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ABSTRACT: Recent developments of plastic scintillators are reviewed, from January 2000 to August 2015. All examples are distributed into the main application, i.e. how the plastic scintillator was modified to enhance the detection towards a given radiation particle. The main characteristics of these newly created scintillators and their detection properties are given.

KEYWORDS: Materials for solid-state detectors; Scintillators and scintillating fibres and light guide

Contents

Introduction

Protection of civilians and facilities against CBRN-E (Chemical, Biological, Radiological, Nuclear, and Explosives) threats represent a true challenge due to the constant increase of world's population movements. According to Dr. El Baradei (1997 – 2009 IAEA Director General), terrorists who are unconcerned about exposing themselves to radiation could easily conceal a source in a truck or a suitcase. *"The danger of handling powerful radioactive sources can no longer be seen as an effective deterrent, which dramatically changes previous assumptions. […] Security of nuclear and other radioactive material has taken on dramatically heightened [in IAEA's work] significance in recent years."*

As an example, a dramatic story happened in late 2013 in Mexico, where a radioactive cobalt-60 source (3,000 Curies, 111 TBq) was stolen from its transportation truck. Fortunately, the material was safely recovered 8 days later [1].

In this context, numerous detectors could be used for NR detection. Among them, we will focus in this Review on plastic scintillators (hereafter abbreviated as PS, see footnote[1]). These materials can be defined as one or several fluorescent probes embedded in a polymer matrix, and the resulting system is able to produce light while interacting with a radioactive source (Figure 1 is an example produced with a UV lamp as excitation). For instance, a typical scintillation cocktail

[1] Standard abbreviations used in this document: α-NPO: 2-(1-Naphthyl)-5-phenyloxazole; AIBN: aso-bisisobutyronitrile; BBO: 2,5-bis-(4-biphenylyl)-1,3,4-oxadiazole; CL: Cathodoluminescence; CQD: carbon quantum dot; DVB: divinylbenzene; FOM: Figure Of Merit; HMPA: hexamethylphosphoramide; IBIL: Ion Beam Induced Luminescence; LiMA: Lithium Methacrylate; Li-Sal: Lithium Salicylate; MEH-PPV: Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylene-vinylene; MPA : mercaptopropionic acid; MOF: Metal Organic Framework; NMP: 1-methyl-2-pyrrolidinone; *p*-T: *p*-terphenyl; PBBO: 2-(4-biphenylyl)-6-phenylbenzoxazole; PBD: 2-(4-Biphenylyl)-5-phenyl-1,3,4-oxadiazole; PEG: poly(ethylene glycol); PL: Photoluminescence; PBMA: Poly(benzyl methacrylate); PMMA: Poly(methyl methacrylate); POPOP: 1,4-Bis(5-phenyl-2-oxazolyl)benzene; PPO: 2,5-Diphenyloxazole; PS: Plastic scintillator; PSD: Pulse shape discrimination; PSt: polystyrene; PVA: Polyvinyl alcohol; PVK: Polyvinylcarbazole; PVT: Polyvinyltoluene; QD: Quantum dots; SEM: Scanning electron microscopy; SSD: Spectral shape discrimination; St: styrene, TTA: Triplet-triplet annihilation.

is made from *p*-terphenyl and POPOP dissolved in polystyrene. The preparation of a plastic scintillator was first described in the late 50's [2].

To detect Special Nuclear Materials (roughly speaking, isotopes of uranium and plutonium), plastic scintillators present several advantages. They are cheap (especially interesting for large size detection systems), sensitive to gamma-rays, can be handled without any specification, reliable, stable and can be prepared in large volumes. More particularly, the choice of the detector will become extremely important in the future due to the combination of the cheapness of PS ($ 2,000 for a 3.8 cm × 36 cm × 173 cm large PVT detector, compared to $ 6,000 for a 5 cm × 10 cm × 41 cm NaI(Tl) inorganic scintillator [3]) and the necessity for some countries to cover at the best their borders with radiation portal monitors.

But some drawbacks have incited several groups to renew with chemical developments of plastic scintillators: they display a poor resolution, were presumed for a long time to be unable to perform fast neutron/gamma discrimination, afford relative low scintillation yields compared with inorganic scintillators and cannot give access to the full energy of an incident gamma.

The most relevant examples will be described in the following sections, which are distributed as a function of the optimization of the PS towards the nature of the ionizing radiation. The Reader is encouraged to refer to the full Review about this topic [4]. Also, this Review is exclusively limited to plastic scintillators and derivatives (composites, sol-gel, etc.). No data will be given regarding improvements on organic single crystals, optical (scintillating) fibres, liquid scintillators and inorganic scintillators.

Optimization of alpha detection

The detection of alpha-emitting radionuclides is always a big challenge, due to the fact that they are highly ionizing and heavy particles. They interact within a few micrometres (e.g. an alpha of energy 5 MeV will penetrate within 35 μm in polystyrene). Contrarily, a 1 MeV gamma would need not less than 14 cm and a 1 MeV electron 4.3 mm to fully release their energy.

Figure 1. Plastic scintillators displaying different emission wavelengths (excitation with UV lamp; © CEA).

In this context, it is noteworthy the work performed by Tarancón et al. for the replacement of liquid scintillators with scintillating microspheres. Contrary to the former, PS microspheres do not generate mixed wastes after the measurement and are particularly suitable for alpha and beta emitters. Scintillation properties of microspheres prepared from PSt doped with various, classical fluorophores: *p*-terphenyl, PPO, POPOP, bis-MSB and naphthalene, with a typical diameter of *ca.* 130 µm were investigated [5].

Although sol-gel materials can be considered as inorganic structures, one has to admit that they are prepared from organic molecules. Also, as they are usually prepared in the form of small monoliths, their size is suitable for alpha measurements. Thus, thin layer scintillators were prepared from various silicates, PPO and POPOP [6]. A modest detection was observed.

Optimization of beta detection

Beta detection can also be performed with microspheres, *via* an alpha/beta discrimination process [5]. Even low-energy beta emitters such as ^{3}H and ^{14}C are detected via scintillation counting. An optimized diameter size was determined for each radionuclide. Detection efficiency values obtained with these synthesized microspheres for ^{3}H, ^{14}C, $^{90}Sr/^{90}Y$ and ^{241}Am sources are better than those obtained using commercial plastic scintillation microspheres (from Saint-Gobain or Detec-Rad).

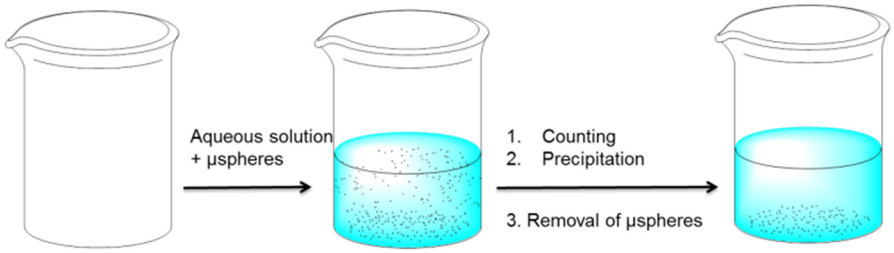

Figure 2. Experimental method for scintillation counting with scintillating microspheres.

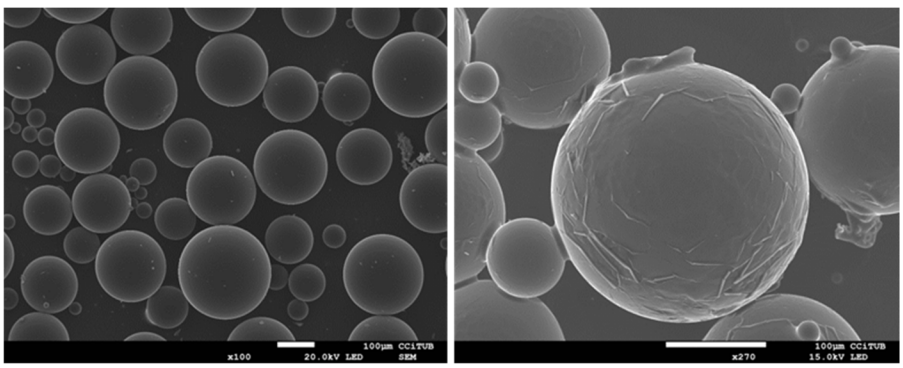

Figure 3. SEM picture of scintillating microspheres (© A. Tarancón, with permission).

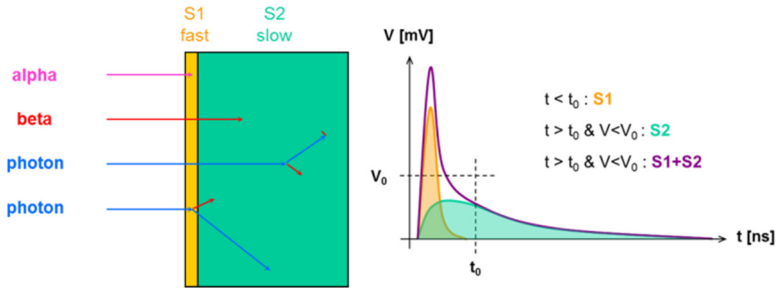

Figure 4. Principle of beta/gamma discrimination using phoswich scintillators.

Another approach using standard size plastic scintillators is based on the phoswich principle. Whereas the electron can deposit its energy in the first (mainly) and the second scintillator, the gamma background will impact both layers, accordingly with their respective volumes. Thus, by comparing the scintillation pulse profiles of the thin – and fast – and the thick – and slow – plastic scintillators, one may discriminate beta from gamma rays [7]. Moreover the two scintillating layers were coupled altogether without glue so as to give access to the full beta energy peak.

Plastic scintillators for gamma and x-rays spectrometry

An important modification of the composition of a plastic scintillator is to make it denser and to increase its effective Z (Z_{eff}) by heavy metal loading. But heavy atoms tend to have a strong fluorescence quenching due to multiple vibrational relaxations. Nevertheless a compromise can be found between higher absorption and lower light output, so as to lead to a pseudo-gamma spectrometry.

Currently most of the developments use tin, lead and bismuth organometallics, and more particularly the two latter metals. Lead, in its lead dimethacrylate form, has been extensively studied these years at the CEA [8] and the loaded plastic scintillator has been implemented for the Laser Mégajoule facility as a hardened imaging system for 10 – 40 keV X-rays [9]. Concerning gamma-rays identification, the best results are obtained with bismuth(III) organometallics as dopants. Triphenyl bismuth, triaryl [10,11] and tricarboxyl bismuth complexes [10,12] have been reported. With such loading, the apparition of the photoelectric peak was observed for high energy X-rays and gamma below 200 keV, as well as in synchrotron conditions [13] by several groups around the world. PE Peak was also claimed to be observable with a ^{137}Cs source (662 keV) [12]. It is noteworthy that some other exotic metals such as hafnium of zirconium – in a nanoparticle form – have also been studied to enhance x-rays detection [14].

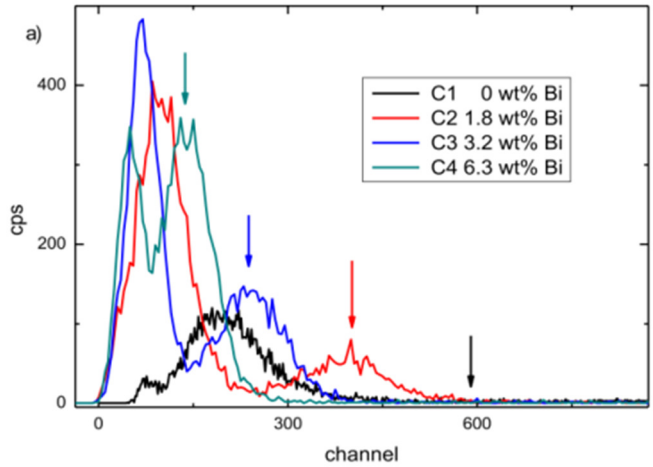

Figure 5. Pulse area spectra of compounds 1 to 4 displaying various $BiPh_3$ loadings when exposed to a ^{57}Co gamma source (0.9 kBq). The arrow shows the full energy peak at 122.1 keV.

Thermal neutron detection

Neutron detection is of primarily importance since the shortage of ^{3}He, the most important neutron absorber for homeland security applications. In this context, several helium-3 alternatives are required. Absorption of thermal neutrons (generated after multiple collisions with hydrogen atoms) is not natural for a standard plastic scintillator and requires different elements other than C, H, N, S or O. The desired isotopes with large cross sections towards thermal neutrons are given in Table 1 with their natural abundances.

TABLE 1. NEUTRON CAPTURE WITH VARIOUS ELEMENTS

Isotope	Reaction of interest	Thermal Cross section (Barns)	Isotope abundance
^{3}He	$^{3}He + n \rightarrow {}^{3}H + 1H + 0.765\ MeV$	5,330	0.00013 %
^{6}Li	$^{6}Li + n \rightarrow {}^{3}H + \alpha + 4.78\ MeV$	940	7.5 %
^{10}B	$^{10}B + n \rightarrow {}^{7}Li* + \alpha + 2.8\ MeV \rightarrow {}^{7}Li + \alpha + \gamma\ (0.48\ MeV)$	3,840	19.9 %
^{113}Cd	$^{113}Cd + n \rightarrow {}^{114}Cd + \gamma's\ (9\ MeV)$	30,000	12.2 %
^{155}Gd	$^{155,157}Gd + n \rightarrow {}^{156,158}Gd* \rightarrow {}^{156,158}Gd + e- + \gamma's\ (8\ MeV)$	60,700	14.7 %
^{157}Gd		254,000	15.7 %

Boron-loaded plastic scintillators are very well documented as they are commercially available (BC-454 or EJ-254 from Saint-Gobain and Eljen Technology, respectively). Due to the low atomic number of boron, there is no gamma rays detection enhancement and the thermal cross section of B allows a moderate, yet efficient loading. Also, as the capture of the thermal neutron releases an alpha particle, pulse discrimination is therefore possible [15].

Usually the carborane molecule is used as dopant in the plastic scintillator series, thanks to its extremely high content of boron inside the whole molecule. However, the use of pyrene modified with pinacolyl borate on four positions [16].The boron-rich pyrene (and benzene) derivatives are prepared by Suzuki conditions using both microwave and traditional heating, affording yields of 40–93%. Plastic scintillators with boron loading as high as 5 wt% are thus obtained.

Lithium has been less studied due probably to the lower cross section and the highly diffusive nature of organolithium compounds. Recently several improvements have been reported, with the use of dopants such as lithium pivalate [17] or lithium (phenyl)salicylate [18], or as co-monomers, in the form of lithium methacrylate [19] or lithium maleate [20] (Figure 7).

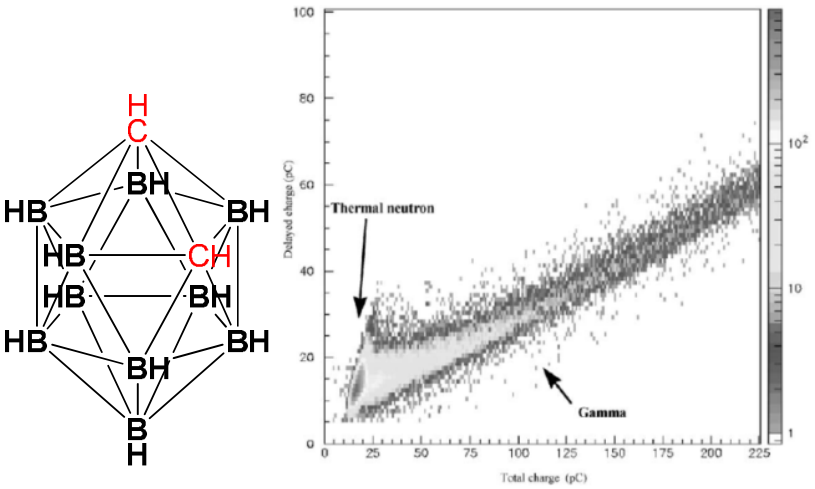

Figure 6. Left: structure of *ortho*-carborane; right: pulse shape discrimination spectrum between thermal neutrons and gamma rays.

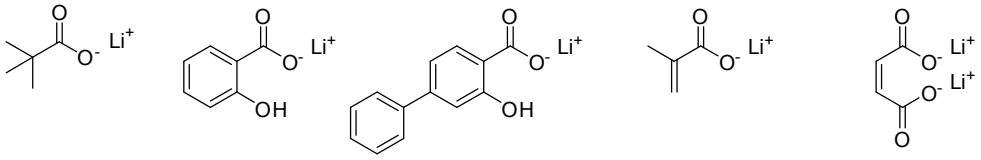

Figure 7. Lithium pivalate, lithium salicylate, lithium 4-phenylsalicylate, lithium methacrylate and lithium maleate.

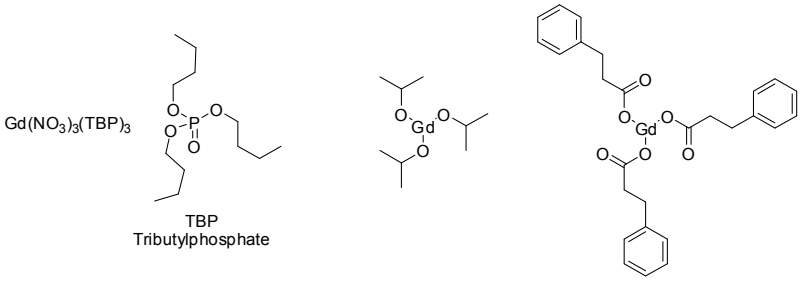

Figure 8. Examples of gadolinium-loaded compounds for potential incorporation into plastic scintillators: gadolinium nitrate tributylphosphate [21], gadolinium tris-isopropoxide [22], gadolinium tris-phenylpropionate [23].

To the best of our knowledge, cadmium loading in its organometallic form in PS- or PVT-based plastic scintillators has never been reported, due to the extreme toxicity of organocadmium compounds.

Last but not least, gadolinium presents the highest cross section of all stable elements, so only a low concentration of Gd is necessary within the scintillator for complete capture of the thermal neutron. Gadolinium is usually added in its organometallic form or as a salt chelated with a suitable phosphate. However, due to the release of electrons and gamma rays upon capture of the thermal neutron, pulse shape discrimination is not possible. But it is still possible to access to the neutron information by compensation [24] or by using the high-energy gamma rays signature emitted by neutron capture with Gd [25]. In this context, we have developed a scintillating sphere – so-called GADOSPHERE™ (Figure 9) – with detection performances close to a ^{3}He-based Bonner sphere of same volume. This relocatable neutron detector presents characteristics as mentioned in the Table 2.

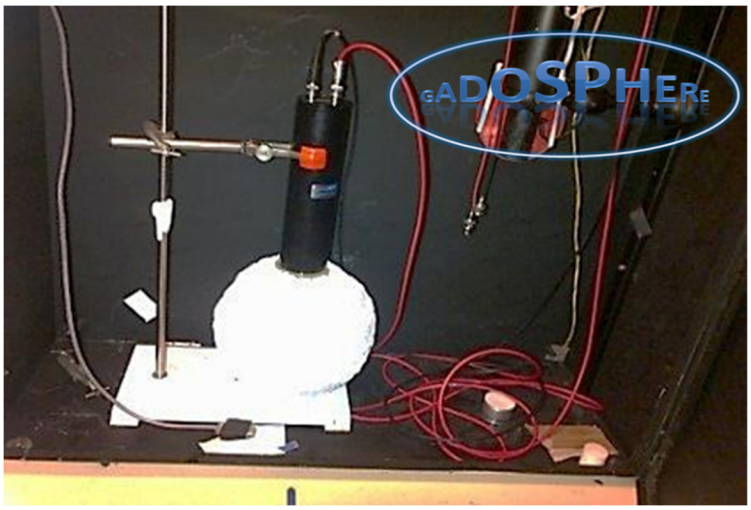

Figure 9. GADOSPHERE™ setup experiment.

Table 2. Main characteristics of GADOSPHERE™ when exposed to a ^{252}Cf source, benchmarked with a Berthold LB6411 probe.

	GADOSPHERE™	Berthold LB6411
Sensitivity ($c.n^{-1}.cm^2$)	0.77 ± 0.13	0.055 ± 0.002
Limit of detection ($n.cm^{-2}.s^{-1}$)	5.0 ± 2.0	2.4 ± 0.8
Gamma background saturation ($\mu Sv.h^{-1}$)	50	10000

Fast neutron detection

The main challenge for fast neutron detection is to dissociate their response from the gamma background. Unlike alpha or beta rays, neutrons have a scintillator penetration range similar to gamma. Only the ionization mechanism is slightly different between gamma and neutrons, and can produce some kind of signal discrepancy. Pulse Shape Discrimination (PSD) was early-on chosen as the technic of choice for observing, separating and attributing individual contributions coming either from a neutron or from a gamma [26].

It was assumed for a long time that plastic scintillators would be unable to efficiently perform PSD. But as early as 1960, Frank D. Brooks prepared a plastic scintillator suitable for fast neutron/gamma discrimination [27]. It was composed on 4-isopropylbiphenyl, a molecule which acted both as a primary fluorophore and "secondary solvent". Added at the concentration of 10 wt%, along with *p*-terphenyl and POPOP inside PSt, it was later on commercialized by Nuclear Enterprise under the trade name NE-150. Unfortunately, the product was discarded after 6 months due to whitening and edge deforming (Figure 10).

Probably based on this preliminary work, different groups noticed that the triplet-triplet annihilation (TTA), which is at the genesis of n/γ PSD, was not probable enough in plastics compared with liquids [28]. Thus, by increasing drastically the loading of primary fluorophore, the probability of triplet-triplet interactions should increase. As their concentration goes up, the likelihood to have a precise overlap of orbitals between two fluorophores increases and they should be able to exchange more excitons, and therefore their triplet – triplet annihilation could occur more efficiently for PSD to be achieved. This assumption was successfully demonstrated with small, lab-scale radioactive sources such as ^{252}Cf or AmBe.

Another strategy consists in the use of triplet-harvesting fluorophores, i.e. integrating fluorophores able to produce light *via* their triplet state. In this context, organometallic, phosphorescent complexes are very promising chromophores for scintillation applications. An example of PSD achieved with an iridium complex in a polymeric matrix was described by Feng *et al.* [29]. The authors prepared a PVT polymer doped with the 0.1 – 0.2 wt% iridium complex $Ir(ppy\text{-}F_2)_2(F_2\text{-}pic)$. When ionized with an AmBe source, the scintillator shows a bi-exponential decay with a prompt and a delayed signal (respectively assigned to the gamma and fast neutrons response). In the same paper, the authors propose an unseen feature: spectral shape discrimination (SSD).

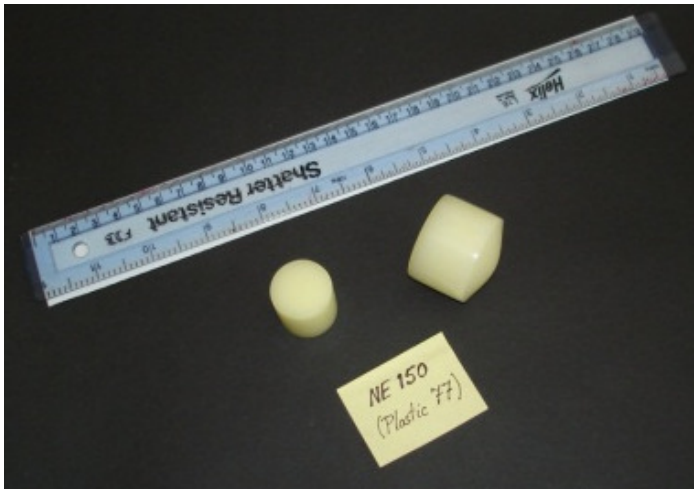

Figure 10; Picture of a degraded NE-150 plastic scintillator (© F.D. Brooks, with permission).

On the same basis, the group of Adadurov reported twice the use of two different fluorophores in PS, one for collecting singlet states (1,4-dimethyl-9,10-diphenylanthracene, with addition of the wavelength shifter Luminophor 59: 1-phenyl-5-(4-methoxyphenyl)-3-(1,8-naphthoenyl-1',2'-benzimidazole)-2-pyrazoline) and Eu(phen)(DBM)$_3$ for collecting triplet states [30]. So herein the large decay time difference between the two dyes is exploited, as the *singlet* fluorophore displays a 13 ns decay time and the *triplet* fluorophore a 370 µs decay time.

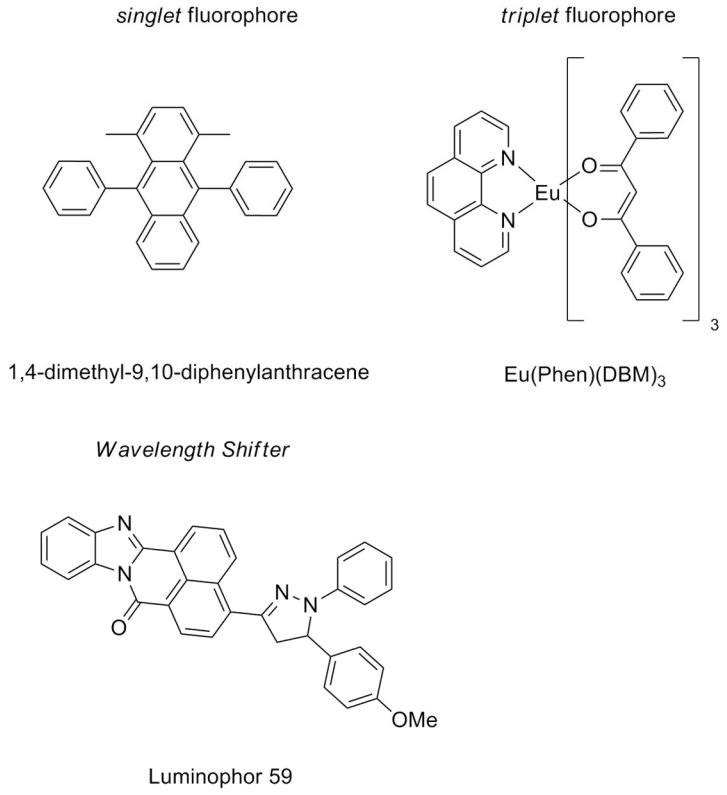

Figure 11. Representation of the singlet (with its wavelength shifter) and triplet fluorophores used by Adadurov et al. [30].

A collaboration between Polish and French teams has led to a proof-of-concept of fast neutron/gamma discrimination using the energy threshold of the reactions $n(^{19}F,^{16}N)\alpha$ or $n(^{19}F,^{19}O)H^+$ for photofission prompt neutron detection [31]. Indeed, the energetic gap allows discriminating neutrons with E > 2.5 MeV with less energetic neutrons and gamma. To reach this, polystyrene has been replaced by poly(2,3,4,5,6-pentafluorostyrene). A very high density was observed (1.56), equal to the Pb-loaded plastic scintillator cited before. Light output was estimated to be close to 3,100 ph/MeV, with a decay time of 3.0 ns, and preliminary results for n/γ discrimination of a PuBe radioactive source were somehow modest but exist (Figure 12), as due to small dimensions of the sample, energy deposition of highly energetic electron from beta decay of ^{16}N and ^{19}O is weak.

Conclusion

To the best of our knowledge, modifying plastic scintillators for fast neutrons detection and discrimination is the most recent example of new material supplied by plastic scintillators manufacturer. Indeed, EJ-299-33 is a promising result for chemists who want to create new solutions for physicists. But still, the road is long for that product to respond to all criteria for Homeland Security applications for example.
Some other challenges are facing chemists; among others, the key of many problems is probably the increase of the resolution and the light yield of the detector, which should lead to many improvements on nuclear detection. Also, the scale up mandatory if these scintillators have to be used in radiation portal monitors, and both chemistry and photophysics issues have to be overcome.

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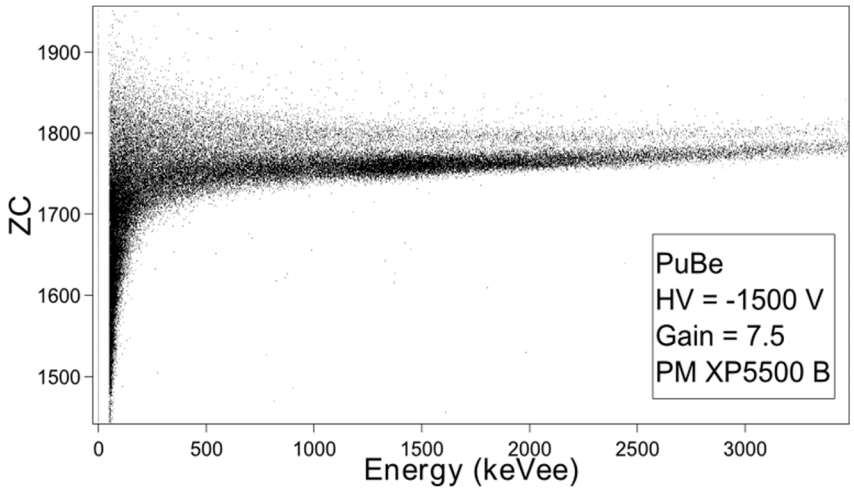

Figure 12. Neutron/gamma discrimination when irradiating the sample with a PuBe source.

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