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Neutron/gamma pulse shape discrimination in plastic scintillators: Preparation and characterization of various compositions

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ABSTRACT

This work deals with the preparation and evaluation of plastic scintillators for neutron/gamma pulse shape discrimination (PSD). We succeeded in developing a plastic scintillator with good neutron/gamma discrimination properties in the range of what is already being commercialized. Several combinations of primary and secondary fluorophores were implemented in chemically modified polymers. These scintillators were fully characterized by fluorescence spectroscopy and under neutron irradiation. The materials proved to be stable for up to 5 years without any degradation of PSD properties. They were then classified in terms of their PSD capabilities and light yield. Our best candidate, 28.6 wt% of primary fluorophore with a small amount of secondary fluorophore, shows promising PSD results and is particularly suited to industrial development, because its preparation does not involve the use of expensive or exotic compounds. Furthermore, even at the highest prepared concentration, high stability over time was observed. As a proof of concept, one sample with dimensions 109 mm ∅ × 114 mm height (≈ 1 L) was prepared.

1. Introduction

From the very beginning of neutron detection, pulse shape discrimination (PSD) between neutrons and gamma rays in plastic scintillators remained one of the holy grails of modern physics, as they were widely considered to be indistinguishable. However, in 1960, Brooks discovered that doping a standard plastic scintillator (*e.g.,* polystyrene+*p*-terphenyl+POPOP) with a so-called "secondary solvent" (herein 4-isopropylbiphenyl) allowed discrimination of fast neutrons from gamma rays [1]. His "Plastic 77" was later marketed under the trade name NE-150 but was, unfortunately, discarded after physical alterations appeared a few months after the production [2]. Since then, several research groups have tried to address this challenge by various means, leading to different levels of success. Their efforts have involved a chemical approach [3], the use of a given geometry that allows gamma rays rejection [4], and mathematical approaches using smart algorithms that identify the sharp difference existing between neutron and gamma signals [5]. Recently, Zaitseva et al., presumably inspired by previous work from Brooks, developed a new plastic scintillator composed from highly concentrated 2,5-diphenyloxazole (PPO) and a wavelength shifter (optional), 9,10-diphenylanthracene (DPA), in polyvinyltoluene (PVT) [6a,b]. They showed that good PSD was observed only when a given PPO concentration threshold was reached. This new plastic scintillator is currently being sold, most likely with slight modifications from what was originally described, by Eljen Technologies under the trade name EJ-299-33 [6c–e],[7]. In 2012, Feng et al. proposed for the first time both spectral and pulse shape discrimination in plastic scintillators, with a triplet harvesting system consisting of an iridium complex [8]. In 2006, a French collaborative effort called Neutromania was initiated for developing new plastic scintillators for this purpose, embedding 5 laboratories from 3 cities and gathering physicists and chemists. This project was the foundation for the work that produced the results presented herein. We will see that a general formulation consisting of {polymer+highly concentrated 1st fluorophore+2nd fluorophore as wavelength shifter} can be used to prepare various plastic scintillators. The present paper determines their ability, or inability, to discriminate neutrons from gamma rays. We present herein our results regarding the preparation and characterization of plastic scintillators with various compositions, displaying good neutron/gamma pulse shape discrimination efficiency in the range of existing discrimination methods.

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2. Experimental

Encapsulated liquid scintillator BC-501A and plastic scintillator BC-408 were obtained from Saint-Gobain Crystals and Detectors (Aubervilliers, France). Plastic scintillator EJ-200 was obtained from Eljen Technologies (supplied by Scionix, Bunnix, The Netherlands). Styrene and vinyltoluene monomers were purchased from Sigma-Aldrich and freshly distilled from CaH_2 prior to use. 1,4-Bis (5-phenyl-2-oxazolyl)benzene (POPOP), 9,10-diphenylanthracene, *p*-vinylbiphenyl, and *p*-terphenyl were also purchased from Sigma-Aldrich. 4-isopropylbiphenyl was purchased from Alfa Aesar. 2,5-Diphenyloxazole (PPO) was purchased from Acros or Sigma-Aldrich. All fluorescent molecules were used as received except *p*-vinylbiphenyl, which was purified by silica gel chromatography. The general procedure for plastic scintillator preparation is as follows: in a flame-dried round bottom flask filled with argon (Ar), the powders were dissolved in the liquids. The gases were then removed using the freeze-pump-thaw technique, and the solution was carefully transferred into a vial for polymerization. After completion, the vial was broken with a mallet and the scintillator was obtained after polishing the raw material. The contours and back were ultimately covered with TiO_2 paint (3 layers, EJ-510 from Eljen Technologies) for better light collection through reflection. The process of preparing these materials has been patented [9].

Pulse shape discrimination was performed by irradiating organic scintillators with an unshielded AmBe $\approx$0.45 GBq, $\approx$30,000 n/s source. The radioactive source was located 5 cm away from the scintillator.

Nuclear experiments using the charge comparison method, which allows light component separation, have been performed. Photophysical reasons justifying this method are explained well elsewhere [10]. The experimental set-up is described in Fig. 1. A range of plastics and one liquid scintillator are characterized using a Hamamatsu H1949-51 photomultiplier (PMT) for light

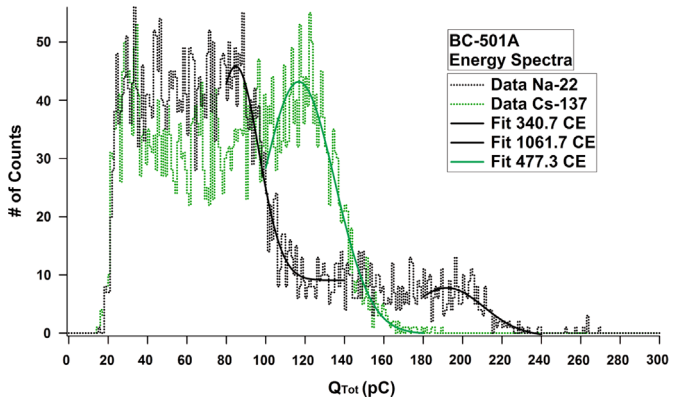

Fig. 2. ^{22}Na and ^{137}Cs energy spectra from BC-501A liquid scintillator.

Table 2
Gamma energy and Compton edges of ^{22}Na and ^{137}Cs.

Gamma sources	Gamma energies E_γ (keV) #1	Gamma energies (E_γ) (keV) #2	Compton edges CE (keV) #1	Compton edges CE (keV) #2
^{22}Na	511	1274.5	340.7	1061.7
^{137}Cs	661.7		477.3	

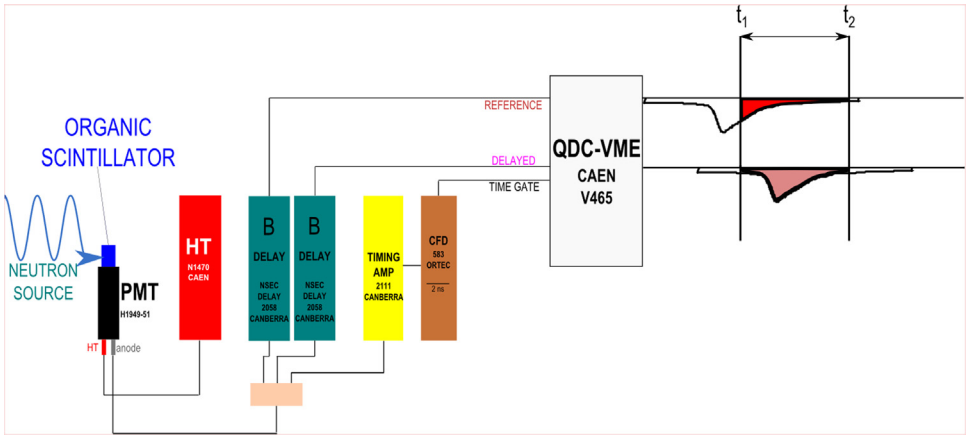

Fig. 1. Experimental set-up of the charge comparison method as the PSD method for n/γ discrimination.

Table 1
Main characteristics of various plastic scintillators prepared.

Sample	Dimensions		1st fluorophore (wt%)	2nd fluorophore (wt%)	Observations
	Diameter (mm) ∅	Thickness (mm)			
#1	49	8	4-isopropylbiphenyl (10) and *p*-terphenyl (3.4)	POPOP (0.05)	From Plastic 77 Brooks' recipe [1]
#2	30	13	4-isopropylbiphenyl (10)	POPOP (0.05)	Equiv. to #1 without p-terphenyl
#3	30	5	4-isopropylbiphenyl (15) and *p*-terphenyl (3.4)	POPOP (0.05)	4-isopropylbiphenyl more concentrated
#4	30	5	4-vinylbiphenyl (10)	POPOP (0.05)	Polymerizable fluorophore
#5	30	9	4-isopropylbiphenyl (10) and *p*-terphenyl (3.4)	POPOP (0.05)	Cross-linked polymer
#6	48	50	*Proprietary (17)*	Yes	Cross-linked polymer
#7	75	75	*Proprietary (17)*	Yes	Identical #6 but bigger
#8	32	27	*Proprietary(29)*	Yes	Cross-linked polymer
#9	32	16	PPO (30)	-	From Zaitseva's recipe [6]
#10	32	16	PPO (30)	DPA (0.2)	From Zaitseva's recipe [6]
#11	103	114	*Proprietary (17)*	Yes	Identical#6 & 7 but bigger