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Polysiloxane Scintillators for Efficient Neutron and Gamma-Ray Pulse Shape Discrimination

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KEYWORDS

1
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3 Plastic scintillator, polysiloxane, pulse shape discrimination, neutron detection, radiation
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7 detection
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16 ABSTRACT

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21 Plastic scintillators based on thermoplastics, such as polystyrene and poly(vinyl toluene)
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24 (PVT), are capable of neutron and gamma radiation detection via pulse shape
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27 discrimination (PSD) when over-doped with select fluorescent molecules. This class of
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31 plastic scintillator has been extensively studied but is limited to applications suitable for
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34 thermoplastics. For applications requiring flexibility, scintillators composed of elastomers,
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37 such as polysiloxanes, offer an alternative to PVT scintillators. Polysiloxane scintillators
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41 are inherently flexible and have short processing time on the order of 3 hours in air and
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44 equivalent or better detection capability at reduced doping concentration (<5 wt%). This
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47 work presents polysiloxane-based scintillators, containing only 1–5 wt% of 2,5 diphenyl-
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51 oxazole (PPO) or 9,9-dimethyl-2-phenyl-9H-fluorene (PhF) as primary dopants and 9,9-
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3 dimethyl-2,7-distyryl-9H-fluorene (SFS) as a wavelength shifter. A 5 wt% PPO
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7 polysiloxane sample had improved neutron and gamma ray PSD and comparable light
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10 yield than EJ-299-33 tested under the same conditions, i.e. FoM of 1.33 ± 0.03 at 450
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13 keV_{ee} and light yield of 94% relative to EJ-299-33. The 5 wt% PhF-polysiloxane sample
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17 had a higher light yield, 144% of EJ-299-33, but lower FoM under the same conditions
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20 (FoM of 1.09 ± 0.03). This work highlights the potential of polysiloxanes as a matrix for
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24 PSD capable plastic scintillators.
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INTRODUCTION

Plastic scintillators are often used in radiation portal monitors as a first line of detection for special nuclear materials (SNM) at international borders and points of entry. Pioneered by Brooks et al. in 1960,¹ modern formulations of plastic scintillators have been developed to distinguish neutrons from gamma rays.^{2–10} The ability to detect neutrons enhances the accuracy of plastic scintillators since only SNM and source materials, such as ²⁵²Cf and alpha-n sources, emit neutrons, while both SNM and benign materials emit gamma rays. One method to distinguish neutrons from gamma rays is pulse shape discrimination (PSD). Thermoplastics, such as poly(vinyl toluene) (PVT) or polystyrene, are capable of PSD if the plastic is over-doped with fluorescent small molecules, such as 2,5-diphenyloxazole (PPO).² This over-doping technique has been commercialized, but the large amount of fluorescent dopant required for PSD (>20 wt.%) leads to unfavorable material properties such as a lower glass transition temperature and dopant precipitation over time.¹¹ Some work to mitigate these effects has been done, including a commercial cross-linked sample from Eljen Technology (EJ-276).^{10–12} EJ-276 showed PSD capabilities similar to a commercial liquid scintillator, EJ-

309, with sources reporting a figure of merit (FoM) above 2 at 500 keV_{ee} and similar light yields to EJ-299-33, at 56% anthracene.^{10,13–15} This proprietary formulation represents a large step towards replacing liquid with plastic scintillators. EJ-276 and other cross-linked formulations of plastic scintillators mitigate many of the undesirable material properties, but the cross-linking strategy may also limit some of the potential applications. The rigidity and high hardness of cross-linked formulations prevents dopant aggregation and is desirable for large scale scintillators but renders them unsuitable for applications requiring flexibility.

Polysiloxanes, also known as silicones, have been considered as an alternative to traditional matrices used in plastic scintillators.^{16–20} As elastomers, polysiloxanes are inherently flexible and easy to process under ambient conditions, offering a complementary set of properties to thermoplastics. Unlike oxygen sensitive vinyl toluene or styrene formulations used for traditional plastic scintillators,^{12,21,22} polysiloxanes can be cured from easy-to-process resins with tunable viscosities in ambient conditions. Recently, Marchi et al. reported that polysiloxane scintillators up to 112 cm³ in volume with low dopant loading (<8 wt% PPO) are capable of PSD.²⁰ A 4 wt% PPO polysiloxane

scintillator (5.1 cm³) had a FoM of 0.65 and light output of 45% of EJ-212 (about 29.3% of anthracene), 3 months after fabrication. A large sample 112 cm³ in volume was also produced, with a FoM of 0.523 and light output of 17% of EJ-200 (10.9% anthracene) two years after fabrication. This was the first time PSD capabilities were shown in large polysiloxane scintillators, even after several months of storage. With these promising results from aged polysiloxane scintillators, we sought to improve the PSD capabilities of polysiloxane scintillators. Polysiloxane scintillators approximately 5.4 cm³ in volume were fabricated with PSD capabilities similar to commonly used PVT plastic scintillators. Based on a modified commercial silicone resin doped with ≤ 5 wt% PPO or an in-house synthesized dopant 9,9-dimethyl-2-phenyl-9H-fluorene (PhF), and a novel wavelength shifter, 9,9-dimethyl-2,7-distyryl-9H-fluorene (SFS), shown in Figure 1,⁹ polysiloxane scintillators with excellent light yields and PSD capabilities were produced in just 3 hours at 150°C in air.^{23,24}

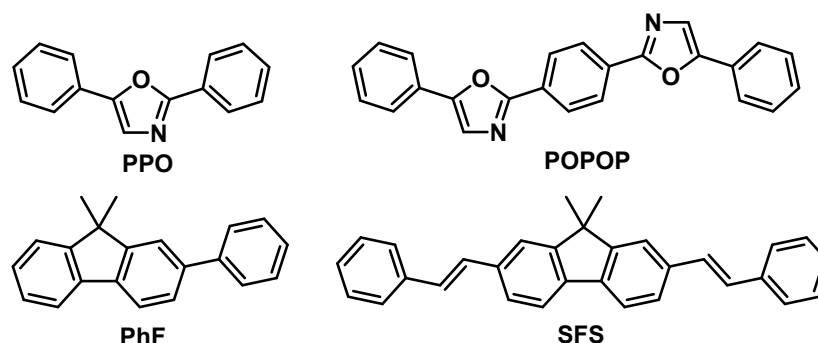


Figure 1. Commercial primary and secondary dopants (PPO and POPOP) and synthesized primary and secondary dopants (PhF and SFS) used in this study.

EXPERIMENTAL METHODS

PhF was synthesized via Suzuki coupling; detailed conditions can be found in the Supporting Information. 9,9-dimethyl-2,7-distyryl-9H-fluorene (SFS) was synthesized according to procedures outlined in our previous work.⁹ Purity was assessed via ¹H NMR (500MHz JEOL ECA-500) and GC/MS (Varian CP-3800 GC, 1200 L Quadrapole MS/MS with a Restek Rsi column).

Sample Fabrication

Glass scintillation vials were used as molds and silanized with a 10 v/v% dichlorodimethylsilane (Sigma) solution in dichloromethane, according to our previous work.^{11,25} Polysiloxane scintillators (6 g) were prepared by dissolving the primary and secondary dopant in an equal mass of xylenes (Sigma-Aldrich) under gentle heating in

the glass vial. Once the dopant was completely solubilized, the two-part commercial polysiloxane (Wacker LUMISIL 579) was added and the vial was capped shut and vortexed for 30-45 seconds to achieve a homogenous mixture. The polysiloxane sample was cured in the capped vial at 150°C for three hours, in air. Samples measuring approximately 1.9 cm in diameter and 1.9 cm in height were removed from the oven and carefully broken out of vials before cooling.

Radiation Testing

Experiments were carried out using a CAEN DT5730 14-bit digitizer operating at 500 MS/s. DPP-PSD firmware together with CAEN CoMPASS software were used to obtain waveforms associated with particle interactions in the scintillator. Samples were wrapped in PTFE tape and coupled to the Hamamatsu R6095 PMT with RX600k optical grease (Rekon Components, Inc) without additional polishing, then exposed to a 40-mCi AmBe source and the waveforms were integrated using long and short gates to calculate the FoM.² To ensure consistency between each of the samples, the same PMT was used as well as the same digitizer, power supply, blackbox, and method for coupling the scintillator and PMT. Further details on FoM calculation can be found in our

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3 previous work.^{9,11,25} A 273- μCi ^{137}Cs source was used for light output and
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7 spectroscopic comparison with commercially available scintillators. To calculate relative
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10 light yield, the Compton edge was fitted with a Gaussian curve and the half-maximum
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13 was used to estimate the position of the Compton edge. The integrated light outputs of
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17 scintillators, produced over the course of six hours, were compared with that of EJ-299-
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21 33 to get the relative light yield at the Compton edge of ^{137}Cs (480 keV). A second
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24 photon source, a 12-mCi ^{60}Co source, was used in tandem with the ^{137}Cs for relative
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28 energy calibration using the position of the Compton edge of each of these sources.
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31 The two Compton edges (^{60}Co and ^{137}Cs) were used as calibration points to obtain the
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34 electron equivalent energy. Due to the low energy resolution of these samples, the
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37 Compton edge from each photopeak of Co-60 were indistinguishable, so the Compton
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40 edge from selected to be the highest energy edge and assumed to a superposition of
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43 the Compton edges from the 1.33 MeV and 1.17 MeV photons. No collimation or
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47 neutron/gamma attenuators were employed in this work for reproducibility. The source-
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51 to-detector distance was fixed at 15.24 cm (6 inches) for all experimental
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RESULTS AND DISCUSSION

A two-part commercial polysiloxane resin from Wacker (Wacker LUMISIL 579) was selected for high phenyl content and optical clarity. Up to 5 wt% of PPO and 0.1 wt% SFS was added and the resin was cured at 150°C for 3 hours to produce clear polysiloxane scintillators, approximately 1.9 cm in diameter and 1.9 cm in height. SFS, shown in Figure 1, was used as a secondary dopant for its enhanced solubility in the polysiloxane matrix over the commercially available POPOP.⁹ All samples were initially clear but slightly yellow due to the strongly pigmented SFS, as shown in Figure 2. The PPO-polysiloxane samples did not require any additional machining or polishing before testing. Additional surface treatment was not necessary, but best results were achieved after wiping or soaking in isopropanol to remove any residual dopants on the surface. All samples prepared in this work contained 0.1 wt% SFS and thus will be identified only by their primary dopant in the remainder of this manuscript.

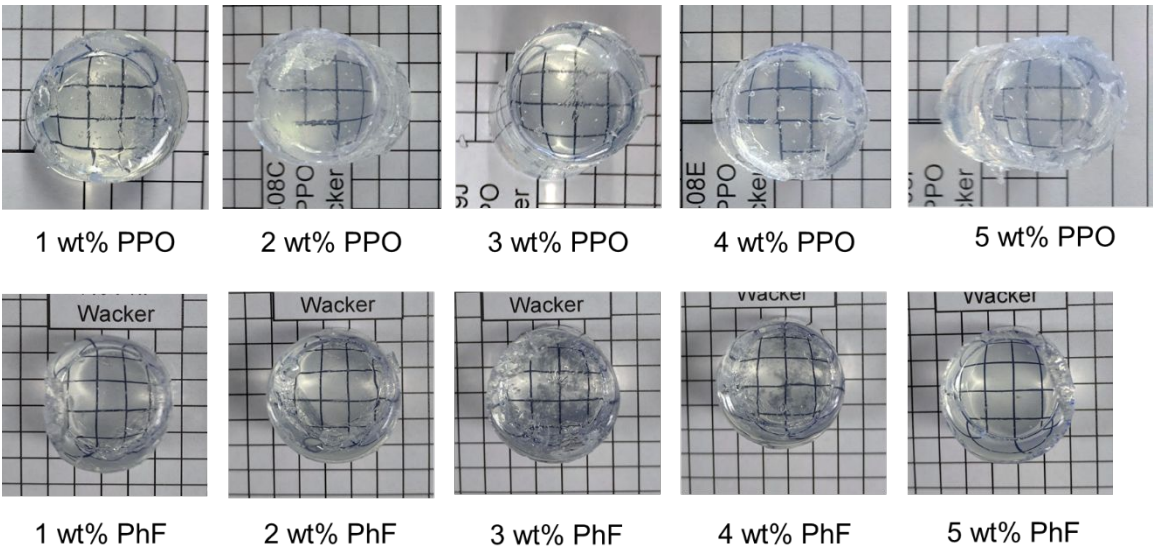


Figure 2. Polysiloxane plastic scintillators with varying concentrations of PPO and PhF, 24 hours after fabrication.

After curing, the PSD capabilities of each sample were quantified by measuring the relative light yield (LY) and figure of merit (FoM). The figure of merit quantifies the separation between neutron and gamma ray signals. A 2 wt% PPO-polysiloxane scintillator showed comparable FoM to a similar sized EJ-299-33 sample at all energies. Above 2 wt% PPO, PPO-polysiloxanes had higher FoM than EJ-299-33 (FoM of 1.13 ± 0.02 at $450 \text{ keV}_{\text{ee}}$), with a 5 wt% PPO achieving a FoM of 1.33 ± 0.03 at $450 \text{ keV}_{\text{ee}}$. Trends in PPO concentration and FoM can be seen in Figure 3 and are also tabulated with associated error in the Supporting Information, Table S1. Despite the high FoM, PPO-polysiloxane had slightly lower LY than EJ-299-33, between 86-94% of EJ-299-33,

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3 as depicted in Figure 4. The ^{137}Cs responses of PPO-polysiloxane samples used to
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7 calculate LY are shown in Figure 5 and the curves are overlaid with their Gaussian fit in
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10 the Supporting Information, Figure S12. The EJ-299-33 sample had a much broader
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13 Compton Edge than the PPO-polysiloxane samples, as seen in Figure 5, possibly due
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17 to increased scattering or re-absorption. This highlights a shortcoming in the widely
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20 accepted method of using the position maximum of the Compton Edge, or some
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23 percentage of the max, to calculate relative LY.^{2,3,26–29} Samples with a very broad
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27 response will lead to artificially high values of relative LY. Some groups have done
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30 Monte-Carlo simulations and analysis to better estimate the position of the Compton
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33 edge,^{26,27} but more consensus within the community is necessary to standardize an
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37 accurate method that accounts for the position and broadening of the Compton Edge
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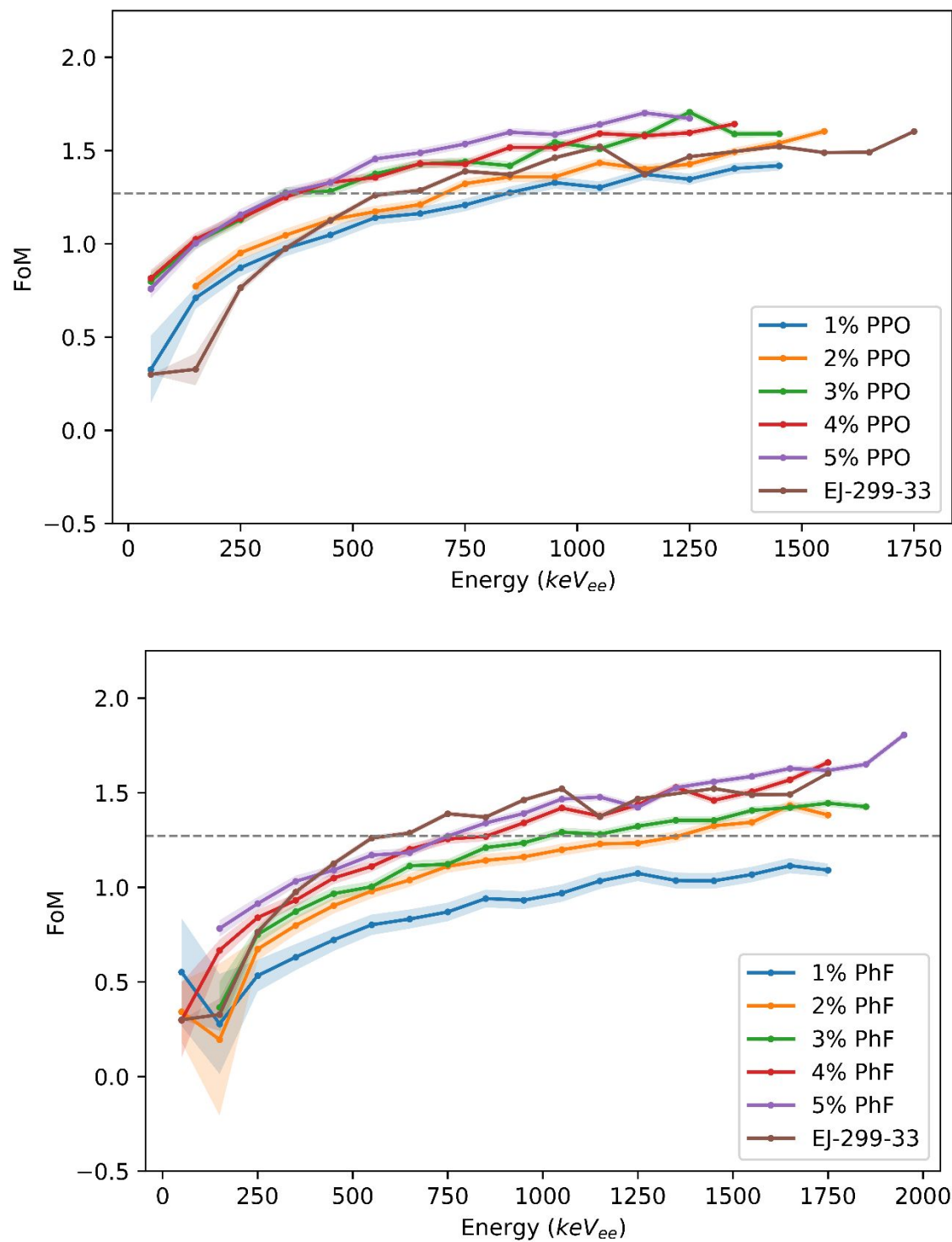


Figure 3. FoM of PPO-polysiloxane (top) and PhF-polysiloxane (bottom) samples at varying energies, compared to commercial sample EJ-299-33. The associated error is represented by the shaded portions and the values are tabulated in the Supporting Information (Tables S1 and S2).

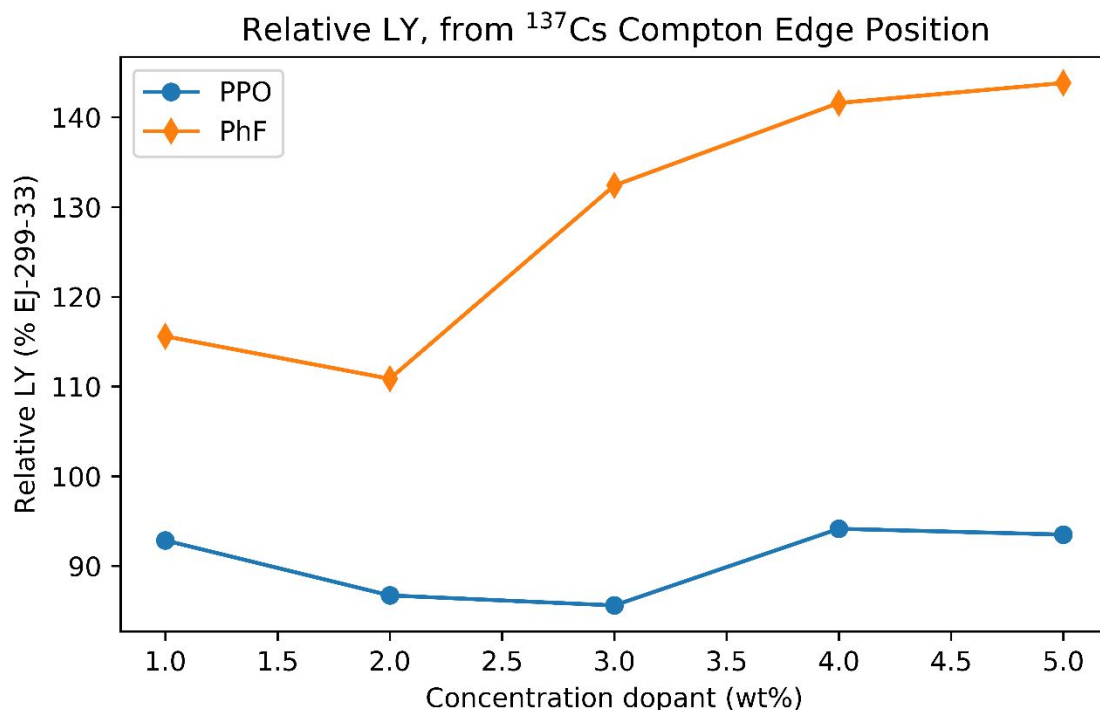
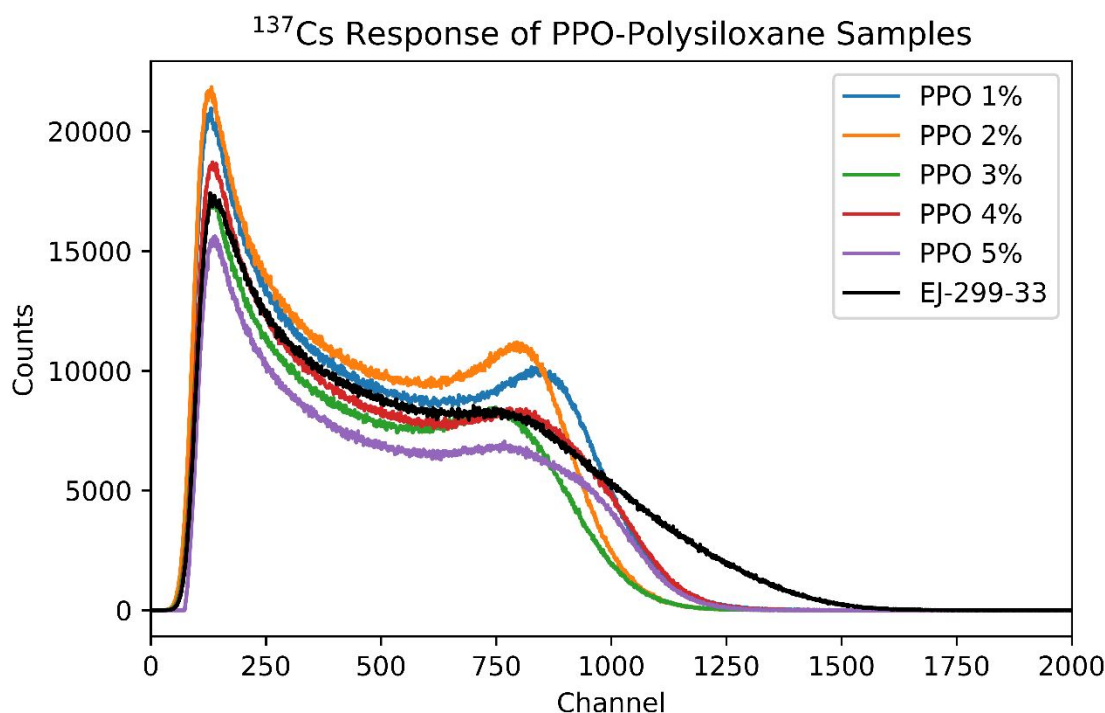


Figure 4. Light yield of PPO- and PhF-polysiloxane scintillators, relative to commercial sample EJ-299-33. The relative LY was calculated by comparing the position of the Compton edge, approximated at 50% of the local maximum intensity. Values are tabulated in the Supporting Information, Table S3.



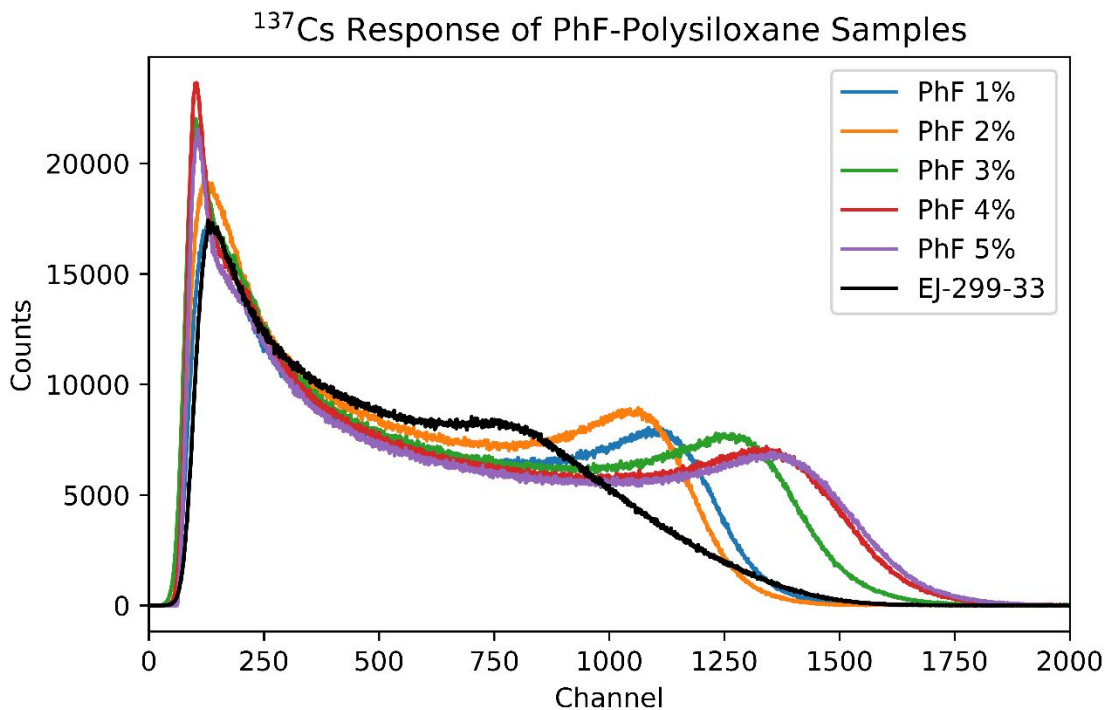


Figure 5. ¹³⁷Cs response of PPO- and PhF-polysiloxane samples. EJ-299-33 (black) shows a broad Compton edge.

Initially, PPO appeared soluble up to 5 wt% in the polysiloxane matrix, as seen in the initial sample pictures in Figure 2. However, over 2 months of storage in ambient conditions, the 5 wt% sample showed visible signs of dopant precipitation, shown in Figure 6. Below 5 wt%, there were no signs of dopant precipitation by visual inspection, implying PPO was soluble only up to 4 wt% in the polysiloxane.

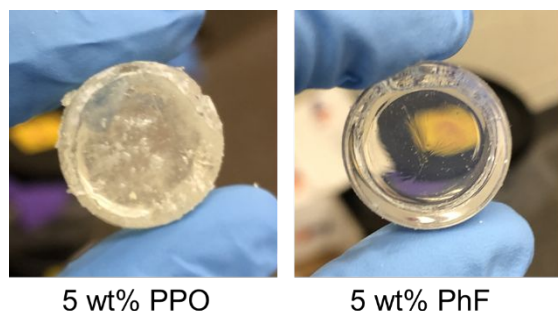


Figure 6. Polysiloxane samples containing 5 wt% PPO (left) and PhF (right) 2 months after fabrication. Visible aggregation and clouding is evident in PPO sample.

As higher concentrations of PPO did not seem stable in the polysiloxane matrix, we explored another primary dopant, PhF, shown in Figure 1. PhF was developed as a primary dopant for PVT plastic scintillators in previous work and had a moderate light yield and FoM but resulted in harder samples than PPO. A 20 wt% PhF-PVT (4.7 cm diameter and 1.2 cm in height) had a FoM of 1.16 between 400 and 600 keV_{ee}, a LY of 70% of BC-408, and a Shore-D of 81. The analogous 20 wt% PPO-PVT sample had better PSD capabilities, with a FoM of 1.61 and LY of 89% of BC-408 but had a Shore-D of only 69.⁹ The better mechanical properties of PhF based scintillators and moderate PSD capabilities made it a promising candidate for polysiloxane scintillators.

Fabricated according to the same method as PPO-polysiloxane scintillators, PhF was soluble in polysiloxanes up to 5 wt%. Above 5 wt%, PhF crashed out during curing.

PhF-polysiloxane scintillators pictured in Figure 2 did not show visible signs of dopant precipitation after 4+ months in ambient conditions. PhF-polysiloxane scintillators were also capable of PSD at low concentrations. A 5 wt% PhF-polysiloxane scintillator had a FoM of 1.09 ± 0.03 at 450 keV_{ee}, similar to EJ-299-33, with a FoM of 1.13 ± 0.02 at 450 keV_{ee}. While the PhF-polysiloxane scintillator FoM was lower than PPO-polysiloxane analogues, PhF-polysiloxane samples had much higher relative LY, ranging between 111% and 144% of EJ-299-33 as shown graphically in Figure 4. This trend was not observed in PhF-PVT analogues, as PhF had a lower LY than PPO-PVT samples as mentioned above. The ¹³⁷Cs response of PhF-polysiloxane samples, depicted in Figure 5 and the Supporting Information Figure S13, shows much sharper Compton edges at higher energies than EJ-299-33. The high LY and lack of dopant precipitation in PhF-polysiloxane scintillators renders PhF interesting for further study as a primary dopant in polysiloxane scintillators despite the slightly lower FoM.

PPO-polysiloxane scintillators based on phenylmethyl polysiloxanes have shown PSD capabilities, but had lower FoM and LY than EJ-299-33A.²⁰ We suspect the high

phenyl content, implied by the high refractive index (1.54) of the Wacker formulation may contribute to the high FoM and LY in PPO and PhF-polysiloxane samples. While the exact composition of the Wacker formulation is proprietary, we know it contains vinyl, hydride, and alkoxy terminated siloxanes and oligomers. Thus, the cross-linking density and potential of condensation reactions may also contribute to the enhanced PSD capabilities of these samples over previously published work. Work is currently underway to fabricate custom formulations to test these hypotheses.

In addition to the high phenyl content, the low solubility of the dopants in the polysiloxane matrix may also lead to enhanced PSD. Preliminary solid-state emission spectra of PPO in polysiloxane (without any wavelength shifter) shows a redshift and broadening between 1 and 4 wt% PPO. The 4 wt% PPO-polysiloxane sample had a emission spectra that closely matched the shape of a 20 wt% PPO-PVT sample, leading us to believe that PPO aggregated similarly and formed excimers.²⁹ Further experiments to compare the decay time are being considered to confirm the formation of excimers.

PhF-polysiloxane scintillators also exhibit a red shift and broadening when dopant concentration increased from 1 to 4 wt%. Increasing PhF from 1 to 4 wt% in polysiloxane and 1 to 20 wt% in PVT resulted in similar red-shifted and broadened emission (Figure 7, Table 1). As with PPO, the similarities in spectral shape of a 20 wt% PhF-PVT and 4 wt% PhF-polysiloxane emission spectra suggest that favorable aggregation and excimer formation may occur to promote PSD at low loadings of PhF in polysiloxanes. The red shift in the PhF-PVT spectra compared to the PhF-polysiloxane spectra may be due to the non-planarity of the molecule. The dihedral angle between the fluorene core and the phenyl ring in the 2nd position is reported as 41.1°. ²⁹ Rotations or deviations from this calculated dihedral angle can be caused by different solvents or matrices such as polysiloxane and PVT. This effect may not be observed with PPO as its ground state geometry is predicted as planar.⁹

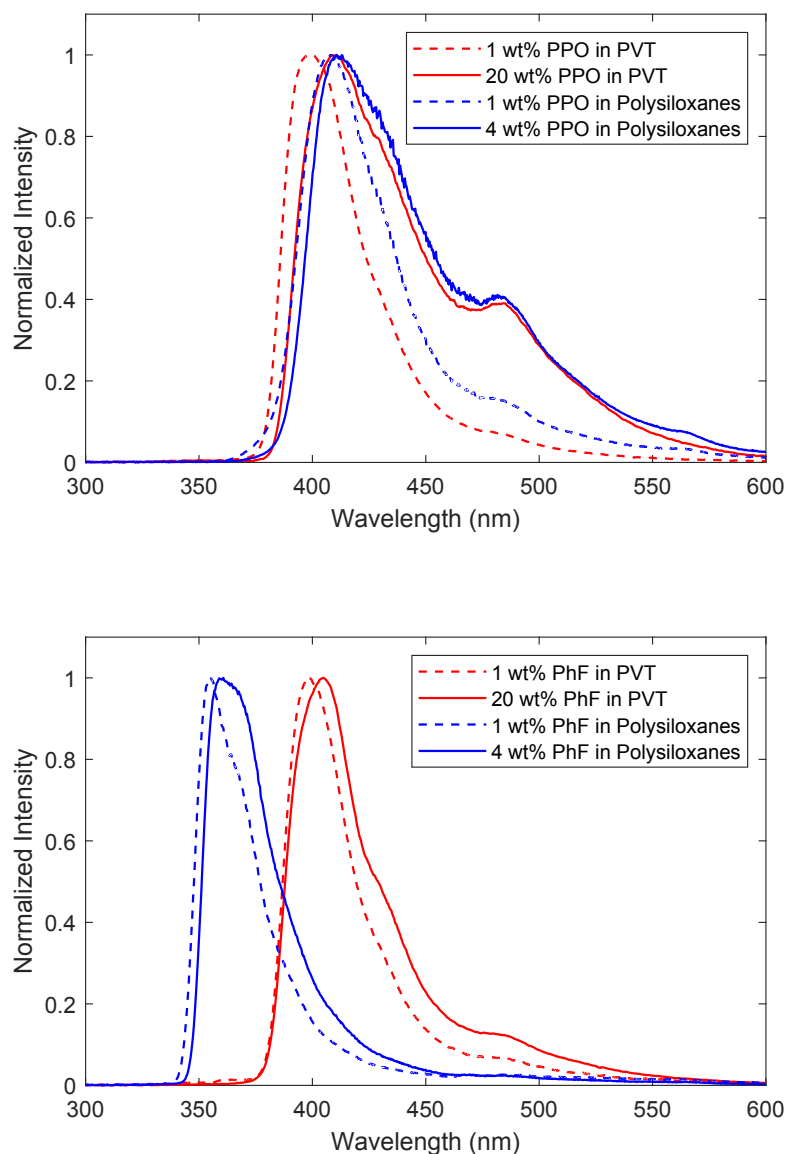


Figure 7. (Top) Emission of PPO in PVT and polysiloxane at varying concentrations. A similar broadening and redshift is observed in PVT and polysiloxanes with increasing concentration of PPO. (Bottom) Emission of PhF in PVT and polysiloxanes. A red-shift and broadening is observed with increasing concentration of PhF.

Table 1. Wavelength of maximum emission of PPO and PhF in PVT and polysiloxanes

	$\lambda_{\text{max, PPO}}$ [nm]	$\lambda_{\text{max, PhF}}$ [nm]
1 wt% in Polysiloxanes	407	356
4 wt% in Polysiloxanes	411	360
1 wt% in PVT	400	398
20 wt% in PVT	408	405

CONCLUSIONS

Plastic scintillators composed of a polymer matrix with various dopants offer a route to customizable radiation detectors, including systems for neutron and gamma ray discrimination. Traditionally, thermoplastic scintillators based on polystyrene or PVT have been doped with greater than 20 wt% of PPO to distinguish neutrons from gamma rays via PSD. The undesirable properties such as softening, low T_g , and dopant aggregation have been addressed by techniques such as cross-linking, and Eljen Technologies has commercialized a formulation rivaling the performance of liquid scintillators (EJ-276). One area traditional thermoplastics, especially cross-linked formulations, fall short is in applications requiring flexibility. In this work, we demonstrated that flexible, elastomer polysiloxane based plastic scintillators are capable of PSD at low dopant loadings. Polysiloxane scintillators (6g) were prepared in

3 hours in air from a commercial siloxane resin with PPO or PhF as primary dopants and SFS as a wavelength shifter. A 5 wt% PPO polysiloxane sample produced a higher FoM than a commercial PVT scintillator but slightly lower light yield, with an FoM of 1.33 ± 0.03 at $450 \text{ keV}_{\text{ee}}$ and relative LY of 94% of EJ-299-33. PhF-polysiloxane scintillators were also capable of PSD with a FoM of 1.09 ± 0.03 at $450 \text{ keV}_{\text{ee}}$ and LY of 144% compared to EJ-299-33. Despite a lower FoM than PPO-polysiloxane analogues, PhF-polysiloxane scintillators are still of interest due to the high LY and lack of dopant precipitation. Polysiloxane scintillators may achieve good PSD and LY at low concentrations of dopants due to a lower solubility of selected dopants leading to favorable aggregation and enhanced excimer formation. Work to thoroughly characterize material properties and optimize curing conditions is underway. By no means a replacement to thermoplastic scintillators, polysiloxane scintillators offer a flexible alternative that is still capable of PSD on par with some thermoplastic scintillators.

ASSOCIATED CONTENT

SUPPORTING INFORMATION

The following files are available free of charge.

Synthesis of 9,9-dimethyl-2-phenyl-9H-fluorene, 2D PSD plot for commercial sample

EJ-299-33, 2D PSD plot for 1 wt% PPO-polysiloxane sample, 2D PSD plot for 2 wt%

PPO-polysiloxane sample, 2D PSD plot for 3 wt% PPO-polysiloxane sample, 2D PSD

plot for 4 wt% PPO-polysiloxane sample, 2D PSD plot for 5 wt% PPO-polysiloxane

sample, 2D PSD plot for 1 wt% PhF-polysiloxane sample, 2D PSD plot for 2 wt% PhF-

polysiloxane sample, 2D PSD plot for 3 wt% PhF-polysiloxane sample, 2D PSD plot for

4 wt% PhF-polysiloxane sample, 2D PSD plot for 5 wt% PhF-polysiloxane sample,

tabulated FoM for PPO- and PhF-polysiloxane samples, ^{137}Cs response of PPO- and

PhF-polysiloxane samples, tabulated light yields of PPO- and PhF- polysiloxane

samples (PDF)

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AUTHOR CONTRIBUTIONS

A.L. and A.S. contributed to the fabrication of polysiloxane scintillators. A.L. synthesized and characterized dopants and measured optical properties of scintillators. J.A. tested radiation response of all samples. A.S. and A.E. designed the research project. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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NOTES

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