs'. Many of the excited molecules that are produced along the track of the primary particle interact with other species such as free radicals, ions and non-excited (ground state) molecules. These types of interactions lead to the non-radiative de-excitation of the excited molecules. This is referred to as 'track quenching'.

Both ions and excited molecules can lead to fluorescence. An appreciable part of the fluorescence (about 60%) is the result of ion recombination.[2,3] Most

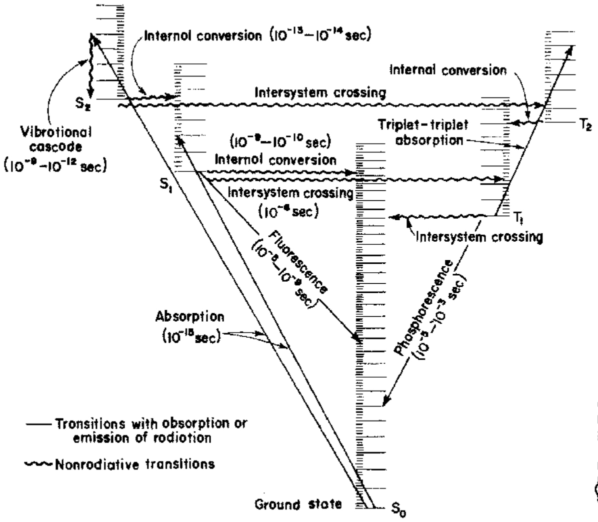

Fig.1. A modified Jablonski diagram showing the various energy levels and the processes of energy absorption and release of energy (radiatively and non-radiatively) by an organic molecule.

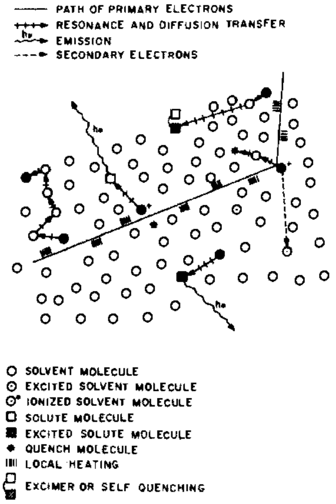

Fig.2. A simplified diagram of some of the processes which occur in a liquid scintillator solution (solvent and solute) when an energetic electron interacts with the molecules of the solution.

of the primary excitations lead to excited solvent molecules in their upper excited singlet energy states. The upper excited singlet states rapidly undergo an internal conversion by non-radiative processes to give finally the first excited singlet energy level. It is this state which is responsible for the energy transfer and migration. However, recent studies have shown that some energy transfer can occur from upper excited states of the solvent molecules.[3,4]

Energy transfer between solvent molecules involves neighbouring solvent molecules. Two theories are presently in debate as to the mode of the solvent-solvent energy transfer. Birks et al.[5] explain the transfer as being due to the formation and dissociation of solvent excimer molecules with the energy transfer to the previously unexcited solvent molecule when the excimer breaks apart. Voltz et al.[6] theorise that the energy actually jumps from one solvent molecule to its neighbour by a non-radiative process.

The solvent-solute energy transfer is quite different. First, it is not necessary that the solvent (donor) molecule and the solute (acceptor) molecule be adjacent. Secondly, the intra-molecular partial de-excitation (vibrational relaxation) of the excited solute molecule traps the energy on the solute molecule, thus preventing the reverse transfer process (solute to solvent). Finally, the solvent-solute transfer occurs by a non-radiative process at moderate solute concentrations.[7] (At very low solute concentrations there is evidence of a radiative transfer process.[8,9]) The solvent-solute energy transfer efficiency is dependent on the solute concentration as is shown in Fig. 3. The energy transfer is essentially 100% at the optimum solute concentration.

If a second solute is present in the scintillator solution there can be an energy transfer between the solute molecules. The primary solute (higher concentration, higher energy singlet excited level) will transfer its energy by a non-radiative process to the secondary solute (lower concentration, lower energy singlet excited level). The transfer is quantitative at the optimum concentrations of the two solutes.

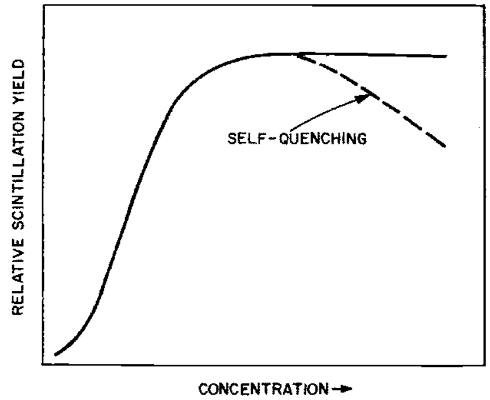

Fig.3. The effect of solute concentration on the relative scintillation yield. This shows the relationship for a solute which self-quenches at high concentrations (i.e. PPO) and a solute which shows no self-quenching at any concentration (i.e. butyl-PBD).

Figure 4 shows a schematic representation of the scintillation processes in a liquid scintillator solution with a primary and a secondary solute.

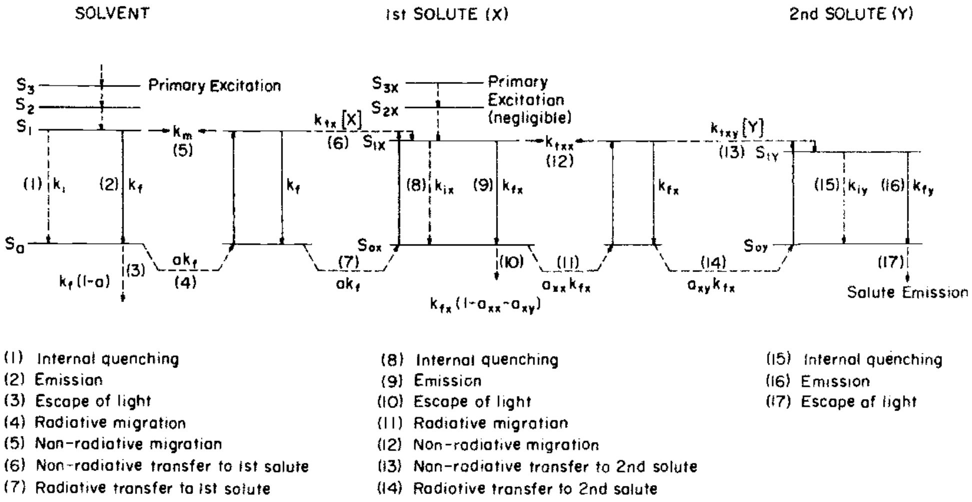

Fig.4. A diagram showing the many steps involved between energy absorption by the solvent molecules and the emission of photons by the solute molecules.

Scintillation efficiency

The scintillation efficiency is defined as the fraction of the energy of the exciting particle which is converted into photon energy, nhv, where n is the number of photons and hν is the energy of the photon. However, there is a whole spectrum of photon energies and the total photon energy is given by the equation

$$E_{ph} = h\int_{\nu_1}^{\nu_2} n(\nu)d\nu$$

where n(ν) is the number of photons of frequency ν and the integration limits ν_1 and ν_2 are the minimum and maximum frequencies of the fluorescence spectrum of the scintillator solute. The scintillation efficiency, S_x, is the ratio of total photon energy, E_{ph}, to excitation energy, E_{ex}, given by the equation

$$S_x = E_{ph}/E_{ex} = \left(h\int_{\nu_1}^{\nu_2} n(\nu)d\nu\right)/E_{ex}$$

Certain approximations can be made by assuming that the average number of photons produced is $\overline{N}_{ph}$ and that the energy of a photon with the average frequency of the fluorescence spectrum is $\overline{E}_{ph}$. Using these approximations the equation for S_x becomes

$$S_x = \frac{(\overline{N}_{ph})\ (\overline{E}_{ph})}{E_{ex}}$$

The solvent used in the scintillation solution determines the value of S_x. Lipsky and co-workers[10] determined the value of S_x for Carbon-14 beta particle excitation of a benzene solution. Hastings and Weber[11] determined the value of S_x for Carbon-14 and Hydrogen-3 beta particle excitations of toluene scintillator solutions. Table 1 summarises their data.

Table 1. Experimentally determined values of the scintillation efficiency, S_x.

Solvent	S_x for excitation with the radionuclide Hydrogen-3	 Carbon-14
Benzene	—	0.042[a]
Toluene	0.044[b]	0.052[b]

[a] Reference 10.
[b] Reference 11.

The solute (or solutes) determine the energy (wavelength) distribution of the emitted photons. Using the fluorescence spectra reported by Berlman[12] it is possible to calculate the average photon energy. For the scintillator solute PPO the average wavelength of the fluorescence spectrum is about 380 nm. The energy of a photon of this wavelength is calculated:

$$\begin{aligned} E &= h\nu = hc/\lambda \\ &= (4.143 \times 10^{-15}\ \text{eV-s})\ (2.99 \times 10^{10}\ \text{cm/s})/(3.8 \times 10^{-5}\ \text{cm}) \\ &= 3.2\ \text{eV} \end{aligned}$$

For a toluene solution the average number of photons, using the data of Hastings and Weber[11] in Table 1, can be equated to

$$\bar{N}_{ph} = 13.7\ E_{ex}\ \text{(in keV)} \qquad \text{For Hydrogen-3}$$

$$\bar{N}_{ph} = 16.3\ E_{ex}\ \text{(in keV)} \qquad \text{For Carbon-14}$$

The scintillation efficiency is not constant with the energy of the electron as can be seen in Fig. 5 taken from Horrocks.[13] Above 300 keV electron energy the scintillation efficiency is constant and is about 1.2 times greater than the scintillation efficiency for an electron energy of 50 keV, the average energy of a Carbon-14 beta spectrum. The value of S_x above 300 keV is 0.062 which gives the relationship to the average number of photons as

$$\bar{N}_{ph} = 19.4\ E_{ex}\ \text{(in keV) for } E_{ex} \geq 300\ \text{keV}$$

The scintillation efficiencies, average number of photons, average number of photoelectrons at 28% photocathode efficiency and a theoretical coincidence counting efficiency as a function of electron energy are listed in Table 2.

Table 2. Scintillation data and counting predictions for a toluene scintillator solution with PPO as the scintillator solute for different energy electrons.

Electron energy (keV)	S_x	$\bar{N}_{ph}$	Number of photoelectrons (at 28% eff.)	Theoretical coincidence counting eff.%
1000	0.062	20,600	5,768	100
500	0.062	10,300	2,884	100
158	0.058	3,060	857	100
50	0.052	802	225	100
5	0.042	66	18	100
1	0.031	10	2.8 (0.94)[a]	63[b]
0.5	0.024	4	1.1 (0.67)[a]	0[b]

[a] Poisson probability of measuring a pulse from the designated number of photoelectrons.

[b] Probability of a coincidence pulse calculated from the relationship. Probability $= 1 - 2^{1-Y}$ where Y is the expected number of photoelectrons. Probability is zero for any value of Y less than two, i.e. one photoelectron in each multiplier phototube.

Responses for different types of particles

Different types of ionising particles, i.e. beta, alpha, protons, etc., induce different responses per unit energy in typical liquid scintillator solutions. As a general rule the higher the specific ionisation of the exciting particle the lower the photon yield per unit energy (scintillation efficiency). Figure 6 shows the response of a typical scintillator solution to alpha and beta particles as a function of particle energy. Figure 7 shows the response for electrons of energy less than 100 keV. The responses for several particles are shown together in Fig. 8.

Gamma rays and X-rays can be measured with liquid scintillator solutions only through their interaction by way of scattering which leads to the production of electrons (Compton scattered electrons) or, if the energy is very low, by the total absorption (photoelectric effect). Neutrons are measured through the transfer of all or part of the neutron energy to hydrogen atoms in the solution by a scattering process which gives energetic protons.

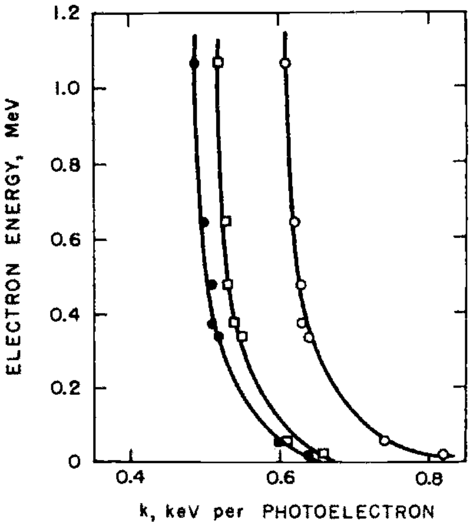

Fig. 5. The average energy required to produce a photoelectron as a function of the energy of the electron which excites the liquid scintillator solution.[1 3]

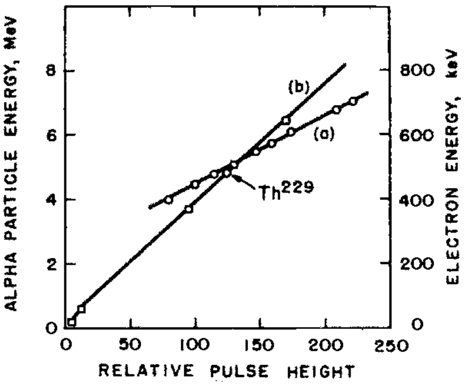

Fig. 6. Pulse height-energy relationship of a liquid scintillator solution for excitations by (a) alpha particles and (b) electrons. Note the factor of ten in the two energy scales.

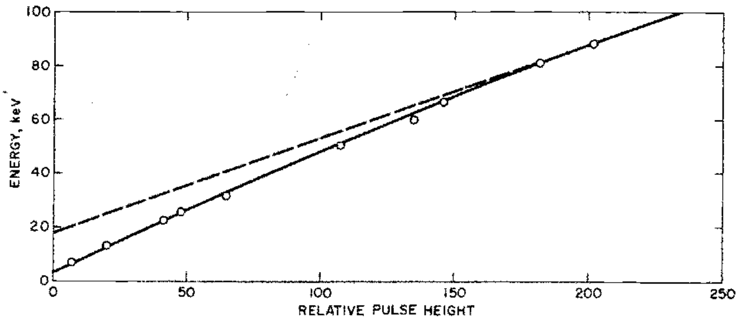

Fig. 7. Pulse height-energy relationship of a liquid scintillator solution for excitation by electrons with energies less than 100 keV.

SPECIAL APPLICATIONS

Determination of absolute disintegration rates

(a) Alpha particles Alpha particles can be counted with 100% efficiency in a liquid scintillation solution. The alpha emitting radionuclide has to be intimately in contact with or dissolved in the solution to count with 100% efficiency. If the radionuclide is in the form of a solid or on the surface of a solid support which is suspended in the solution the counting efficiency will possibly be less than 100%.

Because most naturally occurring alpha emitting radionuclides are present with anywhere from one to several daughter radionuclides which themselves emit beta and alpha particles, the activity of the parent radionuclide can not be determined by merely counting the sample in a liquid scintillator system. Under conditions where the parent radionuclide is in total equilibrium with its daughter

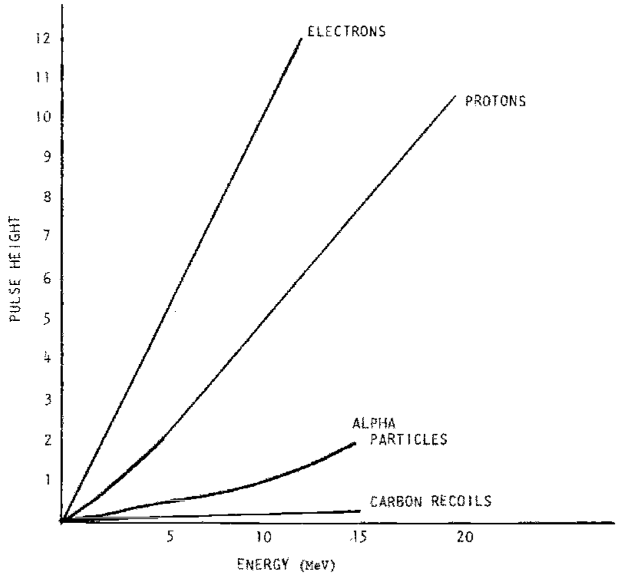

Fig. 8. Relative pulse heights for excitations of equal energy electrons, protons, alpha particles and recoil carbon atoms. (From R. Batchelor, W.B. Gilboy, J.B. Parker and J.H. Towle, Nuc. Instr. and Meth. 13, 70 (1961).

radionuclides and the decay scheme of all the daughter radionuclides is known, a simple total count can be used to calculate the decay rate of the parent radionuclide. Usually there is no total equilibrium and the pulses due to the parent and daughter radionuclides are separated by measurement of the pulse height distribution with a multichannel analyser. Figure 9 shows a pulse height distribution for a sample of thorium dissolved in a liquid scintillator solution.[14] The radionuclide Thorium-232 produced only a part of the total pulses measured.

The determination of small amounts of normal uranium in water and urine[15] illustrates some of the problems encountered when daughter radionuclides grow into secular equilibrium very slowly. The growth of the radionuclides Thorium-234 and Protactinium-234m, daughters of Uranium-238, occurs with a 25 day half life. This is too long a period to wait until the daughters reach equilibrium (in about 170 days the growth will be 99.2% of the true secular equilibrium). Some of the beta particles from Protactinium-234m produce the same pulse height response as the 4.2 and 4.8 MeV alpha particles and a simple counting of pulses of that amplitude would lead to a too high evaluation of the uranium content of the sample. The beta emitters produce a continuum of pulse heights while the single energy alpha particles produce a peaked pulse height response. Figure 10 shows the measured pulse height distribution from a sample of uranium dispersed in an emulsion type liquid scintillator solution.[15] The beta pulse distribution can be obtained by interpolation of the beta continuum and this subtracted from the total pulses to obtain the alpha decay rate.

(b) Beta emitters Radionuclides which decay the emission of beta particles only can be counted very efficiently in a liquid scintillator solution. By dissolving the radionuclide in the scintillator solution it is possible to determine the maximum sample count rate by the integral count method[16] and the use of wide open counting channels. For most radionuclides which have an E_{max} of the beta spectrum

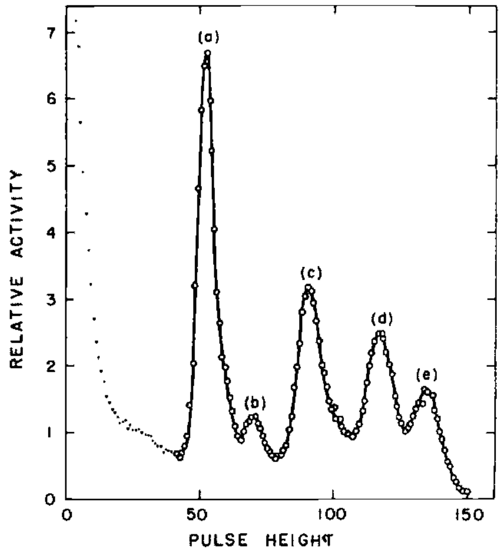

Fig. 9. Differential pulse height spectrum for a sample of thorium dissolved in a liquid scintillator solution. The various peaks are due to (a) 4.0 MeV alpha particles of Thorium-232, (b) 4.7 MeV alpha particles of Thorium-230, (c) 5.3 MeV alpha particles of Thorium-228 plus 5.7 MeV alpha particles of Radium-224 (unresolved), (d) 6.3 MeV alpha particles of Radon-220 and (e) 6.8 MeV alpha particles of Polonium-216.

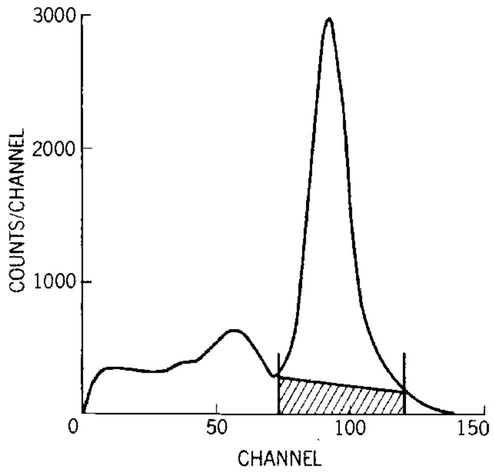

Fig. 10. Differential pulse height distribution for a sample of 1.0 mg of normal uranium in 1.0 ml of water which is dissolved in an emulsifier containing liquid scintillator solution.

greater than 150 keV, the extrapolated integral count rate will be within ±2% of the true sample disintegration rate. Since all beta emitters produce a continuum of beta particle energies from zero to the E_{max}, there are always some electrons which have insufficient energy to produce enough photons to be measured. The fraction of beta particles which have energies below this threshold is not only dependent upon E_{max} but also upon the shape of the beta continuum. Beta spectra weighted towards the low energy particles (called forbidden transitions) have a greater fraction of their beta particles below the threshold. These radionuclides will be measured with a lower counting efficiency.

When the shape of the beta spectrum of the radionuclide is known,[17] it is possible to predict theoretical maximum counting efficiency. Several investigators have developed theoretical methods for the calculation of the disintegration rate of beta emitters from the measured integral count rate and the known beta spectral shape when the energy required to produce a photoelectron in the counting system is known.[18-22]

(c) Conversion electron sources Certain radionuclides which decay by gamma ray emission will for part of the time release their decay energy as kinetic energy of an extranuclear electron in the process called internal conversion. The emitted electrons have a given energy equal to the difference between the energy of the gamma transition and the binding energy of the extranuclear electron:

$$E_e = E\gamma - BE_e$$

The radionuclide Indium-113m decays from an energy level of the nucleus of 393 keV. A gamma ray is emitted in 65% of the decay events while an internal conversion electron is emitted in the other 35% of the events. These approximately 369 keV electrons are counted with 100% efficiency. The decay rate of an Indium-113m sample is equal to the conversion electron count rate divided by 0.35. Figure 11 shows a pulse height distribution obtained for a sample of Tin-113—Indium-113m dissolved in a liquid scintillator solution. The 369 keV electrons are easily detected.

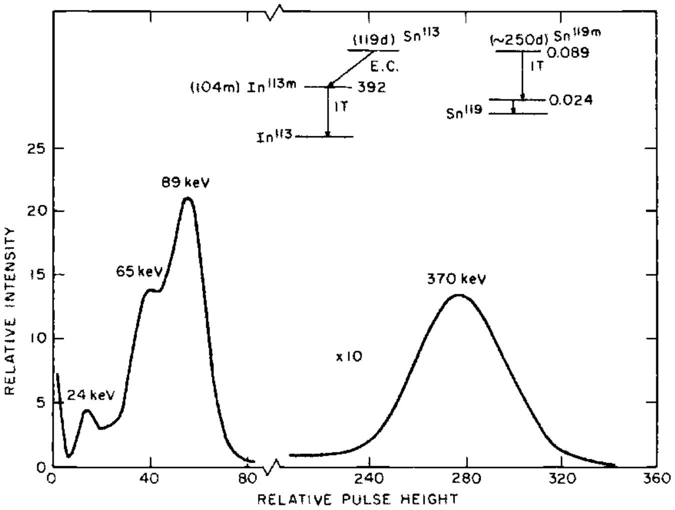

Fig. 11. Differential pulse height distribution for a sample of Tin-113 - Indium-113m dissolved in a liquid scintillator solution showing the 369 keV conversion electrons from the decay of Indium-113m.

(d) Other sources Gamma ray sources can be counted by their production of electrons in the liquid scintillator solution by the Compton scattering and photoelectric effects. The counting efficiencies for gamma ray sources are normally very low for at least two main reasons; (1) the probability of gamma ray interaction with the materials of the solution is low because of the low atomic numbers of the atoms in the material which makes up the solution, and (2) when the source is outside the solution the geometry for the gamma rays passing through the solution is low. The counting efficiency can be increased, especially for low energy gamma rays and X-rays, by the addition of a compound which contains an atom with a high atomic number to the solution.[23,24] In this way the electron density of the solution is increased and the probability of scattering or stopping a gamma ray is increased. Another technique increases the geometry by placing the gamma ray source, usually contained in a second smaller tube, inside the scintillator solution.

Neutrons are detected by the neutron-proton scattering process or by thermalising the neutrons and absorbing them in atoms with a high cross section which have been added to the scintillator solution.[25-28]

Even fission fragments can be counted in liquid scintillator solutions. By dissolving a radionuclide in the solution which will undergo fission, either spontaneously or by external stimulation, the fission fragments will excite the solution causing it to scintillate. In one report[29] a source of Californium-252, which decays by both alpha particle emission and spontaneous fission, was dissolved in a scintillator solution and it was shown that the fission fragments were counted with 100% efficiency.

Determination of beta spectra E_{max}

From the pulse height-energy relationship for electrons in a liquid scintillator[30-32] it is possible to convert a normal pulse height distribution into an energy distribution for beta particles from a given radionuclide. These energy distributions are used to obtain Kurie plots from which it is possible to obtain the E_{max} value of the beta spectrum. In some cases it is necessary to use a second order derivative correction to correct for the energy resolution effect of the liquid scintillator solution.[33] Table 3 lists several values of E_{max} for give radionuclides as determined from liquid scintillation counting. Figures 12, 13 and 14 show Kurie plots and the extrapolated values of E_{max} for Calcium-45, Nickel-63 and Sodium-22, respectively. It is worth noting that Sodium-22 is a positron emitter for which a positron spectrum was measured.

Table 3. The E_{max} values of the beta spectra of several radionuclides as determined by liquid scintillator methods.

Radionuclide	Mode of decay	Accepted[a] E_{max} (keV)	Measured E_{max} (keV)		Ref.
Rubidium-87	β^-	275	272	3	46 ±
Sodium-22	β^+	541	533	7	47 ±
Calcium-45	β^-	254	253	3	46 ±
Nickel-63	β^-	67	67	2	48 ±
Silicon-32	β^-	210	213	7	49 ±
Carbon-14	β^-	156	154	5	49 ±
Phosphorus-32	β^-	1707	1690	30	50 ±

[a] From Reference 45.

Pulse shape discrimination

The intensity of photons from a de-aerated scintillator solution as a function of time can be divided into at least two components; a fast (prompt) and a slow component. Figure 15 shows a typical intensity-time distribution. The fast component is the result of normal fluorescence from the excited molecules. The slow component has all the fluorescence properties of the fast component except it is produced at a time delayed from the initial excitation process. While the prompt component has a decay time equal to the fluorescing molecule (usually 1-2 ns), the slow component has a decay time independent of the fluorescing species and at least two orders of magnitude longer; 200-300 ns.

The slow component is due to the diffusion of two triplet excited molecules until they contact each other and undergo an annihilation reaction leading to the production of a singlet excited molecule which then proceeds to fluoresce:

$$T + T \rightleftharpoons S_1^* + S_0$$

In aerated solutions this delayed fluorescence is not observed because the oxygen

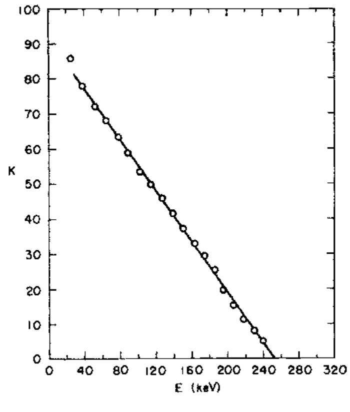

Fig. 12. A Kurie plot for a Calcium-45 sample in a liquid scintillator solution; the extrapolated E_{max} value is 253 keV.

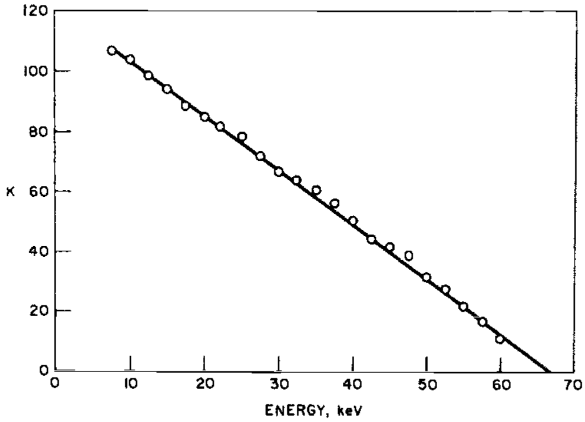

Fig. 13. A Kurie plot for a Nickel-63 sample in a liquid scintillator solution; the extrapolated E_{max} value is 67 keV.

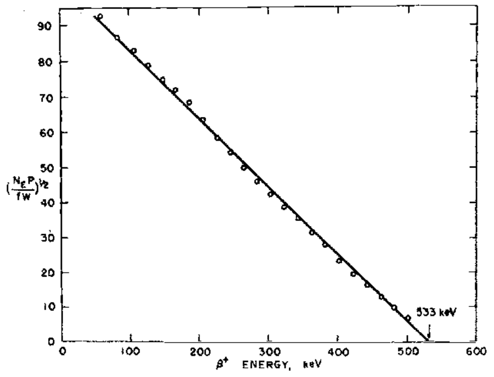

Fig. 14. A Kurie plot for a Sodium-22 sample in a liquid scintillator solution; the extrapolated E_{max} value is 533 keV. Sodium-22 is a positron emitter.

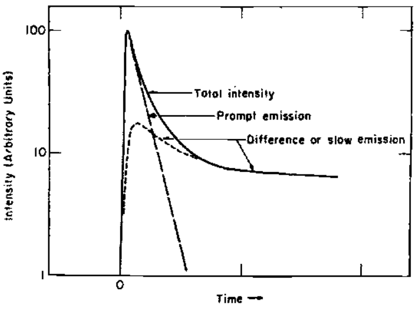

Fig. 15. A representation of the scintillation intensity from an organic liquid scintillator solution as a function of the time after the start of the scintillation event showing the division of the total intensity into a prompt and a slow component.

will quench the triplet excited molecules before they can undergo the annihilation reaction.

The amount of triplet excited molecules and hence the amount of delayed fluorescence is related to the specific ionisation of the exciting particle. Electrons which have a low specific ionisation produce much less triplet excitation than the greater specific ionisation particles like protons and alpha particles. For pulses which give the same intensity in the fast component, the intensity of the slow component is much greater for the proton excitation compared to the electron excitation, as shown in Fig. 16.

The integrated intensity as a function of time shows that the electron generated pulses reach their maximum intensity much faster than the proton generated pulses, as shown in Fig. 17. In the first 50 ns the electron produced pulse has reached 90% of its total integrated intensity while the proton produced pulse has reached only about 75% of its total integrated intensity. The proton produced pulse does not reach 90% of its total integrated intensity until 180 ns after the initiation of the pulse.

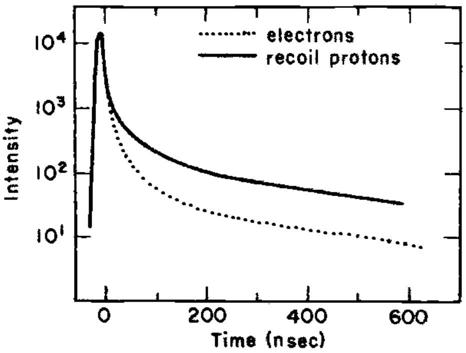

Fig. 16. The intensity of the scintillation as a function of time for a liquid scintillator solution which is excited by neutrons which produced recoil protons in the solution and gamma rays which produced Compton scattered electrons in the solution. The intensities are normalised at the peak intensity of the prompt emission.

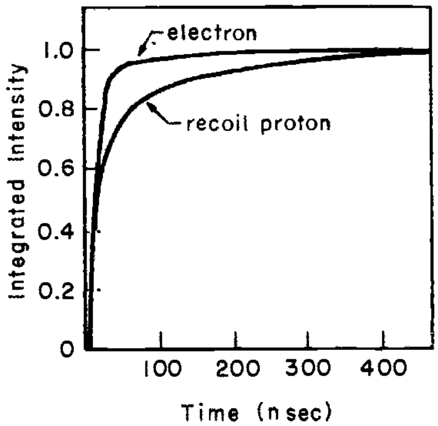

Fig. 17. Integrated intensity curves showing time dependency upon the exciting particle: electrons or recoil protons. The intensities are normalised at the maximum value at a time long after the start of the scintillation.

This difference in pulses produced by different types of particles is the basis for the method of pulse shape discrimination. There are at least three different types of electronic circuits that are used for distinguishing between different types of particles which are being counted at the same time in the same scintillator solution. Three of these methods are based on:

(a) Measurement of the amplitude of the slow component relative to the amplitude of the fast component of a single scintillation pulse.[34]

(b) Measurement of the time required for a single pulse to reach a specific fraction of its total integrated intensity.[35,36]

(c) Measurement of the zero amplitude cross-over time for a single pulse using a double differentiated dynode pulse of the multiplier phototube.[37-40]

Pulse shape discrimination techniques are used to detect and measure recoil protons, alpha particles, fission fragments and other particles with high specific ionisation in the presence of electrons and vice versa. Figure 18 shows the pulse height distribution (total integrated intensity) from a liquid scintillator solution which is being excited by alpha particles and Compton scattered electrons from a gamma ray source. The alpha particles come from a radionuclide dissolved in the scintillator solution. The gamma rays come from a radionuclide which is external to the scintillator solution vial. When the pulse generated by the pulse shape discriminator circuit is used in a coincidence mode, only the alpha particle produced pulses are allowed to be counted, as shown by the distribution obtained in Fig. 18(b). On the other hand, when that pulse is used in an anticoincidence mode, only the Compton scattered electron generated pulses are counted, as shown by the distribution obtained in Fig. 18(c). The pulse shape discriminator circuit was adjusted so that it generated a pulse only for an alpha particle which was determined by the relative amplitude of the slow component of the scintillation pulse.

Pulse shape discrimination techniques have been used extensively to measure the relative contributions of gamma rays and neutrons to the background around an operating nuclear reactor. The gamma rays are counted through the Compton scattered electrons produced in the liquid scintillator solution. The neutrons are often thermalised (by passing through a hydrogenous material like paraffin) and absorbed by an isotope of boron (Boron-10) which undergoes a fission-like event with the emission of an energetic alpha particle. These alpha particles are counted and are proportional to the neutron flux in the area. Figure 19 shows the pulse height spectra obtained with no pulse shape discrimination and with pulse shape discrimination circuit set to reject all electron generated pulses. The spectrum obtained is identical with that obtained when only a neutron source was present.

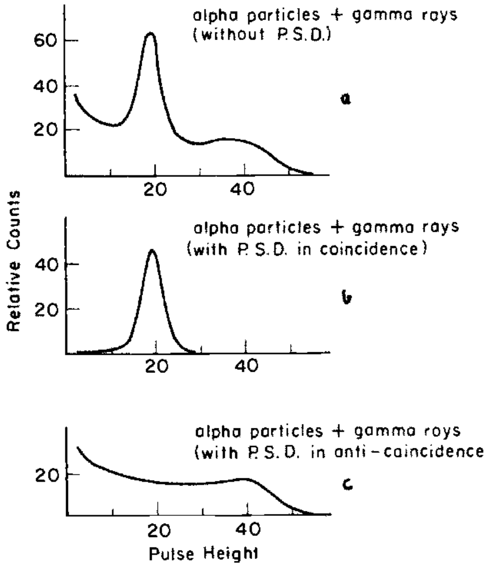

Fig. 18. Relative pulse height spectra from a liquid scintillator solution which was excited by electrons and alpha particles showing the use of the pulse shape discrimination output in coincidence to eliminate electron pulses (curve b) and in anticoincidence to reject alpha particle pulse (curve c).

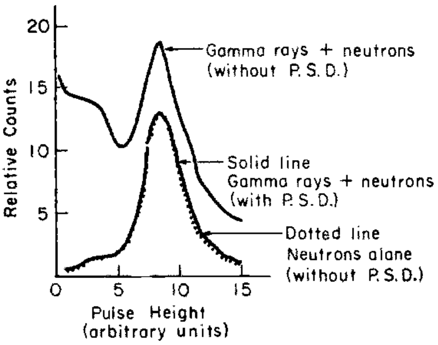

Fig. 19. Pulse height spectra showing the use of pulse shape discrimination to accept or reject pulses produced in a liquid scintillator solution containing boron when a neutron is absorbed.

A more detailed discussion of the pulse shape discrimination technique is given in a review article by Horrocks.[41]

Counting conversion electrons

The conversion electron process gives rise to mono-energetic electrons. The internal conversion process is competitive with gamma ray emission. Indeed, there are many radionuclides which decay by emission of both gamma rays and conversion electrons. These mono-energetic electrons will give a constant scintillation yield per electron. A pulse height spectrum of a conversion electron decay process will have a peak or peaks for each mono-energetic electron. If the electrons are sufficiently different in energy to be resolved, two peaks in the pulse height spectrum will be measured.

The radionuclide Xenon-131m decays by gamma ray emission 3% of the time and by the internal conversion process 97% of the time. The internal conversion will give rise to 134 keV conversion electrons part of the time and 164 keV conversion electrons for the remaining time. Figure 20 shows a pulse height distribution obtained for a sample of Xenon-131m dissolved in a liquid scintillation solution.[42] This detector system (a single multiplier phototube) had sufficiently good resolution that the two energy groups could be distinguished.

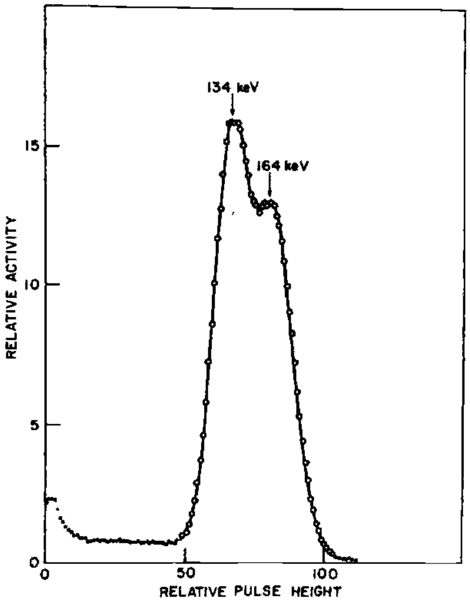

Fig. 20. Differential pulse height spectrum of a Xenon-131m sample dissolved in a liquid scintillator solution.

Another radionuclide which decays partly by the internal conversion process is Indium-113m. Figure 21 shows the pulse height distribution obtained for a liquid scintillator solution containing Indium-113m and its parent radionuclide Tin-113.[43] The 369 keV conversion electrons can easily be measured. The lower energy peaks are due to Auger electrons as a result of the electron capture processes of the radionuclide Tin-113. In one case, the 369 keV conversion electrons were used as a monitor of the amount of quench present in a liquid scintillator solution. Figure 22 shows the shift in the pulse height distribution peak as the amount of quench in the solution changes.[44]

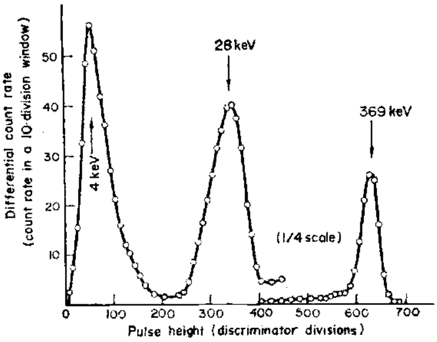

Fig. 21. Differential pulse height spectrum of a sample of Tin-113 – Indium-113m dissolved in a liquid scintillator solution. Pulse height scale is proportional to the logarithm of the energy.

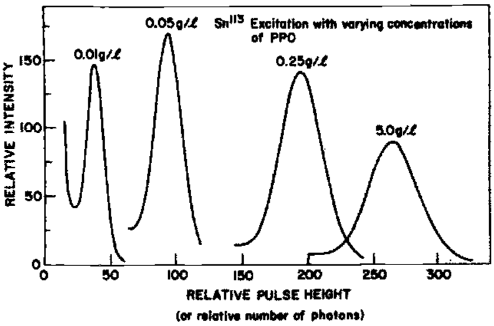

Fig. 22. Differential pulse height spectra for the 369 keV conversion electron peak as a function of the scintillation yield of a liquid scintillator solution.

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DISCUSSION

R. Berthold: What is the present state regarding the energy resolution when measuring α-particles? In the proceedings of the San Francisco conference, the resolution reported with commercial instruments is only around 25%, while you reported around 6% in an earlier paper with a special single tube instrument.

D.L. Horrocks: There has not been much progress in improving resolution with commercial instruments, and there cannot really be expected to be much due to the light being divided between two photomultipliers. This dual system of light collection leads to a very much broadened pulse height spectrum.

A.R. Ware: You implied in your lecture that there was a difference in the energy transfer scheme between samples containing oxygen and those from which oxygen had been removed. For practical purposes, when great numbers of samples are being counted, how important is de-aeration when one bears in mind that the quantity of oxygen present will vary with time as a result and in any case quench corrections will probably be applied?

D.L. Horrocks: For routine work, with only screw cap type vials, oxygen may diffuse back into samples and de-aeration should not be used as the counting efficiency would depend upon the amount of oxygen that had diffused into the sample.

P.E. Stanley: The overlap of the fluorescence and absorption spectra for PPO is much worse than has been indicated and this is because of the relatively high concentrations of PPO used in scintillation fluors. Thus the O.D. is less than around 1.0 for wavelengths above 350nm. These photons are absorbed and re-emitted at wavelength within the fluorescence wavelength of PPO. I was also interested to learn of the change in scintillation efficiencies with beta-energy. This bears out what we have found recently from a mathematical model we have been building of the liquid scintillation process (for colour and chemical quenching as well as vial and detector response). Thus to obtain the observed efficiencies we need to use about 11 photons per keV for Carbon-14 and around 13 photons per keV for Hydrogen-3. This is approximately in the ratio 0.044 : 0.052 that you mentioned.

D.L. Horrocks: The change in scintillation efficiency is shown in more detail in Table 2 of this paper. Actually the scintillation yield varies over a factor of two between the E_{max} of tritium (18 keV) and 1 keV.