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# Comparative analysis of new crystal and plastic scintillators for fast and thermal neutron detection

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**Abstract.** This paper considers scintillation and physical properties of efficient organic scintillators tested as single crystals and as components of plastics with pulse shape discrimination (PSD). For the first time, single crystals of 9,9-dimethyl-2-phenyl-9H-fluorene (PHF) were grown for studies of the basic scintillation properties of this new compound. Comparison to classical organic crystals, like anthracene, *trans*-stilbene, and *p*-terphenyl, and to more recently introduced organic glass showed that the new crystal belongs to a group of the most efficient scintillators, with light output exceeding that of *trans*-stilbene and PSD comparable to that of *p*-terphenyl. Additional studies were conducted to evaluate scintillation performance and physical properties, like hardness and dye leaching, of plastic scintillators prepared with PHF, organic glass, and liquid diisopropylnaphthalene (DIPN) that were considered as examples for potential replacement of PPO (2,5-diphenyloxazole) in current commercial PSD plastic scintillators. Comparison of the highest performing plastic scintillators prepared with these dyes shows that PHF formulations produced a record LO increase of 69% relative to EJ-200. Similar improvements obtained with <sup>6</sup>Li-loaded formulations showed that future development of PSD plastics should not be limited to use of PPO but must involve the search and exploration of new efficient dyes that may lead to discovery of much brighter organic scintillators with improved physical properties required for fast and thermal neutron detection, fast neutron spectroscopy, and antineutrino detection applications.

**Keywords:** Organic crystals; 9,9-dimethyl-2-phenyl-9H-fluorene; Plastic scintillators; <sup>6</sup>Li-loaded scintillator; Pulse-shape discrimination, Neutron detection.

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## Introduction

Organic scintillators belong to an important class of materials widely used in physics research and radiation detection. The fast response time to different types of nuclear radiation, ability for the discrimination between different types of nuclear particles, and possibility of production in large volumes and different configurations at a reasonably low cost make them suitable for many applications, such as special nuclear materials detection, analysis of the mixed radiation fields, coincidence counting, neutron spectroscopy, radiation portal monitors, large-area neutron imaging detectors, etc. [1, 2]. The unique performance of organic scintillators is based on the properties of aromatic compounds that produce efficient fluorescence induced by different types of nuclear radiation, high content of hydrogen for specific  $n,p$  interaction with high-energy (fast) neutrons, a possibility of loading with capture nuclei (e.g.  $^{10}\text{B}$  or  $^6\text{Li}$ ) for low-energy (thermal) neutron detection, and two-component fluorescence decay that gives origins to the pulse shape discrimination (PSD) techniques [3, 4]. The scintillation properties of many aromatic molecules were extensively studied in the last half of the previous century [5], and the most efficient molecules found in those studies have been used for preparation of liquid, single crystal, and plastic scintillators. From the early times of first discoveries, the list of single crystal organic scintillators, characterized by the highest efficiency and excellent scintillation properties, practically hasn't changed. Among crystals, it still includes traditional anthracene that represents the highest scintillation standard for all organic scintillators, *trans*-stilbene long known for its superior PSD [3], and *para*-terphenyl that, according to some publications, may possess light output (LO) and PSD close or even superior to those of anthracene or *trans*-stilbene [6, 7]. In more recent years, new developments have been made mainly with non-crystalline organic scintillators advanced by the addition of high-flash-point liquid scintillators [8], plastic

scintillators with PSD capability [9], heavy-atom loaded plastics for gamma spectroscopy [10], and organic glass (OG) [11]. Among all these materials, plastics represent the most practical and available class of organic scintillators. Compared to single crystals, which require a practiced hand in crystal growth and often a laborious growth process, plastics can be easily produced in large volumes and configurations. However, the problem of the current plastic scintillators is a substantially lower LO, rarely reaching ~50% LO of the best crystals. Eljen Technology [8] lists the LO of their current commercial PSD plastic, EJ-276D, as 14% lower compared to traditional non-PSD plastic (EJ-200) that is typically used as a plastic scintillation standard. It should be noted that, from the first introduction, commercial PSD plastic formulations have been based only on PPO (2,5-diphenyloxazole) that was found to produce the most efficient neutron/gamma discrimination compared to other tested primary dyes. The physical and scintillation properties (e.g., insufficient hardness, dye leaching, performance instability, and initially moderate LO) related to the nature and high concentration of this dye have been eventually tuned using small variations in different additives, like crosslinkers, antioxidants, or secondary dyes [12, 13]. However, recent studies showed that replacement of PPO by less known, non-traditional primary dyes can affect not only PSD but also influence the LO and physical properties of plastics. One example was demonstrated by the replacement of PPO in polystyrene mixtures by an OG component reported to exhibit up to 54% higher LO compared to EJ-200 [14]. More modest but still convincing evidence was presented in studies of new highly soluble fluorophores, some of which, when dissolved in polystyrene, produced higher LO than PPO-based analogs [15]. In these cases, replacements of PPO belonged to a group of highly efficient fluorene derivatives intensively studied in recent years for organic light-emitting diode (OLED) applications [16]. However, while reporting the possibility of achieving high LO [17-20], studies of these

compounds did not present sufficient advantages that could lead to practical applications and potential replacement of PPO.

In this work, we undertook a detailed analysis of scintillation and physical properties of one of the reported fluorene derivatives, 9,9-dimethyl-2-phenyl-9H-fluorene (PHF) that showed the best performance among the molecules synthesized and first tested by the group of Prof. A. Sellinger from Colorado School of Mines [15]. The choice of PHF for the present work was determined by commercial availability and simple structure that enables crystal growth used to demonstrate basic properties comparable or even exceeding the properties of the best-known single crystal and OG scintillators. Another attractive feature of PHF is high solubility that allows for preparation of regular PSD and  $^6\text{Li}$ -loaded plastics and makes possible comparison to PPO and other alternative dyes. More dyes that were compared in this work include OG and DIPN (diisopropylnaphthalene), which were used to demonstrate potential advantages of further exploration and studies of new scintillation dyes that may lead to production of the new generations of crystal and plastics organic materials with highly advanced scintillation and physical properties.

## **Experimental**

### *2.1. Materials*

PHF powder for initial crystal growth and plastic preparation experiments was purchased from Aablocks (95% purity) and purified by double crystallization from ethanol. An additional supply of purified material for larger crystal growth and multiple plastic preparation was provided by Eljen Technology. Monomers of styrene, vinyltoluene, and DVB (divinylbenzene used as a crosslinker) were purchased from Aldrich. PPO (> 99% purity) and liquid DIPN (obtained from Eljen) were used as received, without additional purification. All secondary dyes, Exalite E404,

bis-MSB (1,4-bis(2-methylstyryl)benzene), and Coumarin C460 (7-diethylamino-4-methylcoumarin) were purchased from Exciton/Luxottica. Stilbene crystals used for comparison were grown at LLNL by solution growth technique [21, 22], while *p*-terphenyl crystals were provided for measurements by Texas A&M University. Two versions of organic glass (OG) were purchased from BlueShift Optics [23]. A regular OG composition containing bis-MSB as a wavelength shifter was used for comparison to crystals, while a pure version of OG with no addition of the wavelength shifter was used as a primary dye for the preparation of plastics with controlled concentration of secondary dyes. Both versions were uncoated and measured before surface crystallization, typical for OG compositions, was observed.

## *2.2. Crystal growth*

PHF single crystals studied in this research were grown using solution growth technique. Anisole was used as an organic solvent in which PHF was found to have high solubility changing from 50% to 65% in the temperature range of 25 – 40°C. Initial seeds for crystal growth were obtained by the slow evaporation from PHF solutions in anisole. The growth of a larger bulk crystal was conducted in a 1 L volume crystallizer using the temperature reduction technique in a range from 35°C to RT (~ 25°C). Details of both growth techniques are described in a previous publication for an example of another organic crystal of 1,3,5-triphenylbenzene [24].

## *2.3. Plastic preparations and hardness measurements*

To prepare plastic scintillators, monomers of styrene, vinyltoluene, and DVB were de-inhibited by eluting through a column of basic alumina on top of silica. Dissolved oxygen was removed by sparging with dry nitrogen. The precursor solutions prepared as mixtures of styrene, DVB, scintillation dyes, and initiator (Luperox 231) were made in 20 mL glass vials in a nitrogen-filled glovebox. It should be noted that all concentrations listed below are expressed in weight %

relative to the total weight of a plastic sample. 5% DVB addition was used for all plastic compositions unless shown differently for OG-based formulations that, according to the original report [14] do not require crosslinking for the enhancement of hardness. One 30% PHF plastic was made without DVB for comparison of hardness. The vials were sealed and transferred to a nitrogen-purged oven for polymerization. The starting temperature of 60°C was then increased by 5°C/day over 3 days and continued at 75°C for an additional 7 days. After polymerization, the resulting solidified plastic cylinders were retrieved by breaking the glass containers and polished to standard size cylinders of Ø25 mm x ~18 mm (thickness) used (unless shown differently) in most tests. For accurate comparison, commercially purchased Ø1" x 1" (Ø25 mm x 25 mm) EJ-200 and EJ-276D were cut and polished to the same Ø25 mm x 18 mm size, to match the thickness of all LLNL-prepared samples.

#### *2.4.Characterization techniques*

Absorption and photoluminescence (PL) spectra were measured using an Agilent Cary 3500 UV-Vis spectrophotometer and a Horiba Jobin Yvon Fluoromax-4 spectrometers, respectively.

Measurements of hardness were conducted using a Shore D Scale durometer with three measurements made per sample; the variation between individual measurements did not exceed 10%. The decay time of crystals was measured with the modified time correlated single photon counting (TCSPC) technique [25]. In this technique, TCSPC analysis was performed using beta excitation from a Sr-90 radioactive source. The system start time was provided by a beta passing through a thin plastic scintillator coupled to a MICROFJ-SMA-60035-GEVB SiPM evaluation board. The beta then passed into an optically separate chamber containing the small scintillator sample. Single photons from the interrogated sample were sensed with a Hamamatsu H5784 PMT with low optical coupling to the sample. A Picoscope 6404B was used to digitize the pulses



from the detectors, and offline software was used to produce the single photon time spectra for characterization of the scintillation decay.

The scintillation performance was characterized by using a Hamamatsu R6231-100 photomultiplier tube (PMT) connected to a GAGE Compuscope 14200 digitizer, which records the interaction waveforms at 200 MS/s with 14bit voltage resolution. The scintillation LO was obtained from the position of the Compton Edge in the  $^{137}\text{Cs}$  spectra, in which 477 keV<sub>ee</sub> (electron-equivalent energy) was defined by 50% of the Compton Edge (CE) peak.

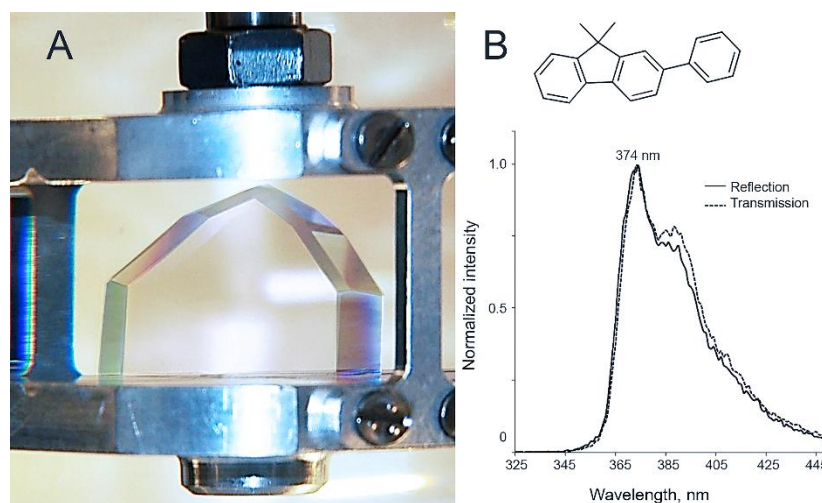
Neutron/gamma PSD was evaluated with a  $^{252}\text{Cf}$  source using tail-to-total charge integration measurement techniques described previously [12]. Integration times were optimized for individual measurements, but typically the tail charge window started  $\sim 45$  ns after the peak of the voltage pulse and ended  $\sim 750$  ns after the peak. Quantitative evaluation of PSD was made by comparison of Figures of Merit (FoM) calculated using a standard formula of  $\text{FoM} = S/(\delta_{\text{gamma}} + \delta_{\text{neutron}})$ , where  $S$  is the separation between gamma and neutron peaks, and  $\delta_{\text{gamma}}$  and  $\delta_{\text{neutron}}$  are full widths at half maximum (FWHM) of the corresponding peaks. The FWHM and mean values for the gamma and neutron distributions were extracted from Gaussian fits. For regular PSD samples FoM was calculated over the energy range 450 – 510 keV<sub>ee</sub>. With  $^6\text{Li}$ -containing samples, PSD was measured using the same  $^{252}\text{Cf}$  source shielded by 1"-thick high-density polyethylene (HDPE). In this case, FoM was obtained by fitting the distributions over the energy region  $\pm 2.5\sigma$  around the centroid of the thermal neutron spot to a Gaussian function. It should be emphasized that for accurate comparison of different scintillators, all samples that had the same size and shape were equally wrapped by the 3 layers of the Teflon tape and attached to the PMT using optical grease. The uncertainty of LO measurements was evaluated as 2.8% based on greater than thirty long-term measurements of a 10 g EJ-200 sample, which showed no

systematic trend. Since FoM varies approximately as the square root of light output, the uncertainty for FoM may be less, but we conservatively apply the same 2.8% uncorrelated systematic error.

### 3. Results and discussion

#### 3.1. PHF single crystal

Unlike most inorganic crystals, where molecules are held together by strong ionic or covalent forces, organic crystals are solids formed by close packing of individual molecules connected by weak intermolecular (Van der Waals) interactions [26]. This major property determines difficulties, especially evident at crystallization of aromatic compounds that, due to their complex and branched structure may not form large and perfectly arranged packing systems. Fortunately, PHF represents a rare example of a pure aromatic crystal that can be relatively easily grown by a classical solution growth technique, like that applied previously to growth of 1,3,5-triphenylbenzene or stilbene crystals [21, 22, 24, 27].



*Fig. 1. A – Photo of a PHF faceted crystal taken during growth from anisole solution. The horizontal size of the crystal is 5 cm; B – PHF molecular formula and photoluminescence spectra measured with a 7 mm-thick crystal. Coinciding spectra taken in reflection and transmission modes indicate very low self-absorption of the crystal.*

The possibility of obtaining single crystals like that shown in Fig. 1A, provides a convenient and reliable way for the initial evaluation of physical, optical, and scintillation properties of the new material. X-ray diffraction analysis and crystal structure refinement conducted with the first, high optical quality crystals grown from anisole solutions showed that PHF forms crystals that belong to the *Pbca* space group of the orthorhombic system that probably determines the relative simplicity of growth compared to the majority of other aromatic compounds crystallizing in lower monoclinic or trigonal symmetry. The corresponding photoluminescence (PL) spectra presented in Fig. 1B show peak energy at 374 nm that is well matching the wavelength sensitivity of the common blue-region PMTs. This means that, like anthracene or stilbene, the PHF crystal can be characterized as a pure one-component system, without need for wavelength shifters required for the originally shorter wavelength scintillators, like *p*-terphenyl, which original PL peak of ~ 340 nm is shifted to ~ 392 nm by addition of 1,4-diphenyl-1,3-butadiene [28], or OG that uses bis-MSB for shifting the initial emission peak from ~ 328 nm to ~ 425 nm [29]. Another feature shown by the spectra is a uniquely small difference between the reflection and transmission spectra that must be explained by the very low self-absorption that once more indicates no need for any wavelength shifting additions.

The first evaluation of scintillation properties of the new crystal was made by comparison to stilbene. It should be noted here that, due to the limited number of PHF crystals grown in this research, it was difficult to find precisely same shape and size crystals needed for the accurate comparison. However, large availability of stilbene crystals grown in our lab and ease of their cutting and polishing allowed for obtaining any size and shape samples that were used as reference standards for evaluation of scintillation performance of other scintillators relative to

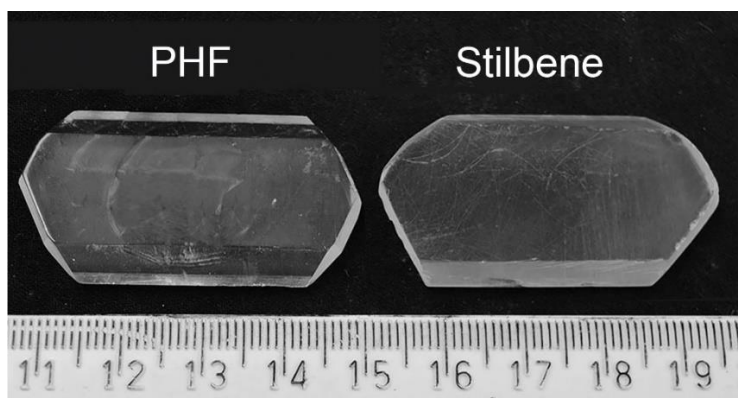


Fig. 2. Photo of PHF and stilbene single crystals used for the initial evaluation of PHF scintillation properties. The naturally faceted PHF crystal is shown as grown. The corresponding stilbene sample was cut out and polished from a larger single crystal to match the size and shape of PHF.

stilbene. Examples of a pair of such crystals are shown on Fig. 2. Results of the measurements obtained with these crystals (Fig. 3) showed that the major scintillation characteristics of PHF are close to those of stilbene; PHF crystal exhibits practically the same LO, with slightly lower but still high PSD typical only to the most efficient known organic scintillators.

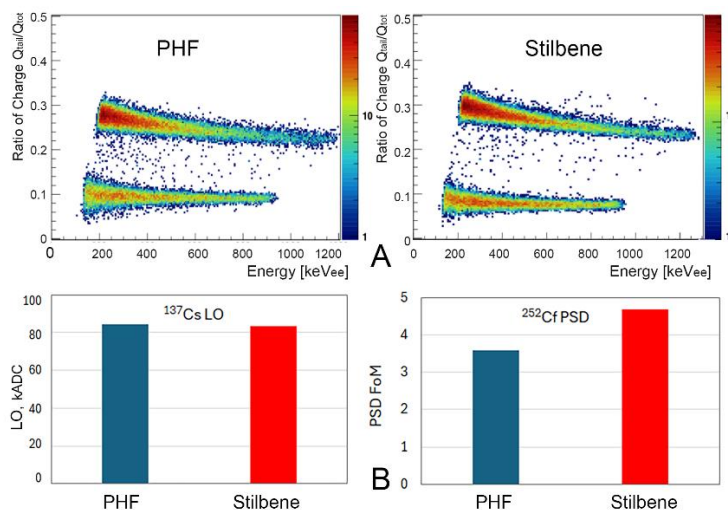


Fig. 3. A – Experimental  $^{252}\text{Cf}$  PSD distributions measured with the crystals shown in Figure 2; B – Corresponding parameters showing that PHF has high LO and efficient PSD comparable to those of the stilbene crystal.

Another similarity between PHF and stilbene was obtained from measurements of decay times that showed close values of 3.27 ns and 3.46 ns for PHF and stilbene, respectively (Fig. 4).

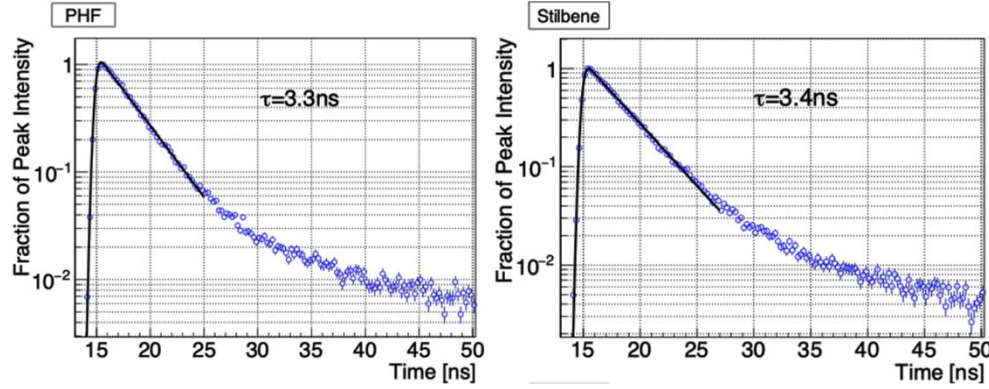


Fig. 4. Decay characteristics of PHF and stilbene measured with the same size crystals of 1.5 mm thickness. The mean values of decay obtained from the exponential fit of experimental data.

Direct comparison of PHF to other highly efficient organic scintillators, like anthracene, *p*-terphenyl, or organic glass, is more problematic because of the practical impossibility of getting any of these materials at equal shapes and dimensions. Therefore, the evaluation of the new

Sample	$^{137}\text{Cs}$ CE [kADC]	PSD FoM
<b>Ø40 mm x 11 mm cylinders</b>		
Stilbene	78.8	4.71
Anthracene	90.8	2.12
<b>20 mm x 20 mm x 20 mm cubes</b>		
Stilbene	76.4	4.81
<i>p</i> -terphenyl	73.2	1.14
<i>p</i> -terphenyl	87.6	3.87
<b>12 mm x 12 mm x 50 mm</b>		
Stilbene	77.2	4.4
Stilbene	78.4	4.58
Organic glass*)	80.4	2.94
Organic glass*)	78	2.76

Sample	$^{137}\text{Cs}$ CE [kADC]	PSD FoM
<b>47 mm x 20 mm x 5 mm</b>		
Stilbene	79.6	4.66
PHF	82.4	3.68
<b>40 mm x 20 mm x 5 mm</b>		
Stilbene	82.9	4.69
PHF	84.4	3.58
<b>10 mm x 10 mm x 10 mm cube</b>		
Stilbene	76.4	4.67
<i>p</i> -terphenyl	80.4	3.42
Organic glass *)	78	2.83
PHF	80	3.74

\*) Uncoated samples before crystallization appeared

Table 1. LO and PSD measured with crystals and OG of different dimensions. Comparison of the same-sized samples shows that PHF crystal belongs to the brightest scintillators with PSD superior to PSD in anthracene, *p*-terphenyl, or organic glass. kADC values correspond to the position of CE for each measured sample.

crystal relative to these scintillators was done indirectly, through their comparison to the same size stilbene cut from home-grown crystals to any desired configurations. Results of such comparisons are presented in Table 1. Note that, due to the difference in sample dimensions, the

LO was expressed in ADC units corresponding to the CE position of the respective crystal. Analysis of results show that, at various sizes, PHF crystals exhibit similar or higher LO compared to the same size stilbene or OG. In respect to PSD, stilbene remains superior to all measured scintillators. However, the PSD performance of PHF is noticeably higher than that of OG, being closer to the *p*-terphenyl crystal, historically known and used as the second-best material for efficient neutron/gamma discrimination [30]. The crystal is stable, non-hygroscopic, and preserves its high performance being stored under ambient conditions. An advantage of PHF crystals can be also consistency of scintillation parameters that, like in stilbene, show less sample-to-sample variations compared to *p*-terphenyl that may exhibit large fluctuation of PSD measured in different samples (see examples of 20-mm cubes in Table 2). This discrepancy is probably determined by the fact that both stilbene and PHF crystals are single-component crystals that provide high performance without doping, while *p*-terphenyl crystals contain a dopant for obtaining higher LO and longer wavelength of emission. As shown previously [31], the molecules of such doping, that must have lower energy of excited states, can work as triplet ( $T_1$ ) traps that, at low concentrations, may substantially affect PSD of mixed or doped crystals. Results of Table 1 obtained with relatively small samples of different crystals cannot be taken as an accurate basis for practical applications that can be limited due to the difficulties in preparation, high cost, or low availability of the compared materials. However, it should not be forgotten that major studies of organic scintillators started in most cases with crystals. In this respect, the high LO and excellent PSD of the PHF crystal make it a valuable addition to a small family of most efficient organic scintillators. The well-known fact that in aromatic crystals fluorescence (and scintillation) is produced by individual molecules may explain the high LO of the brightest crystal systems composed of highly efficient aromatic molecules. However, the

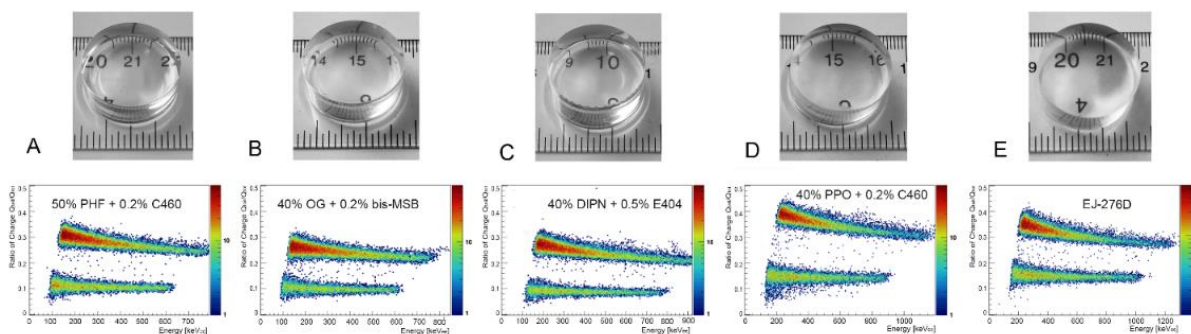
question remains unclear why pure, one-component crystals, like anthracene, stilbene, PHF, or OG that have similar high LO, produce distinctly different PSD. Based on the current knowledge, it is also hard to explain what conditions in the molecular or crystal structure of stilbene determine its superior PSD compared to all currently known organic scintillators. Our results do not offer answers to these questions. However, they point in the direction of further theoretical and experimental studies needed for better understanding the details of PSD mechanisms.

### *PSD plastic formulations alternative to PPO*

#### *s3.2.1. Scintillation properties*

Since the 2012 publication [9], PPO became the main primary dye used for preparation, research, and commercial production of PSD plastics. The possibility of obtaining PSD by dissolution of other highly soluble, traditionally known dyes, like oxadiazoles, biphenyl, or *m*-terphenyl, in aromatic polymers was demonstrated in later publications [32-34]. However, because none of those dyes belong to the class of the brightest scintillation compounds, they typically produced PSD that was lower than in plastics with PPO, with the LO rarely exceeding that of EJ-200 standard. A more recent publication [14] demonstrated that replacement of PPO by OG in plastics may lead to a substantial enhancement of the LO and improvement of physical properties. In that publication, the higher LO and better physical performance of tested compositions were attributed to the special properties of OG that presumably forms unique formulations (blends) that are different from regular PSD plastics. Large solubility and high scintillation efficiency of PHF demonstrated in our crystal growth experiments provided an opportunity to evaluate the performance of this compound in plastics that could be compared to both OG and traditional PPO formulations. To consider the effects produced by the state of the

primary dyes on physical properties, our collection of studied formulations included comparison to one more non-traditional composition prepared using not a glassy or crystalline but a liquid primary dye, DIPN, which is a well-known, low-flash point solvent in the formulation of EJ-309 liquid scintillator. A surprising result obtained with this compound showed that, despite the initial liquid state, it still can be used as a primary dye for the preparation of solid, transparent plastics with relatively efficient LO and PSD. This and other examples shown in Fig. 5 illustrate the impressive ability of aromatic polymers to withstand the high loading of various compounds of different properties or physical state. It should also be expected that at high concentrations, used for the preparation of these formulations, the nature of the primary dyes may profoundly affect the scintillation and physical properties of the final materials.



*Fig. 5. Examples and corresponding  $^{252}\text{Cf}$  PSD distributions measured with highly-loaded PSD formulations prepared by dissolution of different primary dyes in polystyrene: A – PHF; B – Organic glass; C – DIPN; D – PPO; E – EJ-276D. All plastics were prepared with 0.2 % addition of C460 used as a secondary dye, and 5% DVB, except for the OG glass made without addition of the crosslinker. Note that DIPN is a liquid at RT that as a primary dye produces solid plastic.*

More detailed quantitative data obtained with the tested compositions are presented in Table 2. The results allow for a comparison of the effects of primary dye concentration, selection of the secondary dye, and addition of a crosslinker, within and between the individual groups, as well as to the scintillation standards represented by the same-size commercial EJ-200 (LO standard),



EJ-276D (PSD standard for plastics), and a solution-grown stilbene crystal (for comparison of plastics to crystals).

Composition	<sup>137</sup> Cs LO	PSD FoM	Hardness (Shore D)	Leaching
<b>PHF</b>				
30%PHF+no sec.dye	1.2	2.34	87	No
30%PHF+0.2%C460-no DVB	1.54	3.09	85	No
25%PHF+0.2%C460	1.46	2.37	87	No
30%PHF+0.2%bis-MSB	1.45	2.6	87	No
30%PHF+0.2%C460	1.51	2.74	85	No
40%PHF+0.2%C460	1.62	3.3	84	No
50%PHF+0.2%C460	1.69	3.66	78	No
<b>OG (no DVB added)</b>				
30% OG + 0.2% bis-MSB	1.41	2.12	87	No
30% OG + 0.2% C460	1.48	2.07	87	No
40% OG + 0.2% C460	1.54	3.09	84	No
40% P2 +0.2% bis-MSB *)	1.38 *)	n/a **)	n/a	n/a
50% P2 +0.2% bis-MSB *)	1.45 *)	n/a **)	n/a	n/a
<b>DIPN</b>				
30% DIPN + 0.5% E404	1.35	2.83	54	No
30% DIPN + 0.2% C460	1.12	2.45	67	No
30% DIPN + 0.2% bis-MSB	1.02	2.14	70	No
40% DIPN + 0.5% E404	1.41	3.37	31	Slight (liquid)
<b>PPO</b>				
30%PPO+no sec.dye	0.78	2.56	81	Yes
25% PPO + 0.2% C460	1.05	2.86	83	No
30%PPO+0.2%bis-MSB	1.2	3.12	62	Slight
30%PPO+0.2%C460	1.2	3.2	77	Slight
40%PPO+0.2%C460	1.3	3.44	60	Strong
<b>EJ-276D ***)</b>	1.23	3.03	79	No
<b>EJ-200 ***)</b>	1	n/a	84	No
<b>Stilbene ***)</b>	1.87	4.41	n/a	n/a

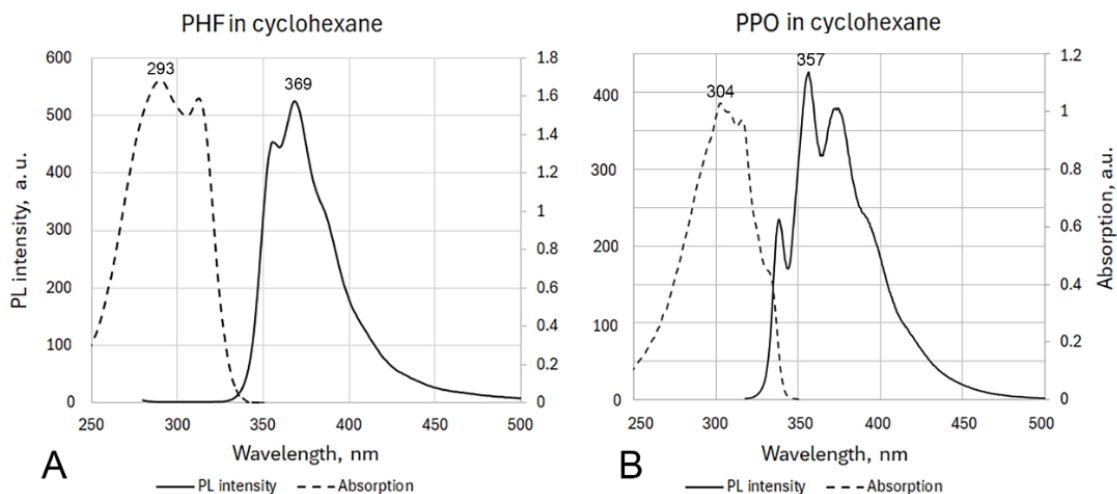
\*) Literature data [14]

\*\*) Cannot be compared because of different conditions of measurements in [14]

\*\*\*) Same size cylinders as all plastics

*Table 2. LO (relative to EJ-200), PSD, and physical parameters of plastics prepared with tested primary dyes of different concentrations. Comparison shows that new compositions may exhibit almost 70% higher LO compared to the EJ-200 standard, with PSD that can accede the levels typical for current commercial EJ-276D. All samples are cut and polished as Ø2.5 cm x18 mm cylinders.*

Analysis of the scintillation parameters presented in Table 2 shows that, at any equal concentration, the use of PHF produces much brighter plastics than PPO. Another advantage of PHF over PPO is that, even without any secondary dye, a 30% PHF plastic exhibits about 20% higher LO relative to EJ-200. Under the same conditions, the LO of the 30% PPO plastic is lower by 22% relatively to EJ-200, while OG or DIPN plastics prepared without secondary dyes lose practically all their LO and PSD efficiency. This unique feature of PHF must be determined by the practical absence of self-absorption already witnessed in crystals and further confirmed by the measurements of the molecular spectra that are more relevant to the polymer solutions (Fig. 6).



*Fig. 6. Overlay of the molecular absorption and photoluminescence spectra measured in diluted cyclohexane solutions: A – PHF showing a negligible self-absorption that must contribute to the efficient LO of PHF in plastics compared to B – Respective spectra of PPO.*

Comparison of PHF to OG compositions may be less accurate because for these formulations, the commercial OG was used instead of the P2 compound reported in [14]. However, since the OG samples prepared and measured in this work did not show any lower LO compared to those

literature data, also measured relative to EJ-200, it can be suggested that PHF plastics can produce similar or even higher LO than the OG analogues. The same can be said about PSD that in 30% PHF plastics with C460, reaches PSD of EJ-276D, while 40% is required for the same PSD in OG formulations. These numbers could change if the weight fractions are replaced by the molecular concentrations that, unfortunately, cannot be calculated due to the unknown molecular weight of the commercial OG used in these studies. However, considering that at high weight concentrations of 40-50% FoMs of both PHF and OG plastics are coming close to the maximum values measured with the corresponding crystal and OG, it can be suggested that under the same conditions PHF molecules also have higher capability for PSD production in plastics.

A common trend that can be observed with the tested plastic compositions is a noticeable enhancement of LO and PSD at increasing concentration of all primary dyes. In a practical sense, it shows that the high concentration of a primary dye is not a limiting factor that unavoidably leads to the deterioration of scintillation properties. On the contrary, under conditions of suitable physical properties, it may be used as a tool for further enhancement of the scintillation performance. In general, the observed trend can be explained by the increasing efficiency of the excited singlet ( $S^I$ ) and triplet ( $T^I$ ) state interactions and energy transfer at decreasing intermolecular distances produced by the high dye concentrations. The fact that, at 40-50% loading, PSD in both PHF and OG plastics approaches the values typical for pure corresponding crystals or glass indicates that at these high concentrations the molecules of primary dyes come to the distance sufficient for maximal  $T^I T^I$  annihilation. This may indicate that PSD comparable to that of crystals and OG may be achieved in plastics. Less positive projections can be made about the LO of plastics that, as shown by comparison to the stilbene sample, does reach the high LO of crystals or OG. The explanation of this difference may relate to the different mechanisms

of  $S^I$  energy transfer. In pure crystals, the interaction of an  $S^I$ -excited molecule with its neighbors leads to a rapid migration of the electronic excitation [3]. It can be suggested that a similar rate of energy transfer can take place in closely packed amorphous OG. In polymers that have relatively large and disordered spacing between multi-component aromatic segments, a larger fraction of the initial excitation energy can be lost resulting in the decrease of the final LO. The theoretical consideration of these processes that are out of scope of the present work can be complicated by the many parameters within real systems. However, further experimental studies of such unique systems as PHF or OG, that can be equally used for preparation of crystals, amorphous glass, and polymer solutions may provide a more certain answer to whether high levels of the LO observed with crystals or OG can be achieved in plastic scintillators.

### *3.2.2. Physical properties*

As can be seen from the data of Table 2, despite the relatively low LO, PPO-based plastics still produce better PSD compared to other dyes at all equal concentrations. There are, however, limits to the practical use of the highest PSD that can be achieved with PPO. These limitations relate to the specific problems imposed by the high concentration on the physical properties of current PPO-based plastics. Among these problems, the most serious are dye leaching and relative softness that become pronounced at high PPO concentrations (see data of Table 2). During PSD plastic development, these problems have been partially mitigated by the addition of a crosslinker and use of C460 as an alternative secondary dye with antioxidant properties [12]. However, as shown by the data of Table 2, for the complete prevention of the negative effects of leaching and insufficient hardness, the PPO concentration in plastics should be decreased to about 25%. It is obvious that, in combination with the modest LO, this limitation may produce a negative impact for deployment of the PPO-based PSD plastics in applications that require

maximization of the LO and PSD. Such applications can benefit from the replacement of PPO with PHF that is free of these disadvantages. As shown by the parameters of Table 2, at 40-50% it allows for the preparation of highly efficient LO and PSD plastics that, being sufficiently hard, do not show any signs of leaching at any periods of observations.

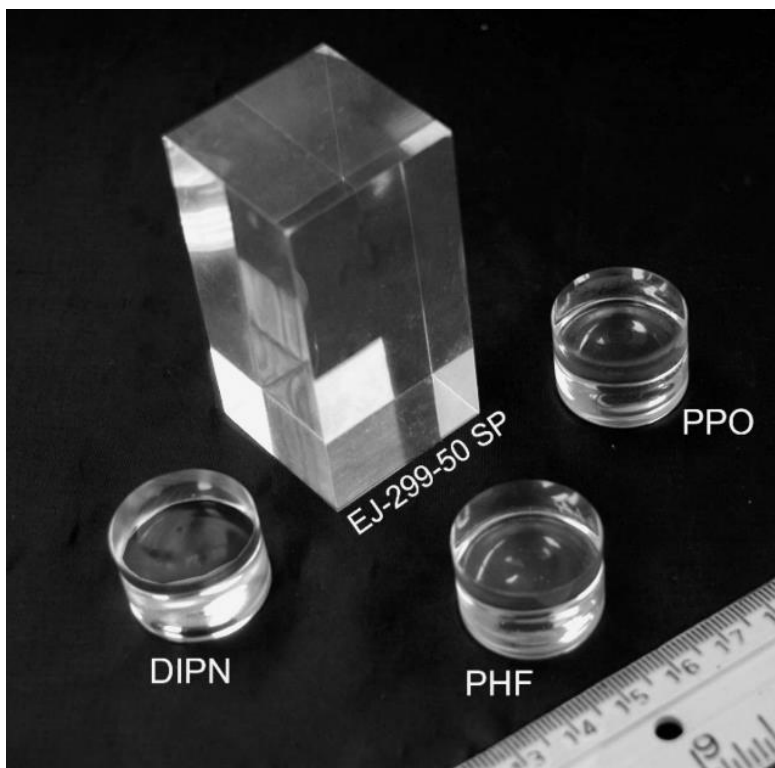
OG formulations that, compared to PHF have a small disadvantage of a lower PSD, can be other obvious candidates for potential PPO replacement. However, in our opinion, there is no obvious reason to refer compositions obtained by dissolution of OG in a polymer to a separate type of organic materials called blends. The unique properties ascribed to the blends, such as no need for crosslinker, high hardness, absence of leaching, or temporal stability were defined based on a sole comparison to PPO. Meanwhile, results of this work show that such properties may be equally achieved by using other dyes in plastic scintillators. As illustrated by Table 2, non-crosslinked PHF formulations do not show any signs of leaching and can be successfully prepared with hardness equal to OG compositions. Our extensive studies of scintillation performance stability [35] showed that degradation and yellowing may not be observed with many other plastics prepared with alternative to PPO primary dyes like e.g., *m*-terphenyl or DIPN. As in the case of DIPN, it is not surprising that at very high concentrations, the nature of primary dyes can strongly affect variations of molecular bonding that must determine plasticization or hardness, while the phenomenon of leaching can be largely influenced by the molecular size and shapes that control the diffusion in porous polymer structures. Such an assumption has been confirmed by observations that leaching phenomena are more pronounced with smaller and more compact molecules, like, e.g., biphenyl [34]. It also explains less excessive PPO leaching in PS compared to PVT that, due to the presence of methyl groups in its structure, has a bigger fraction of free spacings favorable for the diffusion of the dye molecules.

From this point of view, none of the formulations tested in this work contradicts a classical definition of plastics as physical systems produced by the dissolution of certain compounds in polymer matrix, and therefore there is not a clear distinction that would separate blends of OG from highly loaded plastic scintillators that contain alternative primary dyes. However, further studies and exploration of OG compounds that first showed a possibility of the significant enhancement of LO in plastics seems to be one of the promising venues for future developments.

### 3.2.3. *<sup>6</sup>Li-loaded plastics*

Examples of PHF and OG show potential benefits of compounds alternative to PPO in applications that may be affected by poor physical properties of the current PSD formulations. Among such potential applications can be the production of high ratio pixelated arrays for neutron imaging [36] or recently emerging photopolymerization and 3D printing [37] that suffer from precipitation of PPO on the surfaces or in the volume of the scintillator parts. While the use of new dyes in such applications is still in the early stage of development, a positive effect of PPO replacement is already demonstrated with <sup>6</sup>Li-loaded plastics currently tested for commercial production and use in fast neutron spectroscopy [38] and antineutrino detection [39]. According to the current technology, the dissolution of <sup>6</sup>Li in non-polar polystyrene or PVT requires addition of solubilizing agents (organic acids) that lead to a severe LO loss that negatively affects both the resolution of neutron spectroscopy and attenuation in large-scale scintillators needed for the antineutrino detection. Our previous studies [40] suggested that the main reason for this LO decrease is caused by unfavorable interactions between the polar acids and PPO that contains O and N atoms in its molecular structure. This suggestion, first illustrated by the replacement of PPO by a pure hydrocarbon (*m*-terphenyl) [41] finds some confirmation with DIPN and PHF that, like *m*-terphenyl, do not have any substitution atoms in their

hydrocarbon structures. Unfortunately, we failed to prepare any  $^6\text{Li}$ -loaded plastics with OG that did not produce transparent samples for further comparison. However, both DIPN and PHF were found compatible with  $^6\text{Li}$  salt of isovaleric acid ( $^6\text{LiIVA}$ ) that, dissolved in concentration of 0.1 atomic % of  $^6\text{Li}$ , allows for easy preparation of clear transparent scintillators (Fig. 7).



*Fig. 7. Examples of  $^6\text{Li}$ -loaded plastics studied in present work. Small cylinders were prepared with 30% of the different primary dyes indicated by the corresponding legends. The bigger rectangular plastic is a test prototype of PHF-based plastic produced by Eljen Technology under the provisional name of EJ-299-50SP*

Scintillation and physical parameters measured in this work with the studied  $^6\text{Li}$ -loaded compositions are summarized in Table 3. The results illustrate the main problems typical for current, PPO-based plastics. The first problem is a big LO drop that reaches 35-37% compared to the analogous unloaded compositions (see Table 2). This brings the brightness of  $^6\text{Li}$ -loaded PPO plastics to a level much below that of EJ-200. Another problem is dye leaching that in PVT-based plastics used for commercial developments may be pronounced more than with PS. As seen from

Composition	$^{137}\text{Cs}$ LO	PSD FoM	$n_{\text{th}}^{\circ}$ spot E, keV <sub>ee</sub>	Hardness (Shore D)	Leaching
<b>PPO</b>					
30% PPO + 0.2 % bis-MSB in PVT	0.75	2.26	400	82	Slight
30% PPO + 0.2 % C460 in PVT	0.78	2.58	440	84	Slight
<b>DIPN</b>					
30% DIPN + 0.5 %E404 in PS	1.15	3.55	570	75	No
30% DIPN + 0.2 % bis-MSB in PS	0.95	2.65	565	74	No
30% DIPN + 0.2 % C460 in PS	0.94	2.08	520	78	No
<b>PHF</b>					
30% PHF + 0.2 % bis-MSB in PVT	1.28	3.08	465	88	No
30% PHF + 0.2 % C460 in PVT	1.36	3.01	460	87	No
EJ-200	1.0	n/a	n/a	84	No

*Table 3. Scintillation and physical parameters measured with  $^6\text{Li}$ -loaded plastics containing 30% of different primary dyes dissolved in PS or PVT matrices, with addition of 5% DVB and 6.5 % of PMAA (polymethacrylic acid). All plastics are cylinders of the same size ( $\varnothing 25$  mm x 18 mm). PSD FoMs correspond to the keV<sub>ee</sub> range of the respective thermal spot positions.*

comparison, these effects are noticeably milder for DIPN plastics that, being not very bright initially, experience a much smaller LO decrease of 8-14% compared to their unloaded counterparts. However, the most remarkable result can be observed with PHF that, for the first time for  $^6\text{Li}$ -loaded plastics, produced an outstanding 36% higher LO measured relative to the EJ-200. Another advantage of PHF over PPO is increased hardness that is especially important for large-scale assemblies that incorporate large-scale, 1 meter-long  $^6\text{Li}$ -loaded bars [39].

Similar positive effect of PPO replacement is also observed on PSD that, opposite to the unloaded plastics, show higher FoM numbers in PHF and DIPN compositions. Like with LO, this effect may relate to the less pronounced reactions of DIPN and PHF with the polar components that affect excited singlet and triplet interactions in plastics with different primary dyes. Another positive outcome of the PPO replacement is a change of the thermal neutron spot positions that in DIPN and PHF plastics shift to the region of higher energy. These results show



that the replacement of PPO by alternative primary dyes may influence not only LO but may produce a noticeable effect on the ionization quenching that leads to the corresponding improvements of energy resolution, especially strongly pronounced in DIPN-E404 composition (Fig. 8).

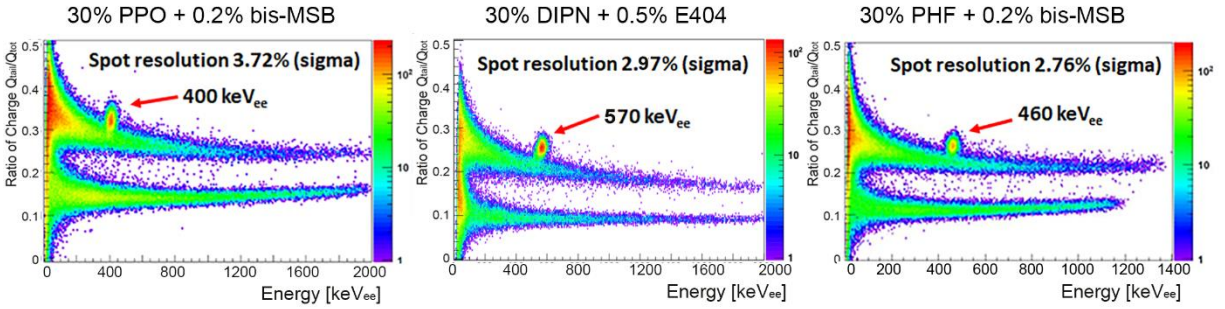


Fig. 8. Experimental distributions measured with plastics prepared with 30% of PPO, DIPN, and PHF. Comparison shows that the position and the energy resolution of the thermal neutron spot, that correlates with the light quenching properties, changes depending on the primary dyes. The distributions were obtained using  $^{252}\text{Cf}$  source shielded by 1'' HDPE.

Deeper understanding of these phenomena will require more investigations with a broader variety of non-traditional dyes that can be found in further developments. Such work with PHF and other promising dyes still involves development of new synthesis, complicated purification procedures, and adjustments of costs, but potential production of new, improved  $^6\text{Li}$  formulations will be beneficial for all applications that involve combined fast/thermal neutron detection, neutron spectroscopy, and antineutrino detection.

#### 4. Conclusion

One of results obtained in this research is growth of a new single crystal of recently reported, non-traditional scintillation compound, 9,9-dimethyl-2-phenyl-9H-fluorene (PHF). This compound represents a rare example of a pure aromatic crystal that can be relatively easily grown by a classical solution growth technique. Initial characterization of the scintillation

properties showed that the new crystal belongs to a family of the most efficient single crystal organic scintillators. Comparison to the classical crystals of anthracene, *trans*-stilbene, and *p*-terphenyl, and to more recently introduced organic glass (OG) showed that PHF crystal may produce LO exceeding that of *trans*-stilbene and PSD of *p*-terphenyl known as the second highest PSD standard for all organic scintillators. Optical characterization of PHF single crystals showed relatively long wavelength of emission (374 nm), that is well matching the wavelength sensitivity of the common blue-region PMTs, and uniquely low self-absorption that, like in *trans*-stilbene or anthracene crystals eliminate the need of wavelength shifter additions.

Another advantage of PHF is the high solubility in organic solvents, that makes it suitable not only for crystal growth but for the use as a primary dye in PSD plastic formulations. This allowed for the preparation and evaluation of scintillation and physical properties of non-traditional plastic compositions prepared with PHF, OG, and liquid diisopropylnaphthalene (DIPN). These dyes were considered as examples for possible replacements of PPO used as a primary dye in current commercial PSD plastics that suffer from modest LO and physical problems expressed in relative softness and dye leaching caused by easy migration of the dye to plastic surfaces. Comparison of plastics prepared with different dyes showed the highest performance of PHF that, at 50% concentration, produced a record LO increase of 69% relative to EJ-200 and PSD approaching that of PHF single crystals. Moreover, high hardness and absence of dye leaching equally observed with plastics containing OG and PHF prepared with and without additions of a crosslinkers indicated that enhanced scintillation and physical properties are not a unique attribute of OG-based type of materials but can be obtained with a broader variety of dyes that determine scintillation and physical properties of the final plastics. These overall results showed that future development of PSD plastics should not be limited to

sole use of PPO. On the contrary, it must involve search and exploration of new efficient dyes that can be used for adjustments of scintillation and physical performance required for different applications. The possibility of such adjustments is demonstrated with  $^6\text{Li}$ -loaded plastics that currently have insufficient LO and poor physical properties introduced by unfavorable reactions of PPO with polar  $^6\text{Li}$  additions. As found in presented experiments, the replacement of PPO with less reactive DIPN or PHF can substantially moderate these reactions and hence preclude deleterious influence on resultant LO, PSD, and neutron energy resolution required for detection of thermal neutrons, fast neutron spectroscopy, and antineutrino detection applications.

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