The Sheet Scintillation Counting of Tritium in Surface Films*

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Because of the extremely soft nature of β-rays, tritium is expected to be the most effective radioactive tracer for elucidating the mechanism of such surface-chemical processes as the adsorption of detergents at the solution/gas interface, as well as for studying the nature of solid surfaces. Thus, the adsorbed amount of tritiated sodium dodecyl sulfate was determined with a 2π proportional counter, with humidified hydrocarbon gas flowing over the detergent solution.[1] The surface roughness of solid samples was determined by measuring the radioactivity of a tritiated specimen.[2] Similar techniques have been employed in studying the behavior of tritiated lauric acid in thin layers on metal[3] and that of hydrocarbons on metallic[4] and aqueous[5] hypophases.

All these experiments have been carried out with a windowless flow counter of a limited size. In these experiments, therefore, the experimental devices had to be small enough to fit into the counter; they can not include, for example, a trough for insoluble monolayers. In addition, the surface is inevitably exposed to the hydrocarbon gas, which in some cases dissolves in the hypophase.[6] To avoid these difficulties, an attempt has been made to develop a method of scintillation counting which does not require a flowing gas and where the space for the sample is not limited.

Experimental

The counting probe consisted mainly of a sheet of plastic scintillator and a photomultiplier tube (EMI 9536s). To minimize the optical loss due to total reflection, coupling of the sheet to the tube window was achieved with silicone oil of the same refractive index, 1.60, as that of the scintillator sheet. The assembled probe was placed, at a distance of 1 mm., face to face with a thin film (0.17—0.85 μg./cm^2) of an extremely pure sample of stearic-9, 10-^{3}H, ^{3}H acid (9.5 c./mol.)[7] spread over the circular range (1.13 cm^2) of a concentric aluminum disk. The whole assembly was thermostated to $\pm$0.5°C throughout the measured temperature range of -5 to 10°C.

The output pulses led to a linear amplifier (Aloka LA-10; maximum amplification, 30000), then to a pulse height analyzer (Aloka PA-22, 2 channel) for counting.

Two types of plastic scintillator sheets were used in the experiments. The sheets of the Pilot Scintillator B, which contained *p*, *p'*-diphenylstilbene as a scintillator, were obtained from the Pilot Chemicals, Inc., Watertown, Mass., U. S. A. The other type, containing terphenyl, was kindly supplied by the Matsushita Denko K. K., Osaka.

Results and Discussion

The background values were generally high, because of the low discrimination levels which we inevitably chose for counting the pulses of tritium β-rays. Therefore, attempts were made to overcome the difficulty by the aid of optical coupling

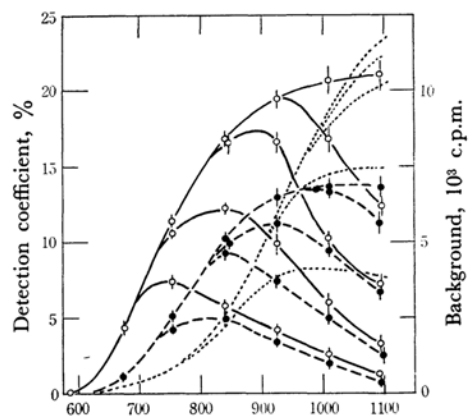

Fig. 1. Voltage dependences of background (dotted lines) and of detection coefficient with terphenyl sheet with (full) or without (chained) optical coupling.
Temperature, 10°C; window width, down to up, 5—12.5, 5—25, 5—50, 5—100 and 5—∞ V.
Each point expresses the average from 5—10 independent experiments.

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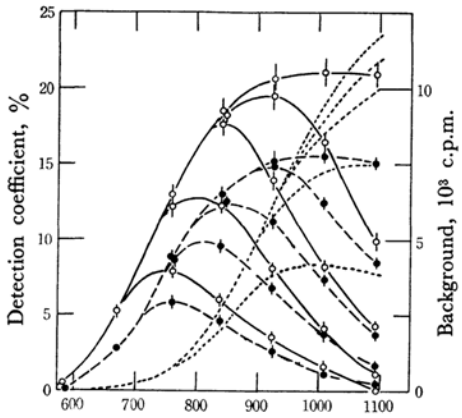

Fig. 2. Voltage dependences of background (dotted lines) and of detection coefficient with the Pilot Scintillator B sheet with (full) or without (chained) optical coupling.

Temperature, 10°C; window width, down to up, 5—12.5, 5—25, 5—50, 5—100 and 5—∞ Volts.

Each point expresses the average from 5—10 independent experiments.

for the efficient transformation of the optical scintillae into electronic pulses. Figures 1 and 2 show the counting characteristics for terphenyl and the Pilot Scintillator B sheets, respectively, with and without an optical coupling at 10°C. It can be seen in the figures that the coupling has caused a shift of the curve to lower voltage and has increased the detection coefficient. Accordingly, the background value at a 10% detection coefficient, for example, was decreased by the coupling from 2600 to 650 c.p.m. with the terphenyl sheet (Fig.1) and 1000 to 330 c. p. m. with the Pilot Scintillator B sheet (Fig. 2).

Precise experiments using 0.01—0.25 mm.-thick scintillator sheets have shown that the counting characteristics do not depend on the thickness.

The lowering of the temperature causes a remarkable decrease in the background, especially in the high voltage region, but it has little influence upon the detection coefficient. For obtaining a 10% detection coefficient, for instance, the background values were 90 c.p.m. at −5°C and 700 c.p.m. at 10°C. Similar or more exaggerated difference was found for obtaining another value of the detection coefficient. Very low temperature, however, could not be used, especially for adsorption experiments with an aqueous system.

Figure 3 shows the effect of sample thickness on the count rate, obtained at the applied voltage in order to give the maximum detection coefficient for each window width using a Pilot Scintillator B at 10°C. The linearity is fairly good, regardless of the counting conditions. If we assume the 20 Å²/molecule to be the cross-sectional area of

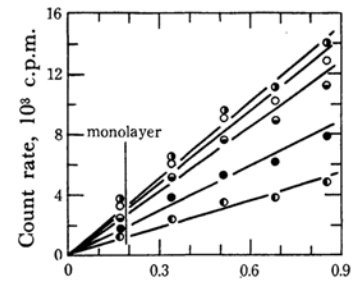

Fig. 3. Effect of sample thickness on count rate. Temperature, 10°C; scintillator, optically coupled Pilot Scintillator B.

Window width and applied voltage; ◐, 5—12.5 V. and 755 V.; ●, 5—25 V. and 800 V.; ◒, 5—50 V. and 840 V.; ○, 5—100 V. and9 25V.; and ◑, 5—∞ V. and 1010 V.

stearic acid, 1200 c.p.m. for a 5—12.5 V. window width would, for example, be obtained as the net count rate of a monolayer of tritiated stearic acid spread over a circle of 1.13 cm.² in area. Therefore, if we would have a monolayer of the same substance with the same specific activity spread on a surfac wider than the tube window, we could expect ca. 21600 c.p.m. as the net count rate, A, due to the tritium in the monolayer,* the value being much more than the background, B, 490 c.p.m. in this case. Figure 4 illustrates the values of A and B/A as a function of B under counting conditions which give a maximum detection coefficient for each window width. The

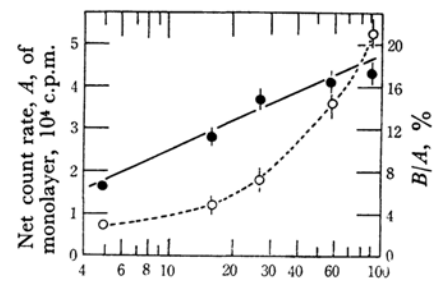

Fig. 4. Net count rate, A, (●) of stearic acid monolayer and the B/A ratio (○) as a function of background, B, at 10°C. Scintillator, optically coupled Pilot Scintillator B; window width, left to right, 5—12.5, 5—25, 5—50, 5—100 and 5—∞ V.

Each point expresses the average from 5 independent experiments.

* It was confirmed that the geometric factor for a point source does not depend on its radial position, almost up to the edge of the scintillator sheet. The constancy of the factor is due to the short distance (1mm.) between the sheet and the sample and, possibly, to the structure of the Venetian-blind-type phototube to which our photomultiplier belongs.

figure shows that the value of B/A increases remarkably with an increase in the background, showing the better conditions obtainable with a narrower window width and, correspondingly, with a lower voltage. It must also be mentioned that the temperature dependence of the background is decreased markedly by a decrease in the applied voltage and by a narrowing of the window width.

The sheet scintillation, which was first reported for counting tritiated hydrogen[8] and for the low-background counting of hard β-rays[9] will thus be of great use in determining the amount of a tritiated substance present in very thin layers at the air/water interface of any size available. It will enable us to determine simultaneously the radioactivity and such other properties as surface pressure and potential.

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