Review of Plastic Scintillators for Neutron Detection

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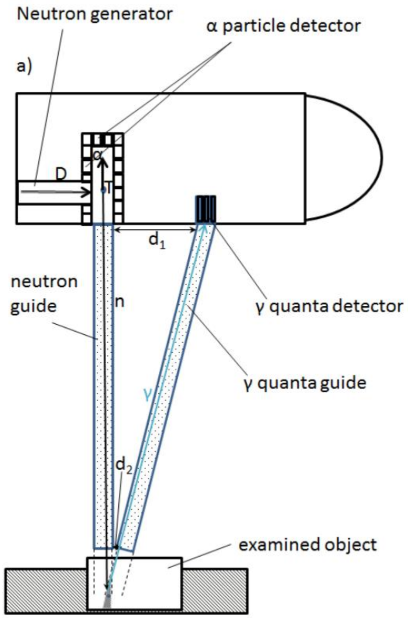
Outline

1) Light emission mechanism in plastic scintillators
2) Emission and absorption spectra
3) Radical bulk polymerization
4) Chemical components in standard plastic scintillators:
   - polymers;
   - UV fluorescent additions;
   - blue fluorescent additions.
5) Chemistry of plastic scintillators for neutron detection with:
   - boron;
   - gadolinium;
   - puls shape discrimination.
6) Summary

Motivation

Develop fast plastic scintillators loaded with elements interacting with neutrons as boron or gadolinium for neutron detection in:

- SABAT (Stoichiometry Analysis by Activation Techniques) project for non-invasive detection of hazardous materials in the aquatic environment;
- homeland security portal monitors installed in seaports, airports or at borders crossing points for detection of explosives and illicit drugs;
- physics experiments using time of flight, fast neutron counting, thermal neutron detection and pulse shape discriminating of gamma and fast neutron signals.

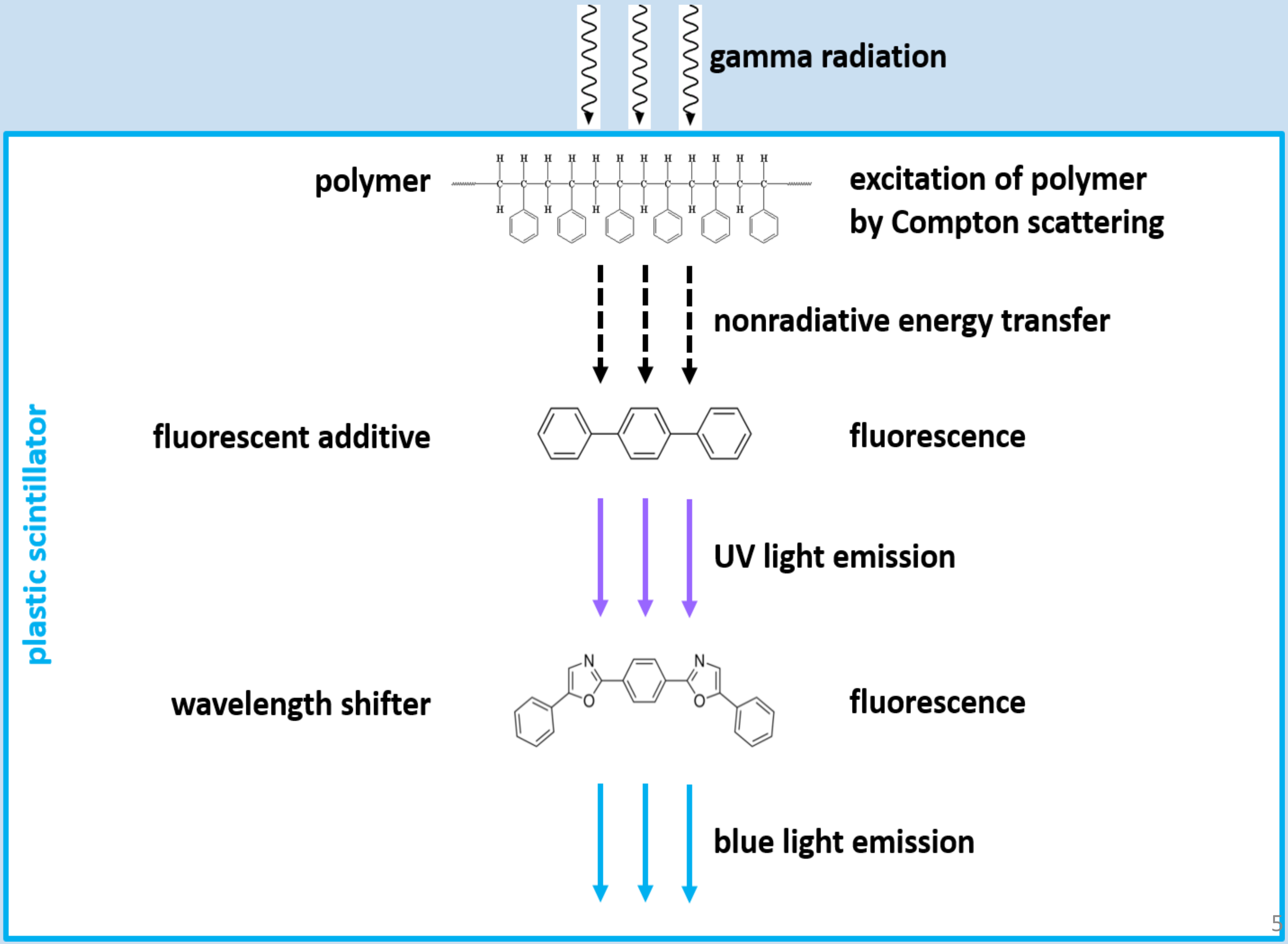


M. Silarski et al., Project of the Underwater System for Chemical Threat Detection, Acta Phys. Polon. A 127 (2015) 1543

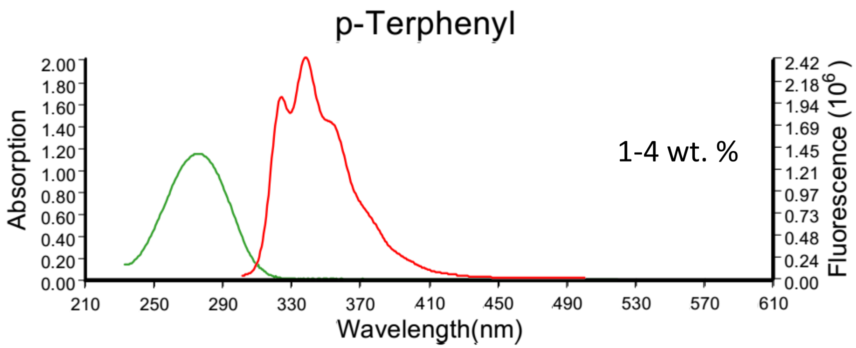
Properties of selected scintillators

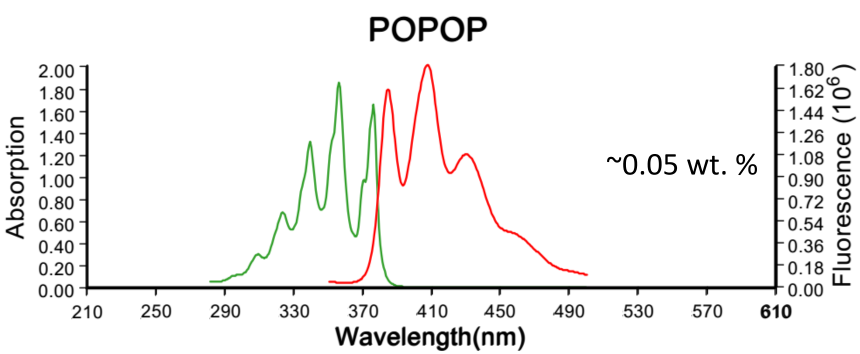
Scintillator	Light yield [ph/MeV]	Decay time [ns]	Wavelength of maximum emission [nm]	Refractive index	Density [g/cm^3]	Price [US$/cm^3]
Inorganic						
NaI(Tl)	37 700	230	415	1.85	3.67	6
^{6}Li glass	2 000	60	390-430	1.56	2.60	1 500
BaF_2	10000/1400	630/0.8	315/220	1.50	4.88	15
YAP	18 000	27	350	1.94	5.55	100
LSO	30 000	40	420	1.82	7.40	60
LYSO	32 000	40	420	1.81	7.10	70
BGO	8 200	300	480	2.15	7.13	35
$PbWO_4$	100/31	10/30	420/425	2.20	8.28	6
$LaBr_3$(Ce)	63 000	16	380	1.90	5.08	500
Organic plastic						
BC-420	12 240	1.5	391	1.58	1.03	0.11-0.32
BC-422Q	2 200	0.7	370	1.58	1.03	
BC-452	6 400	2.1	424	1.58	1.08	3
BC-454	9 600	2.2	425	1.58	1.03	11-12
Organic liquid						
BC-505	16 000	2.5	425	1.50	0.88	0.07-0.25
BC-509	4 000	3.1	425	1.40	1.61	
BC-521	12 000	4.0	425	1.50	0.89	
Organic crystalline						
Stilbene	14 000	3.5	390	1.64	1.22	-
Anthracene	20 000	30	445	1.62	1.25	-
P-terphenyl	27 000	3.7	420	1.65	1.23	-

Scintillation principles: energy transfer and light emission mechanism

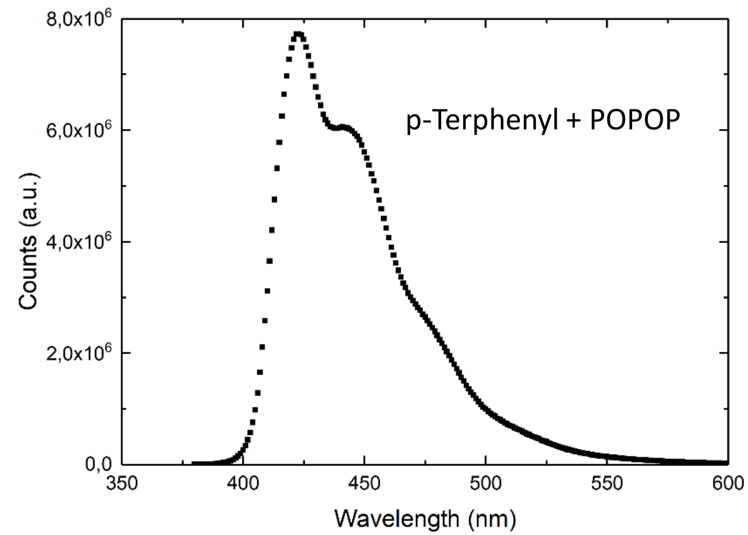


Scintillation principles: emission and absorption spectra





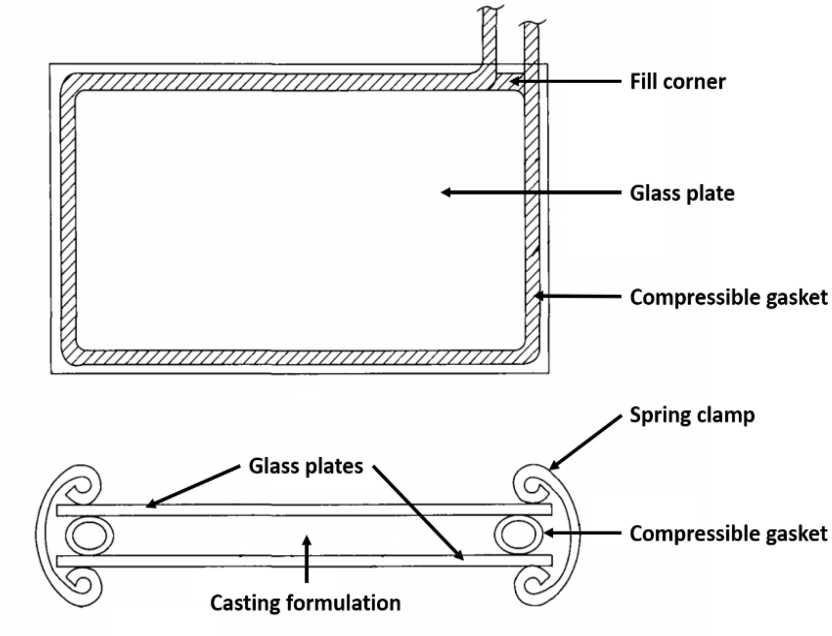
Absorption (green) and emission (red) spectra of two fluorescent substances commonly used in plastic scintillators

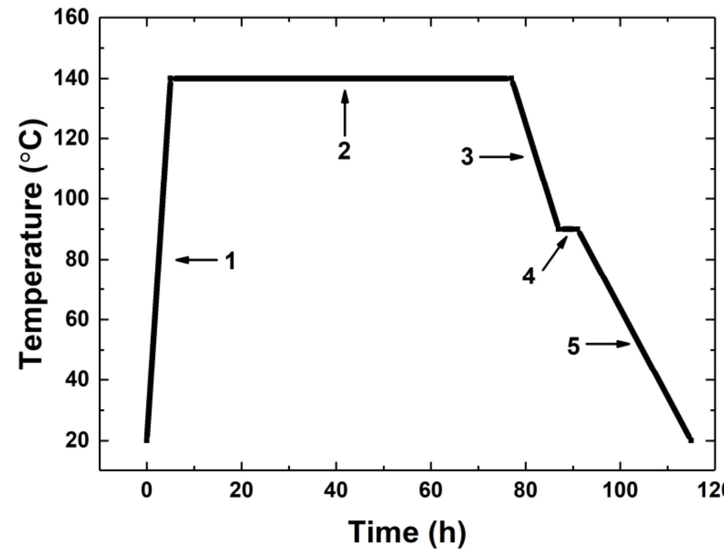


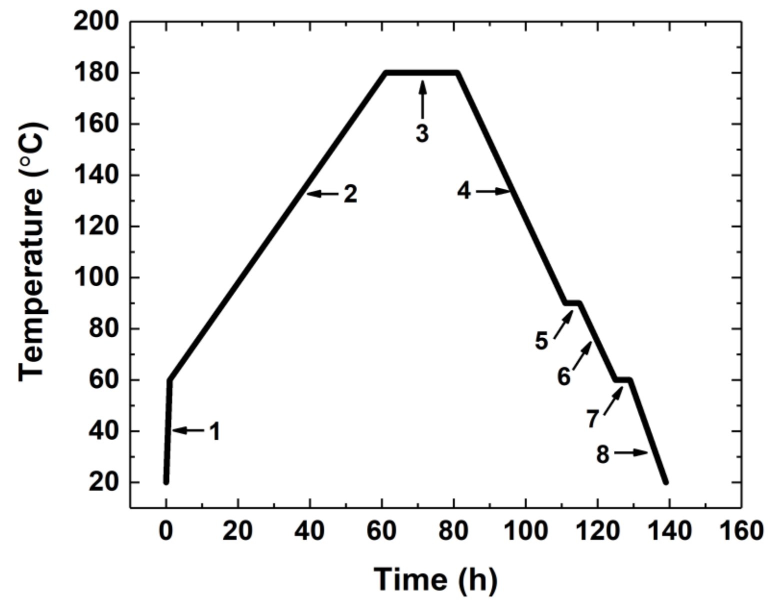
Emission spectrum of plastic scintillator with p-Terphenyl and POPOP

p-Terphenyl emission spectrum
overlaps with
POPOP absorption spectrum

Radical bulk polymerization: cell casting







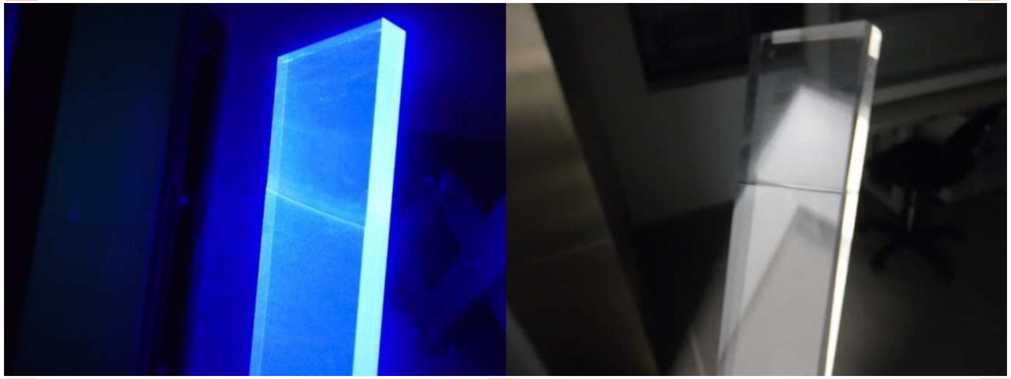
Ł. Kapłon et al., Plastic scintillators for positron emission tomography obtained by the bulk polymerization method, BAMS 10, 27 (2014)

What is inside plastic scintillators for neutron detection?

polymer
60-98%

cross-linker
0.01-5%

polymerization initiator
0.01-1%

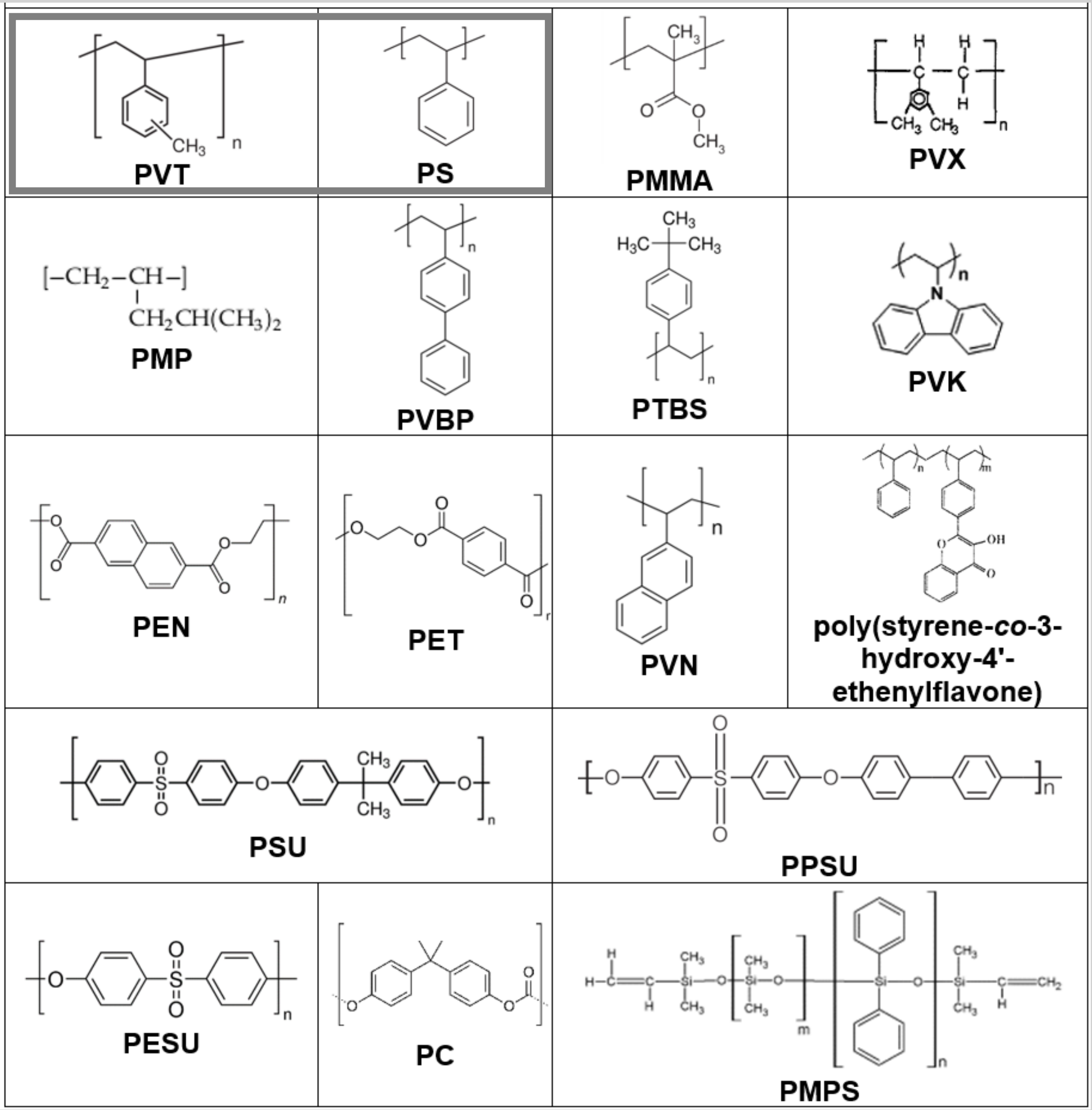


UV
fluorescent
addition
1-36%

blue
fluorescent
addition
0.03-0.2%

compound
with boron
or gadolinium
5-15%

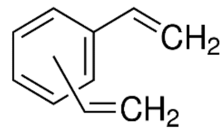
Chemical components in plastic scintillators: polymers



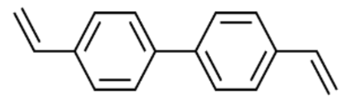
Main features:

- amorphous;
- transparent to visible light;
- contain aromatic unit;
- glass transition temperature 100-220 °C;
- density 1.00-1.36 g/cm^3;
- refractive index 1.46-1.68;
- manufactured by bulk polymerization.

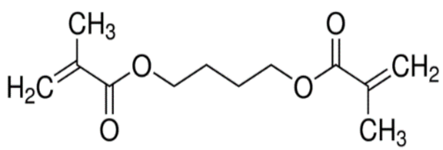
Chemical components in plastic scintillators: cross-linkers



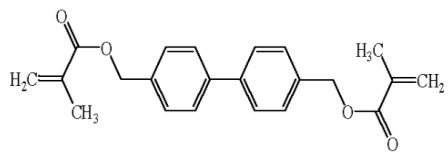
divinylbenzene



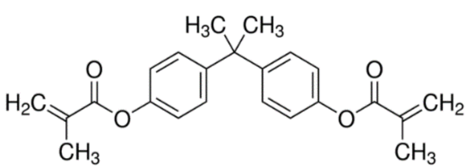
4,4-divinyl-p-biphenyl



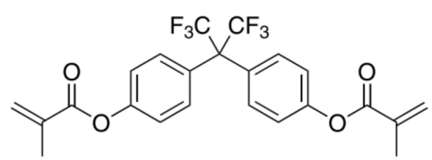
1,4-butanediol dimethacrylate



4,4-bis-methylene-2-methacrylate-biphenyl



bisphenol A dimethacrylate

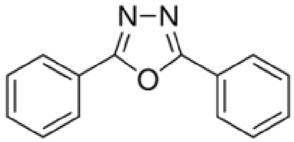
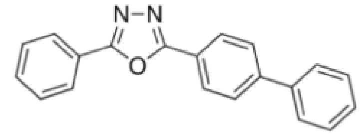
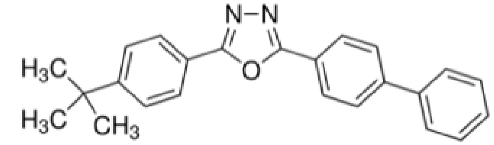
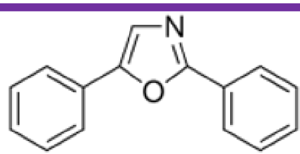
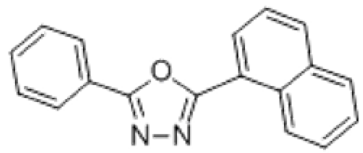
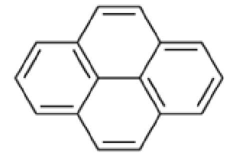
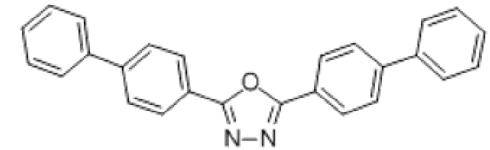
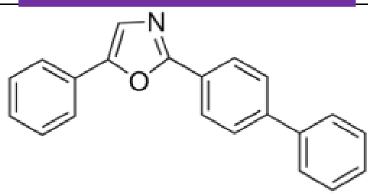
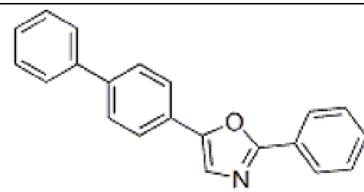
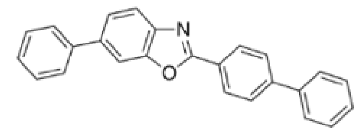


4,4’(hexafluoroisopropylidene) diphenyl dimethacrylate

Main features:

- **improves mechanical properties (hardness) of polymers;**
- **increase glass transition temperature;**
- **transparent to visible light;**
- **contain aromatic units with two double bonds.**

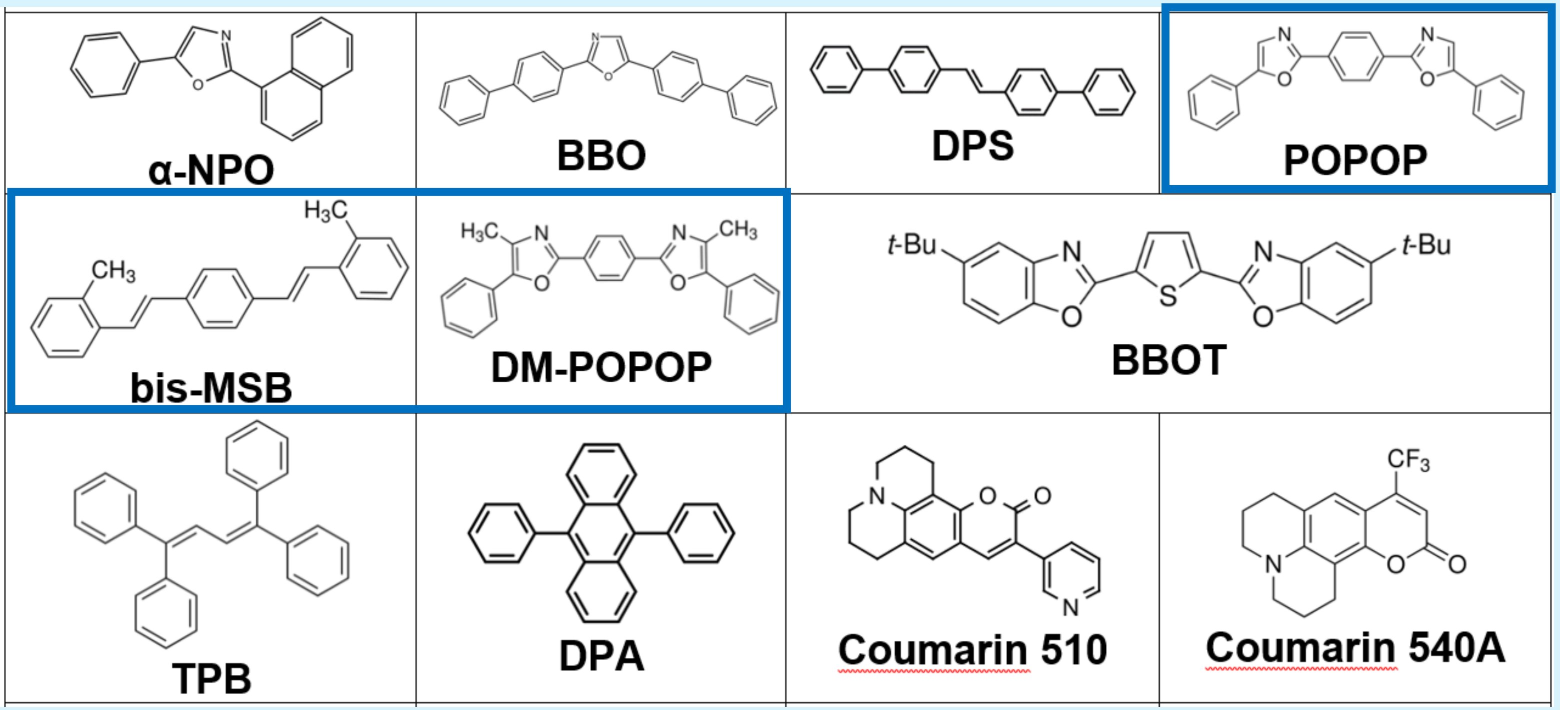
Chemical components in plastic scintillators: UV fluorescent additions

 PPD	 PTP	 PBD	 BPBD
 PPO	 α-NPD	 pyrene	 BBD
 BPO	 PBO	 PBBO	

Main features:

- contain aromatic units;
- absorb energy from polymer ~ 280-330 nm;
- emitt UV light ~ 330-400 nm;
- hight fluorescence quantum yield up to 100%;
- fast decay time near 1-2 ns;
- high solubility in monomer for high energy transfer (2-4 wt. %);
- chemical stability and temperature tolerance in polymerization process at 80-180 °C.

Chemical components in plastic scintillators: blue fluorescent additions



Main features:

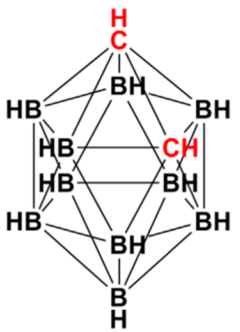
- contain aromatic units;
- absorb energy from UV additions ~ 340-370 nm;
- emitt blue light ~ 400-490 nm;
- hight fluorescence quantum yield up to 100%;
- fast decay time near 1-2 ns;
- moderate solubility in monomer for emission shift (0.03-0.2 wt. %);
- chemical stability and temperature tolerance in polymerization process at 80-180 °C.

Elements with a high cross section for thermal neutron capture

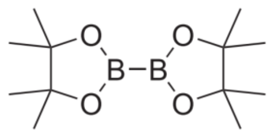
Interaction	Energy T_n	Cross section (b)	Q-value (MeV)	Products
$^{1}H(n, n')$	100 keV – 10 MeV	0.7–28	–	proton
$^{3}He(n, p)$	Thermal	5330	0.764	proton, triton
$^{10}B(n, \alpha)$	Thermal	3840	2.792	alpha, lithium ion
$^{6}Li(n, \alpha)$	Thermal	940	4.78	alpha, triton
$^{157}Gd(n, \gamma)$	Thermal	254000	7.937	photons, electrons
$^{155}Gd(n, \gamma)$	Thermal	60900	8.536	photons, electrons
$^{113}Cd(n, \gamma)$	Thermal	20600	9.04	photons, electrons

J. Dumazert et al., Nuclear Inst. and Methods in Physics Research, A 882 (2018) 53

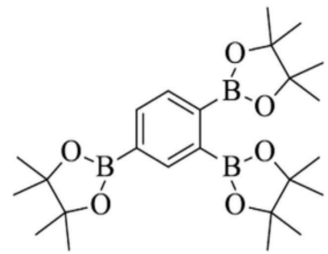
Compounds with boron for thermal neutron capture



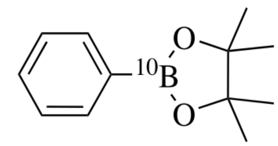
ortho-carborane



bis(pinacolato)diboron



benzene derivative with 3 boron atoms

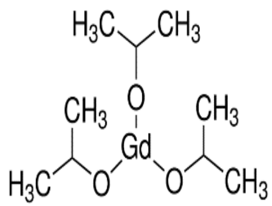


benzene derivative with 95% enriched B-10

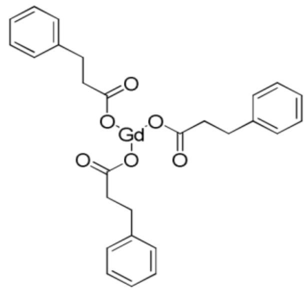
Main features:

- natural boron content in compound 7-75 atomic %;
- colorless substances, do not decrease fluorescence in scintillator;
- hight solubility in monomer (5-15 wt. %);
- maximal boron content in scintillator up to 5%;
- chemical stability and temperature tolerance in polymerization process at 80-180 °C;
- 12-30% of light output reduction compared to unloaded scintillator.

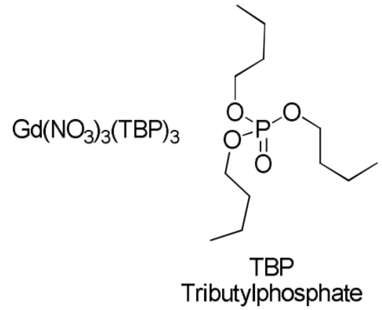
Compounds with gadolinium for thermal neutron capture



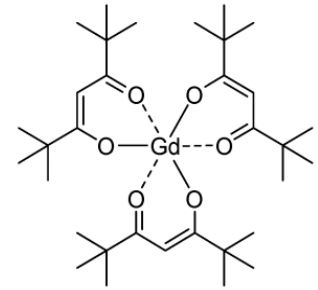
gadolinium
tris(isopropoxide)



gadolinium
phenylpropionate



gadolinium
nitrate tributylphosphate



gadolinium
tris-tetra-methylheptanedionate

Main features:

- **natural gadolinium content in compound 12-47 atomic %;**
- **colorless complexes, decrease fluorescence in scintillator;**
- **hight solubility in monomer (6-28 wt. %);**
- **maximal gadolinium content in scintillator up to 4%;**
- **chemical stability and temperature tolerance in polymerization process at 80-180 °C;**
- **18-60% of light output reduction compared to unloaded scintillator.**

Compounds with gadolinium for thermal neutron capture

	Matrix	Dopant	wt% Gd	Dimensions	Light yield
Glodo et al. [69]	$GdI_3:Ce^{3+}$	–	29 (Matrix)	10 × 20 × 1 mm	58000 ph/MeV
Czirr [71]	PVT	$Gd(Hba)_3$ (homogeneous)	0.1–0.5	2.54 (ø) × 15.24 cm	15%–42% of unloaded NE120 (0.1–0.2 wt% Gd)
Brudanin et al. [72]	PMMA	$Gd(NO_3)_3$ (homogeneous)	3	30 (ø) × 10 mm	51% of unloaded plastic scintillator
Nemchenok et al. [73]	PMMA	$GdCl_3$ (homogeneous)	4	27 (ø) × 10 mm	67.6% of unloaded plastic scintillator
Velmozhnaya et al. [74]	PS	$Gd(PhV)_3$ (homogeneous)	4	–	60% of unloaded plastic scintillator
Watanabe et al. [75]	Bisphenol A resin	Unknown dopant	0.1	20 × 200 × 3 mm	56.5% of unloaded plastic scintillator~ 700 ph/MeV
Bell et al. [76]	PVT	$Gd(NO_3)_3(TBP)_3$	1.5	2.5 (ø) × 1 cm	60% of unloaded plastic scintillator (1 wt% Gd)
Ovechnika et al. [77]	PS	$Gd[OCH(CH_3)_2]_3$ (homogeneous)	2.5	14 (ø) × 6 mm	76% of unloaded plastic scintillator
Bertrand et al. [78]	PS	$Gd(TMHD)_3$ (homogeneous)	2	16 (ø) × 8 mm	50% of unloaded plastic scintillator~ 5000 ph/MeV

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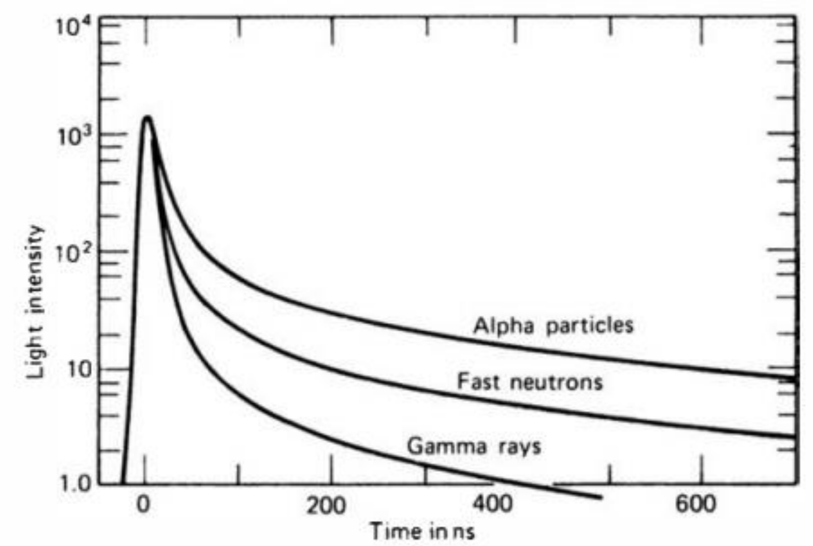
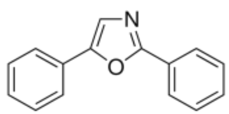
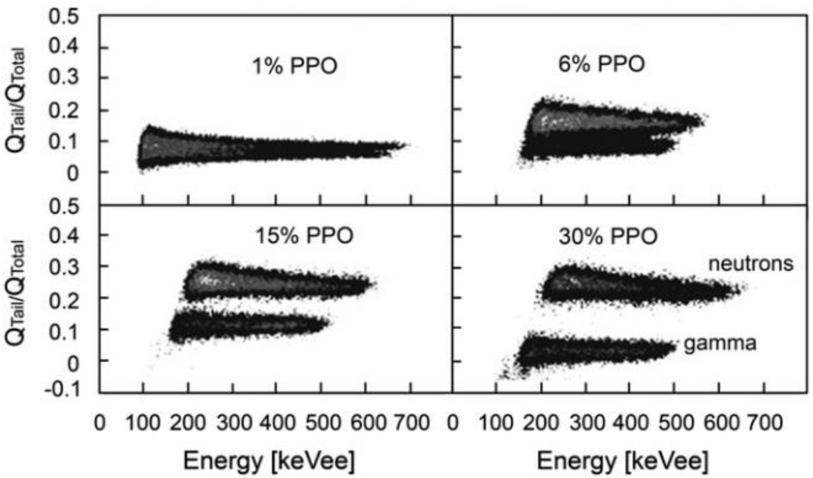
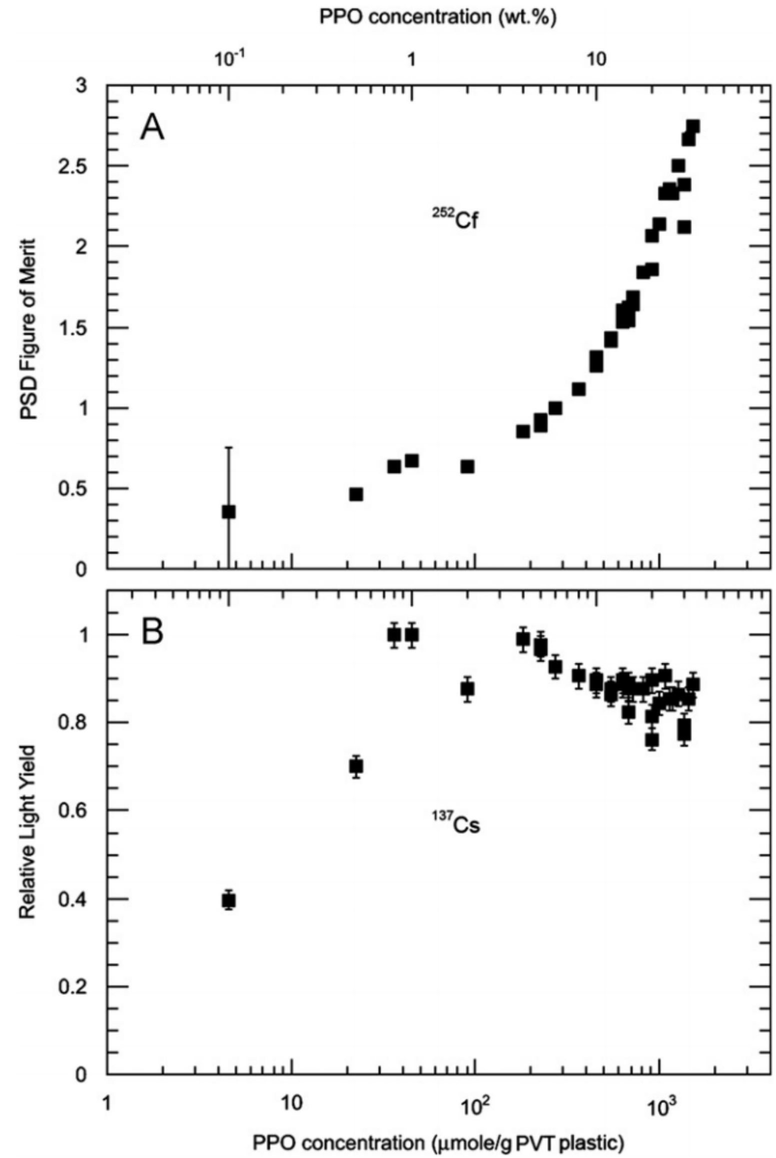
Plastic scintillators with pulse shape discrimination (PSD)









N. Zaitseva et al., Nuclear Instruments and Methods in Physics Research A 668 (2012) 88

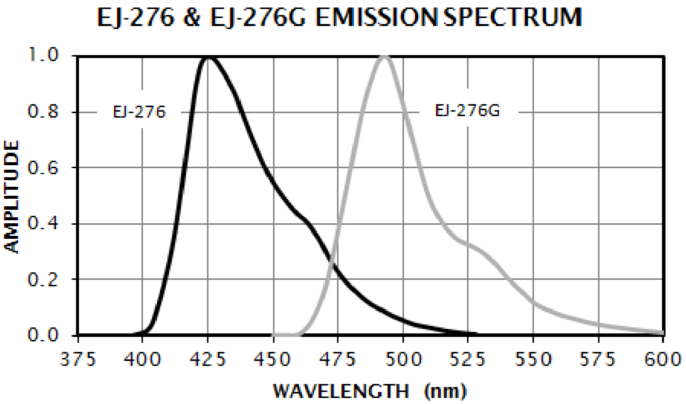
Plastic scintillators with pulse shape discrimination (PSD)

Ref.	Strategy	Biggest size (∅, h)	FOM (at a given energy) (keVee)	Scintillation yield (ph/MeV)	Decay time (ns)	Emission wavelength (nm)
[40] **b**	Organometallics	25 mm × 15 mm	1.4 (400)	7,300	800	515
[41] **b**	Organometallics	16 mm × 10 mm	1.37 (250)	n.d.	370,000	590–620
[43]	1st fluo highly concentrated	25 mm × 25 mm	n.d.	n.d.	n.d.	≈ 420 (assumption)
[44] **e**	1st fluo highly concentrated	103 mm × 114 mm	1.25 (30 × 5 mm, 200)	3,400–4,500	13	420
[45] **b**	1st fluo highly concentrated	50 mm × 50 mm	3.31 (25 × 25 mm, 480)	n.d.	n.d.	≈ 430
[48, 49]**g**	EJ-299-33	127 mm × 150 mm	0.84 (50 × 50 mm, 100)	8,600	≈ 5	420
[46]	1st fluo highly concentrated	25 mm × 15 mm	1.05 (25 × 15 mm, 300)	≈ 9,000 (relative)	n.d.	≈ 420 (assumption)
[47] **c**	1st fluo highly concentrated	390 cm^3	2.25 (50 × 50 mm, 1000)	≈ 13,000	< 10	≈ 440
[51] **c**	Stilbene single crystals in silicone	200 mm × 20 mm	1.00 (500)	n.d.	4.5 (assumption)	410 (assumption)
[52] **b**	*p*-T or stilbene single crystals in Sylgard	50 mm × 50 mm	1.41 (*p*-T, 600) 1.19 (stilbene, 600)	≈ 9,900 (*p*-T) ≈ 5,700 (stilbene)	n.d.	420 (*p*-T); 395 (stilbene)
[55]	Ionic liquids	Micrometers	n.d.	n.d.	< 50 (assumption)	≈ 380 (assumption)

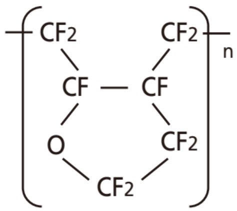
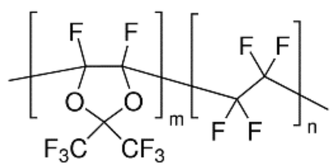
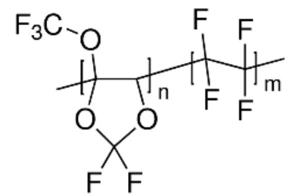
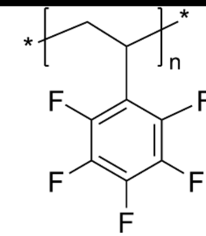
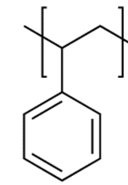
G. Bertrand et al., Nuclear Instruments and Methods in Physics Research A 776 (2015) 114

Commercial plastic scintillators with boron and PSD

Company, PS name	Light Output [% anthracene]	Wavelength of Max. Emission [nm]	Decay Time [ns]	Density [g/cm^3]	Refractive Index	Polymer Base and Softening Point [°C]
Eljen Technology EJ-276	56	425	13	1.096	1.60	-
Eljen Technology EJ-276G	52	490	13	1.096	1.57	-
Eljen Technology EJ-254 (5% B)	48	425	1.5	1.026	1.58	PVT 75
Eljen Technology EJ-254 (2.5% B)	56	425	1.5	1.023	1.58	PVT 75
Eljen Technology EJ-254 (1% B)	60	425	1.5	1.021	1.58	PVT 75
Saint-Gobain BC-454 (5% B)	48	425	2.2	1.026	1.58	PVT 60



Polymers with fluorine for fast neutron detection

Polymer	Cytop S type	Teflon AF 1600	Hyflon AD 40	Polypentafluorostyrene	Polystyrene
Molecular formula					
Density [g/cm³]	2.03	1.78	1.98	1.55	1.06
Glass transition temperature [°C]	108	160	90	107	100
Refractive index	1.34	1.31	1.33	1.48	1.59
Price [EUR/g]	43	98	160	2.5	0.02

Threshold activation detection (TAD) technique:
neutron interaction with ^{19}F leads either to ^{16}N or ^{19}O that undergo β^- and gamma ray decays with half-lives of 7.1 s and 26.9 s, respectively.

Diameter (mm)	32.9
Thickness (mm)	3.6
Weight (g)	4.741
Density (g/cm³)	1.55
λ_{max}^{em} (nm)	416
$\lambda_{max}^{radiolum}$ (nm)	425
Fluorine content (atoms/cm³)	3.73×10^{22}
Hydrogen content (atoms/cm³)	2.24×10^{22}
F/H ratio	1.66
Number of photoelectrons (N_{phe}/MeV)	870 ± 50
Light output (ph/MeV)	3100 ± 300
Light output ratio $^{241}Am/^{137}Cs$ (%)	76 ± 3
Decay time (ns)	3.0 ± 0.3

M. Hamel, P. Sibczynski et al., Nuclear Instruments and Methods in Physics Research A 768 (2014) 26

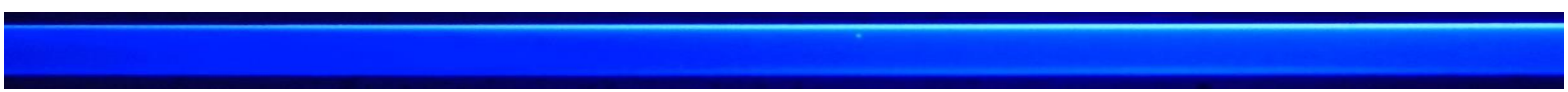
Summary

- Plastic scintillators based on PS and PVT possess the best properties and are commercially avialible at low price: 200-1000 USD/kg.
- Scintillators can be tuned by changing fluorescent substances: influence on light output, decay time and emission spectra.
- Modification of plastic scintillators by adding boron or gadolinium compounds or overdoping fluorescent additions can increase neutron detection capabilities.
- Possible ways of improving scintillators properties:

  - design of a new fluorescent additions: organic chemistry;

  - adapting chemical compounds from other fields like LED and OLED devices;

  - research on different polymers.



Thank you