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FABRICATION OF PLASTIC SCINTILLATOR USING POLYSTYRENE MATRIX BASED

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ABSTRACT

Experiments on making a plastic scintillator with polystyrene as a base material mixed with fluorescent compounds (primary and secondary dopants) have been performed. Primary dopants (PTP) used to produce emission at wavelengths of visible light and secondary dopants (POPOP) to shift the visible wavelengths to wavelengths that could be detected by Photomultiplier Tube (PMT)) were carried out. Experiments were performed on the melting points condition of these materials, which had previously been subjected to a thermo-mechanical analysis using a Thermogravimetric Analysis-Differential Scanning Calorimetry (TGA-DSC) machine, where the melting point was in the range of 200-240°C. Furthermore, the fabrication was carried out using the extrusion technique, where polystyrene pellets mixed with PTP (1.5% by weight) and POPOP (0.05% by weight) were fed into an extrusion machine which has four hot areas to obtain a thin plate plastic scintillator. The plates were then analyzed with a UV-Vis Spectrophotometer to determine the absorption spectrum and Fluorescence Spectrophotometer to determine the emission spectrum. From the results of the analysis, it was found that the samples that went through scintillation pellets and without the addition of antioxidants had absorption spectrum data of 330 nm and emission spectrum of 421 nm. These values are in accordance with the characteristics of plastic scintillators on the market.

Keywords: plastic scintillator, polystyrene, a fluorescent compound, extrusion

INTRODUCTION

The plastic scintillator is an instrument to detect the presence of radiation made from the organic matrix material. Organic scintillator materials such as polystyrene (PS) or polyvinyl toluene (PVT) polymers will be mixed with primary and secondary dopants to produce a good plastic scintillator. Plastic scintillators can be applied to the medical and security fields, including Radiation Portal Monitor, medical imaging, detection of ionizing radiation, muon tomography, and spectroscopy. From various research, it was stated that plastic scintillator is more durable than liquid scintillator, can be formed quickly, have fast rise and decay time, high optical transmission, have low production cost, and can be made in large dimensions [1-3] Due to the characteristics possessed, the development of plastic scintillators has been widely carried out.

Plastic scintillator with PS matrix based has been widely developed. This type of plastic scintillator can be fabricated in large dimensions but also can be made in tiny and thin sizes depending on the need. In its fabrication, plastic scintillator uses primary and secondary dopants as fluorescent compounds. These are examples of primary dopants, i.e., 2,5-Diphenyloxazole (PPO), p-Terphenyl (PTP), and 2-(4-tert-Butylphenyl)-5-(4-biphenyl)-1,3,4-oxadiazole (b-PBD), while for secondary dopant, i.e., 1,4-bis-[2-(5-Phenyl)oxazolyl]benzene (POPOP), 9,10-Diphenylanthracene (DPA), 1,4-bis(2-Methylstyryl) benzene (Bis-MSB), and 1,4-Bis(4-methylstyryl)benzene (Me-MSB). Based on the research conducted by Jun Zhu et al. [4], from all the primary and secondary dopant types mentioned above, PPO and POPOP were the most efficient additive dopants for polystyrene-based plastic scintillators with mass ratios of 0.2% and 0.02%, respectively.

However, the weakness of PS-based plastic scintillator is the efficiency of the plastic scintillator on the scintillation response, which can be decreased on dosimetry with photon energy less than 260 keV [5]. The decrease in scintillation response at low energy occurs due to the low effective atomic number of the scintillated plastic compared to the effective atomic number of water. The decrease in the scintillation response at low energies is also partly due to the cooling effect of ionization, in which the emission of light around excited or ionized molecules produced by solid ionizing radiation is inhibited [6]. To determine the energy response of several commercial plastic scintillators, Whittaker et al. [7] carried out measurements using low-energy X-rays in the range of 30 to 150 kV_p with HVLs between 0.2 - 8 mm. The obtained results showed a decrease in the relative response at an effective energy of 0.014 MeV when compared to the relative response at an effective energy of 0.061 MeV.

The fabrication process of a plastic scintillator can be done through polymerization or extrusion techniques [8]. Both techniques require primary materials, such as monomers and

polymers, and additional materials, such as primary and secondary dopants. Polymerization requires liquid monomer as the main ingredient, which has been previously purified, and then primary and secondary dopants are added. The polymerization process is carried out at a low temperature of 30-78°C to a high temperature of 120-160°C for 218-300 hours in the absence of oxygen [3,9]. The extrusion process uses polymer as the primary material and is carried out at high temperatures to eliminate the purification process [10].

In this study, a thermo-mechanical analysis was carried out to determine the melting point of the plastic scintillator materials, namely PS as the primary material, PTP as the primary dopant, POPOP as the secondary dopant. Furthermore, the extrusion technique performed the fabrication of a plastic scintillator made from the above materials. The absorption and emission spectra of the plastic scintillator were analyzed to determine its characteristics.

METHOD

The experiment was started by conducting a thermo-mechanical analysis using a Thermogravimetric Analysis-Differential Scanning Calorimetry (TGA-DSC). TGA-DSC are two different methods of thermal analysis that can be operated in combination or separately, yet combination usage of TGA-DSC was recommended [11]. This analysis was performed to determine the melting point of PS, PTP, and POPOP.

Furthermore, the fabrication of a plastic scintillator with the primary material of PS pellets was carried out by extrusion techniques at PT. Intera Lestari Polymer, Balaraja. Experiments were carried out by making 3 (three) plastic scintillator samples with the addition of PTP as the primary dopant and POPOP as the secondary dopant, as well as the addition of antioxidants. The composition of each sample can be seen in TABLE 1.

No	Sample Name	Polystyrene		РТР		POPOP			
		% wt	Weight (gram)	% wt	Weight (gram)	% wt	Weight (gram)	Additional Information	
1	PS 01	100	3000	1.5	45	0.05	1.5	Without antioxidant, going through scintillating pellets	
2	PS 02	100	3000	1.5	45	0.05	1.5	Adding some antioxidants, going through scintillating pellets	
3	PS 03	100	3000	1.5	45	0.05	1.5	Without antioxidant, not going through scintillating pellets	

TABLE 1.Plastic scintillator sample composition

In sample PS 01, PS pellets mixed with PTP and POPOP were fed into extruder machine number 1. Inside the extruder machine number 1, the mixture was heated to a temperature of 240°C and stirred with a screw for 2-3 minutes. From machine number 1, PS 01 samples were produced into scintillating pellets in the form of threads with a thickness of about 2-3 mm. The PS 01 sample scintillating pellet was then chopped into small pieces to an extruder machine number 2 to be molded into thin plates with a thickness of 2 mm.

PS pellets mixed with PTP and POPOP on PS 02 samples were added with antioxidants. This mixture was then fed into extruder machine number 1 and number 2. At the same time, the PS

03 sample, a mixture of PS pellets, PTP, and POPOP without antioxidants, was added and directly inserted into machine number 2.

These processes can be seen through the schematic diagram in FIGURE 1 below.

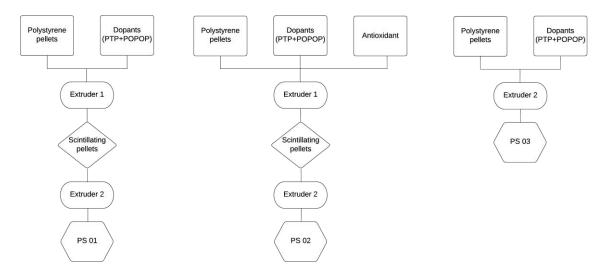


FIGURE 1. Schematic diagram of PS 01, PS 02 and PS 03 fabrication processes.

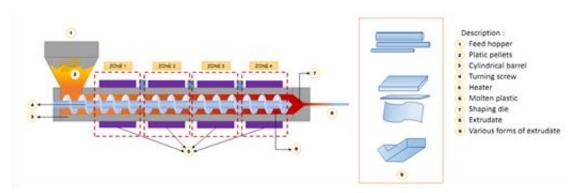


FIGURE 2. Schematic diagram of extruder machine number 1.

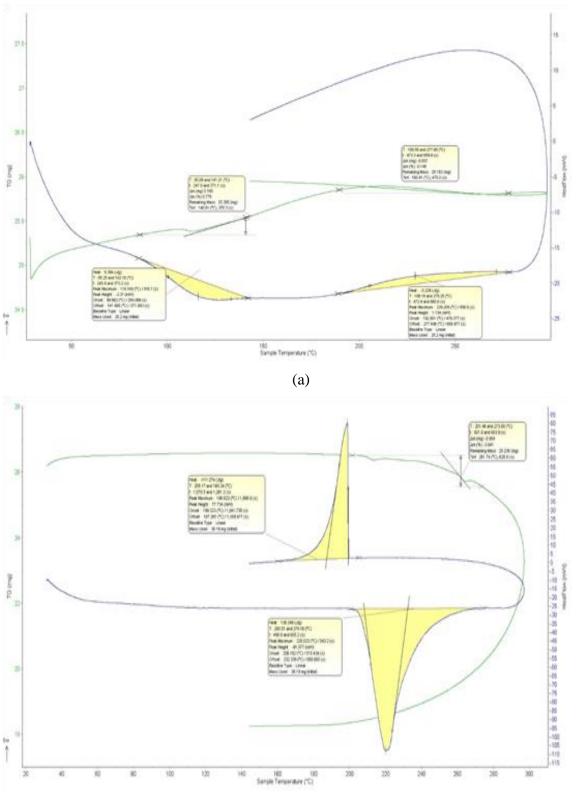
Extruder machine number 1, which produced scintillating pellets in the form of threads, was consists of feed hopper, turning screw, heater, and shaping die. In FIGURE 2 above, we can see the component of extruder machine number 1. The extruder machine also had a heater with four (4) hot areas.

The thin plates of PS 01, PS 02, and PS 03 samples were then analyzed using a UV-Vis Spectrophotometer to determine the absorption spectrum and HORIBA's Fluorescence-Spectrophotometer to determine the emission spectrum.

RESULT AND DISCUSSION

Thermo-mechanical analysis was carried out to determine each plastic scintillator material's melting point to maximize the process temperature. Since this research was still in the early

stages of the experiment (preliminary process), the melting point of the material was very important to be performed. These melting point information were used to estimate the temperature range of the process.



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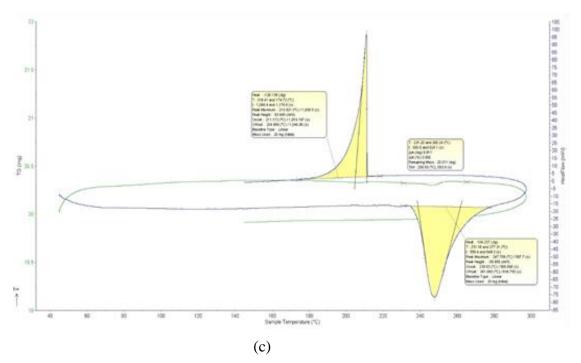


FIGURE 2. Thermo-mechanical analysis result using TGA-DSC machine for (a) of polystyrene, (b) PTP and (c) POPOP

From the FIGURE 3 above, can be seen that the melting point of PS was in the range of 189.59°C to 277.88°C; The melting point of PTP was in the range of 200.51°C to 274.18°C and the melting point of POPOP was in the range of 231.16°C to 277.31°C. So that in later experiments, the process temperature was set in the range of 200°C to 240°C.

On extruder machine number 1, there were 4 hot areas on the heater with a temperature of 210°C (heater 1) to 240°C (heater 4), as can be seen in Table 2. The temperature of the heater zone was adjusted according to the results of the thermo-mechanical analysis of the three materials (PS, PTP and POPOP).

TABLE 2. Hot zone on extruder machine number 1

Heater's hot zone	1	2	3	4	
Temperature, °C	210	220	230	240	

Antioxidants were added to the PS 02 sample to reduce the effect of oxidation on the sample. It was expected that with the addition of this antioxidant, the color of the sample would not turned into yellowish.



FIGURE 4. Thin plates from extrusion process irradiated by UV light

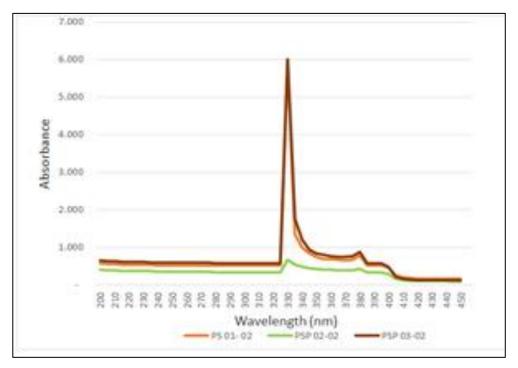
From the extruder machine, we obtained transparent white samples of the PS01, PS02 and PS03 samples. If the scintillation plates were exposed to UV light, it will emit blue light as can be seen in FIGURE 4.

From the results of the absorption spectrum analysis, as shown in FIGURE 5 (a), it was found that the absorption spectrum of the samples PS01, PS02, and PS03 had the same value at 330.24 nm. PS02 sample has the lowest absorbance value compared to other samples. This is because of the addition of antioxidants into the mixture of plastic scintillator materials, causing weak absorption of UV rays which results in weak excitation rays.

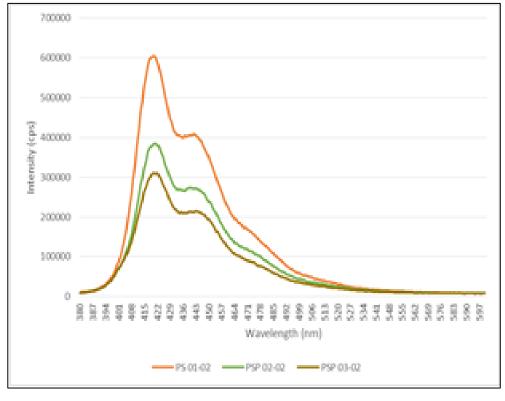
The results of the emission spectrum analysis, as shown in FIGURE 5 (b), show that the emission spectrum values of the PS01, PS02, and PS03 samples had almost the same value at 420 nm. While the average intensity value in the PS03 sample was the lowest. In this experiment, the PS, PTP and POPOP pellets in the PS 01 and PS02 samples were heated twice at their melting point, in the extruder machine number 1 and extruder machine number 2, resulting in a more homogeneous mixture. This had an effect on the intensity values of the two samples which were higher than the PS03 which only underwent one time heating.

Plastic scintillator with dopants or wavelength shifter absorbs the radiation energy and emits it in UVwith the help of primary dopant into blue light at $400 \sim 500$ nm with the help of secondary dopant, so that it can be detected by PMT [8,12].

In plastic scintillator with wavelength-shifter, the emission wavelength is approximately about 380 – 640 nm [8, 13]. In the commercial plastic scintillator with polyvinyltoluene based such as BC 408 and BC 412 (Saint Gobain Crystal) the wavelength of maximum emission are 425 nm and 434 nm, respectively [14]. While plastic scintillator with polystyrene based such as UPS 923 A (Amcrys) has wavelength of maximum emission of 425 nm [15].







(b)

FIGURE 3. Spectrum analysis on the PS 01, PS 02 and PS 03 samples (a) Absorbtion spectrum and (b) Emission spectrum

From the results obtained, it could be concluded that the emission wavelength of PS01, PS02, and PS03 samples were lower than the commercially available plastic scintillator and had values within the range for plastic scintillator with wavelength-shifter.

The result of absorption spectrum and emission spectrum from the experiment had a different value, which absorption spectrum value was lower than the emission spectrum value i.e 320 nm and 420 nm, respectively. These were in accordance with the Stokes law where the wavelength of emission would be greater than the wavelength of absorption [12].

CONCLUSION

Fabrication of plastic scintillator using extrusion technique based on PS matrix based not only can skip the purification process of styrene monomer, but also shorten the production process time. The addition of antioxidants to the PS02 plastic scintillator samples made its absorption to UV light keep lower. Since PS01 and PS02 underwent two (2) times heating at their melting point, it produced a more homogeneous dopants mixture which affected the intensity results. Direct fabrication using extruder machine number 2, the PS 03 sample had a low emission spectrum value, since it only went through one heating process so that the mixture was less homogeneous.

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REFERENCES

- [1] B. Liu and X. Ouyang, "Optical improvements of plastic scintillators by Nanophotonics," *in Plastic Scintillators*, Springer Nature Switzerland, 2021.
- [2] T. J. Hajagos *et al.*, "High-Z Sensitized Plastic Scintillators : A Review," *Advanced Materials*, vol. 30, no. 27, 2018.
- [3] C. H. Lee *et al.*, "Characteristics of Plastic Scintillators Fabricated by a Polymerization Reaction," *Nuclear Engineering and Technology*, vol. 49, no. 3, pp. 592-597, 2017.
- [4] J. Zhu *et al.*, "The impact of fluorescent dyes on the performances of polystyrene-based plastic scintillators," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 835, pp. 136-141, 2016.
- [5] A. Frelin *et al.*, "Comparative Study of Plastic Scintillators for Dosimetric Applications," *IEEE Transactions on Nuclear Science*, vol. 55, no. 5, pp. 2749-2756, 2008.
- [6] J. B. Birks, "The Theory and Practice of Scintillation Counting," *International series of monographs in electronics and instrumentation*, Elsevier, vol. 27, 2013.
- [7] C. A. Whittaker *et al.*, "Evaluating the energy dependence of various polystyrene based plastic scintillators," *Radiation Measurements*, vol. 122, pp. 57-62, 2019.

- [8] T. Ardiyati *et al.*, "PENGKAJIAN TEKNOLOGI PEMBUATAN DETEKTOR PLASTIK SINTILASI," *PRIMA-Aplikasi dan Rekayasa dalam Bidang Iptek Nuklir*, vol. 17, no. 1, pp. 50-57, 2020.
- [9] R. Rahmanifard *et al.*, "Synthesis and development of a vinyltoluene-based plastic scintillator," *Journal of Luminescence*, vol. 194, pp. 456-460, 2018.
- [10] A. Pla-dalmau, A. D. Bross and K. L. Mellott, "Low-cost extruded plastic scintillator," Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip., vol. 466, no. 3, pp. 482-491, 2001.
- [11] J. Yang and N. Hedin, "Advances of lab-scale analytical methods for solidification/stabilization technologies," in Low Carbon Stabilization and Solidification of Hazardous Wastes, Elsivier, pp. 483-495, 2022.
- [12] A. Wieczorek, "Development of novel plastic scintillators based on polyvinyltoluene for the hybrid J-PET / MR tomograph," Jagiellonian University, 2017.
- [13] C. Dujardin and M. Hamel, "Introduction-Overview on Plastic and Inorganic Scintillators," *in Plastic Scintillators*, Topics in Applied Physics Springer Nature Switzerland, pp. 3-33, 2021.
- [14] Saint-Gobain Crystal, "Organic Scintillation Materials and Assemblies," 2021.
- [15] Radcore-Amcrys, "Scintillation Materials and Detectors," 2008.