

## PRINCIPLES AND PRACTICE OF PLASTIC SCINTILLATOR DESIGN

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### ABSTRACT

Plastic scintillators are used for ionizing radiation detection and measurement. A brief review is given of the standard plastic base materials, typical classes of fluorescent compounds, and some processing methods for producing plastic scintillators. The available design parameters that determine the resultant scintillator performance are discussed.

### KEYWORDS

Aromatic, fluorescence, plastic scintillator, polystyrene, polyvinyltoluene, radiation detection

### INTRODUCTION

The most common type of plastic scintillator is composed of an aromatic plastic base, which has the benzene ring as a pendent group along the polymer backbone. Aromatic structures are necessary for the fluorescence processes of absorption and emission in the area of interest of plastic scintillators. The common commercial materials are polystyrene (PS) and polyvinyltoluene (PVT). The monomeric unit of PS is shown in Fig. 1. Other aromatic plastic bases studied include phenyl silicones (Bowen, 1988), and epoxies (Markley, 1968). The non-aromatic plastic polymethylmethacrylate (PMMA), commonly called acrylic, is sometimes used as a scintillator base by adding an aromatic co-solvent, typically naphthalene (10-25% by weight). However, this scintillator is generally not as efficient as using aromatic polymers as the base.

Fluorescence occurs in these plastics when energy from ionizing radiation is deposited. This process is covered in detail in the standard reference texts (Birks, 1964, Berlman, 1971, Turro, 1978), and has been reviewed in several recent publications (Davis *et al.*, 1989, Flournoy, 1988, Kauffman, 1988, Majewski *et al.*, 1992).

The plastic itself, though, is not useful as a scintillator because its fluorescent yield is very low, it is not transparent to its own emission over any appreciable distance (<1cm), and its emission spectrum (300-350nm) is too short to match the common photodetectors.

The addition of about 1% by weight (it can range from 0.3-4%) of certain aromatic compounds, called primary fluors, makes an efficient scintillator. At this high concentration, a non-radiative energy transfer (Forster transfer) from the plastic to the primary fluor occurs, with subsequent fluorescence, generally in the UV (350-400nm). This scintillator, now much brighter (30-50% of that obtainable using crystalline anthracene), still has limited light transmission (<0.1 meters). To increase the emission-light bulk attenuation length to up to 2 meters and longer, an additional fluorescent compound, called a spectrum shifter, is added at about 0.01% by weight (it can range from 0.001-0.100%). The effect of various shifters on attenuation length of plastic scintillators has recently been discussed (Hurlbut, *et al.*, 1991). At this concentration, the energy transfer between the primary and the spectrum shifter is radiative, with subsequent emission of the shifter—typically in the blue to blue-green (400-500nm).

Some of the classes of compounds used as primary fluors and shifters are shown in Fig. 2. The oligophenylenes, oxazoles, oxadiazoles and benzoxazoles all contain several compounds useful in plastic scintillator formulas, with each class contributing both primaries and shifters.

The photophysical properties of a compound are determining factors in its selection for use in a scintillator formula. Figure 3. shows the absorption and emission spectra of an example scintillator

consisting of PS, a primary fluor PBD and a spectrum shifter dimethyl POPOP. Overlap of the primary fluor absorption spectrum with the emission of the plastic base, as well as overlap of the absorption spectrum of the spectrum shifter with the emission of the primary fluor and subsequent emission in the blue, is shown.

Some compounds, such as PMP (Gusten, 1983), can absorb energy from the plastic base and emit in the blue in one step. Other fluors, such as 3-hydroxyflavone (Renschler *et al.*, 1984) by means of intramolecular proton transfer (IPT), can absorb energy from the plastic base and emit in the green in one step. These compounds can provide high light transmission (6-9 meters, Ruchti, private communication), and also are in some of the most radiation-tolerant plastic scintillator formulas to date (Zorn *et al.*, 1988).

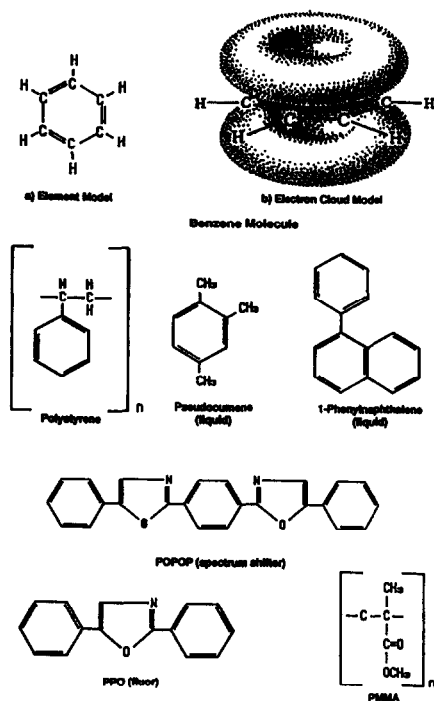
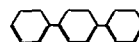


Fig. 1. Organic Scintillator Chemicals

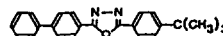
### Oligophenylenes

p-terphenyl 345 nm  
p-quaterphenyl 374 nm



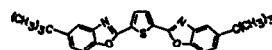
### Oxazoles & Oxadiazoles

butyl-PBD 365 nm  
POPOP 410 nm



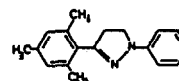
### Benzoxazoles

BBOT 435 nm



### Pyrazolines

PMP 425 nm



### Proton Transfer

3HF 530 nm  
HBT 530 nm

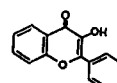


Fig. 2. Some classes of fluorescent compounds used as primary fluors and spectrum shifters

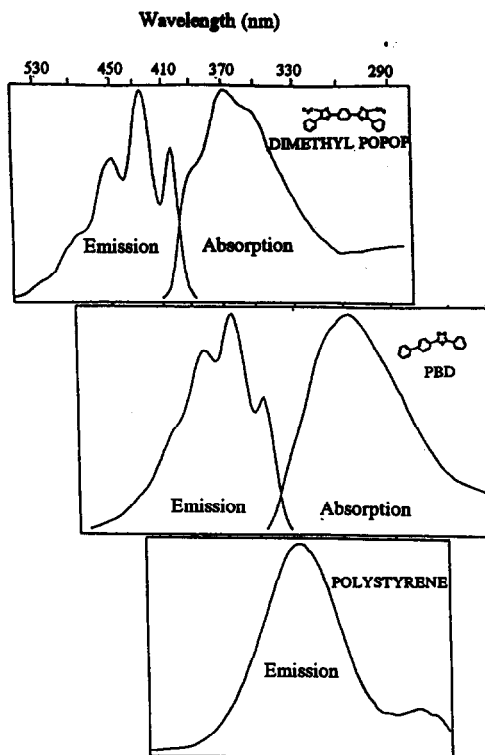


Fig. 3. Overlap of emission and absorption spectra of a typical blue emitting plastic scintillator.

## PERFORMANCE FACTORS

### Base

Scintillation efficiency, obtained by the aromatic structures previously discussed, is the key requirement from the base. Scintillation light output numbers (relative to anthracene) in part depend on the type of base plastic used. A comparison of scintillators made with PVT, PS and PMMA plus naphthalene, with equivalent fluor concentrations, produced relative signal intensities of 1.0, 0.88 and 0.38 respectively (measured in the author's laboratory). Transparency is another requirement for the base material as the scintillation photons may be required to travel several meters through the polymer to the read-out device.

### Fluors

There are several performance requirements for fluors which have been covered in depth in some recent reviews (Cahill, Kauffman, this proc.). Essentially, they are: suitable optical spectra; low self-absorption for high light transmission; high extinction coefficient ( $> 10,000$ ) to minimize the amount required for good energy transfer; high quantum yield ( $> 0.6$ ) again for efficient energy transfer and brightness; short decay time (2-3ns for blue emitters and up to 12ns and longer for green emitters); and radiation hardness. How rad hard a fluor is determined to be depends on the matrix (liquid or plastic) and test conditions.

## PLASTIC SCINTILLATOR PREPARATION

### Polymerization From the Monomer

**Ampoule Method.** For preparing small samples to test new materials, vacuum ampoules are often used. The fluors and any other additives are dissolved in the monomer, then transferred to the ampoule, degassed and sealed, and put through a thermal cycle. Polymerization and final cure is done in the ampoule. The plastic is removed from the ampoule and machined to size.

**Batch Reactor Method.** Again, the highly purified fluors (from commercial suppliers or custom synthesized) and other additives are dissolved in highly purified monomer. This high purity is to minimize chemical quenching and impurity-induced color which would adversely affect light transmission. This solution is then transferred to a reactor to begin the polymerization. Examples of additives are mold releases (to aid in the release of the cured plastic from molds) and antioxidants (to

protect the organic materials both during processing and during storage and use of the finished plastic). A partially polymerized resin is then transferred to cells of parallel glass panes, and the final cure is completed in an oven. The polymerization in the reactor and oven is designed to achieve the desired Molecular Weight Distribution (MWD). The proper MWD is important in achieving good mechanical properties and scintillation performance. The resulting plastic sheets are removed from the cells and machined to size. The plastic is made against glass to obtain highly reflective surfaces that result in high light transmission. This method is used for high volume production of long sheets (up to 5 meters) and thick sheets (> 40 cm). Also, rods and annuli of large diameter (up to 1 meter) can be produced.

In variations and combinations of the above methods, some process additives are used, such as initiators to polymerize at low temperatures and chain transfer agents to control molecular weight. Cross-linking agents can be added to increase heat-distortion resistance and surface processability, such as machining. However, these additives may decrease clarity due to increased color generated by reaction by-products.

### Compounding

The fluors and other additives are mechanically blended into polymer pellets. This mix is then molded or extruded into the desired configuration. This method can be used for high volume production of sheets of moderate thickness.

## **PROCESS FACTORS**

### Plastic Base

The selected material must be able to be purified and processed in quantities of hundreds to thousands of kilograms at a reasonable cost (factory resources and labor must be taken into consideration).

### Fluors

The volume need of fluors is smaller and is more amenable to custom synthesis, but other properties required are outlined below.

High Solubility. For primaries, at least 10 grams per liter, and for shifters at least 0.1 gram per liter.

Temperature Tolerance. Must be able to withstand processes going through 100-170 deg. C.

Chemical Stability. Must be compatible with all the other components in the mix. Must not be susceptible to radical attack during the polymerization itself.

Photostability. Many fluors are light sensitive, and this must be taken into account when they are being handled.

Cost. Again, be able to acquire all the above requirements and be obtainable at an acceptable cost.

## **PLASTIC SCINTILLATOR DESIGN PERSPECTIVES**

Traditional concerns have been high light output and long attenuation lengths, but plastic scintillators may be designed with specific properties enhanced to meet the requirements of a particular application. For example, organic compounds that contain lead or boron may be added to the matrix to increase its sensitivity to x-rays or thermal neutrons respectively. A 10% loading of lead increases the sensitivity to x-rays less than 50 keV by 400% or more. Standard plastic scintillators are insensitive to thermal neutrons, but the addition of boron containing molecules provides a capture cross-section when none existed before. However, when a particular property is enhanced, there are usually design compromises that must be made. These considerations include decreased signal amplitude, decreased transparency, and increased costs. Figure 4. shows the decrease in light output versus increased loading of element. At some point the signal gain from the element present is offset by the decrease in light with increased percent loading.

### Design Parameters

The design parameters available in scintillator formulating are summarized below.

Base Materials. Many commercial and custom polymers are potential candidates, including copolymers. Must be compatible with the other components in the formula and with the process method selected. The intended application is a major consideration in the base material selection. For

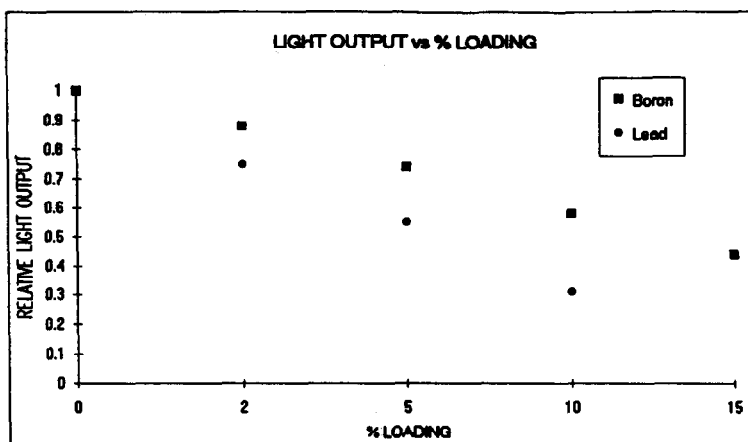


Fig. 4. Illustration of the loss of signal amplitude with increased element loading

example, PS is the material of choice for plastic scintillating fibers because it can be pulled into a fiber with outstanding light transmission. This is in contrast to PVT, but since PVT produces the highest scintillation output of the most common commercial materials, it is often the material of choice for cast sheets and rods.

**Fluors.** Many different types and combinations are available. Must be selected on the basis of which wavelength shifter or photodetector it is to be matched up with.

**Additives.** Compatibility of any process additives has to be considered both a function of the materials above and, also, the intended application of the scintillator. As an example, some of the traditional additives cannot be used in a high radiation environment due to radiation induced discoloration (Clough, 1988).

**Process Method.** Not all scintillation materials are processible by all the methods covered, such as silicones or epoxies. These polymers are usually made from prepolymerized resins, which are fully polymerized by the addition of catalysts and/or initiators and then set up in cross-linked networks. This network may now have desired properties of increased thermal stability and increased solvent resistance, but is no longer processible thermally. Also, the original prepolymerized materials may limit which fluorescent additives can be used due to solubility.

**Cost.** Again, the necessity of acceptable cost of materials, process time, etc.

At present there is particular emphasis in increasing radiation tolerance in plastic scintillators. The design goals are to maintain high light output and transparency while exposed to high doses over a lifetime of several years. Some of the means to achieve this involve modifications to the plastic base, to the processing methods employed, to fluor selection, and use of radiation stabilizing additives. In conclusion, the demands of designing to high radiation levels has an impact on every design parameter.

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