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Tuning the decay time of liquid scintillators

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Abstract

The choice of an appropriate viscosity of a liquid scintillator solvent can lead to the fine tuning of its decay time. We present herein the photophysical and scintillation properties of liquid scintillators containing pyrene and 9,10-diphenylanthracene as the primary and secondary fluorophores, respectively, being dissolved either in toluene, diisopropyl naphthalene or mixtures of these two solvents. By modifying the dynamic viscosity, the photoluminescence decay time can be easily chosen in the range 40 – 120 ns. For all liquid scintillators the light yields were satisfying, and the scintillation decays were slightly different, ranging from 36 up to 125 ns.

Keywords

Liquid scintillator; Pyrene; TCSPC; Radiation detection

1. Introduction

Thanks to their carbon-based nature, organic – and in the context of this document, liquid – scintillators can be chemically fully modified, provided the fact they keep their scintillation efficiency.¹ The concept of liquid scintillator appeared in 1950 in two seminal papers that were published nearly at the same time.^{2,3} Immediate interest was granted from the community⁴ and the next decades have seen the positioning of liquid scintillators (LSs) at the central place of radiation detectors with numerous applications, from fundamental physics research (such as Borexino⁵ or SNO+⁶), metrology of the activity,⁷ dosimetry,⁸ environmental assays⁹ and various R&D activities looking at new, promising media.¹⁰ Five different advantages of LSs can be highlighted:

1. the possibility to perform pulse shape discrimination of particles displaying different dE/dx ratios,
2. the detection of beta-emitters with very low energy such as ³H,
3. the possibility to load the scintillator with various elements from the periodic table,
4. their radiochemical purity which is paramount for the detection of rare radioactive decays,

38 5. an outstanding access up to cubic meter volumes which is particularly suitable for large
39 experiments where almost no other radiation detector can compete in terms of effective size.

40 Unfortunately, LSs do not represent the Holy Grail in terms of detectors. Since they are composed of
41 hydrogenocarbon molecules, they are not suitable for gamma spectroscopy due to their low effective
42 atomic number, unless they are loaded with heavy elements (although this can be advantageous when
43 performing beta counting in the presence of high energy gamma rays). They are also tricky to handle
44 and depending on the nature of their solvent they can be hazardous or even toxic at some point. Above
45 all, oxygen is a very efficient quencher of the scintillation light,¹¹ which imposes to use LSs constantly
46 degassed and maintained or encapsulated under neutral atmosphere to have the best detection
47 efficiency.

48 Chemically speaking, a liquid scintillator is a formulation of one or several fluorescent molecules that
49 are dissolved in an organic solvent. Eventually, some additives can be added such as organometallics
50 or chelators. All these stages have been extensively studied in the past. Since the solvent is the main
51 component of the scintillator, several developments have been performed regarding the effect of
52 impurities, substituting hydrogen with deuterium for neutron detection, and an important work has
53 been performed on the quest for safer liquid scintillators so as to replace small organic molecules such
54 as xylenes or toluene by linear alkyl benzene (LAB) or diisopropylnaphthalene (DIN). This allows
55 scintillators with less toxicity, higher flash point and eventually biodegradability feature. The solvent is
56 also the medium where the initial radiation/matter interaction happens, hence the start of the
57 scintillation process. Not to reduce to a small piece their influence and the huge work on their choice
58 and modifications, primary and secondary fluorophores have less impact on the scintillation efficiency,
59 as they will mainly govern the photophysical properties such as emission wavelength and decay time
60 provided the fact that their concentration is adequate in the solvent. Particularly efficient molecules
61 are for instance 2,5-diphenyloxazole (PPO) and 1,4-bis(5-phenyl-2-oxazolyl)benzene (POPOP), which
62 act as primary and secondary fluorophores, respectively.

63 Usually, the main decay time of LSs (in principle the fastest component when the decay is double- or
64 even multi-exponentially fitted) falls within the range 1 – 10 ns.^{12,13} Such timing resolution is very fast
65 so one has to adapt the electronics that are dedicated to the signal processing for a good digitization
66 of the fast pulse. Various research programs were conducted to accelerate this decay time for fast
67 timing applications, leading to ultra-fast liquids,¹⁴ but to the best of our knowledge extending the decay
68 time to longer values has only been barely studied.¹⁵ Here in 15 all the liquid scintillators that have
69 been tested are from Nuclear Enterprises and their formulation is proprietary. On the basis of the
70 Authors' work, only three liquids display decay times higher than 10 ns: NE 321 (15.70 ns), NE 227
71 (17.02 ns) and NE 317 (20.21 ns). NE 321 is a boron-loaded LS presumably based on 4-
72 isopropylbiphenyl but the fluorophores are not revealed,¹⁶ and the two other scintillators are still
73 unknown to us in composition. In this context and following our preliminary studies with the extension
74 of the decay time of plastic scintillators,¹⁷ we propose herein several new formulations or possibilities
75 that allow tuning the decay time of scintillating liquid in a range between 36 and 125 ns.

76

77 2. Experimental

78 Pyrene was purchased from Sigma-Aldrich and was carefully purified by sublimation prior to use. 9,10-
79 diphenylanthracene was purchased from Alfa AesarTM and used without purification. Spectroscopic
80 toluene was purchased from Carlo Erba and used as received. Diisopropylnaphthalene (DIN) was
81 purchased from Perkin-Elmer, was purified on silica gel chromatography and kept in the dark prior to

82 use. BC-501A was purchased from Saint-Gobain Crystals and Detectors. Prior to each experiment, the
83 liquid scintillator was carefully saturated with argon during 8 minutes.

84 Fluorescence spectra were recorded at 90° angle with a Horiba Jobin Yvon Fluoromax-4P device,
85 monitored with FluorEssence software. The photoluminescence decay characteristics of the liquid
86 scintillators were investigated with the Time-Correlated Single Photon Counting (TCSPC) module
87 available on the Fluoromax-4P spectrofluorometer, with an excitation provided by a laser diode
88 operating at 274 nm which allows exciting the entire formulation of the scintillator. Another NanoLED
89 emitting at 368 nm was also used to probe the secondary fluorophore only. Decay spectra were fitted
90 using DAS6 software (Horiba Jobin Yvon), the quality of the fit was determined so that the χ^2 value was
91 less than 1.30.

92 To record scintillation properties, a scintillation vial containing 15 cm³ of the fully degassed liquid
93 sample was placed in the center of a white chamber which was connected to a Hamamatsu R6231-100
94 photomultiplier tube operating at +1400 V. A ¹³⁷Cs source (530 kBq, 02/15/2016) was used to excite
95 the scintillator. Pulses coming from the PMT were sorted and treated with a CAEN DT5730B electronic
96 board during an acquisition time of 20 minutes. The high voltage was kept constant for all tested
97 scintillators. Determination of the Compton edge (CE) position was evaluated from the top of the
98 Compton edge with a relative uncertainty of 5% of its channel value. The relative scintillation yield was
99 calculated using a reference spectrum (BC-501A sample), whose scintillation yield is given as
100 12,000 ph/MeV.¹⁸ This comparison is possible due to the fact that the tested scintillators emit in the
101 same wavelength range as the reference material. By rule of thumb, the light yield of the prepared
102 plastic scintillator sample is determined using this equation:

$$103 \quad R_{sample} = R_{BC-501A} \times V_{CE,sample} / V_{CE,BC-501A}$$

104 where R is the scintillation yield, and V is the channel of the Compton edge position. The response
105 linearity evaluation was performed by plotting the Compton edge position against the Compton edge
106 energy connected with the corresponding gamma ray emitter. For the energy calibration, the following
107 gamma-emitting sources with activities were used: ²⁴¹Am (1.7 MBq, 06/13/2016), ¹³⁷Cs (530 kBq,
108 02/15/2016), ⁶⁰Co (101 kBq, 05/22/2018), ²²Na (1.28 MBq, 05/22/2018). The same strict experimental
109 setup was used as for light yield evaluation.

110 Scintillation pulses were recorded using the above-mentioned ⁶⁰Co source which was used for the
111 excitation, and the setup was identical as before as well, except that the CAEN digitizer was replaced
112 by a digital oscilloscope LeCroy wavepro 7300A operating at 500 MS/s. 5,000 pulses were collected
113 and averaged, inverted (since the pulses incoming from the photomultiplier tube are negative), and
114 the resulting pulse was fitted with the same DAS6 software used for TCSPC.

115

116 3. Results and discussion

117 Photophysical properties

118 Decay time characterization of liquid scintillators has been the main subject of numerous
119 publications.^{12,15,19,20} As already mentioned, having access to long-lived (*i.e.* 10 – 100 ns)
120 photoluminescence decay times in liquid media is not straightforward. First, the choice of organic,
121 simple molecules able to decay with lifetimes higher than 10 ns is rather narrow. In the context of our
122 studies, naphthalene and pyrene have been chosen thanks for their decay, but also for their efficiency
123 as scintillating molecules. A recent paper from Biller et al. introduced the use of acenaphthene as slow
124 fluorophore in linear alkyl benzene for neutrinoless double beta decay,²¹ so this might be another
125 molecule choice. Besides, one has to keep in mind the low cost of a new liquid scintillator, so complex

126 molecules are to be avoided, or at least for a first approach study. Here, naphthalene will not be
127 presented and acenaphthene has not been tried. In the world of photoluminescence, pyrene is one of
128 the most exciting fluorophore thanks to several of its prominent photophysical characteristics: the
129 fluorescence spectrum strong dependence on its chemical environment,²² a long decay time and an
130 excited pyrene emission vs. excimer emission.²³ It seems that pyrene in polystyrene appeared as early
131 as 1953,²⁴ but J. B. Birks was the first scientist that extensively studied this molecule in the field of
132 organic scintillation counting.²⁵ Almost sixty years later, the pyrene moiety is still under interest in the
133 scintillation domain.²⁶

134 Since the mobility of pyrene can affect its decay time, we decided to check the influence of the viscosity
135 of the solvent upon its photoluminescence. In other words, mixing a viscous, scintillation-grade solvent
136 to another fluid, scintillating solvent would afford liquid scintillators with various decay times. Mixing
137 at least two solvents to prepare liquid scintillators is somehow peculiar in this field. Mixed solvent such
138 as linear alkyl benzene and pseudocumene have already been described, here to benefit from
139 advantages of each solvent to some extent.²⁷ Although not related to the scintillation field, Wolff et al.
140 have noticed that adding viscous solvent (which is in this case diisopropylnaphthalene – DIN) to a fluid
141 solution containing 9,10-dimethylantracene leads to increasing its apparent decay time from 12 up
142 to 30 ns.²⁸ Other applications have already studied the viscosity dependence on the fluorescence
143 lifetime, such as in the field of molecular rotors,^{29,30} and protein fluorescence kinetics.³¹ Interestingly
144 in 29 the Authors list fluorophores that are sensitive to viscosity and/or temperature. Table 1 gives an
145 overview of various solvents that are suitable for liquid scintillation counting, along with their dynamic
146 viscosity given at room temperature. This list is sorted from more fluid (toluene) to more viscous
147 solvents, and the two limits thus constitute our choice in the preparation of the scintillating mixtures.
148 Obviously more viscous solvents than DIN exist but the choice was restricted to scintillating-grade
149 solvents only.

150 The abbreviation of DIN as reported in many publications may be the cause of several mistakes due to
151 the number of isomers of diisopropylnaphthalene. Herein we talk about the DIN which is currently
152 used in several commercial LS cocktails: it is in fact a mixture of isomers and is registered under the
153 CAS [38640-62-9]. Its dynamic viscosity at 20 °C is equal to 12.67.³²

154

155 *Table 1 Dynamic viscosities for selected scintillation-grade solvents at 25 °C (unless otherwise stated)*

Solvent	Dynamic viscosity η (mPa.s)
Toluene	0.59
<i>p</i> -Xylene	0.65
Benzene	0.66
<i>o</i> -Xylene	0.81
Pseudocumene	0.9
Linear alkyl benzene	4.00
PXE	≈ 6.8
DIN	12.67 (at 20 °C)

156

157 The two viscosity limits being established, we decided to study the photoluminescence decay
158 dependence of scintillators in this range. Pyrene and 9,10-diphenylantracene (9,10-DPA) were chosen
159 as the fluorescent probes, since they are highly referenced in the scintillation field and they are often
160 matched. Thus, toluene-based or DIN-based liquid scintillators were prepared from the appropriate
161 solvent containing pyrene and 9,10-diphenylantracene. From these stock solutions, combinations
162 were performed from 100% toluene-based LS to 100% DIN-based LS. Starting with the more fluid

163 solvent (toluene, $\eta = 0.59$ mPa.s at 25 °C) to the more viscous solvent (DIN, $\eta = 12.67$ mPa.s at 20 °C),
164 the viscosity of solvents mixture is given by:

$$165 \quad \text{Log}(\eta_{\text{mixture}}) = X_{\text{toluene}} \times \text{Log}(\eta_{\text{toluene}}) + X_{\text{DIN}} \times \text{Log}(\eta_{\text{DIN}})$$

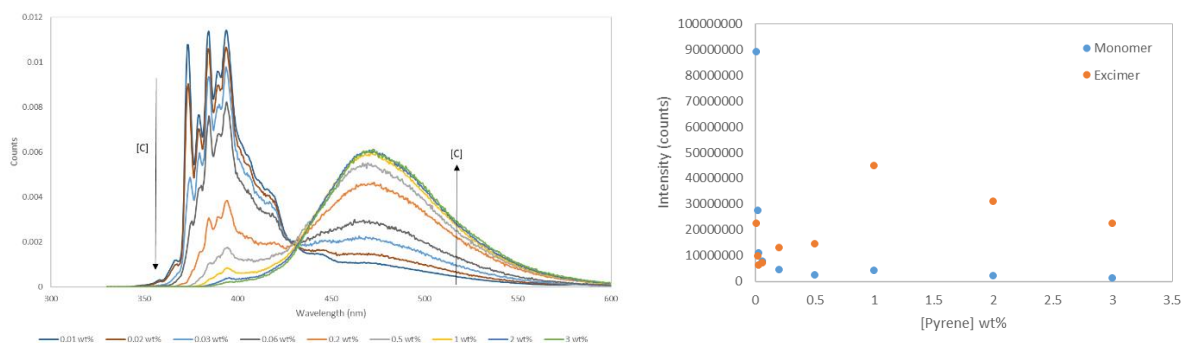
166 where X_i is the molar fractions of molecules and η_i the dynamic viscosity of the solvent i .

167 Noteworthy, the decay shape of the couple between pyrene in its excimer form and 9,10-
168 diphenylanthracene against the viscosity of the solvent has already been studied,³³ where the two
169 combined solvents were benzene and cyclohexanol. The dynamic viscosity of cyclohexanol is very high:
170 56 mPa.s at 25 °C. However, it is not a good solvent for scintillation counting.

171 Then the appropriate quantity of dyes giving the scintillator the best scintillation properties was
172 determined. No optimization was performed on 9,10-DPA in this study, and a typical 0.03 wt%
173 concentration was considered. Also, the same concentrations of dyes were used for all mixtures of
174 solvents. Pyrene has a very strong luminescence dependence with its concentration, both in terms of
175 intensity and emission wavelength. It is noteworthy the rather unusual, low concentration of pyrene
176 as the primary fluorophore we have chosen. At a glance, the pyrene in its excimer form would be the
177 best choice since its emission is centered around 480 nm, an emission wavelength which is interesting
178 for the purpose of scintillation counting. As such, pyrene under its dimer form could be used as the
179 only solute in a liquid scintillator, which means with no need of secondary fluorophore.³⁴ Thus, toluene
180 containing 3 wt% of finely sublimed pyrene was tested as a binary liquid scintillator. This solution
181 revealed good scintillating properties but a long decay time was never reached. The result of the pulse
182 height spectrum when excited with gamma rays of ¹³⁷Cs gave a light yield of $\approx 6,500$ ph/MeV (relative
183 to BC-501A), but the monoexponential fit of the decay time was found to be 50 ns.

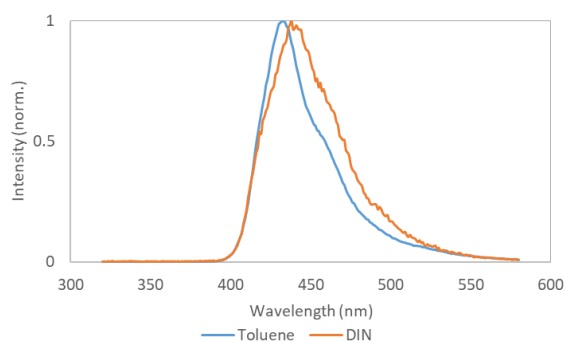
184 So we decided to focus on the photoluminescence properties of excited pyrene in its monomer form
185 (therefore at low concentration), and 9,10-DPA is associated to afford a ternary liquid scintillator.
186 Figure 1 left resumes the photoluminescence spectrum of pyrene in toluene at various concentrations.
187 As already depicted, the pyrene excimer starts to be predominant at rather low concentrations for a
188 fluorophore usually used for scintillation. The isobestic point between the monomer and the dimer
189 area-normalized emissions is clearly visible at 432 nm on Figure 1 left. Even if the excimer's
190 luminescence is still visible, we decided to choose the pyrene weight concentration at 0.03 wt%, which
191 is $1.29 \cdot 10^{-3}$ mol.L⁻¹. To get rid of all excimer emission, various pyrene concentrations are reported:
192 from 10^{-3} mol.L⁻¹³⁵ or lower than $5 \cdot 10^{-5}$ mol.L⁻¹.³⁶ This last concentration would be too low for an
193 appropriate use in scintillation counting. With the same excitation and emission parameters, Figure 1
194 right shows the resulting fluorescence integrals against the pyrene concentration. Interestingly, there
195 is a decrease of the fluorescence intensity when the two species are present in solution, then the
196 intensity rises, with highly diluted 0.01 wt% solution being the brightest among the others. However,
197 we preferred to keep the concentration of pyrene at 0.03 wt%, where the integral of the monomer
198 emission is 1.7 times higher than the excimer's. Figure 2 shows the photoluminescence difference
199 between toluene-based and DIN-based pyrene + 9,10-DPA solutions at the same concentration. The
200 respective emission maxima are 432 and 428 nm, a difference which is not significant for scintillation
201 counting, given the fact that standard photomultiplier tubes have almost the same quantum efficiency
202 for both 432 and 428 nm wavelengths. This figure also shows that the viscosity influence on the LSs
203 decay time is not due to an increase part of excimer but a true monomolecular emission.

204



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Figure 1. Left: area-normalized photoluminescence dependence of pyrene with concentration. Solvent: toluene; $\lambda_{ex} = 310$ nm, same experimental parameters for all. Right: integral of the photoluminescence curves with pyrene concentration. The delimitation to calculate monomer or excimer intensities is defined at the isobestic point.



209
210
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Figure 2. Normalized fluorescence spectra of toluene-based and DIN-based liquid scintillators containing pyrene and 9,10-DPA, both at concentrations 0.03 wt% ($\lambda_{ex} = 300$ nm).

212

213 Then the mixtures of solvents containing 0.03 wt% of pyrene and the same concentration of 9,10-
214 diphenylanthracene were evaluated in terms of Time-Correlated Single Photon Counting,
215 photoluminescence spectroscopy and gamma-ray pulse height spectrum. For the TCSPC recordings, a
216 274 nm NanoLED from Horiba Jobin Yvon was used to excite the liquid scintillator. Figure 3 reports the
217 mean decay values of the biexponential fits against the viscosity of the mixtures of solvents containing
218 the two fluorophores. The mean decay values are calculated according to: $\langle \tau \rangle = \tau_{fast} \times \%_{fast} + \tau_{slow} \times$
219 $\%_{slow}$. Table 2 gives the full details of the decay fitting. A linear dependence of the mean decay time on
220 the viscosity of the solvent combination can be observed. BC-501A is added on the last line of Table 2
221 for comparison sake: it is a reference liquid scintillator from Saint-Gobain Crystals and Detectors. Its
222 data has been added since it will be useful later on for the scintillation performance characterization.
223 Here as well, a biexponential fitting is privileged, with the values which are of the same order as
224 previously reported.³⁷

225

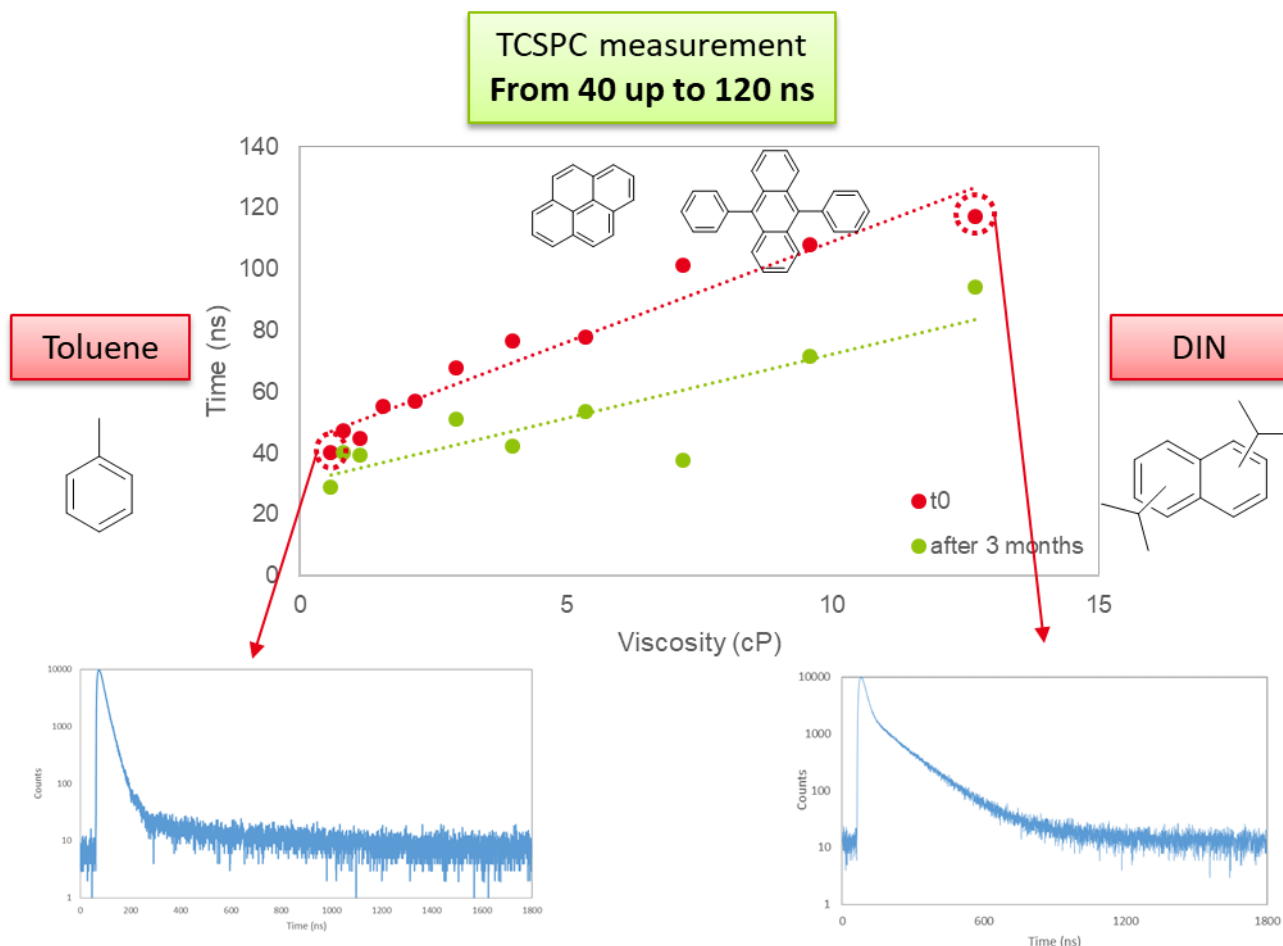
226 Table 2 Full data of the biexponential fits of the decays for various formulations. BC-501A is added. $\lambda_{ex} = 274$ nm, $\tau_{obs} =$
227 430 nm. The decay times given in the table are governed by pyrene only.

Solvent mixture (%) + pyrene & DPA at 0.03 wt%	Viscosity η (mPa.s)	Biexponential fitting $\tau_{fast} (\%_{fast}) + \tau_{slow} (\%_{slow})$	Mean value $\langle \tau \rangle$ (ns)	χ^2
PhMe 100 – DIN 0	0.59	16.1 (16.8) + 45.2 (83.2)	40.3	1.18
PhMe 90 – DIN 10	0.82	40.1 (88.8) + 105.9 (11.2)	47.5	1.21
PhMe 80 – DIN 20	1.14	37.0 (86.5) + 95.1 (13.5)	44.8	1.29
PhMe 70 – DIN 30	1.58	25.4 (21.4) + 63.5 (78.6)	55.3	1.09
PhMe 60 – DIN 40	2.16	22.8 (18.8) + 64.8 (81.2)	56.9	1.08

PhMe 50 – DIN 50	2.95	24.3 (20.0) + 78.5 (80.0)	67.7	1.19
PhMe 40 – DIN 60	3.99	21.1 (16.8) + 87.6 (83.2)	76.5	1.19
PhMe 30 – DIN 70	5.37	19.6 (14.2) + 87.5 (85.8)	77.9	1.19
PhMe 20 – DIN 80	7.19	18.4 (13.4) + 114.4 (86.6)	101.5	1.05
PhMe 10 – DIN 90	9.57	17.1 (12.2) + 120.9 (87.8)	108.2	1.06
PhMe 0 – DIN 100	12.67	18.5 (21.6) + 144.6 (78.4)	117.4	1.20
BC-501A	n.d.	2.72 (87.3) + 23.7 (12.7)	5.4	1.16

228 n.d.: not determined.

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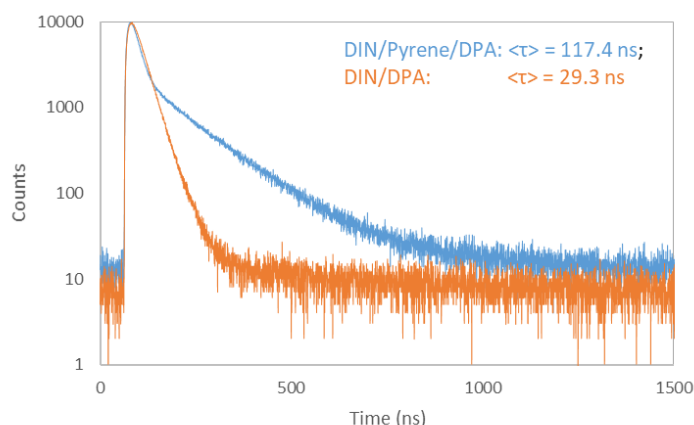
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231 *Figure 3. Mean decay value of pyrene and 9,10-DPA in mixtures of solvents, according to the viscosity. $\lambda_{ex} = 274 \text{ nm}$; $\lambda_{obs} =$*
 232 *430 nm. Time evolution at 3 months of PhMe 70 – DIN 30 and PhMe 60 – DIN 40 were not recorded.*

233

234 The observed decay times given in Table 2 point at a dual emissive system. As the primary fluorophore
 235 (pyrene) and secondary fluorophore (DPA) are present at almost the same concentrations, two
 236 emission pathways should be considered. The direct excitation of DPA gives the fast components and
 237 the classic cascade of pyrene toward DPA energy transfer gives the slow component. The decay time
 238 variation shows steady fast component which makes the slow component the only responsible for the
 239 decay tuning. This exclude non-specific quenching, which should have an effect on all the components.
 240 It implies that either, specific molecular quenching, pyrene exciton dynamics and/or pyrene to DPA
 241 energy transfers are the sources of the decay variation. A preliminary conclusion is that the variation
 242 is governed only by the pyrene behavior in a solvent with changing viscosity.

243



244

245 *Figure 4. TCSPC of DIN-based liquid scintillator including either pyrene and DPA (blue) or DPA only (orange). $\lambda_{ex} = 274 \text{ nm}$, λ_{obs}*
 246 *= 430 nm.*

247

248 It is also noteworthy that our values do not correspond to the reported decay value of excited pyrene
 249 monomer in organic solvent at room temperature. In a binary mixture of pyrene in DIN, a 282 ns decay
 250 time was observed, in agreement with literature, so adding DPA to the solution prevents keeping this
 251 decay time.³⁸ Also, this decay is highly sensitive to oxygen quenching and might be an issue for our
 252 scintillators. Here the purpose of our work is to prepare liquid scintillator cocktails having in mind these
 253 formulations could be prepared by nuclear physicists, so without easy access to chemical facilities
 254 allowing low oxygen content within the scintillator. Table 3 shows two things. First is the reproducibility
 255 of the bubbling procedure. It is clearly seen the possibility to reuse the same scintillating solution after
 256 a while, simply by bubbling once again with neutral gas. However, the same (slowest) scintillator gives
 257 mean decay fittings with a pretty wide variation: from 102 ns up to 142 ns, related with the “quality”
 258 of the bubbling step. Second is the evolution of the TCSPC with time for the fastest and the slowest
 259 formulations and Table 3 gives the complete dataset. At t_0 both scintillators were carefully saturated
 260 with neutral gas, the scintillator vial was sealed with Parafilm® then the decay values were recorded
 261 from time to time. For the DIN-based liquid scintillator, a TCSPC recording was performed each minute
 262 during the first twenty minutes and virtually no variation was observed. After four days, the decay was
 263 decreased to about 30 % the initial value, but this (small) decrease leads way enough time for
 264 radioactive assays. The results we show herein are also the most penalizing ones, since on another try
 265 performed on a 100 days’ timeline, the decrease of the decay was 34 % only (94 ns instead of 142 ns –
 266 bubbling #2). The results of decay time evolution after 100 days for almost all scintillators is shown in
 267 Figure 3. All the scintillators’ decay decrease, but the slope is not the same as for the neat materials.
 268 Scintillators with fastest decays are less affected than slowest.

269

270 *Table 3 Evolution of the decay time of two liquid scintillators with time. $\lambda_{ex} = 274 \text{ nm}$, $\lambda_{obs} = 430 \text{ nm}$.*

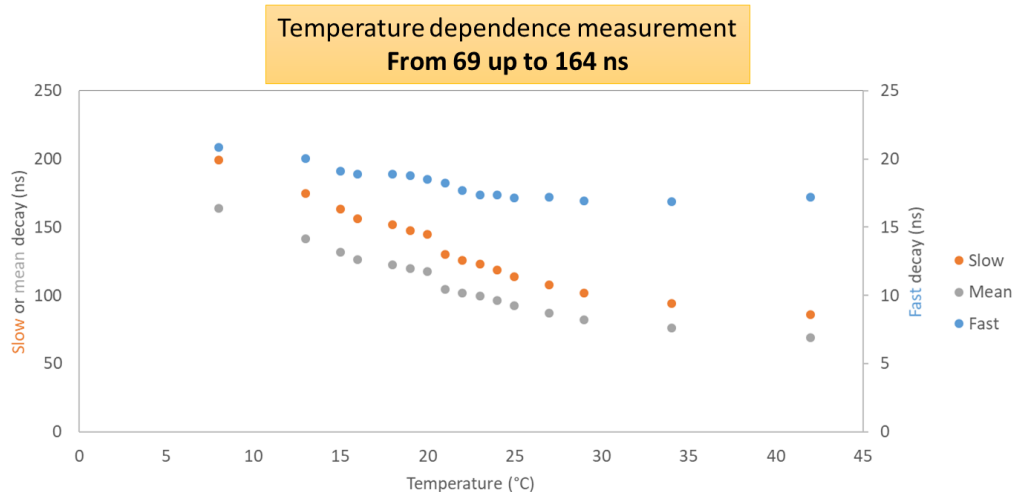
Solvent mixture (%) + pyrene & DPA at 0.03 wt%	Time of recording	Biexponential fitting	Mean value $\langle \tau \rangle$ (ns)	χ^2
		τ_{fast} (%fast) + τ_{slow} (%slow)		
PhMe 100 – DIN 0	t_0	16.1 (16.8) + 45.2 (83.2)	40.3	1.18
	$t_0 + 1 \text{ day}$	16.7 (31.1) + 40.1 (68.9)	32.8	1.15
PhMe 0 – DIN 100	Bubbling #1	18.3 (25.9) + 131.6 (74.1)	102.3	1.14
	Bubbling #2	19.4 (17.8) + 169.3 (82.2)	142.6	1.14
	Bubbling #3	18.5 (21.6) + 144.6 (78.4)	117.4	1.20
	Bubbling #4	17.8 (22.4) + 132.2 (77.6)	106.6	1.19
	Bubbling #5 – t_0	18.1 (23.3) + 136.6 (76.7)	108.9	1.16

$t_0 + 20 \text{ min}$	18.1 (23.4) + 135.3 (76.6)	107.8	1.14
$t_0 + 2 \text{ hours}$	17.9 (24.5) + 123.7 (75.5)	97.8	1.20
$t_0 + 1 \text{ day}$	15.1 (30.9) + 95.5 (69.1)	70.7	1.23
$t_0 + 2 \text{ days}$	16.2 (29.4) + 92.9 (70.6)	70.3	1.11
$t_0 + 3 \text{ days}$	16.2 (31.4) + 90.2 (68.6)	67.0	1.13
$t_0 + 4 \text{ days}$	16.2 (27.9) + 93.7 (72.1)	72.0	1.21

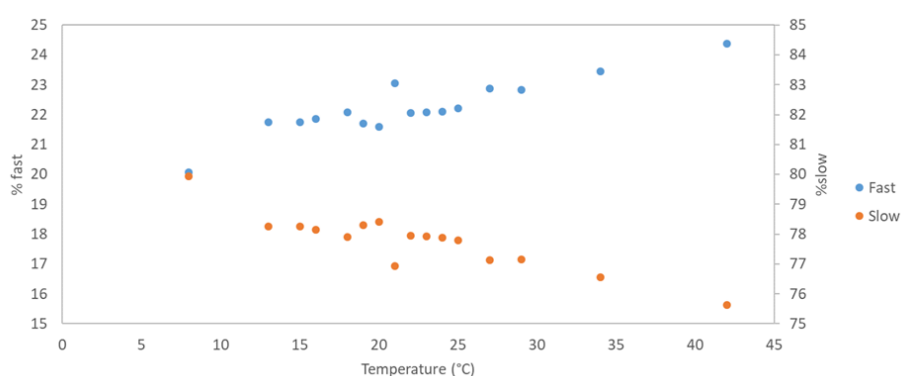
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272 Figure 5 shows the impact of temperature on the decay time of the longest-decaying liquid mixture,
 273 with temperatures ranging from 8 up to 42 °C. Since the fitting of the TCSPC curves is biexponential, it
 274 is represented in Figure 5 with fast component (blue), slow component (orange) and the mean value
 275 τ_{mean} (grey). The mean component is based on the relative weight of each components. By changing
 276 the temperature in this range, the mean value of the decay time spans from 163.6 ns down to 69.2 ns,
 277 so almost 100 ns can be covered just by adjusting the temperature of the sample. It is interesting to
 278 see that only the slow component of the photoluminescence is actually influenced by the temperature
 279 variation. Considering the whole dataset, when the mean value of τ_{fast} is 18.2 ns with a standard
 280 deviation of 1.2 ns, the mean value of τ_{slow} is 133.6 ns with a standard deviation of 31.0 ns. This
 281 information is critical for the detection of radionuclides, especially for choosing the best coincidence
 282 parameters or to insert it in a physical model. This difference of behavior of the slow and fast
 283 components fit with the explanation of the fast components coming from the direct excitation of the
 284 DPA and the slow components from the pyrene + DPA system. The strong variation of the slow
 285 components suggest that the pyrene is the one being the most influenced by the temperature changes.
 286 Several hypotheses can be made on the cause of this temperature dependence and we will try to
 287 narrow it down. Figure 5 bottom shows that the relative weight of the slow and fast components are
 288 behaving as expected, meaning the higher the temperature the more fast transition you get. However
 289 going in the right direction the range of this phenomenon variation is not enough to explain the 100 ns
 290 difference between high and low temperature decay time. An explanation based only on molecule
 291 specific quenching is thus impossible and other effect must be considered. Hence, our hypothesis is
 292 that the majority of this decay change is due to a dual effect. An indirect one is the influence of the
 293 temperature on the viscosity, and a direct one is the efficiency of the pyrene transition, as specific or
 294 non-specific quenching are not enough to explain the overall trend. This will require further study; as
 295 such a strong dependence on temperature ($\pm 40^\circ\text{C} \rightarrow \pm 100 \text{ ns}$) can be a crucial parameter in the many
 296 precise application of liquid scintillators. As a conclusion, it is another possibility to tune the decay time
 297 of the liquid scintillator, not by changing the formulation but the experimental temperature.

298



299



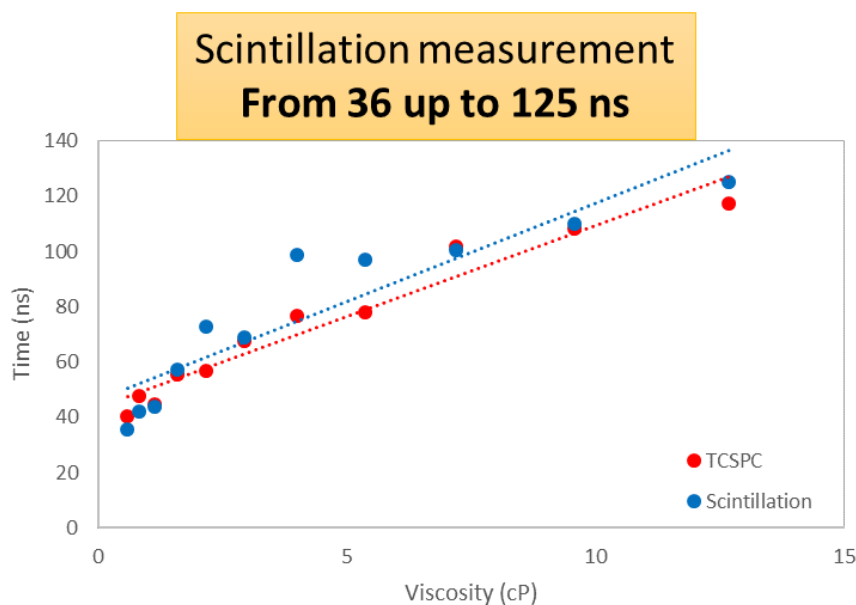
300

301 *Figure 5. Top) Time-Correlated Single Photon Counting of a liquid scintillator composed of 0.03 wt% of pyrene, 0.03 wt% of*
 302 *9,10-DPA in DIN as a function of temperature. $\lambda_{ex} = 274$ nm; $\lambda_{obs} = 430$ nm. Note the vertical axis which is the same for slow*
 303 *and mean decay values (left) and fast decay value (right). Bottom) Evolution of the relative weight of fast and slow*
 304 *components as a function of the temperature.*

305

306 Scintillation properties

307 After having determined the photophysical parameters, the as-prepared liquid mixtures were assessed
 308 in terms of scintillation efficiency. All the previous decay data were collected on TCSPC, that is to say
 309 under 274-nm UV excitation. This wavelength is suitable for excitation of the solvents mixture, but the
 310 complex scintillation mechanism may not be fully represented from these TCSPC measurements.
 311 Therefore, the next set of experiments consisted in recording the scintillation pulse of each liquid
 312 scintillator combination. TCSPC using radioactive sources is possible but is not straightforward since it
 313 usually requires very strict experimental conditions and days or even weeks of continuous recordings.³⁹
 314 This timescale is higher than the potential stability of our materials. Thus, we decided to use the
 315 simplest method, which is recording thousand pulses and measure the time profile of the pulse. The
 316 average of 5,000 pulses obtained after excitation of the liquid scintillator with a gamma-ray emitting
 317 ⁶⁰Co source was processed using the same software we used to fit the data obtained from TCSPC
 318 measurements, so the comparison is easier. Figure 6 summarizes the values observed from TCSPC and
 319 from scintillation measurements. As one can see, a global trend between the two experiments is
 320 respected. This undoubtedly confirms the possibility to calculate the decay time of a liquid scintillator
 321 from a UV excitation, at least for this range of decay times, TCSPC with UV being much more of practical
 322 use than scintillation decay times.



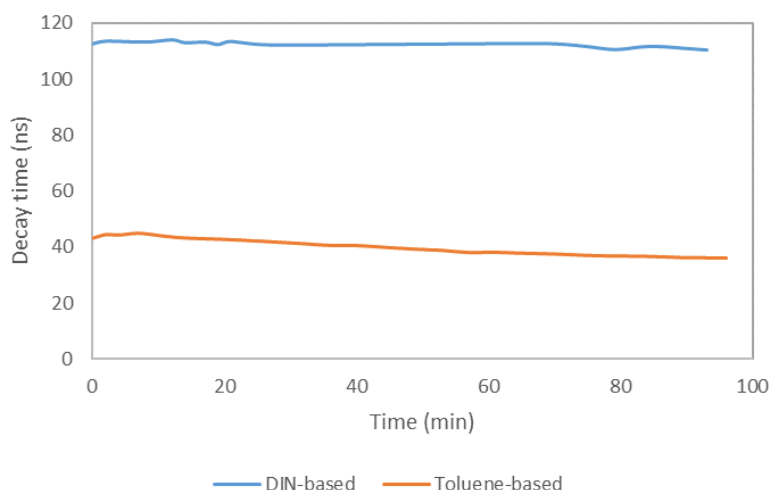
324

325 *Figure 6. Decay times obtained from TCSPC (red) or scintillation (blue) measurements. The dashed lines are drawn to guide*
 326 *the eye.*

327

328 The two scintillators that have the decay time limits were also checked for pulse shape variations
 329 within a given time. Thus, the scintillators were flushed with argon and the pulses were recorded from
 330 time to time within a ≈ 100 minutes range, then approximately 18 hours in closed scintillation vial. The
 331 results are given Figure 7. Linear decrease of the mean decay time is observed for both scintillators,
 332 but with different kinetics. The slowest scintillator displays outstanding stability over these 100
 333 minutes, with a decrease of 0.02 ns/min. The fastest scintillator decay decreases more rapidly with a
 334 decrease of 0.10 ns/min. These values are stable enough for radionuclide assay experiments with such
 335 scintillators. After 18 hours, the mean decay values are 51.6 ns and 94.0 ns for these two liquid
 336 scintillators. To confirm the slow diffusion of air into the liquid leading to the quenching of the
 337 scintillation, these two scintillators were stirred in open flask for 30 minutes, then another run of
 338 experiment was performed. The corresponding decay times are still different, with τ_{mean} of 16.1 ns and
 339 62.0 ns for the fastest and slowest scintillators, respectively.

340



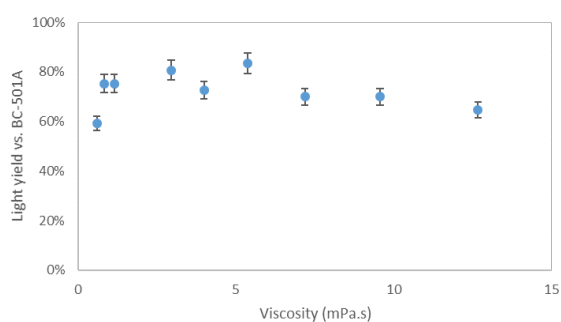
341

342 *Figure 7. Mean scintillation decay time over time.*

343

344 Another important factor is to keep the scintillation yield as high as possible for all mixtures, which
 345 cannot be predicted *a priori*. Among other parameters, the concentration of pyrene (0.03 wt%) is very
 346 low regarding the typical concentration one can find for all other regular primary fluorophores
 347 (typically 1 – 3 wt%). All scintillators were thus exposed to a gamma-ray emitting ^{137}Cs radioactive
 348 source and the pulse shape spectra were recorded. A commercial liquid scintillator, namely BC-501A
 349 was used as the reference. This scintillator is known to display 12,000 ph/MeV (78 % of anthracene
 350 single crystal).⁴⁰ The results are given Figure 8. Interestingly, the observed light yields were better than
 351 expected due to the low content of pyrene and fall in the range 7,000 – 9,900 ph/MeV, with the worst
 352 values observed for the 100 % toluene-based scintillator.

353



354

355 *Figure 8. Relative ^{137}Cs gamma light yield against BC-501A reference as a function of viscosity. The vertical uncertainties are*
 356 *given at 5 % of the value.*

357

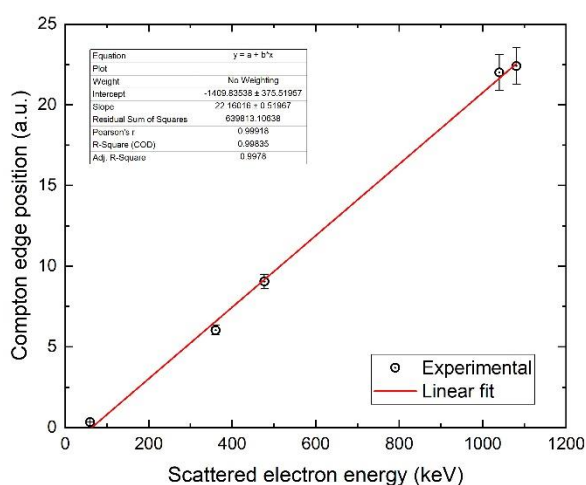
358 The last experiment was performed with the slowest decaying liquid scintillator only. Most of organic
 359 and inorganic scintillators are known to feature non-linear response towards the impinging energy.
 360 Especially, gamma rays with energy below 100 keV do not fall within the same slope as for $E_\gamma > 100$ keV.
 361 Thus, the DIN-based liquid was exposed to four gamma sources with various energies, ranging from
 362 ^{241}Am ($E_\gamma = 59$ keV) to ^{22}Na ($E_\gamma = 1274$ keV, accompanied by an electron/positron annihilation of
 363 511 keV). Backscattered Compton electrons from gamma interactions within the scintillator with a
 364 maximum energy is given by the following formula:

365

$$E_{CE} = \frac{2E_{\gamma}^2}{m_e c^2 + 2E_{\gamma}}$$

366 with m_e the electron mass (511 keV/c²) and c the celerity in the vacuum. In this calculation is also
 367 neglected the binding energies of the electrons in the molecules of the scintillator. In the case of ²⁴¹Am,
 368 the energy of the photoelectric peak (i.e. $E_{\gamma}^{241Am} = 59 \text{ keV}$) was considered at the expense of the
 369 Compton edge since the gamma ray is fully absorbed within the material's volume. The results are
 370 given Figure 9. Without more available gamma sources, it is difficult to conclude on the linearity of the
 371 scintillator's response below 59 keV.

372



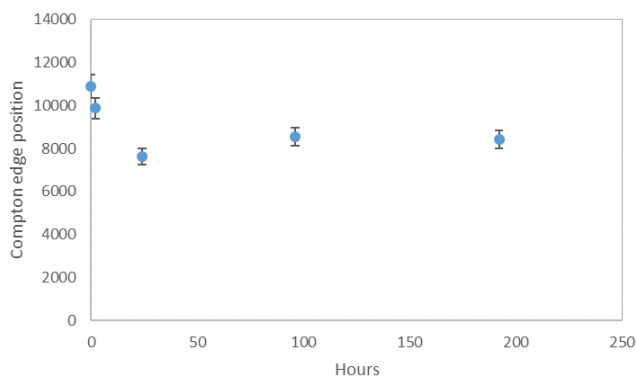
373

374 *Figure 9. Energy response against the Compton edge position of four gamma sources. The line is drawn to guide the eye.*

375

376 Along with the decay time stability, the light yield evolution with time is a key-parameter. Once again
 377 for the slowest scintillator, ¹³⁷Cs pulse height spectra were performed from time to time on the same
 378 DIN-based scintillator that was saturated with argon at the beginning of the experiment (Figure 10).
 379 The light yield falls quite rapidly over the first ten hours, with 91 % of the initial light output 2 hours
 380 after bubbling. This fast decrease, probably due to oxygen diffusion, may be one of the main drawbacks
 381 of the slowest formulation. After one day it seems to stabilize in the range 70 – 79 % of the maximum
 382 value obtained at t_0 .

383



384

385 Figure 10. ^{137}Cs Compton edge position with time. The uncertainty is set at 5 % of the position of the Compton edge position.

386

387 4. Conclusion

388 In the prolongation of the tuning of the decay time of plastic scintillators,¹⁷ we present herein a
 389 modulation of the decay time for liquid scintillators. Compared with plastics, another strategy was
 390 developed based on varying the viscosity of the solvent used for preparing the scintillator. Thus, pyrene
 391 and 9,10-DPA were chosen as primary and secondary fluorophores thanks to their apparent long decay
 392 time when oxygen is removed efficiently and a 430 nm-centered desired emission wavelength. The
 393 concentration of pyrene was reduced to an unusual rate for a primary fluorophore so that mainly the
 394 monomer species is able to emit light.

395 Then various mixtures of toluene-based and DIN-based liquid scintillators containing the same molar
 396 concentrations of pyrene and 9,10-DPA were prepared. A linear dependence of the decay time was
 397 observed with the viscosity of the solvent mixture. The decay times observed by TCSPC fall in the range
 398 40 ns – 120 ns for the less and the most viscous solutions, respectively. The quality of the bubbling
 399 procedure (or in other words, the remaining diluted oxygen in the solution) may affect the decay time,
 400 more particularly for the slowest materials.

401 All scintillators displayed good ^{137}Cs gamma detection properties, with relative light yields in the range
 402 7,000 – 9,900 ph/MeV. TCSPC measurements were then compared with scintillation decays and the
 403 results were satisfactory. All scintillators decay times are within the range 36 – 125 ns. An in-depth
 404 study performed on the slowest scintillator revealed a minor drawback with a light output decrease
 405 within the first two hours. However, one should consider this 36 – 125 ns range as *ideal*. Degrading
 406 slopes of not more than 0.10 ns/min after bubbling with argon should be taken into account for the
 407 nuclear physicist, so that they would narrow this range. No obvious energy non-linearity was observed,
 408 however within a limited energy range.

409 An obvious criticism of such method could be the relationship of the viscosity of a solvent with
 410 temperature⁴¹. The literature example of toluene gives a 34 % variation of its viscosity in the range 293
 411 – 333 K (thus a 40 °C range). In our case, the time-response of liquid scintillators versus temperature
 412 can represent another possibility to perform an efficient and accurate tuning of the decay time, as was
 413 exemplified with DIN-based liquid scintillator. Its decay time varied from 163 ns down to 69 ns when
 414 increasing the temperature in the range 8 – 42 °C. Interestingly, only the slow component of the
 415 biexponential fit is affected by temperature variations. So this drawback could be seen as a strength if
 416 the scintillator is carefully thermalized while in use. This 36 to 125 ns range could thus be widely
 417 extended thanks to temperature adjustments of the LS. Considering regular, fast formulations, one can
 418 have now in in hands all the possibilities from ultra-fast liquids up to 163 ns.

419 The next step will be to evaluate these new scintillators as liquid scintillation cocktails. Various
420 radioactive sources will be evaluated for environmental assays.

421

422 Conflict of interest

423 The authors declare that they have no known competing financial interests or personal relationships
424 that could have appeared to influence the work reported in this paper.

425

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430 References

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- ¹ G. H. V. Bertrand, M. Hamel, and F. Sguerra, *Chem. – Eur. J.*, 2014, **20**, 15660-15685. <https://doi.org/10.1002/chem.201404093>
- ² G. T. Reynolds, F. B. Harrison, and G. Salvini, *Phys. Rev.*, 1950, **78**, 488. <https://doi.org/10.1103/PhysRev.78.488>
- ³ H. Kallmann, *Phys. Rev.*, 1950, **78**, 621-622. <https://doi.org/10.1103/PhysRev.78.621.2>
- ⁴ E. Rapkin, *Int. J. Appl. Rad. Isotop.*, 1964, **15**, 69-87. [https://doi.org/10.1016/0020-708X\(64\)90052-3](https://doi.org/10.1016/0020-708X(64)90052-3)
- ⁵ D. Franco, G. Bellini, J. Benziger, D. Bick, G. Bonfini, D. Bravo, M. Buizza Avanzini, B. Caccianiga, L. Cadonati, F. Calaprice, C. Carraro, P. Cavalcante, A. Chavarria, D. D'Angelo, S. Davini, A. Derbin, A. Etenko, K. Fomenko, C. Galbiati, S. Gazzana, C. Ghiano, M. Giammarchi, M. Göger-neff, A. Goretti, L. Grandi, E. Guardincerri, S. Hardy, A. Ianni, A. Ianni, A. Kayunov, V. Kobychhev, D. Korablev, G. Korga, Y. Koshio, D. Kryn, M. Laubenstein, T. Lewke, E. Litvinovich, L. Ludhova, B. Loer, F. Lombardi, P. Lombardi, I. Machulin, S. Manecki, W. Maneschg, G. Manuzio, Q. Meindl, E. Meroni, L. Miramonti, M. Misiaszek, D. Montanari, P. Mosteiro, V. Muratova, L. Oberauer, M. Obolenksy, F. Ortica, K. Otis, M. Pallavicini, L. Papp, L. Perasso, S. Perasso, A. Pocar, R. S. Raghavan, G. Ranucci, A. Razeto, A. Re, P. A. Romani, A. Sabelnikov, R. Saldanha, C. Salvo, S. Schönert, K. Simgen, M. Skorokhvatov, O. Smirnov, A. Sotnikov, S. Sukhotin, Y. Suvorov, R. Tartaglia, G. Testera, D. Vignaud, R. B. Vogelaar, F. Von Feilitzsch, J. Winter, M. Wojcik, A. Wright, M. Wurm, J. Xu, O. Zaimidoroga, S. Zavatarelli, and G. Zuzel, *Nucl. Phys. B*, 2013, **237-238**, 104-106. <http://dx.doi.org/10.1016/j.nuclphysbps.2013.04.068>
- ⁶ J. Maneira, and the SNO+ Collaboration, *J. Phys.: Conf. Ser.*, 2013, **447**, 012065. <https://doi.org/10.1088/1742-6596/447/1/012065>
- ⁷ R. Broda, P. Cassette, and K. Kossert, *Metrologia*, 2007, **44**, S36-S52. <https://doi.org/10.1088/0026-1394/44/4/S06>
- ⁸ L. Beaulieu and S. Beddar, *Phys. Med. Biol.*, 2016, **61**, R305-R343. <https://doi.org/10.1088/0031-9155/61/20/R305>
- ⁹ L. Salonen, L. Kaihola, B. Carter, G. T. Cook, and C. J. Passo, in *Handbook of Radioactivity Analysis (Third Edition)*, edited by M. F. L'Annunziata (Academic Press, 2012), pp. 625-693. <https://doi.org/10.1016/B978-0-12-384873-4.00009-8>
- ¹⁰ E. Montbarbon, F. Sguerra, G. H. V. Bertrand, É. Magnier, R. Coulon, R. B. Pansu, and M. Hamel, *Chem. – Eur. J.*, 2016, **22**, 12074-12080. <http://dx.doi.org/10.1002/chem.201601749>
- ¹¹ H.-L. Xiao, J.-S. Deng, and N.-Y. Wang, *Chin. Phys. C*, 2010, **34**, 571-575. <https://doi.org/10.1088/1674-1137/34/5/011>
- ¹² T. Marrodán Undagoitia, F. von Feilitzsch, L. Oberauer, W. Potzel, A. Ulrich, J. Winter, and M. Wurm, *Rev. Sci. Instrum.*, 2009, **80**, 043301. <https://doi.org/10.1063/1.3112609>
- ¹³ P. Lombardi, F. Ortica, G. Ranucci, and A. Romani, *Nucl. Instr. Methods A*, 2013, **701**, 133-144. <http://dx.doi.org/10.1016/j.nima.2012.10.061>
- ¹⁴ R. Lauck, M. Brandis, B. Bromberger, V. Dangendorf, M. B. Goldberg, I. Mor, K. Tittelmeier, and D. Vartsky, *IEEE Trans. Nucl. Sci.*, 2009, **56**, 989. <https://doi.org/10.1109/TNS.2008.2009449>
- ¹⁵ J. Kirkbride, E. C. Yates, and D. G. Crandall, *Nucl. Instrum. Methods*, 1967, **52**, 293-297. [https://doi.org/10.1016/0029-554X\(67\)90234-0](https://doi.org/10.1016/0029-554X(67)90234-0)

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- ¹⁶ H. E. Jackson, and G. E. Thomas, *Rev. Sci. Instrum.*, 1965, **36**, 419-425. <https://aip.scitation.org/doi/pdf/10.1063/1.1719592>
- ¹⁷ M. Hamel, M. Soumaré, H. Burešová, and G. H. V. Bertrand, *Dyes Pigm.*, 2019, **165**, 112-116. <https://doi.org/10.1016/j.dyepig.2019.02.007>
- ¹⁸ <https://www.crystals.saint-gobain.com/products/bc-501a-bc-519>
- ¹⁹ R. K. Swank and W. L. Buck, *Rev. Sci. Instrum.*, 1955, **26**, 1516. <https://doi.org/10.1063/1.1771230>
- ²⁰ P. G. Sjölin, *Nucl. Instrum. Methods*, 1965, **37**, 45-50. [https://doi.org/10.1016/0029-554X\(65\)90335-6](https://doi.org/10.1016/0029-554X(65)90335-6)
- ²¹ S. D. Biller, E. J. Leming, and J. L. Paton, *Nucl. Instrum. Methods*, 2020, **972**, 164106. <https://doi.org/10.1016/j.nima.2020.164106>
- ²² L. Rocha, C.-M. Păiuș, A. Luca-Raicu, E. Resmerita, A. Rusu, I.-A. Moleavin, M. Hamel, N. Branza-Nichita, and N. Hurduc, *J. Photochem. Photobiol. A*, 2014, **291**, 16-25. <http://dx.doi.org/10.1016/j.jphotochem.2014.06.018>
- ²³ J. Duhamel, *Langmuir*, 2012, **28**, 6527-6528. <https://doi.org/10.1021/la2047646>
- ²⁴ L. Pichat, P. Pesteil, and J. Clément, *J. Chim. Phys. Phys.-Chim. Biol.*, 1953, **50**, 26-41. <https://doi.org/10.1051/jcp/1953500026>
- ²⁵ (a) J. B. Birks, *J. Phys. Chem.*, 1963, **67**, 2199-2200. <https://doi.org/10.1021/j100804a054>; (b) J. B. Birks, *J. Phys. Chem.*, 1964, **68**, 439-440.
- ²⁶ H. A. Yemam, A. Mahl, U. Koldemir, T. Remedés, S. Parkin, U. Greife, and A. Sellinger, *Sci. Rep.*, 2015, **5**, 13401. <https://doi.org/10.1038/srep13401>
- ²⁷ Z. Zheng, J. Zhu, X. Luo, Y. Xu, Q. Zhang, X. Zhang, Y. Bi, and L. Zhang, *Nucl. Instr. Methods A*, 2017, **850**, 12-17.
- ²⁸ T. Wolff, K. Pfanner, and C. Springob, *J. Photochem. Photobiol. A*, 1993, **74**, 247-253. [https://doi.org/10.1016/1010-6030\(93\)80123-Q](https://doi.org/10.1016/1010-6030(93)80123-Q)
- ²⁹ A. Vyšniauskas, M. Qurashi, N. Gallop, M. Balaz, H. L. Anderson, M. K. Kuimova, *Chem. Sci.*, 2015, **6**, 5773-5778. <https://doi.org/10.1039/C5SC02248G>
- ³⁰ E. Blatt, F. E. Treloar, K. P. Ghiggino, R. G. Gilbert, *J. Phys. Chem.*, 1981, **85**, 2810-2816. <https://doi.org/10.1021/j150619a022>
- ³¹ F. H. Alturkistany, K. Nichani, W. Li, J. P. Houston, "Effect of Viscosity on Fluorescence Lifetime Measured Using Flow Cytometry," 2018 Conference on Lasers and Electro-Optics (CLEO), San Jose, CA, 2018, pp. 1-2. <https://ieeexplore.ieee.org/document/8426955/>
- ³² [DIN MSDS](#) (in French).
- ³³ J. M. G. Martinho and V. R. Pereira, *J. Lumin.*, 1988, **40-41**, 673-675. [https://doi.org/10.1016/0022-2313\(88\)90384-5](https://doi.org/10.1016/0022-2313(88)90384-5)
- ³⁴ J. M. Marchant, B. B. Dolezal, M. L. Nabholz, D. P. Howard, B. W. Baumbaugh, J. E. Clecker, L. F. Hernandez, C. R. Hurlbut, and R. C. Ruchti, *IEEE Nuc. Sci. Symp. Conf. Rec.*, 2009, **N25-134**, 1555-1557. <https://doi.org/10.1109/NSSMIC.2009.5402277>
- ³⁵ U. K. A. Klein, R. Frey, M. Hauser, and U. Gösele *Chem. Phys. Lett.* 1976, **41**, 139-142. [https://doi.org/10.1016/0009-2614\(76\)85266-9](https://doi.org/10.1016/0009-2614(76)85266-9)
- ³⁶ C. Bohne, E. B. Abuin, and J. C. Scaiano *J. Am. Chem. Soc.* 1990, **112**, 4226-4231. <https://doi.org/10.1021/ja00167a018>
- ³⁷ F. T. Kuchnir and F. J. Lynch, *IEEE Trans. Nucl. Sci.*, 1968, **15**, 107-113. <https://doi.org/10.1109/TNS.1968.4324923>
- ³⁸ J. B. Birks, D. J. Dyson, and I. H. Munro, *Proc. R. Soc. Lond. A*, 1963, **275**, 575-588. <https://doi.org/10.1098/rspa.1963.0187>
- ³⁹ Y. Abe, H. Hosoda, Y. Arikawa, T. Nagai, S. Kojima, S. Sakata, H. Inoue, Y. Iwasa, K. Iwano, K. Yamanoi, S. Fujioka, M. Nakai, N. Sarukura, H. Shiraga, T. Norimatsu, and H. Azechi, *Rev. Sci. Instrum.*, 2014, **85**, 11E126. <https://doi.org/10.1063/1.4896957>
- ⁴⁰ <https://www.crystals.saint-gobain.com/sites/imdf.crystals.com/files/documents/bc501-501a-519-data-sheet.pdf>
- ⁴¹ C. Leite Paiva, R. Silva Pinheiro, F. Xavier Feitosa, and H. Batista de Sant'Ana, *J. Chem. Eng. Data*, 2020, **65**, 540-548. <https://doi.org/10.1021/acs.jced.9b00718>.