

SYNTHESIS AND CHARACTERIZATION OF PLASTIC SCINTILLATORS FOR THE TOTAL-BODY J-PET SCANNER*

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The aim of the research was to develop a polystyrene scintillator for use in the novel time-of-flight Jagiellonian Positron Emission Tomography (J-PET) scanner being elaborated for the whole-body imaging. To achieve this goal, polystyrene-based plastic scintillators with the different chemical compositions were produced and characterized. Light output, decay time and emission spectra were measured to develop the best composition of the polystyrene scintillator.

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1. Introduction

The Jagiellonian Positron Emission Tomograph (J-PET) is a time-of-flight PET scanner based on plastic scintillators [1]. Description of first two prototypes (a small and a full-scale ones) of J-PET scanners can be found in a formerly published paper by Niedźwiecki *et al.* [2]. The main objective of the J-PET Collaboration is to build a modular, low weight and portable PET scanner for the total body examination [3] and positronium imaging dedicated for research and clinical examinations [4, 5]. Currently, the main effort of the J-PET Collaboration is to build prototype modules consisting of 500 and 1000 mm long plastic scintillator strips with silicon photomultipliers coupled at both ends [6] and the layer of wavelength shifters [7].

Blue emitting plastic scintillators used for J-PET scanner construction usually consist of two kinds of fluorescent compounds dissolved in amorphous transparent-to-visible light polymer base. First fluorescent substance

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absorbs energy deposited by radiation in polymer matrix and emits ultraviolet light. Second substance, called wavelength shifter, converts ultraviolet light to blue light which is more convenient to be detected by light detectors and allows light propagation at long distances inside scintillator by internal reflections.

During our research, plastic scintillators were developed with short rise and decay times needed in time-of-flight PET detectors [8]. Scintillators described above are based on polyvinyltoluene, 2,5-diphenyloxazole and 2-(4-styrylphenyl)benzoxazole as a polymer base, first and second additions, respectively. In this work, scintillators are based on polystyrene and a few common fluorescent additives selected to obtain scintillator with short decay time and high light output comparable with formerly used polyvinyltoluene scintillators.

2. Synthesis of plastic scintillators

Styrene as a monomer was chosen to manufacture plastic scintillators. A styrene monomer is a low cost compound, is commercially available, and scintillators produced on this base have almost the same properties as polyvinyltoluene [9]. In order to measure spectroscopic properties of plastic scintillators, small cylinders were synthesized with the following dimensions: 25 mm in diameter and 25 mm height. Polymerization inhibitor 4-tert-butylcatechol present in styrene was removed on glass column filled with activated alumina [10]. Primary and secondary solutes were purchased from Sigma-Aldrich and were not purified (> 99% purity grade, suitable for scintillation).

Synthesis of samples was performed in glass ampoules 25 mm in diameter. Before polymerization, the ampoules were for 12 hours treated with 30% solution of dichlorodimethylsilane in chloroform to prevent sticking of scintillator samples to glass. After this procedure called silanization [11], the ampoules were rinsed in turn with chloroform, acetone, distilled water and were dried in an oven. A prepared solution of fluorescent substances in styrene was poured into a glass ampoule and argon gas was blown through Teflon pipe submerged in solution for 15 minutes to remove oxygen which inhibits polymerization and quenches fluorescence. The ampoules were sealed under argon gas atmosphere using flame burner. Thermal activated bulk polymerization process was performed in electric furnace with automatic control of temperature with an accuracy of 1°C. Polymerization was conducted within several days with a temperature cycle with maximal temperature of 140°C as described in a previous study by Kapłon *et al.* [12].

Fluorescent compounds for polystyrene scintillator synthesis were selected from many common fluorescent substances used in plastic scintillator manufacturing [13]. From such a group, the best fluorescent substances

were selected with the highest fluorescence quantum yield close to 1.0 and the shortest decay time with value less than 2 ns [14]. As the primary additive, the following substances were selected: 2,5-diphenyloxazole (PPO), p-terphenyl (PTP) and 2-(4-tert-butylphenyl)-5-(4-biphenyl)-1,3,4-oxadiazole (BPBD). As the secondary additive (wavelength shifter), the following substances were selected: 1,4-bis(5-phenyl-2-oxazolyl)benzene (POPOP), 1,4-bis(4-methyl-5-phenyl-2-oxazolyl)benzene (DM-POPOP) and 1,4-bis(2-methylstyryl)benzene (bis-MSB). Primary and secondary additives possess different absorption and emission spectra thus mixing a few similar compounds could increase energy transfer from polymer to wavelength shifter and enhance light output of scintillators.

Mentioned wavelength shifters were used because they have maxima of emission spectra around 420 nm. Wavelength of maximum emission in that region is well-matched with highest quantum efficiency of light detectors used in first J-PET scanner prototypes. Vacuum photomultiplier tubes have usually maximum quantum efficiency around 400 nm [15] and silicon photomultipliers for PET application have this value about 450 nm [16].

3. Measurement of optical properties

Light output of samples was measured with photomultiplier tube (PMT) and oscilloscope LeCroy SDA 6000A. Cylindrical shape scintillator samples with diameter 25 mm and height 25 mm (polished all sides) were wrapped with three layers of white Teflon tape and coupled to photomultiplier R5320 (Hamamatsu Photonics) with bialkali photocathode using optical grade silicone grease EJ-550 (Eljen Technology). Radioactive source Cs-137 was placed on top of the black light tight cover with photomultiplier and scintillator setup. Amplitude spectra from synthesized polystyrene scintillator samples were compared with spectra obtained from stilbene crystal of the same shape and sizes, whose light yield 14 000 photons/MeV [17] was taken as 100%.

To measure decay time of plastic scintillators the time-correlated single photon counting (TCSPC) was used. Fluorescence decay time was acquired with spectrofluorometer FluoroLog-3, (Horiba Jobin-Yvon) using NanoLED diode with 340 nm emission maximum and 1.3 ns pulse full width at half maximum (FWHM). In plastic scintillators consisted of two fluorescent substances [18], the distribution of the time of photon emission followed by the interaction of the gamma quantum at time Θ is given by the following convolution of two exponential and Gaussian terms:

$$f(t) = K \int_{\theta}^t \left(e^{-\frac{t-\tau}{t_d}} - e^{-\frac{t-\tau}{t_r}} \right) e^{-\frac{(\tau-\theta-2.5\sigma)^2}{2\sigma^2}} d\tau, \quad (1)$$

where following symbols denote: K — normalization constant, Θ — time of the gamma quantum interaction with scintillator, t_d — decay time of the final light emission, t_r — average time of the energy transfer to the wavelength shifter, σ — standard deviation reflects total uncertainty of light emission from scintillator and TCSPC components: excitation diode, PMT and electronics. Decay time was determined by fitting equation (1) to the experimental data with K , Θ , t_d , t_r , and σ treated as a free parameters of the fit. Gaussian part of equation (1) reflects effectively a spread of the signal due to the light emission from scintillator, density distribution of photons emitted by UV diode excitation, transit time spread in PMT detector and broadening of the signal due to electronics in TCSPC measurement.

Photo-induced emission spectra were measured using the Horiba Jobin–Yvon FluoroLog-3 spectrofluorometer. The emission signal was detected using a PMT R928P (Hamamatsu Photonics). Measurement was done in reflecting mode. Plastic scintillators samples disc with diameter 25 mm and 2 mm thickness were excited on surface by continuous wave xenon source with monochromator located at 45 degree from the normal to the surface. Emission light was collected by PMT at the opposite side of surface at the same angle.

4. Results and conclusions

Table I contains summarized results from synthesized polystyrene scintillators. First series of samples contain fixed at 2% content of first addition PPO and various combinations of wavelength shifters at 0.06% concentration. Second series of samples have fixed at 0.06% concentration of wavelength shifter POPOP and a few combination of first additions with concentration at 2%.

Obtained values of light output, decay time and wavelength of maximum emission spectra are very similar to properties of commercially available plastic scintillators based on polyvinyltoluene. Polystyrene scintillator with 2% PPO primary solute and 0.03% bis-MSB and 0.03% POPOP wavelength shifters is characterized by the best light output of over 11 200 photons per MeV. Decay time of 1.51 ± 0.02 ns of the fastest polystyrene scintillator is for composition with 2% BPBD primary solute and 0.06% POPOP wavelength shifter. Wavelengths of maximum emission of produced plastic scintillators are in the range from 422 nm to 435 nm.

The most significant result of this study are manufactured polystyrene-based plastic scintillators with high light output. Values of light output of five best samples are exceeding 10 000 photons/MeV which are better than for many commercial polyvinyltoluene based plastic scintillators. Polystyrene as a scintillators base is also cheaper than polyvinyltoluene. Synthesized

TABLE I

Optical properties of synthesized polystyrene scintillators.

Chemical composition	Wavelength of maximum emission [nm]	Decay time [ns]	Light output [photons/MeV]
2% PPO, 0.06% POPOP	422	1.66	9370
2% PPO, 0.06% DM-POPOP	435	1.96	7151
2% PPO, 0.06% bis-MSB	429	1.62	9809
2% PPO, 0.03% POPOP, 0.03% DM-POPOP	434	1.81	8845
2% PPO, 0.03% DM-POPOP, 0.03% bis-MSB	433	1.81	7946
2% PPO, 0.03% bis-MSB, 0.03% POPOP	423	1.80	11212
2% PPO, 0.02% POPOP, 0.02% DM-POPOP, 0.02% bis-MSB	433	1.80	10672
2% PPO, 0.06% POPOP	—	1.76	9346
2% PTP, 0.06% POPOP	—	1.59	9611
2% BPBD, 0.06% POPOP	—	1.51	10772
1% PPO, 1% PTP, 0.06% POPOP	—	1.58	10141
1% PTP, 1% BPBD, 0.06% POPOP	—	1.62	8727
1% BPBD, 1% PPO, 0.06% POPOP	—	1.87	10336
0.67% PPO, 0.67% PTP, 0.67% BPBD, 0.06% POPOP	—	1.66	9846

polystyrene scintillators possess short decay time in the range from 1.5 to 2 ns and its properties are suited for time-of-flight radiation detectors construction. Wavelengths of maximum emission of polystyrene scintillators obtained in this study are matched with highest quantum efficiency of light detection of vacuum photomultiplier tubes and silicon photomultipliers for PET application. Further directions in the next studies will research optimum concentrations of fluorescence additives.

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