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PAPER

Characterization of plastic scintillator samples produced by a university-SME Collaboration

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Abstract

Scintillator samples are synthesized by a university-SME collaboration and the light yield, light emission and light transmission properties are studied with the aim of determining the fluor content that gives the highest light yield. Three plastic scintillator samples with different fluor additives are produced and their optical properties are found to be comparable with a high-light-yield EJ-204 reference sample. Amongst the three, the sample with 0.75% PPO + 0.75 %PTP and 0.04% POPOP + 0.04% Bis-MSB provides the highest light yield. The authors plan to use the same fluor additive concentration to produce application-specific scintillators that are not commercially available for nuclear reactor monitoring and medical applications.

1. Introduction

Detection of radiation is important to improve critical applications in high-tech fields; such as in medical imaging, national security, nuclear reactor monitoring, agricultural and environmental areas and basic sciences. While various radiation detection materials exist, plastic scintillators are often preferred due to their availability in large sizes at low cost and ease of manufacturing. Plastic scintillators are polymer-based materials that emit light when interacted with ionizing radiation. The initial emission is in the ultraviolet (UV) range, the UV light is then turned into visible light when doped with wavelength shifters [1]. Since the main purpose is to convert ionizing radiation into light, the synthesized scintillator must perform well with respect to the light emission spectrum, light transmission factors and light yield.

Recently, many improvements in plastic scintillator composition have been reported on, resulting in the increase in sensitivities to particular types of radiation detection. New types of plastic scintillator have been produced by adding organometallics [2–5], nanoparticles [6–8] and high concentrations of organic fluor for pulse shape discrimination [9–11]. While many companies produce plastic scintillators, e.g. Luxium Solutions [12], Eljen Technology [13], Epic Crystal [14], Rexon [15], they do not normally provide gadolinium or lithium doped products that enrich thermal neutron sensitivity. Gadolinium doped plastic scintillators are being considered for nuclear reactor monitoring in the future [16].

CRONUS Technology [17] in Turkey has started to produce commercial plastic scintillators, with and without gadolinium organometallic doping. Given the importance of determining the optimal concentrations

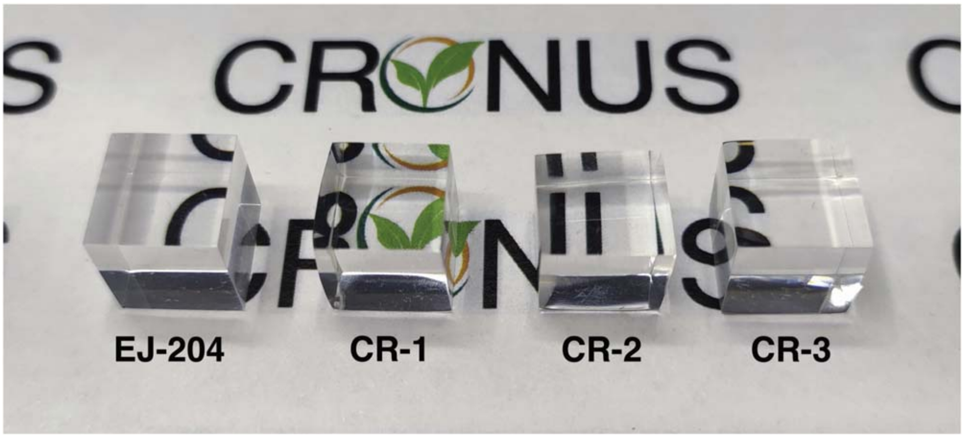

Figure 1. The synthesized plastic scintillator samples and the reference sample.

Table 1. List of scintillator samples and their incorporated additives.

Sample Name	Type and Concentration of primary additive (wt%)	Type and Concentration of secondary additive (wt%)
CR-1	1.5% PPO	0.08% POPOP
CR-2	1.5% PTP	0.08% Bis-MSB
CR-3	0.75% PPO + 0.75 %PTP	0.04% POPOP + 0.04% Bis-MSB

of flour to maximize the light yield, three samples with different flour content are analyzed in the present study. Their synthesis and the measurement of the optical properties are presented in the following sections.

2. Synthesis of plastic scintillator samples

The common approach to plastic scintillator production is thermal polymerization of a solution of liquid monomer containing specific additives. A typical plastic scintillator consists of three components: polymer base, primary fluor (first additive), and wavelength shifter (second additive). Incident particles excite molecules in the polymer matrix, the energy is transferred non-radiatively through the matrix and is absorbed by the fluorescent additive. The fluorescent additive releases the energy as a radiative emission in the UV region, the UV light is absorbed by the wavelength shifter and blue light is emitted.

The most used polymer bases for plastic scintillator synthesis are polystyrene and polyvinyltoulene. Since the latter provides slightly better light yield [18], it is chosen as the polymer base in the produced scintillator samples. The selected monomer is purified with activated alumina sorbent to remove inhibitor and water. Two different primary fluors (2,5-Diphenyloxazole (PPO, CAS:92-71-7) and p-Terphenyl (PTP, CAS:92-94-4)) and two different wavelength shifters (1,4-bis(5-phenyl-2-oxazolyl)benzene (POPOP, CAS:1806-34-4) and 1,4-Bis(2-methylstyryl)benzene (Bis-MSB, CAS:13280-61-0)) are compared. The additives are poured into the volume of purified monomer and stirred with a magnetic stirrer for 6 hours at 40 °C to ensure that the components dissolve homogeneously. High purity Argon gas is blown through the homogenous solution for two minutes for oxygen removal. Finally, the mixture is poured into a glass jar and sealed under argon atmosphere. It is then kept at 100-120 °C for 5 days. The obtained bulk scintillator samples are cut and polished into a cubic shape with a side length of 10 mm. To ensure reproducibility, cubic glass molds with dimensions of 10 mm will be used in the future. Figure 1 shows the synthesized plastic scintillator samples labelled CR-1, CR-2, CR-3, together with the reference plastic scintillator sample produced by Eljen Technology labelled EJ-204. The list of synthesized plastic scintillators with the details of incorporated additives is presented in table 1.

3. Optical properties of plastic scintillator samples

3.1. Transmission and emission measurement

The optical properties of the synthesized plastic scintillator samples in the visible light wavelength range is investigated using a Jasco V-730 UV-Vis spectrophotometer and compared with a EJ-204 plastic scintillator reference sample of the same geometrical shape. The transmission factors as a function of light wavelength are

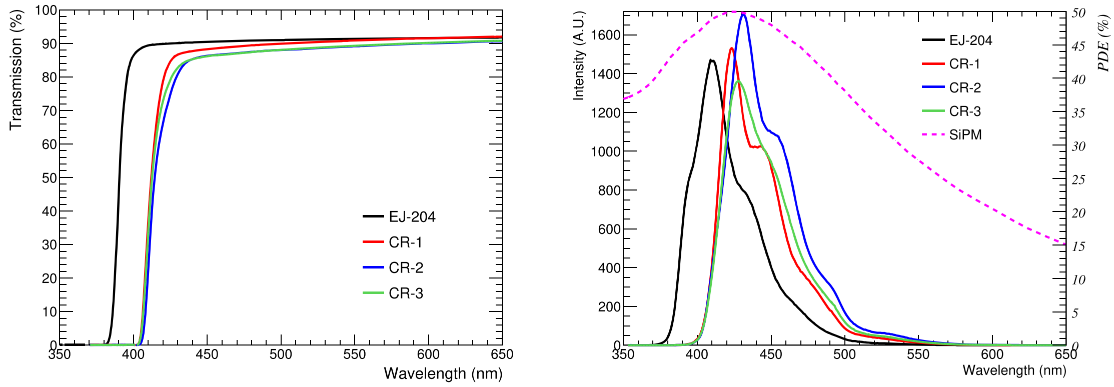

Figure 2. Transmission (left) and emission (right) distributions of EJ-204 and the produced plastic scintillator samples.

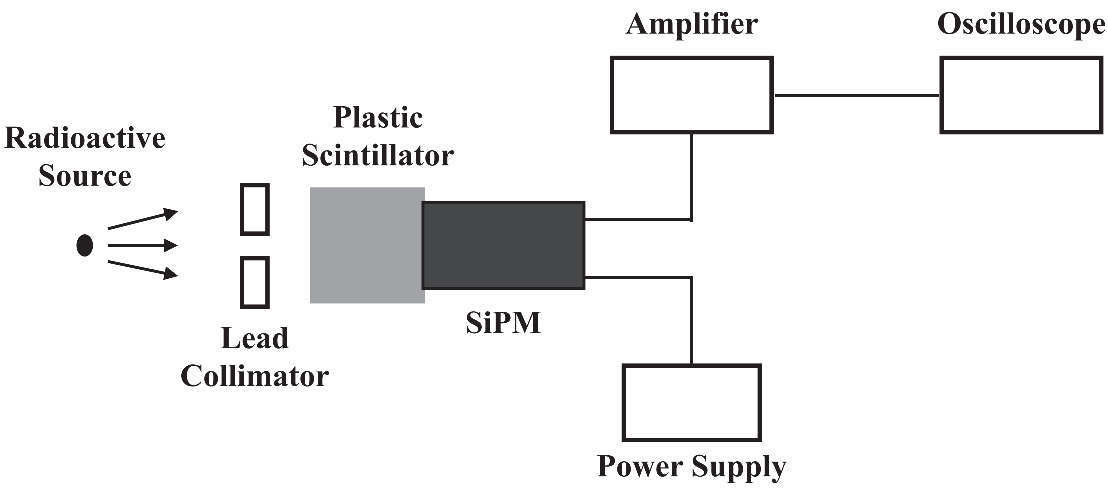

Figure 3. A schematic view of the experimental setup for measuring light yield.

presented on the left side in figure 2. The transmission factors of all the produced samples are comparable with EJ-204 and surpass 86% after 440 nm. In addition, the emission wavelength distribution of the produced plastic scintillator samples and the EJ-204 sample are shown on the right side in figure 2. The emission wavelength distributions are measured using a Jasco FP-8300 Spectrofluorometer by scanning excitation wavelengths from 200 nm to 400 nm. Since the samples consist of different types of primary and secondary fluor, the peak of the emission wavelengths show up at different values: 410 nm for EJ-204, 423 nm for CR-1, 431 nm for CR-2 and 428 nm for CR-3. The photon detection efficiency (PDE) curve of the Onsemi J-Series silicon photomultiplier (SiPM), which peaks at 430-440 nm, is also presented in the emission distribution [19]. The PDE of the scintillator samples at the emission peak values differ about 1%.

3.2. Light yield measurement

The light yield (LY) of a plastic scintillator corresponds to the number of produced photons per unit of deposited energy in the scintillator. It is one of the most important features of a scintillator since higher LY values provide better energy resolution. In order to determine the LY of a scintillator, a reference radioactive source with a known energy and a reference plastic scintillator sample with a known LY are needed. The main approach is to use the Compton-edge of the Compton spectrum, which is the maximum energy transfer value between the photon and the electron. The schematic diagram of the experimental setup for the light yield measurement is shown in figure 3. An Onsemi J-series SiPM with the size of 6×6 mm (MICROFJ-60035-TSV) is used as a photodetector and 10×10 mm plastic scintillator samples are coupled with it using Eljen EJ-550 optical silicon grease [20]. The optical silicon grease has a refractive index of 1.46 and its transmission is about 99% for a thickness of 0.1mm for 430 nm and higher. A low-voltage power supply is connected to the SiPM biasing board. The output signal is amplified via a CAEN N978 fast amplifier and the signal shape is digitized and recorded by a

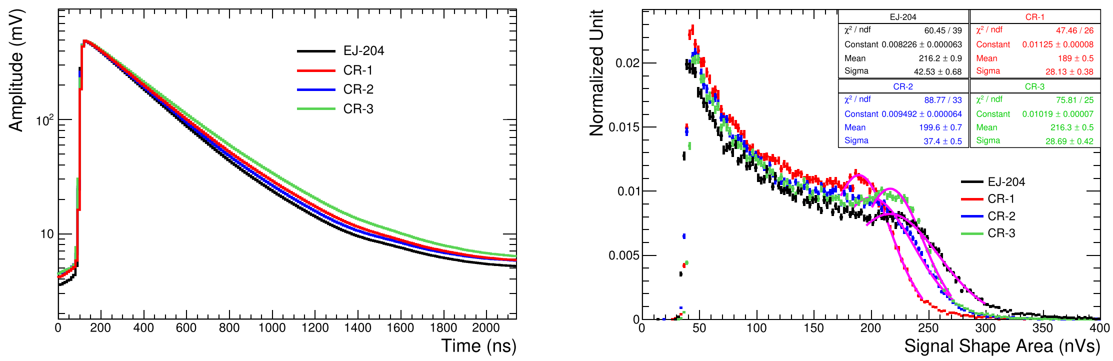

Figure 4. Signal shapes (left), and signal shape areas and fit parameters (right) of EJ-204 and the produced plastic scintillator samples.

Table 2. The RLY and LY values of the produced plastic scintillator samples with respect to EJ-204 with mean squared error estimation.

Sample Name	RLY at 90%	RLY at 80%	RLY at 70%	RLY at 60%	RLY at 50%	Average RLY	LY (photons/MeV)
EJ-204	100	100	100	100	100	100	10 400
CR-1	85.7	85.0	84.5	83.9	83.4	84.5 ± 0.4	8788 ± 42
CR-2	91.9	91.8	91.6	91.5	91.5	91.7 ± 0.1	9537 ± 10
CR-3	97.3	96.2	95.3	94.6	94.0	95.5 ± 0.5	9932 ± 52

Keysight MXR254A oscilloscope. ^{137}Cs is selected as the radioactive source since it has a clear Compton-edge at 477.3 keV. The plastic scintillators are wrapped with aluminum foil and plastic tape and located inside a wooden box with a wall thickness of 2 cm. The ^{137}Cs source is positioned 20 cm away from the scintillator.

The signal shapes for the prepared scintillator samples and EJ-204 are shown on the left side in figure 4. Peak normalization is applied to the signal shape distributions. Since the fluors in the plastic scintillator samples have different timing properties, the signal width of the scintillator samples is not identical. Therefore energy deposition estimation was performed by measuring the area under the pulse shapes instead of by measuring pulse height.

Since organic scintillators don't show photo-peaks due to their low-Z content, the determination of LY is performed by measuring the Compton-edge location. For this purpose, the edge region in the energy distribution is fitted with a Gaussian function, the height of the Gaussian peak is determined, and the edge location is taken to be the energy corresponding to a certain percentage of the peak height [7]. The suggested values in the literature vary from 66% to 89% but the optimum value changes depending on the detector and scintillator. Since choosing a specific percentage of the local Compton maximum is not an ideal way to represent the Compton-edge location [21], the percentage values are chosen from 90% to 50% in 10% steps to determine the relative light output. We consider evaluating the relative light yield output with different percentage values is a more conservative approach than what is suggested in the literature. The fit ranges of the Gaussian function of each distribution are chosen in such a way that once the fit is performed and has successfully converged, the peak value of the distribution stays within the fit range and the lower-end of the range is at least half a sigma away from the peak. We also note that changing these lower-ends by ± 10 nVs affect the relative light yields by less than 1% for all of the fits. The Gaussian fits are shown on the right side in figure 4. The relative light yields of the prepared plastic scintillator samples are determined from the EJ-204 commercial scintillator sample produced by Eljen Technology by $RLY_{sample} = RLY_{EJ-204} \times A_{CE,sample}/A_{CE,EJ-204}$, where RLY is the relative light yield and A is the signal shape area of the selected Compton-edge position. Since the light yield information of EJ-204 is already given by Eljen Technology as 10 400 ph/MeV, the relative light yield of EJ-204 is taken as 100 at each selected percentage value and the relative light yield of the prepared plastic scintillator samples are calculated based on it. The obtained relative light yield values at each selected percentage values are listed in table 2. The measurements show that the maximum deviations from the relative light yield at 90% to 50% are 2.7% for CR-1, 0.4% for CR-2, and 3.3% for CR-3. Finally, the light yield values of the prepared samples are estimated by multiplying the average relative light yield with the light yield value of EJ-204.

4. Conclusion

Producing application-specific scintillators such as Gadolinium doped plastic scintillators is crucial since they are not commercially available. For any such application, determining the optimum flour content for best light yield is important. In this study three plastic scintillator samples with different fluor additives have been produced in cubic shape with 10 mm length by a university-SME Collaboration. The LY, light emission and light transmission measurements of the scintillator samples have been performed along with an EJ-204 sample of the same size. The transmission factors of all the produced samples are comparable with EJ-204 and surpass 86% after 440 nm. The peak of emission wavelengths are 423 nm for CR-1, 431 nm for CR-2 and 428 nm for CR-3. The PDE of the scintillator samples at the emission peak values differ about 1% when used with a Onsemi J-series SiPM. The measurements show that the maximum deviations from the relative LY at 90% to 50% are 2.7% for CR-1, 0.4% for CR-2 and 3.3% for CR-3. The LYs are estimated by taking EJ-204 as a reference and found to be 8788 ± 42 for CR-1, 9537 ± 10 for CR-2 and 9932 ± 52 for CR-3. The results indicate that CR-3 (0.75% PPO + 0.75 %PTP and 0.04% POPOP + 0.04% Bis-MSB) has the highest LY value among the produced samples. We conclude that the CR-3 combination is the most suitable for a future gadolinium doped scintillator production.

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Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://cernbox.cern.ch/s/kHT6gfJGbI4nJ1q.

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