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Synthesis and development of a vinyltoluene-based plastic scintillator

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ABSTRACT

In this paper, the effects of some synthesis parameters such as type and concentration of primary and secondary dopants, temperature and time of polymerization on luminescence properties of a vinyltoluene based plastic scintillator are investigated. Thermal polymerization was used to polymerize the samples. After synthesizing, the samples were cut into dimensions of 2.5 cm diameter by 1 cm thickness and were evaluated compared to the commercial sample BC-400. The results of gamma-ray spectroscopy under cobalt-60 and cesium-137 sources as well as photoluminescence spectroscopy showed that some synthesized plastic scintillators have a higher light yield with respect to commercial one. The measurements showed that the increase of dopant larger than optimum concentration degraded the performance of plastic scintillator due to concentration quenching. It was also found that pre-treatment at the temperature of 78 °C led to about 6% light yield loss compared to direct treatment at 160 °C, due to more non-uniform structure acquired during former treatment which might attenuate the efficient transfer of energy.

1. Introduction

Radiation detectors are instruments which can identify the presence of energetic particles, radiations emitted from nuclear decay and cosmic rays. Scintillation detectors are the most common type of detectors which can measure ionizing radiation energy by meaning photoluminescence effect excited by incident radiation. Generally, the scintillator materials are utilized in liquid, crystalline and plastic states. The ease of fabrication, the possibility to produce different shapes in large dimensions, desirable mechanical stability, fast decay time, fair efficiency and relatively low manufacturing cost are the features making the plastic scintillators as a suitable alternative for liquid and crystalline ones [1,2]. In addition, the problems of handling, sealing, toxicity, flammability, disposal and sensitivity to dissolved oxygen in liquid scintillators [3,4] and difficulty of preparation in large volume, complicated growing and machining processes and high cost in crystalline scintillators have accelerated the development of plastic scintillators and their application rather than liquid and crystalline systems [5,6]. A plastic scintillator is comprised of a polymer as base material and dopants whose selection is vital to achieve the desirable luminescent properties. Since the presence of aromatic groups is necessary for fluorescence of the base material, aromatic polymers such as polystyrenes, polyesters, polysiloxanes, polyepoxides and polyvinyl xylene were utilized [6–9], however, amongst which, polystyrene and poly (vinyltoluene) are the most prominent [1,2,6–9]. It was found that the latter has a higher scintillation efficiency [10]. The dopants are also incorporated into the polymeric base to improve the scintillation mechanism, wavelength shift and increased light output [11]. In terms of dopant, its nature and concentration are important to reduce the effect of self-absorption, to prevent the significant efficiency loss during the energy transfer process and to increase the wavelength of light output. Hamel et al. [12] have attained an increase of light yield with increasing of primary dopant. Nakamura et al. [13] demonstrated that photon yields can be controlled by the fluor concentration. The studies conducted by Basile et al. [14] on the light output of poly(vinyltoluene)-based plastic scintillator containing 2-phenyl-5-(4-biphenylyl)-1,3,4-oxadiazole (PBD) as a primary dopant and diphenylstilbene (DPS) as a secondary dopant, indicated that an increase in the concentration of PBD up to 4 wt% could lead to the improved light output. Destruel et al. [15] have also investigated the effect of 1-phenyl-3-mesityl-2-pyrazoline (PMP) concentration as a dopant upon the performance of poly(vinyltoluene) polymer. Their results showed that the materials containing 0.05 wt% of PMP had a scintillation efficiency and the decay time comparable with those of commercial plastic scintillator; NE110. Generally speaking, there is an optimum dopant concentration. Sun et al. [16] showed that the concentration quenching occurred for the samples with PBD concentration larger than 1% in polystyrene. The most efficient light output was also reported by Zaitseva et al. [17] at relatively low dye concentrations about 1 wt% of 2,5-diphenyloxazole (PPO) where PPO changed from 0.1 to 30 wt% in poly(vinyltoluene)

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matrix loaded with 9,10-diphenylanthracene (DPA) as a secondary dye.

The evaluation of Zhu et al. [18] on the incorporation of different primary dopants such as PPO, *p*-terphenyl and butyl-PBD into polystyrene-based plastic scintillator revealed that PPO could show a stronger fluorescence intensity and light yield. They also indicated that with increasing of PPO concentration, the light yield was increased when it was less than of 2 wt%. van Loef et al. [11] were also found that light yield and scintillation decay time were remarkably improved compared to BGO scintillator when PPO concentration was increased up to 30 wt% in polystyrene. In terms of dopant type, there are also some reports that showed the detection efficiency was successfully enhanced by the incorporation of zirconia nanoparticles [19] and quantum dots (QDs) [8]. However, surface chemical modification or incorporation in a pre-polymer has been suggested to prevent luminescence self-quenching due to particle aggregation [8].

Secondary dopant concentration is another important parameter which can affect luminescent properties of a plastic scintillator. The studies done on polystyrene-based scintillator doped with *p*-terphenyl [20] and PPO [18] as primary dopants and POPOP (1,4-bis-[2-(5-phenyloxazolyl)]-benzene) as a secondary dopant showed that there was an optimal secondary dopant concentration which could provide a maximum light yield. It is worth to note that this concentration cannot be a fixed characteristic. Adadurov et al. [20] revealed when larger dimensions of a plastic scintillator were used, optimal concentration of POPOP solute was decreased as a result of increased absorption contribution. Polymerization temperature can also influence upon luminescence output. In order to examine the efficiency of plastic scintillator affected by temperature, Funt et al. [21] investigated the synthesis temperature in the range of 80–200 °C for a poly(vinyltoluene)- based plastic scintillator. Their results indicated that synthesis temperatures between 80 and 150 °C yielded a significant light output while the increase of temperature over 175 °C decreased this feature. A higher light output was also obtained by Basile et al. [22] from the samples which were polymerized at the higher temperature.

According to above discussion, the constructing components of a plastic scintillator and synthesis conditions can significantly affect the light response. However, the lack of a systematical and concentrated study on a certain composition is well felt. Therefore, at the present study has been attempted to take into account the effect of different parameters such as type and concentration of dopant and temperature of synthesis on luminescent properties of a poly(vinyltoluene)-based plastic scintillator.

2. Materials and methods

2.1. Synthesis of plastic scintillator

In order to synthesize the plastic scintillator, the vinyltoluene monomer was used with purity of 99.5%. *P*-terphenyl and PPO were used as primary dopants and the secondary dopant was POPOP. All materials were supplied from commercial vendors (Aldrich Co.). At the first step, it was necessary to purify the vinyltoluene from present inhibitors in the monomer. Hereby a mixture of 100cc distilled water and 3 g normal sodium hydroxide (NaOH) was prepared and added to vinyltoluene and was spent an enough time about 10 min to achieve a complete separation. This procedure was repeated three times to reach the accurate purification. Then, the distilled vinyltoluene was mixed with primary and secondary dopants. The mixture was degassed through bubbling method with high purity nitrogen gas (99.9995 wt%) and was poured into a cylindrical vial with diameter of 25 mm by height of 60 mm in an air-free atmosphere provided by a glove box. For the solid samples, at first, the vial was sealed within the glove box and was kept at a constant temperature for 5 days to perform the polymerization. Thus, thermal initiation was used to complete polymerization. Table 1 lists the details of incorporated dopants and synthesis conditions for different samples used in present study. The obtained blank was cut and polished into disks of 25 mm diameter and 10 mm thickness for next investigations. Fig. 1 shows a sample of the synthesized plastic scintillator.

2.2. Scintillation measurement

In order to evaluate energy resolution of the synthesized plastic scintillator and to measure light yield, the samples were excited by collimated gamma rays from cesium-137 and Cobalt-60 sources and energy spectra were collected by Institute of Applied Physics (IAP-MCA-4000) spectroscopy. Fig. 2 shows setup of the measurement device consisting of the source, detector and photomultiplier tube (PMT) coupled to specimen. Each sample was placed in the same position with respect to the source and the photomultiplier. The properties of the gamma sources are observed in Table 2. The specification of the photomultiplier was CR-169 model with a spectral response in the range of 300–650 nm and a maximum wavelength of 420 nm. To reduce the light reflection from the scintillator, it was housed in an aluminum casing. A silica gel was also used between the plastic scintillator and the photomultiplier as an optical coupling medium. At all, the final signal was measured by an oscilloscope device connected to the photomultiplier tube. In order to evaluate the scintillator performance, light yield of the samples was estimated by using Compton scattering edge position. The middle point where the slope drops to one-half of the value of the Compton peak was selected as Compton edge position.

Photoluminescence spectroscopy was performed by a miniature PL-Uvis-2500, PhysTec equipped with a Xenon lamp for excitation a wavelength of 265 nm to determine the extent of dopant-induced wavelength shift compared to the exciting wavelength. The samples whose fluorescence spectra were determined were the same samples that were measured for light output under gamma excitation. The fluorescence spectra were obtained by examining the radiation emitted by the face perpendicular to that which was directly excited. In order to evaluate the luminescent property of the synthesized samples compared with that of a commercial one was used from a cylindrical shaped sample of BC-400 with identical physical dimensions and surface polish as the synthesized sample. The specifications of BC-400 plastic scintillator has been given in Ref. [3].

3. Results and discussion

3.1. Spectral output measurement

Fig. 3 shows pulse height spectra for Cesium-137 and Cobalt-60 of the synthesized plastic scintillator samples containing two kinds of different dopants compared to the commercial one. The behavior of these spectra indicates a strong dependence with nature of dopant. Since the measurement of the Cesium-137 and Cobalt-60 spectra as well as count rate are not only affected by energy and nature of sources [23] but also depend on kind of detector [24,25], hence due to the variation of type and concentration of dopant in the synthesized samples, it is expected a change in measured spectrum. Fig. 3 and Table 1 show that light yield increases about of 14% when PPO dopant is used. This indicates that the nature of first additive is an effective parameter to achieve a higher detection efficiency in a scintillator detector. The difference in scintillation behavior between PS1 and PS2 samples can be attributed to the low solubility of *p*-terphenyl dopant in the matrix. Although, the increased temperature during polymerization improves the solubility of *p*-terphenyl, but it seems that its simultaneous occurrence with polymerization process cannot make a homogeneous distribution of first additive throughout the matrix leading to a reduction of light output. In addition, the results obtained by Basile et al. [8] on the light output measurements of plastic scintillator doped with PBD and *p*-terphenyl showed that light emitted from *p*-terphenyl was well below the range of maximum sensitivity of the photomultiplier when no secondary solute was used in the system, but addition of a small amount