Fabrication and Characterization of Plastic Scintillators with PPO and Coumarin in an Epoxy Matrix

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Abstract. We fabricate plastic scintillators with varying concentrations of 2,5-diphenyloxazole (PPO) as a primary fluor and 7-diethylamino-4-methylcoumarin (coumarin) as a wavelength shifter and characterize their light yield. We demonstrate that effective scintillators can be made, and in one sample we achieve 27% higher light yield than in a commercially available scintillator. However, we also show that spatial variability has a significant impact on measured light yield. We show that higher concentrations of PPO and coumarin generally lead to higher light yields. We also demonstrate that the fabricated plastic scintillators can successfully detect muons in a CosmicWatch detector.

INTRODUCTION

Scintillators are widely used in particle detection and radiation measurement to convert the energy of ionizing radiation into visible light. For example, high-energy photons interact with a scintillator via the photoelectric effect, Compton scattering, or electron-positron pair creation, transferring their energy to electrons in the material. Molecules in the medium are excited by these electrons and subsequently emit deexcitation photons. Charged particles such as muons excite and ionize the molecules directly, triggering the same excitation/deexcitation processes, producing photons with a predictable, material-specific wavelength [1].

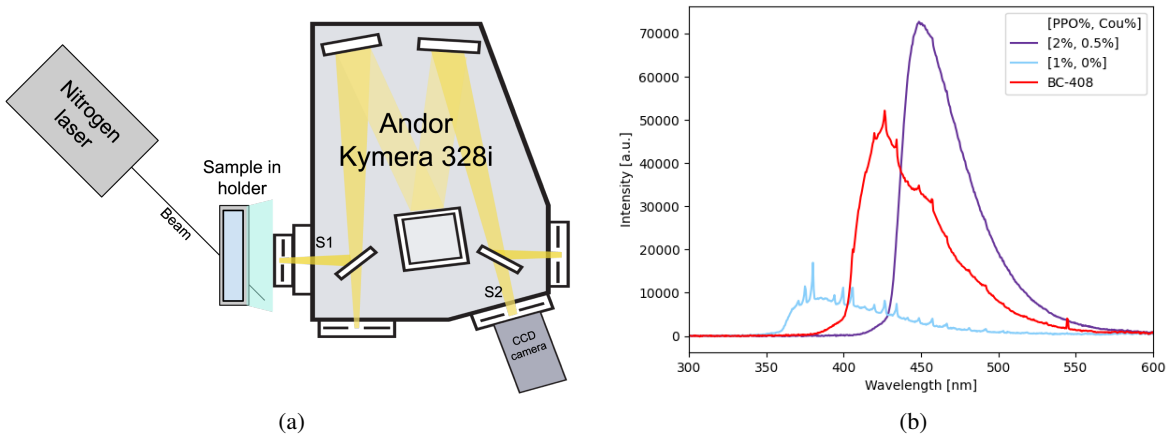
Plastic scintillators are transparent solids usually made from a polymer base such as polystyrene or polyvinyltoluene [1]. An organic fluorescent compound (fluor) is added to the base to make it scintillate [2]. In our case, the primary fluor, 2,5-diphenyloxazole (PPO), emits in the near-UV range, so a secondary wavelength shifter, 7-diethylamino-4-methylcoumarin (coumarin), is added to shift the emission to the visible spectrum since that is where most photon detectors are sensitive [1]. Having two wavelength shifters is a good way to limit the self-absorption within the scintillator by providing a larger Stokes shift, which is the separation between the wavelengths at which light is emitted and absorbed.

Commercial plastic scintillators such as Bicron BC-408 are readily available. BC-408 has a well-characterized emission spectra, peaking around 425 nm, and is frequently used in particle detectors. It has a high light output, fast response time, and good optical transparency [3]. However, we were interested in making our own scintillator to enable greater control of shape, size, and resulting photon emission spectra, since different photon detection systems may have different ranges of light collection capabilities. We particularly wanted a final emission wavelength well-matched to the SiPM chips used for CosmicWatch detectors [4]. We were also interested in demonstrating that it is feasible and inexpensive to make homemade scintillators in an undergraduate laboratory setting. Our highest concentration scintillator had a materials cost of 2.42 USD, compared to 21-25 USD for a similarly sized BC-408 scintillator [3]. Because we had previously used BC-408 with a CosmicWatch detector and were familiar with its characteristics, we used it as a reference for comparisons with the scintillators we fabricated.

This project originated in an upper-level undergraduate experimental techniques course at Wellesley College focused on particle physics detector technology. Our goals were to fabricate epoxy-based plastic scintillators using PPO as the primary scintillator and coumarin as the wavelength shifter and characterize their wavelength-shifting behavior and performance in a CosmicWatch muon detector [4].

METHODS AND MATERIALS

We chose to make our scintillators $5 \times 5 \times 1\,\mathrm{cm}^3$ so that we could use them in a CosmicWatch muon detector. To achieve the necessary volume, we used epoxy and hardener in a ratio of $1 : 0.83$ as specified by the manufacturer (the Epoxy Resin Store), resulting in a calculated weight of 22.51 g. We used concentrations of 0.5%–2% PPO and



(a) (b)

FIGURE 1: (a) Measurement setup. The laser beam is incident on the center of the sample but not the entrance slit of the spectrometer (S1). The dispersed light exits the spectrometer at slit S2, where it is imaged by a CCD camera. (b) Emission spectra of homemade (purple) and commercial (red) scintillators with a PPO-only sample (blue) shown for comparison. The spectra peak at 449 nm and 427 nm for the homemade and commercial scintillators, respectively. The roughness and additional peaks correspond to peaks in the nitrogen laser spectra that are above the absorption limit for PPO. We reason that coumarin in the purple sample absorbs additional light between 350 and 430 nm, resulting in a smoother spectrum.

0.01%–0.5% coumarin, as done in similar epoxy-based scintillator projects [5, 6]. We additionally made two reference samples, one with no PPO scintillator and another with PPO but no wavelength shifter. We measured and combined the amounts of epoxy and scintillators, stirring each sample by hand for 5 min until the mixture appeared visually homogeneous, and then added hardener. Samples were then cured on a hot plate for 2 h at around 60°C. Samples were not polished after fabrication.

To excite the PPO but not the coumarin, and therefore better isolate absorption by the primary fluor from absorption and emission by the wavelength-shifter, we ideally would have used a light source with a wavelength between 240 nm and 319 nm [7, 8]. We did not have such a source, so we instead used a nitrogen laser (337 nm). This wavelength still falls within the absorption range of the PPO, but it is long enough to directly excite the coumarin. A comparison of a sample containing only 1% PPO with a sample containing an additional 0.02% coumarin revealed a 72% increase in light yield for the coumarin-containing sample, indicating that direct absorption and emission by coumarin significantly contributes to light yield. We use two wavelength shifters, despite these complications, to give an overall larger shift in wavelength; the gap between PPO's emission and absorption peaks is only about 50 nm, but by adding coumarin, we can increase the gap to approximately 150 nm [7, 8]. The resulting emission wavelength of 499 nm is also better suited to our SiPM than that of just PPO at around 350 nm. As seen in Fig. 1(a), the laser was angled such that the beam was incident on the center of the sample without directly entering the spectrometer (S1). Spectra were averaged over 30 1-s measurements. To find the integrated light yield (ILY), we integrated the spectra over a 350–600 nm range. We defined relative light yield (RLY) as the ratio of each sample's ILY to that of the commercial sample. To test the spatial uniformity of our samples and quantify the effects of uneven mixing during the fabrication process, we recorded emission spectra at three locations (left, middle, right) on selected samples.

To test our scintillators in a particle detection context, we integrated one of our samples into a CosmicWatch muon detector [4].

RESULTS AND DISCUSSION

As seen in Fig. 1(b), the emission from the PPO-only sample peaked at 384 nm, while samples with both PPO and coumarin had a peak at 446 nm. Because there was no residual 384-nm peak in the PPO with coumarin sample spectrum, we conclude that the coumarin efficiently converted the PPO emission photons. Our scintillators have a higher peak emission wavelength than the commercial scintillator, although the emission intensity varies across samples, with RLYs ranging from 0.29 to 1.27.

TABLE 1: Relative light yield of our samples normalized to the commercial scintillator.

PPO (%)	Coumarin (%)	RLY	Name
	0.5	1.27	Sample 5
2.0	0.25	0.824	
	0.02	0.292	
	0.5	0.483	
1.0	0.25	0.455	
	0.02	0.423	
	0.5	0.458	
0.5	0.25	0.485	
	0.02	0.339	Sample 3

TABLE 2: Relative light yield for the different locations on the samples and the resulting spatial uncertainty, compared to the commercial scintillator measured at the center. The spatial uncertainty is defined as

$$\sigma_{\text{spatial}} = \max\left[\frac{L_{\text{left}} - L_{\text{center}}}{L_{\text{center}}}, \frac{L_{\text{right}} - L_{\text{center}}}{L_{\text{center}}}\right].$$

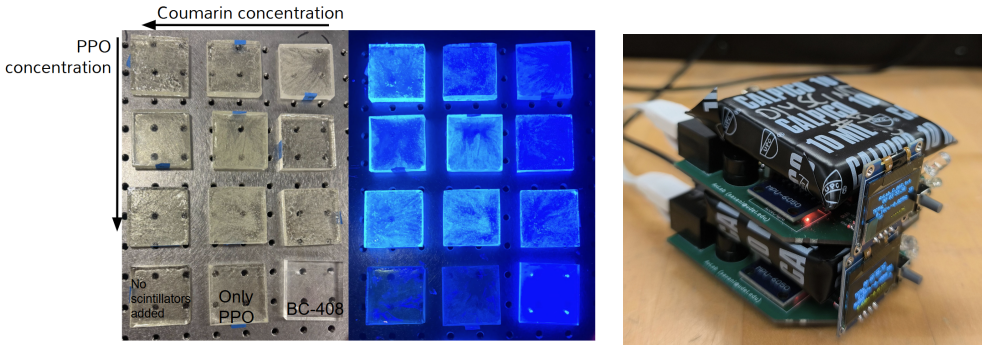
Sample	RLY_{left}	$\text{RLY}_{\text{center}}$	$\text{RLY}_{\text{right}}$	σ_{spatial} (%)
Sample 3	0.43	0.38	0.58	55.7%
Sample 5	0.62	0.64	0.69	8.2%
Commercial	0.98	1.0	1.09	8.8%

To identify a trend between PPO and coumarin concentrations and RLY, we created samples across a range of primary scintillator and wavelength shifter concentrations. The RLY values for these samples are shown in Table 1. These values were found with the laser beam incident on the approximate center of each sample. For a given PPO concentration, RLY increases with coumarin concentration. However, we do not find a monotonic increase of RLY with PPO concentration for a fixed coumarin concentration. This is particularly noticeable for the 0.02% coumarin samples. A likely explanation is that for the samples with the least coumarin (0.02%, corresponding to 5 mg), the 1-mg error associated with measurement and incomplete powder transfer has a larger proportional impact than for samples with higher concentrations of scintillator.

We selected the least and most visually uniform scintillators from our samples, samples 3 and 5, respectively [see Fig. 2(a)], to study the spatial variability in the RLY. We measured RLY with the scintillator illuminated at three different locations to see if the spatial variability impacts our light yield. We found no variation in the emission spectra shape or peak wavelength, but we observe significant variability in the RLY. The results are summarized in Table 2, and the spectra can be seen in Fig. 3. Sample 3 shows the largest spatial variability (56%), consistent with its appearance, while sample 5 and the commercial scintillator show comparable spatial uncertainties (8%), indicating similar levels of uniformity. The spatial variability in light yield suggests our mixing process could be improved. For future work, spatial variability studies could be done with samples of the same concentrations of PPO and coumarin. That would create a controlled comparison with all variables constant except the mixing quality.

These spatial nonuniformity tests had a slightly different setup than the PPO/coumarin concentration tests in Table 1. The differences in the setup were minor: small changes in the angle and distance to the laser. Our unpolished samples, made in a mold, have one flat side and one irregular side, though we did not keep track of which faced the spectrometer for the concentration light yield study. Although the RLY values show high variability resulting from spatial inhomogeneity in the samples, we are confident in the measured trend of RLY with PPO/coumarin concentration. Additional work that results in more homogeneous samples could help quantify the relationships between concentration and light yield. Similarly, we see a strong correlation between the amount of visual spatial nonuniformity of samples 3 and 5 and the measured spatial light yield differences. We note that the PPO and coumarin concentrations differ between the two samples, so their concentration could be a confounding variable.

To confirm that our scintillators were sensitive to ionizing radiation, we integrated sample 5 into a CosmicWatch muon detector. Coincidence data were first collected using two commercial scintillators integrated into two detectors in a stacked configuration. Next, we replaced the top BC-408 scintillator with our sample, otherwise maintaining the same configuration [see Fig. 2(b)]. Coincidence was recorded when both detectors in the stack reported an event at the same time, indicating a throughgoing muon. The coincidence rate is defined as the total number of coincidence events divided by the measurement time. The coincidence rate for the commercial-commercial configuration was $0.19 \pm 0.02\ \text{s}^{-1}$, while that for the sample 5-commercial configuration was $0.030 \pm 0.008\ \text{s}^{-1}$. This means that the muon detection efficiency with sample 5 is 16% of that with two commercial scintillators. This demonstrates that our sample is capable of detecting muons. It is important to note that the smoothness of the surface of the scintillator greatly impacts the photon detection efficiency: a polished surface provides a more uniform optical interface and increases the probability that photons are transmitted toward the detector [4]. The commercial samples were polished before integration into the muon detector; our homemade sample was not. We expect that polishing our scintillators would enhance the muon detection efficiency, though that study is beyond the scope of this work.



(a) (b)

FIGURE 2: (a) Photographs of scintillator samples under room light (left) and under UV illumination (right). Sample 3 is in the top right corner, and sample 5 is in the left column of the third row. (b) CosmicWatch muon detector configuration. Two detectors are stacked vertically, resulting in nearly 2π solid angle coverage on the sky.

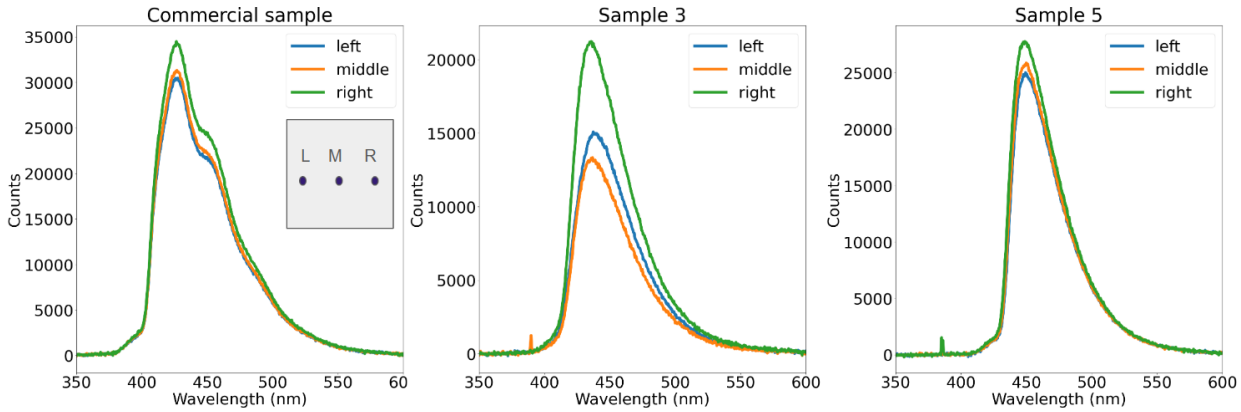


FIGURE 3: Emission spectra for the commercial sample, sample 3, and sample 5. The blue curve shows spectra from the left location, the orange curve the middle, and the green curve the right. The locations on the sample (L = left, M = middle, and R = right) can be seen in an inset in the first plot.

CONCLUSION

We created and characterized epoxy-based scintillators with a range of concentrations of PPO (fluor) and coumarin (wavelength shifter). In doing so, we demonstrated that using PPO and coumarin as primary and secondary scintillators works to convert UV light into visible light. We found that higher concentrations of both PPO and coumarin generally increase light yield, and we produced a sample with a higher light yield than a commercial scintillator. Many of our samples showed high spatial variability, so there is room for further investigation of synthesis methods to ensure more uniform distribution of the fluor and wavelength shifter powders in the epoxy. By integrating one of our samples into a CosmicWatch detector, we successfully measured muon candidates in coincidence, though at a lower detection efficiency relative to a commercial scintillator. This work shows that it is viable to make and characterize epoxy-based scintillators in an undergraduate laboratory.

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