Development of Organic Scintillators Using Molecules Exhibiting Aggregation-Induced Emission

<u>Masanori Koshimizu</u>¹, Atsushi Sato², Akito Watanabe², Yutaka Fujimoto², Keisuke Asai²

koshimizu.masanori@shizuoka.ac.jp

¹Research Institute of Electronics, Shizuoka University, 3-5-1 Johoku, Naka-ku, Hamamatsu 432-8011, Japan
²Department of Applied Chemistry, Tohoku University, 6-6-07 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan Keywords: aggregation-induced emission, plastic scintillator, organic crystalline scintillator

ABSTRACT

We have succeeded in the development of plastic scintillators containing phosphors with aggregationinduced emission characteristics.

1 Introduction

Organic scintillators are categorized into three groups: one is organic crystalline scintillators, which are widely used for fast neutron detection. The other two are plastic and organic liquid scintillators, which are easily fabricated in a large scale. Organic liquid scintillators are also used for detection of radioactive substances as solutes. From the viewpoint of scintillation properties, organic scintillators exhibit faster scintillation decay and lower scintillation light yield than those of inorganic scintillators. One of the cause of the low scintillation light yield in organic liquid and plastic scintillators is insufficient energy transfer from the solvent or host polymer to light emitting molecules: the energy transfer efficiency can be enhanced by containing light emitting molecules at high concentration; however, scintillation light yield would be reduced owing to concentration guenching.

To avoid the concentration quenching, we used light emitting molecules which have aggregation-induced emission (AIE) characteristics. We have developed plastic scintillators containing AIE phosphors [1] and organic crystalline scintillators based on AIE phosphor molecules [2]. Herein, the achievement in both approaches is presented.

2 Experiments

1,1,2,2-tetraphenylethylene (TPE) was used as the phosphor molecule exhibiting AIE properties for the development of plastic scintillators: at high concentration, TPE does not have direct π -stacking structure and hence suppress the non-radiative relaxation channel. Polystyrene (0.45 g) was first dissolved in tetrahydrofuran (5.0 g), and subsequently 0.10, 0.50, 1.0, 2.0, 4.0, ro 6.0 mol% of TPE was added to the solution and dissolved. The solutions were dried at room temperature for 5 d to obtain solid-state scintillator samples.

Organic crystalline scintillators were developed based

on 1,1,2,3,4,5-hexaphenyl-1H-silole (HPS), which is commercially available molecule exhibiting AIE. The crystals were grown in a slow evaporation method.

Photoluminescence excitation-emission maps and quantum yields were obtained using an absolute quantum yield spectrometer. Pulse height spectra of scintillation detectors equipped with the sample scintillators were recorded using 662-keV gamma-rays from ¹³⁷Cs for the plastic scintillators containing TPE and 59.8-keV gamma-rays from ²⁴¹Am for the HPS crystal. X-ray-induced radioluminescence spectrum was obtained using an X-ray generator equipped with a Cu-target X-ray tube operated at 40 kV and 40 mA in combination with a CCD-based spectrometer.

3 Results and Discussion

Figure 1 presents the photoluminescence quantum yield of TPE in the plastic scintillators as a function of concentration. The quantum yield does not decrease at the highest soluble concentration of 6 mol%, which indicates that the concentration quenching is suppressed. Excitation-emission map of the plastic scintillator containing TPE at 6 mol% is shown in Figure 2. The excitation peak at 370 nm and the emission peak at 480 nm are attributed to the excitation and emission of TPE. The pulse height spectra of the scintillation detectors equipped with the sample scintillators for 662keV gamma-rays from ¹³⁷Cs. are presented in Figure 3. The high channel edge of the spectra of the sample scintillators shifted toward the higher channel with TPE concentration, which indicates that the scintillation light vield increased with the TPE concentration, although the scintillation light yield seems to be about half of that of the plastic scintillators containing b-PBD as the fluorescent molecule. This is owing to the low photoluminescence quantum yield of TPE.

X-ray-induced radioluminescence spectrum of the HPS crystal is presented in Figure 4. A clear band at 490 nm attributable to the emission of HPS was observed. The scintillation decay profile of the HPS crystal is shown in Figure 5. The decay behavior was fitted with a sum of two exponential decay functions, and the decay time constants were 5.4 and 21 ns, which are attributed to the

emission from singlet excited state and triplet-triplet annihilation, respectively. The pulse height spectra of the scintillation detectors equipped with the HPS crystal or EJ-256 are presented in Figure 6. Based on the comparison of the full-energy peak channels, the scintillation light yield of the HPS crystal was estimated to be 11,000 photons/MeV.



Fig. 1. Photoluminescence quantum yield of TPE in plastic scintillators as a function of concentration.



Fig. 2. Excitation-emission map of plastic scintillator containing TPE at 6 mol%.



Fig. 3. Pulse height spectra of scintillation detectors equipped with sample scintillators for 662-keV gamma-rays from ¹³⁷Cs.



Fig. 4. X-ray-induced radioluminescence spectrum of HPS crystal.



Fig. 5. Scintillation decay profile of HPS crystal.



Fig. 6. Pulse height spectra of scintillation detectors equipped with HPS crystal or EJ-256.

Acknowledgements

This research was supported by the Grant-in-Aid for Scientific Research (A) (Grant No. 18H03890, 2018-2021). Part of this research was supported by the Cooperative Research Project of the Research Center for Biomedical Engineering, Ministry of Education, Culture, Sports, Science, and Technology.

References

- Arisa Magi, Masanori Koshimizu, Yutaka Fujimoto, Takayuki Yanagida, Keisuke Asai, Radiat. Meas. 137 (2020) 106401.
- [2] Atsushi Sato, Masanori Koshimizu, Yutaka Fujimoto, Shusuke Komatsuzaki, Shunji Kishimoto, and Keisuke Asai, Mater. Chem. Front. 6 (2022) 1470.