Blue-emitting quantum dots based liquid scintillator for radiation detection

Sujung Min [a,b], Bumkyung Seo [b], Changhyun Roh [b], Sangbum Hong [b*], Jae Hak Cheong [a]
[a]*KyungHee University, Yongin 17104, Korea*
[b]*Korea Atomic Energy Research Institute, Daejeon 34057, Korea*
[*]*Corresponding author: sbhong@kaeri.re.kr*

1. Introduction

There are various methods for detecting radiation using ionization, excitation, luminescence, and chemical reaction. Among them, a scintillation detector using excitation and luminescence reaction is being researched and developed in various fields such as medical, industrial, and national defense by combining a scintillator and an optical sensor. Such a scintillation detector selects a scintillator according to the detection purpose. The scintillator is largely divided into an inorganic scintillator and an organic scintillator. The inorganic scintillator has high luminescence efficiency, high chemical resistance, and high atomic number and density, so it is easy to measure gamma-rays or X-rays. Since organic scintillators are mainly composed of C, H, and O, the probability of photoelectric effect is low, so it is easy to measure beta or alpha. Inorganic scintillators have the disadvantage of being difficult to manufacture and expensive. However, the organic scintillator has advantages such as low price, flexibility, and easy production in various shapes and sizes depending on the purpose of the user. Organic scintillation detectors are generally composed of three materials. The first material is a polymer and absorbs incident radiation. The second material is the primary dopant, which absorbs the energy deposited in the polymer and emits ultraviolet light. Here, a dopant is a light-emitting material constituting an 'organic emission layer (EML) together with a polymer material as a host. The third material is a wavelength shifter that absorbs ultraviolet light and emits blue light. Because the polymer has low quantum efficiency of fluorescence, a fluorescent dye that emits ultraviolet light is added. And a secondary dopant, which is a wavelength shifter, is used to emit light suitable for the photosensor. In this study, we propose the use of quantum dots composed of high atomic numbers not only to solve the problem of secondary dopants but also to increase the interaction probability with incident photons. Also, quantum dots were used to improve detection efficiency with a precise emission wavelength and quantum dots can control the bandgap by controlling the size of the particle. As such, by controlling the wavelength emitted by the quantum dot according to the size of the particle, it is possible to obtain the precise emission wavelength and excellent efficiency required in the sensor.

2. Methods and Results

2.1 Optical properties of quantum dots and liquid scintillator

The first optical characteristic is the decay time (fluorescence extinction time) characteristic of synthesized quantum dots. For the decay time, the time-resolved fluorescence analysis equipment that analyzed the luminescence characteristics was used. Decay time was analyzed for each sample at an 361nm excitation wavelength and is calculated through Equation (1) based on raw data. Equation (1) is an equation for multi-exponential fitting of the spectrum over time, A_n is the amplitude of component nth, and τ_n is the lifetime of component n_{th}. The decay time of the synthesized quantum dots was analyzed to be 77 ns at an excitation wavelength of 316 nm. In general, the decay time of a plastic scintillator is ~ns, and since quantum dots are composed of high atomic number materials, it is analyzed that the decay time is increased due to delayed fluorescence.

$$I=[A_1 \times e^{\frac{-t1}{\tau1}}] + [A_2 \times e^{\frac{-t2}{\tau2}}] \qquad (1)$$

The second characteristic is Photoluminescence (PL) analysis. The PL analysis result is shown in Figure 1. In the case of PPO, a peak was observed around 380 nm, and when QD was added, a peak was also observed around 420 nm. In addition, as the content of QD increases, the effect of PPO decreases and the effect of QD increases. QD has a broad absorption wavelength range, and as the QD content increases, the energy transfer rate from PPO improves and the intensity of fluorescence increases. In other words, it is shown that energy transfer such as FRET (Fluorescence resonance energy transfer) phenomenon is well accomplished. Here, FRET means that when there are two fluorescent materials that receive light of a specific wavelength and emit light of a specific wavelength, the material that gives energy is called a donor, and the material that receives energy is called an acceptor. At this time, if the emission wavelength of the donor matches the absorption wavelength of the acceptor, the energy transfer rate is improved. As such, FRET is a mechanism to explain energy transfer between molecules using two fluorescent materials.

2.2 Evaluation of Gamma detection performance

In this section, the measurement performance evaluation results were described using the liquid scintillator manufactured. First, the detection characteristics of Cs-137 were analyzed as shown in Figure 2. As shown in Figure 2, it was analyzed that the spectrum shifted due to self-absorption as the concentration of quantum dots increased. Here, self-absorption is a phenomenon in which energy emitted from a certain material is absorbed by the material itself, that is, it occurs when the absorption wavelength and the emission wavelength overlap, meaning that the intensity decreases. In addition, in the case of the scintillator to which quantum dots were added, it was analyzed that the Compton edge shifted to the left compared to the spectrum of commercial plastics or scintillators to which quantum dots were not added.

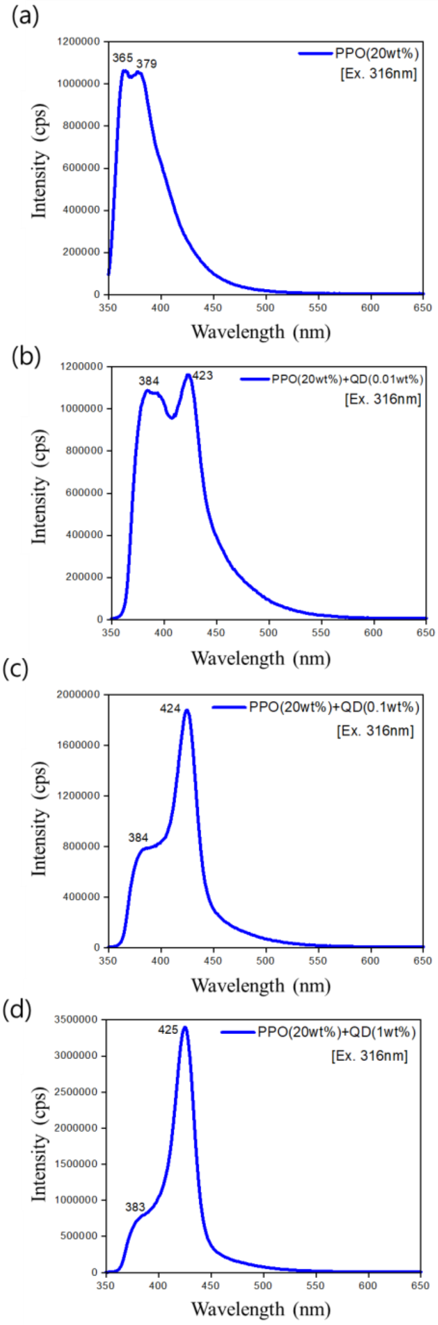

Fig. 1. Photoluminescence analysis result by quantum dot content at 316nm excitation wavelength (a) PPO (20wt%) (b) PPO(20wt%) and QD(0.01wt%) (c) PPO(20wt%) and QD(0.1wt%) (d) PPO(20wt%) and QD(1wt%), The red box indicates the region of the PPO.

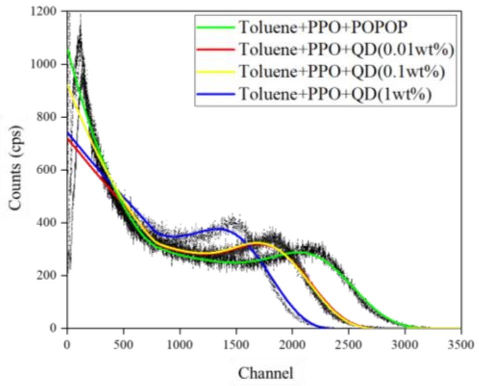

Fig. 2. Measurement results for each sample using Cs-137 source. The black line is raw data, and for easy identification, they are pre-processed by smoothing and ex-pressed as colored lines.

3. Conclusions

CdS/CdZnS/ZnS having a multi-shell structure having a blue wavelength was successfully synthesized through a hot injection method. The wavelength of the synthesized quantum dots is suitable for the response of the photomultiplier tube (PMT) used in this study. It is a wavelength with a quantum efficiency of about 25% or more. When compared with the emission characteristics of POPOP, it was confirmed that a single emission peak was observed with good resolution in quantum dots. In addition, as a result of performing optical property evaluation by fabricating a liquid scintillator containing quantum dots, the quantum dot-based sample showed a tendency to decrease in light yield compared to commercial materials. This is considered to be due to the effect of the primary dye and self-absorption of quantum dots as the quantum dot content increases. However, as a result of evaluating the detection efficiency based on the compton edge region, it was analyzed that the efficiency was improved by up to 7.7% when QD was used compared to the existing commercial material (POPOP). Based on the results of this study, it is judged that quantum dots can replace existing materials through content optimization of quantum dots, surface modification of quantum dots, and component changes.

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