A MELTABLE THERMOPLASTIC SCINTILLATOR

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ABSTRACT. We have developed a meltable thermoplastic scintillator material, Meltilex™, for solid and on-support liquid scintillation counting (LSC). To obtain good physical contact between the sample and scintillator, we melted the material (60–120°C) and counted it after cooling and solidification. The scintillator is especially suited for use in filter LSC, *e.g.*, in ligand-receptor binding assays. Other LS applications include counting of fine-grained powders, sands, ashes and precipitates where fixed and permanent conditions are required. Light output is comparable to conventional scintillators and, in filter applications, ^{3}H-counting efficiency is up to 50%. The material also has good alpha- and beta-particle pulse-shape separation properties, which can be used in our LS counter with a pulse-shape analyzer to produce simultaneous α- and β-particle spectra from the same sample. The material can be molded, and when prepared in sheet form, is a mechanically flexible plastic-like scintillator, easy to cut to proper size, according to the needs of the application. The material is non-toxic and non-volatile. The amount of waste disposal is reduced and liquid waste is eliminated.

INTRODUCTION

A major problem with on-support and solid-sample applications is in unstable counting conditions, including partial elution and sedimentation of the sample. Counting samples deposited on filtermat with liquid cocktail may require an elution procedure, *e.g.*, in ligand-receptor binding (LRB) assays, where samples are harvested onto the filtermat, filtration spots are punched into the vials, liquid cocktail is added and sample eluted. To avoid the addition of cocktail to harvested samples on filtermat, Potter and Warner (1991) proposed a filtermat coated with organic crystals and fluors.

Counting of insoluble solid samples requires careful mixing of finely ground powder so that it is suspended in the cocktail emulsion. Sedimentation can be prevented by using gelling agents, Cab-O-Sil®, aluminum stearate (Peng 1981), water and gelling scintillator cocktail, *e.g.*, Optiphase MP, or liquid scintillators that liquify/solidify upon heating/cooling. The latter is achieved by adding to the liquid scintillator (solvent and dissolved fluors) polyolefines (Benakis 1971), paraffin or surface active agent (Fujii & Takiue 1989). A polymerized solid block with sandwiched samples yields excellent counting efficiencies, but the sample preparation procedure is elaborate and time consuming (Yang *et al.* 1991). We describe here the advantages gained by using Meltilex™, a meltable thermoplastic scintillator.

MELTILEX™ CHARACTERISTICS

Meltilex™ is a new type of organic scintillation material that contains no liquid solvents. Its physical appearance is solid (at room temperature), odorless and translucent, with bluish fluorescence. It is non-volatile and non-toxic, requiring no fume hood for sample preparation. Meltilex™ comprises two polymeric components, a low-molecular-weight aromatic thermoplastic polymer and an ethylenevinylacetate co-polymer that is elastic at room temperature. Paraffin is also included to increase the fluidity of the molten material. The fluors are the commonly used PPO and bis-MSB, which are homogenized by heating to 100°C while mixing. Meltilex™ can be produced in sheet form with different shapes and thicknesses.

Meltilex™ can be used as a melt-on scintillator, or as a lay-on scintillator without melting, by simply using it as a scintillating plate. Figure 1 shows the melt-on method. A Meltilex™ sheet can be cut or punched to the proper size needed for the application. For on-support sample counting, Meltilex™ is laid on a dry sample support and melted. For particulate or powder-sample counting

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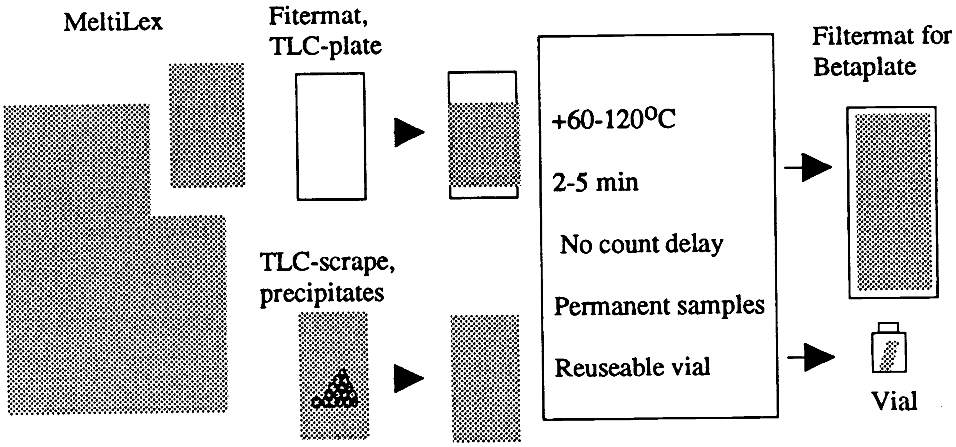

Fig. 1. Meltilex™ melt-on method of sample preparation for counting on-support and solid samples

(*e.g.* TLC scrape, precipitate), the sample is deposited as a thin layer (to minimize self-absorption) on top of one Meltilex™ sheet, then another sheet is laid on it and melted. Meltilex™ can be melted at 60–120°C using, *e.g.*, a heat sealer, hot plate, oven, iron, infrared lamp, heat roller or hair dryer. The melt-on sample preparation time is less than 5 min. On cooling, the scintillator resolidifies, and the solid sample can be measured by a flatbed counter, such as Betaplate™ or a standard LS counter. Meltilex™ has excellent thermal stability. Remelting is possible; after one week at 80°C, we observed no changes in counting performance.

This melt-on method was introduced for biological applications, such as membrane receptor analysis and other on-support LS applications, but it is also applicable to wipe test swabs and solid samples in environmental radiation monitoring (Oikari *et al.* 1991). The lay-on method is a sandwich technique, in which two sheets of Meltilex™ are pressed on either side of the filtermat or fine powder of the solid sample. This concept is used for counting ^{32}P-labeled samples in gel fragments (Potter, Tan & Ratcliffe 1991) and Dot Blots (Potter & LeJeune 1991) in Betaplate™.

Scintillation light output is comparable to that of plastic scintillators. Decay time for the scintillation pulse is *ca.* 4 nsec (main component). ^{3}H counting efficiency is up to 50% for filter applications.

The use of Meltilex™ with a Betaplate™ counter (Potter *et al.* 1986; Warner *et al.* 1985; Warner & Potter 1986; Warner *et al.* 1991)) is an optimized flatbed counting setup. A commercial Meltilex™ heat sealer encloses the sample spots on the filtermat with scintillator. The activity of the spots is then measured with the Betaplate™ counter. Absorption losses are small due to short light absorption paths. Color quench is minimized due to the short light absorption paths in a thin sample and chemical quench is minimized due to the resolidification of the Meltilex™ before the chemical quenchers from the sample support are eluted into the Meltilex™. The sample remains stable with no phase changes and counting conditions are reproducible. Background count rates are low because the sample acts as a thin target in which ionizing radiation events are not intercepted.

METHODS

We measured the detection efficiencies of common isotopes on filtermat and the performance of the lay-on method with a Wallac 1204 Betaplate™ LS counter. We measured the filter samples by a wipe test, direct counting from TLC plate and TLC scrapes with a Wallac 1409 LS counter. The α/β discrimination on Meltilex™ was measured on a Wallac 1411 standard counter with α/β discrimination electronics.

Table 1 compares the relative detection efficiency of Meltilex™ and the melt-on method to the standard liquid cocktail and conventional sample preparation method. We made the measurements with spotted samples on a glass-fiber filtermat. Detection efficiencies are similar for ^{14}C and more energetic β isotopes.

TABLE 1. Relative Detection Efficiencies of Common Isotopes in Filtermat Counting: Meltilex™ *vs.* BetaplateScint™

	^{51}Cr	^{3}H	^{125}I	^{14}C	^{45}Ca	^{32}P
Relative efficiency *vs.* BPS* (%)	61.3	75.6	77.9	99.8	97.1	101.7

*BetaplateScint™ (BPS) is a di-isopropylnaphthalene-based liquid cocktail for non-aqueous samples.

A water solution of known activity was spotted on a controlled wipe test area of 25 x 25 mm. The water was allowed to evaporate and the remaining precipitates on the surfaces were wipe-tested. The test surfaces were aluminum and laminate. Sucrose (6,6(n)-^{3}H and U-^{14}C) from the Wallac internal standard kit was used as a β-labeled contaminant. The contaminant was bound loosely to the surfaces; a suitable wipe medium was a filter swab moistened with water (100 μl/100 mm^{2}). Under these conditions, wipe tests and liquid and solid scintillants can be compared (Table 2).

TABLE 2. Wipe Test Procedures

Dry Meltilex™	Press on the area of interest and count in a 20-ml vial
Melted Meltilex™	Press on the area of interest, melt on with hot air and count in a 20-ml vial
Dry filter + Meltilex™	Wipe the area of interest until the surface is clear, melt on Meltilex™ and count in a 20-ml vial
Wet filter + Meltilex™	Wipe the area of interest until the surface is clear; dry the wipe at 85°C for 20 min; melt on Meltilex™ for 2 min and count in a 20-ml vial
Dry filter + elution in OPHS*	Wipe the area of interest until the surface is clear, then elute the filter with 100 μl water and add OPHS in a 20-ml vial, shake to homogenize and count
Wet filter + elution in OPHS	Wipe the area of interest until the surface is clear, then add OPHS in a 20-ml vial; shake to homogenize and count

*OPHS = Optiphase HiSafe™ 2, a water-miscible di-isopropylnaphthalene-based scintillant.

Table 3 shows the recovery of sucrose based on six samples. Samples were counted within 1 h of the wipe test. Prolonged overnight elution in Optiphase HiSafe™ 2 improved counting recovery of ^{14}C from 84 to 89% in the last case. Good recoveries are obtained with dry filter and Meltilex™ as compared with elution in Optiphase HiSafe™ 2. Results are excellent for Meltilex™ and wet filter, which is more suitable for ^{14}C and sucrose contaminants. Dry and melted Meltilex™ wipe-test results seem to have poor reproducibility with sucrose, whereas deviation of wet samples was small (3–5%) in the case of applied activity in six replicate samples.

TABLE 3. Sucrose Recovery (%)

	Aluminum surface		Laminate	
	^{3}H	^{14}C	^{3}H	^{14}C
Dry Meltilex™	0.25	6.5	0.05	2.0
Melted Meltilex™	0.13	0.55	0.15	2.6
Dry filter + Meltilex™	6.0	50	11	61
Wet filter + Meltilex™	21	81	23	85
Dry filter + elution in OPHS	26	64	33	64
Wet filter + elution in OPHS	36	88	43	84

Counting efficiency of ^{3}H for spot-on samples on filter with Meltilex™ is 23%, and for cocktail, is 47%. Counting efficiency of ^{14}C for spot-on samples on filter with Meltilex™ and cocktail is 86% for each. Prolonged elution times, 24 h or more, should be used to gain maximum, invariable counting efficiency in the elution method. Recovery for wet filter swab and scintillant (Table 3) is the same as the counting efficiency of spot-on samples on filter for ^{3}H and ^{14}C. The Meltilex™ with wetted filter swab was tested further with water soluble mixtures of ^{90}Sr and ^{241}Am chlorides, where high-energy β and α particles give high counting efficiency (Fig. 2). The wipe test gave a recovery of 95%. Wetted filter swab recovery is almost complete for ^{90}Sr and ^{241}Am.

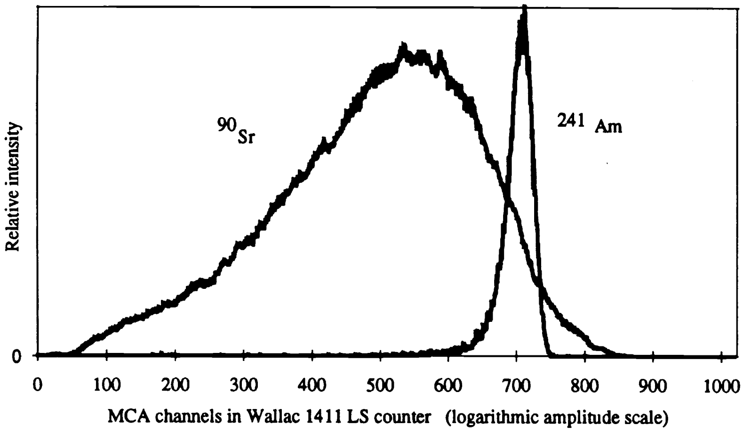

Fig. 2. α/β separation of ^{90}Sr and ^{241}Am in Meltilex™. The sample was a wipe from a laminate surface.

Known activities of ^{14}C(sucrose) and ^{3}H(thymidine) in aqueous solution were spotted on the TLC plate (silica layer thickness 0.2 mm). Plates were dried overnight at room temperature. The identified spots on the TLC plate were scraped off between two sheets of Meltilex™ and melted on. Meltilex™ was also melted directly on a piece of TLC plate. The TLC scrape was measured for comparison with conventional gellified cocktail. The samples were counted in 20-ml vials. Table 4 shows counting efficiencies for six replicates, and illustrates that the counting efficiencies of the two methods were comparable. We measured the same samples with both the lay-on method and the melt-on method. Table 5 shows the results of sandwich measurements for spotted samples on the glass-fiber filtermat.

The lay-on method could be very useful for high-energy β radiation, *e.g.*, ^{32}P, especially when the sample must be recovered for further use. In Meltilex™, as in many other organic scintillators, α particles produce longer pulses than β particles. This property can be utilized in LS counters that have a pulse-shape analyzer to produce simultaneous α- and β-particle spectra from the same sample (Kaihola & Oikari 1991) (Fig. 2). In particulate samples, self-absorption of α particles may cause some broadening of the spectra (Fig. 3).

TABLE 4. Mean Counting Efficiencies (± 1σ) for TLC-Scrape Measurements. Comparison Between Meltilex™ and Gellified Cocktail*

	^{3}H	^{14}C
Melt-on Meltilex™	16.7 ± 0.3	85.9 ± 1.1
Scrape-in Meltilex™	16.5 ± 0.5	87.8 ± 1.0
Scrape-in gel	18.9 ± 0.4	87.7 ± 2.1

*Six parts of cocktail (Lumac/3M: Lumagel®) and 4 parts of de-ionized water

TABLE 5. Comparison of Meltilex™ Detection Efficiency in Lay-on Method and Melt-on Method

Isotope	Lay-on	Melt-on
^{32}P	98.4%	100%
^{14}C	31.0%	100%
^{90}Sr	93.0%	100%

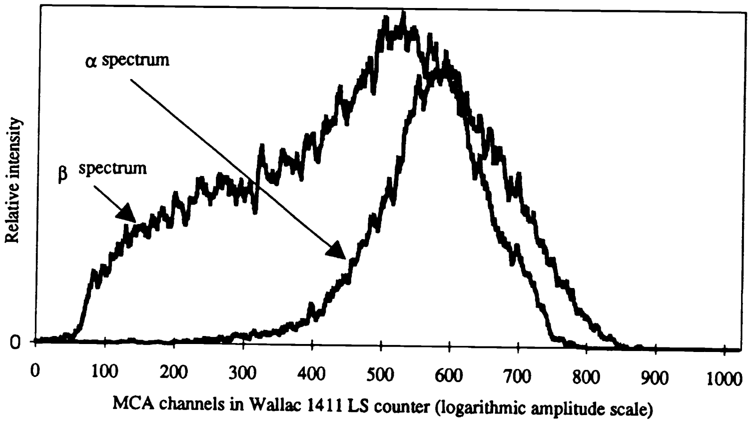

Fig. 3. α/β separation from U-containing sand; the sample was put between 2 Meltilex™ sheets, which were then melted.

For direct counting of radon in air, Kaihola, Oikari and Suontausta (1992, in press) used a standard LS vial coated with Meltilex™ as a radon collection and detection chamber. α background was 0.03 cpm in a Teflon vial with the Quantulus low-level LS counter. The lower limit of detection was 15 Bq m^{-3} with a 100-min counting time (50% error at the lower limit).

CONCLUSION

Meltilex™ is an ideal detection material for scintillation counting of various solid samples, fine-grained powders and on-support samples. Fixed and repeatable counting geometry is achieved with no sedimentation or partial-elution problems. It is possible to store prepared samples for longer periods before counting. Meltilex™ itself is non-volatile and non-toxic; waste disposal is reduced and liquid waste is eliminated.

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