

"AN ULTRA-THIN FILM TRITIUM SOURCE STANDARD FOR INSTRUMENT CALIBRATION"

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1. INTRODUCTION

One requirement called for in consultative documents for revised national legislation on Radiological Protection, following EEC directive L246, is that measuring instruments must be calibrated annually against referable standards. The provision of standard sources for low energy beta emitters, of which tritium is the most familiar, presents special problems because self absorption both distorts the energy spectrum and reduces the number of particles emitted.

We have made a number of sources by the Langmuir-Blodgett (L-B) method and our tests presented here demonstrate that primary and working large-area standards having negligible self absorption for low energy emitters can now be produced which will be of interest to the International Standards Organisation (ISO). Previous investigations carried out elsewhere using radioactively labelled L-B films complement and support our conclusions⁽¹⁾.

2. LANGMUIR-BLODGETT FILMS

2.1 General principles: A monolayer of molecules is conveniently formed when a solution of surface-active material, dissolved in a water-insoluble, volatile organic solvent, is placed drop by drop onto a clean water surface. The drops spread and the solvent evaporates, leaving behind a monomolecular layer of the surface-active material which can then be compressed by moving barriers on the surface, as indicated in figure 1. When a monolayer is compressed into a tightly packed, coherent state it is possible, by passing a carefully prepared solid substrate through it, to transfer the monolayer intact from the liquid to the solid surface, figure 2; this is the L-B technique^(2,3). By repeated passages of the substrate through the monolayer, thicker (multilayer) films can be built up.

It is essential to maintain the monolayer at constant surface pressure throughout the deposition process. The original approach was to use a "piston oil"^(1,2), but modern "L-B troughs" employ motor-driven barrier mechanisms linked to a surface pressure sensor via an electronic feedback system. Provided adequate care is taken the L-B technique allows remarkably pinhole-free, homogeneous films to be prepared with fine control of film thickness to within a few tenths of a nanometer. Furthermore it is possible to modify the molecular structure in order to achieve the required film characteristics, including the incorporation of radioactive nuclei.

2.2 Preparation of L-B tritium sources: Tritium-labelled stearic acid, dissolved in toluene (specific activity 3.6 mCi mm^{-1} and concentration 0.45 mg cm^{-3}), was obtained from Amersham International PLC. It had been prepared by the catalytic reduction of oleic acid $\text{CH}_3(\text{CH}_2)_7 \text{CH}=\text{CH}(\text{CH}_2)_7\text{COOH}$ using tritium gas.

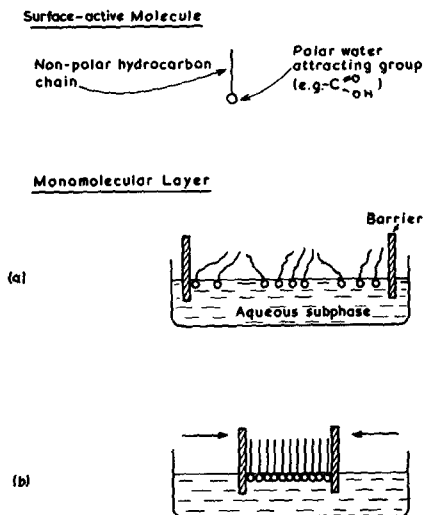


Fig. 1 Schematic showing a monomolecular layer of surface-active material, (a) under low compression and (b) under high compression with the molecules tightly packed.

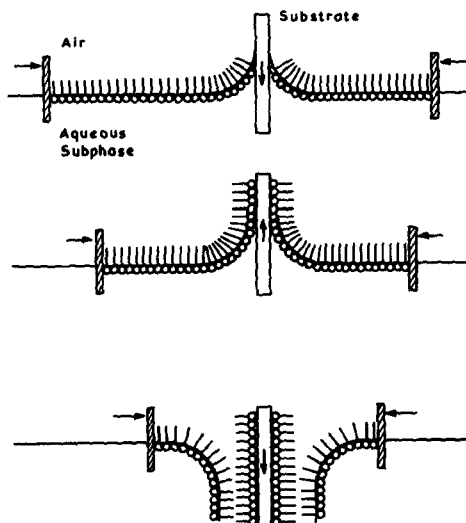


Fig. 2 Transfer of monomolecular layer to hydrophilic solid substrate.

Earthed electrically-conducting substrates are required for absolute emission rate counting. In order to achieve this, aluminium films of thickness $0.15 \mu\text{m}$ were deposited onto Corning 7059 glass plates 5 cm square by thermal evaporation. Substrates were dipped at 0.5 cm min^{-1} to a typical depth of 3.5 cm using a motor-driven micrometer and the monolayer surface pressure was maintained at $30 \pm 2 \text{ mN m}^{-1}$ during the deposition process using an electronic feedback system. The subphase was a $2.5 \times 10^{-4} \text{ M CdCl}_2$ aqueous solution ($\text{pH} = 6.05 \pm 0.05$, $T = 20 \pm 0.5^\circ\text{C}$); purified water (resistivity $18 \text{ M}\Omega \text{ cm}$) was obtained from a Millipore "Milli-Q" filtration system. Under these conditions fatty acids are almost totally ionised and so the monolayers are converted to cadmium stearate.

3. MEASURING INSTRUMENTS

A Whitlock tritium meter WTM5000 was used for source emission measurements. It comprises a 100 cm^2 flat plastic scintillator detector viewed by two photomultipliers and coincidence pulses are digitally displayed. A rubber moulding excludes external light and acts as a distance gauge ($0.50 \text{ mm} \pm 0.12 \text{ mm}$) when the instrument is used for measuring surfaces. A partial vacuum is created in the chamber bounded by the rubber moulding by means of a single-action, manually-operated pump, the reaction spring of which creates a highly reproducible pressure of approximately half an atmosphere.

Emission rates were also measured using a Hurfurth HGZII gas flow proportional detector by mounting and earthing the source on the inside of the mylar window. The activity of material removed on smears was measured using a Nuclear Enterprises LSC1 liquid scintillation counter.

4. SOURCE EMISSION CHARACTERISTICS

The activity cm^{-2} (a_A) of a monolayer source is determined by the specific activity (a_S) and the number of molecules cm^{-2} (n_M) which depends on surface pressure and subphase conditions. In this investigation, $a_S = 3.6 \text{ mCi mM}^{-1}$ and $n_M = 4.9 \times 10^{14}$ molecules of stearic acid cm^{-2} (equivalent to $8.1 \times 10^{-7} \text{ mM cm}^{-2}$). Hence, $a_A = 2.9 \times 10^{-6} \text{ mCi cm}^{-2}$ which should result in 107 dps cm^{-2} (4π geometry). Measurements with the HGZII support this value allowing for a value of 15% for backscatter enhancement, but as yet ignoring Bremstrahlung secondaries.

Measurement of activity for 1, 5 and 9 layer sources using the WTM5000 gave 2.66, 2.86 and 2.77 cps cm^{-2} of monolayer transferred respectively. This demonstrates that, within experimental error ($\sigma \approx 5\%$), (a) self-absorption with respect to total activity is not significant for thicknesses $\leq 22.5 \text{ nm}$, which is in agreement with Beischer⁽¹⁾, (b) L-B films may be built-up in a controllable, uniform fashion, and (c) the WTM5000 has a 5% efficiency in 2π geometry.

5. PHYSICAL TESTS ON SOURCES

In order to assess the suitability of the L-B sources as standards and working sources they were subjected to the tests described below.

5.1 Thermal stability: Sources were held for one hour at various temperatures ($-20^\circ\text{C} \leq T \leq 80^\circ\text{C}$) in an environmental cabinet, then removed and restored to 25°C in air dried over silica gel. Emission rate measurements showed that the monolayer sources did not degrade at 70°C but a 20% permanent loss of activity was observed at 80°C . Multilayer sources showed a 10% permanent loss at 60°C increasing to 20% at 80°C .

5.2 Effect of relative humidity: Environmental cabinet tests using a range of temperatures below 60°C with relative humidity up to 100% resulted in negligible change of emission rate.

5.3 Effect of pressure: The sources were subjected to vacuum tests down to 0.18 mbar at 25°C for periods of one hour during which no change in emission rate was detected. This stability can be expected to be maintained over longer periods of time because cadmium stearate is much less volatile than the free acid which is known to degrade significantly⁽⁴⁾. Furthermore, emission rates were measured at a range of sub-atmospheric pressures and source-to-detector distances and representative results are shown in figure 3. Note the improved sensitivity that can be gained by measuring tritium sources at short range and reduced pressures.

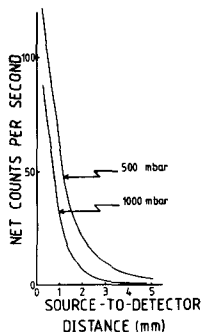


Fig. 3 Emission rate for the 9 layer tritium-labelled cadmium stearate source as a function of source-to-detector distance and air pressure.

5.4 Wipe tests: It is virtually inevitable that any source will be mishandled during its lifetime and so, to be viable, a source must be sufficiently robust to withstand such treatment. Due to the low emission rate from our monolayer sources we anticipated that any small reduction in emission rate resulting from an "average" manual wipe would be difficult to observe. Therefore, for this reason, and in an attempt to employ a quantifiable, more reproducible wipe procedure, we constructed a simple device for wiping sources with cloth wipes ("Nu Con" smears) at a pressure of 25 g cm^{-2} over a fixed stroke of 2.5 cm. A series of wipe tests were carried out on both 1 and 9 layer sources. Source activity was recorded after 5, 10, 50 and 100 wipes using the WTM5000 and the activity on the smears was measured using the LCS1.

The results of this rather severe abrasive test showed that at the pressure used here the loss of source activity was $\leq 2\%$ per wipe for both the monolayer and multilayer sources; the WTM5000 and LCS1 data were self-consistent. A more quantitative analysis regarding the adhesive characteristics of the cadmium stearate layers could not be made due to the removal of some of the supporting aluminium film by the smear. Abrasive resistance could be improved by the use of polished solid aluminium plates.

6. CONCLUSIONS

We believe that, with only little further development, both primary and working standard sources of low energy emitters could be produced by the L-B method to allow calibration of measuring instruments, thereby satisfying the requirements of new legislation, the ISO (ISO/TC85/SC2/WG2) and anyone wishing to study unmoderated emission characteristics.

ACKNOWLEDGEMENTS

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