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Melt-cast organic glasses as high-efficiency fast neutron scintillators



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ABSTRACT

In this work we report a new class of organic-based scintillators that combines several of the desirable attributes of existing crystalline, liquid, and plastic organic scintillators. The prepared materials may be isolated in single crystalline form or melt-cast to produce highly transparent glasses that have been shown to provide high light yields of up to 16,000 photons/MeVee, as evaluated against EJ-200 plastic scintillators and solution-grown *trans*-stilbene crystals. The prepared organic glasses exhibit neutron/gamma pulse-shape discrimination (PSD) and are compatible with wavelength shifters to reduce optical self-absorption effects that are intrinsic to pure materials such as crystalline organics. The combination of high scintillation efficiency, PSD capabilities, and facile scale-up via melt-casting distinguishes this new class of amorphous materials from existing alternatives.

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1. Introduction

An ongoing objective in the field of organic radiation detection materials is to achieve a balance between scintillation performance characteristics and cost. Organic molecular crystals possess superior light-yields and neutron/gamma pulse-shape discrimination (PSD) but possess anisotropic properties and are expensive to produce in large sizes. In contrast, liquid and plastic scintillators are readily scalable to large sizes, although both are characterized by reduced light yields and particle discrimination properties. For example, the brightest (non-PSD) plastic scintillators possess light yields of $\sim 10,400$ photons/MeVee, which is approximately 80% that of solution-grown *trans*-stilbene crystals [1]. The light yield of a commercially-available PSD-capable plastic scintillator (Eljen EJ-299-33A) is ~ 8600 photons/MeVee, while a peak light yield of 13,000 photons/MeVee has been reported for research-grade plastic scintillators [2–4]. For the above reasons, our efforts are focused on developing high-performance solid-state organic scintillators that provide stable performance and have reduced preparation costs.

We previously reported a series of high-symmetry organic crystals that are based upon molecules that possess local C_3 -symmetry [5]. In that work, it was shown that increased molecular and crystallographic symmetry led to significantly higher light yields and PSD relative to their respective low-symmetry parent chromophores. High-symmetry space groups are also

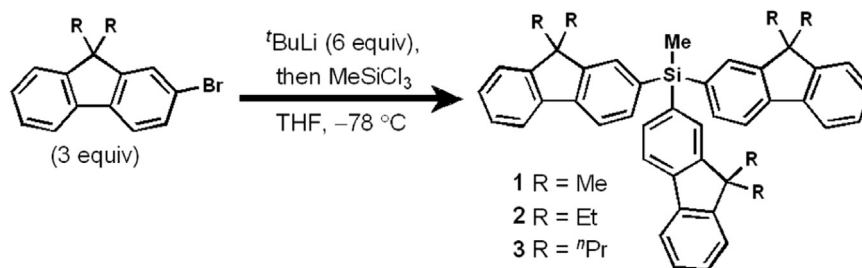
associated with improved crystal growth properties and mechanical robustness due to the presence of a greater number of independent slip systems to dissipate stresses induced by thermal or mechanical stimuli [6]. A similar strategy was employed in this work for compounds based upon three fluorene chromophores arranged around a silicon central atom, as shown in Scheme 1. Initial photophysical characterization of these materials showed that they behave as efficient scintillators in their respective crystalline forms, although an unexpected finding was that they also form stable and highly transparent organic glasses following cooling of the melt (Fig. 1). Photoluminescence, thermoanalytical analysis, and scintillation measurements were performed to assess these materials and benchmark their performance against well-known organic scintillators. The combined results indicate that stable organic glasses present a promising new route towards high light-yield and low-cost PSD scintillators.

2. Experimental

Compounds 1–3 were synthesized via lithiation of the corresponding 9-alkyl-2-bromofluorene using standard organic synthesis techniques. Purification was achieved via flash column chromatography and recrystallization from toluene. Glasses of 1–3 were prepared by melting the powders in a Teflon-lined mold (20 mm \times 20 mm \times 20 mm) or by casting the liquid samples onto a glass slide and allowing to cool (\emptyset 10 mm \times 5 mm). The different samples were prepared to confirm that the light collection efficiency and corresponding light yields remained constant for these small sample sizes in the experimental configuration described

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Scheme 1. General procedure for the synthesis of compounds 1–3.

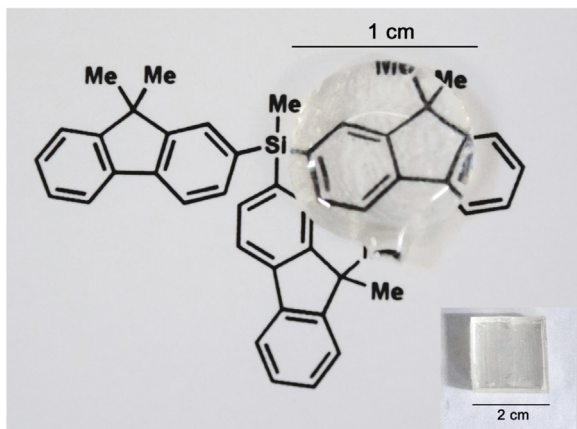


Fig. 1. Photograph of organic glasses of compound 1. The sample shown in the main figure was drop-cast from the liquid, whereas the cube shown in the inset was melted and cooled in a silicone mold.

below. No additional surface treatment was performed on the obtained glasses. Reference samples of EJ-200 plastic scintillator were purchased from Eljen Technologies in dimensions of (10 mm × 10 mm × 10 mm) and (Ø25.4 mm × 25.4 mm). All EJ-200 surfaces were polished to optical quality using a diamond tipped lathe. The obtained pulse-height spectra for different sizes of glasses of 1–3 and EJ-200 revealed light yields that did not vary with the size and geometry of each sample studied. The *trans*-stilbene reference sample of dimensions (20 mm × 20 mm × 20 mm) was prepared according to the technique described by Carman et al., comprising synthetic preparation from styrene, followed by crystal growth from anhydrous anisole [7]. The crystal faces were polished; the face perpendicular to the *c*-crystallographic axis was optically coupled to the PMT. The purity and light-yield of the prepared sample is consistent with the published results, which provided a light-yield more than twice that of a melt-grown *trans*-stilbene crystal obtained from Scinti-Tech, Inc [7].

Steady-state spectra, quantum-yields, and time-resolved photoluminescence measurements were collected using a Horiba Jobin-Yvon Fluorolog FL3-21 fluorescence spectrometer equipped with an integrating sphere attachment. Data were collected on powder samples at 90° incidence for steady-state spectra and time-resolved measurements, and in the front-facing geometry for quantum-yield data. Differential scanning calorimetry data were collected using a Mettler Toledo DSC 822 instrument and processed using the STARe SW 12.10 software package. A heating rate of 10 °C was used during data acquisition. Powder X-ray diffraction data were obtained using a PANalytical Empyrean instrument equipped with a 40 kV Cu K- α source and an XY-rotating sample stage. 33 keV X-ray spectra were obtained using the Barium $K_{\alpha 1}$ and $K_{\alpha 2}$ X-rays emitted from a ^{137}Cs source. The relative

Table 1

Summary of photoluminescence quantum yields and gamma-ray scintillation light-yield data.

Compound	PL quantum yield	662 keV relative L.Y.	33 keV relative L.Y.
<i>Trans</i> -stilbene – Crystal	0.62	1.00	1.00
EJ-200 – Plastic	0.55 (PVT=0.12)	0.78	0.89
1 – Crystal	0.46	^a	1.26
2 – Crystal	0.53	^a	1.21
3 – Crystal	0.78	^a	1.44
1 – Glass	0.39	1.02	0.97
2 – Glass	0.57	0.74	0.83
3 – Glass	0.48	0.59	0.68
(1+0.02% DPA) – Glass	0.89	1.03	0.93
(2+0.02% DPA) – Glass	0.66	1.11	1.11
(3+0.05% DPA) – Glass	0.67	1.23	1.18

^a Not observed due to small crystal size.

pulse heights in Table 1 were evaluated from the position of the 33 keV photopeak. No background corrections were required due to the high relative intensity ratios for these X-ray emissions and the high photoelectric cross sections of organic scintillators in this energy range. 662 keV gamma-ray scintillation pulse-height spectra were similarly obtained for bulk samples using a ^{137}Cs source. All samples were evaluated using an Electron Tubes 9124-QB bi-alkali photomultiplier tube biased at 1000 V. All samples were wrapped on five sides by reflective PTFE tape, coupled to the PMT face, and covered by an aluminum light shield. The ^{137}Cs and AmBe sources were placed directly on the aluminum shield, resulting in a source-to-scintillator distance of ~1 mm. Pulses were digitized using a 600 MHz, 12-bit LeCroy HRO 66Zi oscilloscope and histogrammed using a 500 ns integration time for scintillation pulse-height measurements. Neutron/gamma pulse-shape discrimination data were obtained using a similar experimental configuration but processed off-line using the charge-comparison method [8]. The charge-comparison PSD parameters were optimized for each sample, comprising the prompt/delayed cutoff time and total integration time.

3. Results and discussion

3.1. Photoluminescence spectroscopy

Photoluminescence measurements were performed on glasses of compounds 1–3. Steady-state excitation and emission spectra for 1 are provided in Fig. 2, illustrating properties that are dependent upon the preparation method. Data in the top plot was obtained on a glass prepared via sublimation, whereas the data in the bottom figure was obtained for a melt-cast glass. The emission spectra reveal a 38 nm blue-shift in the peak emission wavelength for sublimed versus melt-cast samples of 1. The presence of fine structure in both spectra suggests that the shift is not due excimer

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