Comparative study of neutron and gamma-ray pulse shape discrimination of anthracene, stilbene, and p-terphenyl

Takayuki Yanagida^{a)}, Kenichi Watanabe^{b)}, Yutaka Fujimoto^{a)}

^a Kyushu Institute of Technology, 2-4 Hibikino, Wakamatsu, Kitakyushu, Fukuoka, 808-0196, Japan

^b Nagoya Univ., Furocho, Chikusa, Nagoya, 464-8603 Japan

Abstract

Solid state organic scintillators, such as anthracene, stilbene, and p-terphenyl were investigated on their basic scintillation properties and neutron-gamma discrimination capabilities. Scintillation wavelengths under X-ray irradiation of anthracene, stilbene, and p-terphenyl were 445-525, 400-500, and 350-450 nm, respectively. Scintillation light yields of anthracene, stilbene, and p-terphenyl under ¹³⁷Cs gamma-ray irradiation were 20100, 16000, and 19400 ph/MeV, respectively. Neutron and gamma-ray events discrimination capabilities were examined and anthracene exhibited the best figure of merit among three organic scintillators.

Keywords: Scintillator; organic; anthracene; stilbene; p-terphenyl; PSD

*¹ Corresponding author. Present address: Kyushu Institute of Technology, 2-4 Hibikino, Wakamatsu, Kitakyushu, Fukuoka, 808-0196, Japan
Tel.: +81 93 695 6049; fax: +81 93 695 6049.
E-mail address: yanagida@lsse.kyutech.ac.jp (Takayuki Yanagida)

1 **1 Introduction**

2 In ionizing radiation detection measurements, scintillators which convert ionizing radiations to visible photons
3 have been widely used [1]. As a viewpoint of the market volume, inorganic crystalline materials possess a dominant
4 position since they are main devices in medical and security applications. On the other hand, organic crystals are also
5 one of the common scintillators and show a significant presence especially in particle detectors from mid-20th century.
6 Organic scintillators are characterized by relatively large light yield **compared with most of inorganic scintillators** and
7 fast decay times with suitable emission wavelength to the most conventional photodetectors, PMTs. The most
8 conventional organic scintillator is anthracene and this material is treated as a standard scintillator in a standard text
9 book of ionizing radiation measurement [2]. Due to a recent technological progress, some new solid state organic
10 scintillators are introduced. **For example, new chemical composition like fluorocarbon materials [3] and new structural**
11 **challenge like microsphere materials [4] have been introduced.** In addition, ³He gas shortage [5] has motivated many
12 research groups to develop organic scintillators for neutron detectors [6-7]. For the next stage of development of
13 organic scintillations, comparison of some famous organic scintillators in neutron detection capability is important.

14 In this work, we compared neutron-gamma discrimination capability of famous organic scintillators such as
15 anthracene (treated as a standard organic scintillator in [2]), stilbene [8], and recently introduced p-terphenyl [9-10]
16 based on the pulse shape discrimination (PSD) technique. Figure 1 demonstrates photographs of three organic
17 scintillators. Anthracene (Tamaroya, Japan) was packaged since it had a sublimability. Other two scintillators, stilbene
18 and p-terphenyl were not packaged and products of Cryos-Beta Ltd., in the Ukraine. Samples had sizes of 30 mm ϕ $\times$
19 10 mm (anthracene) and 10 mm ϕ $\times$ 10 mm (other two crystals).

20 **2 Experimental procedures**

21 Basic scintillation properties of X-ray induced radioluminescence and scintillation decay time profiles were
22 evaluated. In this evaluation, X-ray generator was used as an excitation source. Supplied bias voltage and tube current
23 were 80 kV and 1 mA, respectively. Scintillation photons were detected monochromator (SR163, Andor) equipped
24 CCD (DU420A-BU2, Andor) camera. The detailed description can be seen in past work [11]. Scintillation decay time
25 profiles were evaluated by using pulse X-ray afterglow characterization system in the scintillation mode [12]. **The**
26 **timing resolution of the system in this mode was around 1 ns limited by the response of PMT (Hamamatsu R7400P)**

and emission wavelength was not resolved to simulate actual applications. The pulse X-ray had bremsstrahlung spectral shape and the endpoint of the energy was 30 keV. Pulse height spectra to evaluate scintillation light yields were measured by using a common system. Sample crystals were optically coupled with PMT R7600-200 (Hamamatsu) and anode signal was fed into preamplifier (ORTEC 113), shaping amplifier (ORTEC 572), and multichannel analyzer (Amptek Pocket MCA). The excitation source was ^{137}Cs . Because no photoabsorption peak can be seen due to their low atomic number chemical compositions, the light yields were determined through comparison of Compton edge between the measured and simulated spectra. The simulated spectra were derived using Monte Carlo code EGS 5 and convoluted with a proper energy resolution.

Then, pulse shape discrimination technique was conducted to distinguish neutron and gamma-ray events. The basic concept of our system was published previously [13]. Neutron and gamma-ray sources were ^{252}Cf and ^{60}Co , respectively. The anode signal of PMT was fed into high-speed digitizer (Agilent U1071A). Digitized waveforms were digitally processed with PC in order to discriminate neutron and gamma-ray induced events. By using this system, neutron and gamma-ray induced scintillation decay time profiles were evaluated at the same time. Scintillation decay time profiles were deduced by two components exponential assumption. All experiments were done at room temperature.

3 Experimental results and discussion

Figure 2 represents X-ray induced radioluminescence spectra of three organic scintillators. Emission wavelengths of anthracene, stilbene, and p-terphenyl were 445-525, 400-500, and 350-450 nm, respectively. These emission wavelengths were consistent with previous literatures [2, 9] and emission wavelengths were same with those under UV excitation (photoluminescence). Figure 3 shows X-ray induced scintillation decay time profiles of three organic scintillators. As shown in this figure, p-terphenyl exhibited the fastest response. Scintillation decay times of anthracene, stilbene, and p-terphenyl resulted 27.6+224.1, 3.4+19.4, and 1.9+14.1 ns, respectively. Typical errors in these evaluations were 1 ns which was the timing resolution of the system. Though scintillation decay times depended on excitation sources, observed results of anthracene and stilbene were consistent with previous reports [2]. For example, primary decay times of anthracene and stilbene in the textbook [2] were 30 and 4.5 ns, respectively. Figure 4 shows pulse height spectra of three organic scintillators under ^{137}Cs irradiation superposed with calculations by EGS5. The

shaping time of the amplifier was 0.5 μ s in all cases and sufficient longer than the scintillation decay times. Compton edge was clearly observed in all organic scintillators and simulation data well reproduced experimental results around Compton edge. Although no photoabsorption peaks can be shown in these spectra due to their low atomic number chemical compositions, positions of the photopeaks can be determined by fitting the EGS5 simulation data to the experiments. Based on the pulse heights of the photoabsorption peaks, scintillation light yields of anthracene, stilbene, and p-terphenyl irradiation resulted 20100, 16000, and 19400 ph/MeV, respectively, by taking into account the quantum efficiency of R7600U-200 at each emission peak wavelength based on figure 2. It should be noted that the wavelength sensitivity of our PMT was high at ultraviolet wavelength and pulse height of anthracene looked inferior to others.

Figure 5 (top) demonstrates neutron and gamma-ray induced events as the two-dimensional histogram between the pulse height with fast integrated time window and the ratio of the pulse height under fast and slow integrated time windows. Definition of the fast and slow integrated time windows were tuned to each material and slightly differed. Fast/slow integrated time windows of anthracene, stilbene, and p-terphenyl were 20-40/40-150 ns, 20-40/40-150 ns, and 20-30/30-150 ns, respectively. In this plot, neutron and gamma-ray events were clearly separated. Although we tested the conventional two-dimensional plot on fast and slow integrated time windows plane [14-15], separation looked better in our plotting way. In figure 5 (bottom), scintillation decay time profiles under ^{252}Cf neutron and ^{60}Co gamma-ray selected by two-dimensional plots are exhibited. Fast scintillation component did not show difference at faster time range and a difference was observed in a time range around several hundreds ns. When irradiated by neutrons and normalized at fast emission peak-top, intensity of the slow component increased. As a result, decay times under neutron irradiation of anthracene, stilbene, and p-terphenyl were 32+370, 5.3+50, and 4+31 ns, respectively. Typical uncertainty of this evaluation was around 1 ns. Compared with 30 keV pulse X-ray excitation, scintillation decay times under neutron irradiation became slower in all materials.

Figure 6 illustrates pulse height spectra of neutron and gamma-ray events selected from two-dimensional plot. Unlike Li or B contained materials, organic scintillators do not show a particular peak and all scintillators showed typical spectra under neutron and gamma-ray events. Finally, we compared the neutron-gamma discrimination capabilities of three organic scintillators. Figure 7 shows the PSD index spectra under ^{252}Cf irradiation. The PSD index,

that is the horizontal axis value in Fig. 7, is defined as the ratio of slow/fast components. By this plot, we could observe two peaks and they corresponded to neutron and gamma-ray events. To compare quantitatively, we defined the figure of merit $FOM = S / (FWHM_{\text{neutron}} + FWHM_{\text{gamma}})$, where S meant the distance between neutron and gamma-ray peaks, and $FWHM_{\text{neutron}}$ and $FWHM_{\text{gamma}}$ were full width of half maximum of neutron and gamma-ray peaks in Fig. 7. As a result, FOM of anthracene, stilbene, and p-terphenyl were 1.53, 1.10, 1.26, respectively. Similar plots but with different definition of FOM were examined by many groups [9, 15-18] and to our knowledge all evaluations essentially equal. One of the recent reports, Iwanowska et al., [9] reported PSD capabilities of stilbene and p-terphenyl based organic scintillators and they concluded that p-terphenyl based one was better. This results was consistent with our results. In our study, anthracene showed the best FOM among three organic scintillators in spite of larger physical size which generally degraded scintillation properties.

4 Conclusion

Conventional organic scintillators such as anthracene and stilbene, and relatively new material, p-terphenyl were investigated on their basic scintillation characteristics and neutron-gamma discrimination capability on PSD technique. All organic scintillators exhibited consistent scintillation responses to standard test or previous literatures. When PSD was performed in ^{252}Cf and ^{60}Co mixed field, the most conventional one, anthracene demonstrated the best neutron-gamma discrimination capability among three organic scintillators.

Acknowledgements

This work was mainly supported by a Grant in Aid for Scientific Research (A)-26249147 from the Ministry of Education, Culture, Sports, Science and Technology of the Japanese government (MEXT) and partially by JST A-step. Partial assistance from Nippon Sheet Glass Foundation for Materials Science and Engineering, Tokuyama Science foundation, Iketani Science and Technology Foundation, Hitachi Metals Materials Science Foundation, Mazda Foundation, JFE 21st century Foundation, and The Asahi Glass Foundation, the Cooperative Research Project of Research Institute of Electronics, Shizuoka University and Collaborative Research Program of Institute for Chemical Research, Kyoto University (2014-31) are also gratefully acknowledged.

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31

References

[1] T. Yanagida, Opt. Mater., 35 1987 (2013).
[2] G. F. Knoll, Radiation Detection and Measurement, 3rd eds., Wiley (2001).
[3] M. Hamel, P. Sibczynski, P. Blanc, J. Iwanowska, F. Carrel, A. Syntfeld-Kazuch, S. Normand, Nucl. Instrum. Methods A, 768 26 (2014).
[4] L.M. Santiago, H.Bagán, A.Tarancón n, J.F.Garcia, Applied Radiation and Isotopes 93 18–28 (2014).
[5] R. T. Kouzes, “The ³He Supply Problem,” Technical Rpt. PNNL-18388, Pacific Northwest National Laboratory, (2009).
[6] N. P. Zaitseva, J. Newby, S. Hamel, L. Carman, M. Faust, V. Lordi, N. J. Cherepy, W. Stoeffl, S. A. Payne, Proc. SPIE 7449, Hard X-Ray, Gamma-Ray, and Neutron Detector Physics XI, 744911
[7] M. A. Kovash, B. Daub, J. French, V. Henzl, K. Shoniyozov, J. L. Matthews, Z. Miller, H. Yang, Advancements in Nuclear Instrumentation Measurement Methods and their Applications (ANIMMA), (2011).
[8] P. Harihar, W.J. Stapor, A.B. Campbell, A.R. Knudson, Nucl. Instrum. Methods A, 302 469 (1991).
[9] J. Iwanowska, L. Swiderski, M. Moszynski, T. Szczesniak, P. Sibczynski, N.Z. Galunov, N.L. Karavaeva, J. Instrumentation, 6 P07007 (2011).
[10] M. Angelone, G. Battistoni, F. Bellini, V. Bocci, F. Collamati, E. De Lucia, R. Faccini, F. Ferroni, S. Fiore, M. Marafini, D. Materazzo, I. Mattei, S. Morganti, V. Patera, L. Piersanti, M. Pillon, L. Recchia, A. Russomando, A. Sarti, A. Sciubba, E. Solfaroli Camillocci, C. Voena, IEEE Trans. Nucl. Sci., 61 1483 (2014).
[11] T. Yanagida, K. Kamada, Y. Fujimoto, H. Yagi, T. Yanagitani, Opt. Mat.,35 2480-2485 (2013).
[12] T. Yanagida, Y. Fujimoto, T. Ito, K. Uchiyama, K. Mori, Appl. Phys. Exp., 7 062401 (2014).
[13] A. Yamazaki, K. Watanabe, A. Uritani, T. Iguchi, N. Kawaguchi, T. Yanagida, Y. Fujimoto, Y. Yokota, K. Kamada, K. Fukuda, T. Suyama, A. Yoshikawa, Nucl. Instr. and Meth. A, 652 435-438 (2011).
[14] N.P.Hawkes, G.C.Taylor, Nucl. Instrum. Methods A, 729 522 (2013).
[15] K.A.A. Gamage, M.J.Joyce, N.P.Hawkes, Nucl. Instrum. Methods A, 642 78 (2011).
[16] I. A. Pawelczak, A. M. Glenn, H. P. Martinez, M. L. Carman, N. P. Zaitseva, S. A. Payne, Nucl. Instrum. Methods A, 751 62 (2014).
[17] S. A. Pozzi, M. M. Bourne, S. D. Clarke, Nucl. Instrum. Methods A, 723 19 (2013).
[18] P. N. Zhmurin, V. N. Lebedev, V. D. Titskaya, A. F. Adadurov, D. A. Elyseev, V. N. Pereymak, Nucl. Instrum. Methods A, 761 92 (2014).

Figure caption

Figure 1. Photographs of anthracene (left), stilbene (center), and p-terphenyl (right). The unit of the number in ruler was cm. Anthracene was encased and other two were wrapped by Teflon tape reflector.

Figure 2. X-ray induced radioluminescence spectra of three organic scintillators. Emission intensities were validly scaled to compare emission wavelengths.

Figure 3. X-ray induced scintillation decay time profiles of three organic scintillators.

Figure 4. ^{137}Cs and irradiated pulse height spectra (circle) of anthracene (left), stilbene (center), and p-terphenyl (right) superposed with EGS5 calculations (dotted lines).

Figure 5. (Top) Two-dimensional histogram of anthracene (left), stilbene (center), and p-terphenyl (right) on a plane of the pulse height with fast integrated time window and the ratio of the pulse height under fast and slow integrated time windows. Fast/slow integrated time windows of anthracene, stilbene, and p-terphenyl were 20-40/40-150 ns, 20-40/40-150 ns, and 20-30/30-150 ns, respectively. (Bottom) Scintillation decay time profiles (b) under neutron and gamma-ray excitation of anthracene (left), stilbene (center), and p-terphenyl (right) by event selection.

Figure 6. Pulse height spectra of anthracene (left), stilbene (center), and p-terphenyl (right) in ^{252}Cf and ^{60}Co mixed field. Neutron and gamma-ray induced pulse height spectra were reconstructed by adequate event selection.

Figure 7. Histograms of neutron and gamma-ray events of anthracene (left), stilbene (center), and p-terphenyl (right), plotted on the ratio of slow/fast components. Slow/fast definition was same with figure 5.

Figure1

Antracene

Stilbene

p-terphenyl

Figure2

Figure3

Figure4

Figure5

Figure6

Anthracene

Stilbene

P-terphenyl

Figure7