



## Study of new luminophores for use in modern scintillation cocktails

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### ABSTRACT

This paper deals with the study of 1-phenyl-3-(2,4,6-trimethyl-phenyl)-2-pyrazoline, 2,2',7,7'-tetrakis(N,N-diphenyl-amino)-9,9'-spirobifluorene, 2-(4-biphenyl)-6-phenylbenzoxazole and 9,9'-bifluorenyl as possible replacements of the existing and most widely used luminophore (2,5-diphenyloxazole (PPO)) in contemporary scintillators. The study is focused on the determination of emission wavelength, concentration optimization, the influence of the wavelength shifters POPOP and *bis*-MSB. In addition, the response to alpha and beta radiation were measured, in particular, the pulse length index (PLI) was determined. Finally, yet importantly, the effect of emulsifiers on the shape of the spectrum and the solubility of the aqueous phase was tested. The results obtained were compared with the commercially available Aqualight scintillation cocktail.

### 1. Introduction

Since the development of liquid scintillation technique, those involved in further development and improvement of this analytical method have sought better and more efficient scintillation cocktails. The way of improvement is generally divided into three major areas; solvent, fluor and emulsifiers. The first area is devoted to discovering ideal solvent. The definition of “ideal” can be understood differently, based on the point of view, but nevertheless, there are some criteria, which generally define ideal solvent [1]. Those are: high flash point, low vapor pressure, odorless, low toxicity and irritancy, no permeation through plastics, biodegradability, exceptional fluor solubility, low photo- and chemiluminescence, high counting efficiency, especially for tritium and high color and chemical quench resistance. During the years, many chemicals have been employed; toluene and xylene were initial solvents of choice in 1950's. In addition, dioxan and naphthalene were widely used in this period. Over the next decades, little had happened until pseudocumene was introduced in 1970's. Even it is more than 40 years, it is still used in modern applications [2,3] due to high light output and low price. Because of the high toxicity and the harmful effect on the environment, it was realized a very different solvent would be required. The effort resulted in the discovery of the polyalkylbenzenes, which suffer from low counting efficiency. The answer to this drawback can be found in the utilization of di-isopropyl-naphthalene (DIPN), which is a mixture of position-isomeric di-isopropyl-naphthalene [1,4–6]. The second major area is the fluor component, or generally scintillators, which are responsible for converting photon energy into visible light. This component, which can be

a mixture of different scintillators, plays the main role in counting efficiency, pulse height and pulse duration. The ideal solute should exhibit high fluorescence quantum yield, long emission wavelength, short decay time and possess the ability to distinguish between different interacting particles due to pulse length index (PLI) [7,8]. Furthermore, it has to be highly soluble in the solvent, be transparent and exhibits a low dependence of pulse height on the concentration. Today's best choice fluor mixtures are 2,5-Diphenyloxazole (PPO) and 1,4-Bis(2-methylstyryl)-benzene (*bis*-MSB) [9–12] or 1,4-Bis(5-phenyl-2-oxazolyl)benzene (POPOP) [13–18] acting as a wavelength shifters. Besides the traditional wavelength shifters mentioned above, also new fluors are intensively studied, such as 1,4-bis(4-methoxystyryl)benzene [3,19] Emulsifiers or surfactants have to be employed if, in particular, there is an effort to measure aqueous samples and determine their activity. The improvement of surfactants applied in liquid scintillation cocktails contributed significantly to the simplification of sample preparation procedure. The well known classical sol-gel cocktails [6,20] have been replaced by cocktails exhibiting a continuous clear liquid phase in an application with aqueous samples. This feature is very important, especially when dealing with small counting vials. The increasing demand of applications led to the introduction of cocktails based on nonionic (e.g. alkylphenol ethoxylates) and anionic (succinates, sulfonates, sulfates, etc.) surfactants [21–23].

In this paper 1-phenyl-3-(2,4,6-trimethyl-phenyl)-2-pyrazoline, 2,2',7,7'-tetrakis(N,N-diphenyl-amino)-9,9'-spirobifluorene, 2-(4-biphenyl)-6-phenylbenzoxazole and 9,9'-bifluorenyl were investigated as a potential substituents of today's most popular mixture of PPO and *bis*-MSB. The selection of these luminophores was based both on their

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luminescence properties previously published for different solvents [7] and on their structural similarities with s quaterphenyl and sexiphenyl structures, which were reported as promising scintillators [24]. Furthermore, other luminophores were selected with respect to their maximum emission wavelengths to fit the optimal range of quantum efficiency of commonly used photomultipliers. The determination of emission wavelength, concentration, the influence of the wavelength shifters POPOP and *bis*-MSB as well as the response to alpha and beta radiation expressed by the pulse length index (PLI) were examined. Just as importantly, the effects of Triton X-100 and Triton X-114 emulsifiers on the shape of the spectrum and the solubility of the aqueous phase were tested due to their extensive use as efficient nonionic surfactants.

## 2. Experimental

### 2.1. Devices and equipment

The measurements were carried out using Liquid Scintillation Counter 300SL (Hidex, Finland) with 20 ml polyethylene vials. The setting of the instrument was as follows: the PLI index was set to zero, time of the measurement was set to 300 s, alpha and beta window was set to full width (1-1023 channel). Aqualight (Hidex, Finland) was used as a liquid scintillation counting (LSC) cocktail etalon for standardization and comparative measurement.

Proper shaking was performed by shaker IKA KS 130 basic (IKA, USA). Luminescence decay was determined using LSC counter Triathler (Hidex, Finland) in Luminescence mode and CAMAG UV lamp 4 (CAMAG, CH) as a light source with wavelength emission of 254 nm.

### 2.2. Chemicals

The investigated primary fluors were 1-phenyl-3-(2,4,6-trimethylphenyl)-2-pyrazoline, henceforth called PMP, 9,9'-bifluorenyl, henceforth called BF, (both Tokyo Chemical Industry, Japan), 2,2',7,7'-tetrakis(N,N-diphenyl-amino)-9,9'-spirobifluorene, henceforth called DAS and 2-(4-biphenyl)-6-phenylbenzoxazole, henceforth called BPB, (Sigma-Aldrich, USA). The 2,5-diphenyloxazole (Sigma-Aldrich, USA) was utilized as a standard primary fluor used in commercial liquid cocktails. 1,4-Bis(5-phenyl-2-oxazolyl)benzene (Fluka Chemie GmbH, Germany) and 1,4-Bis(2-methylstyryl)-benzene (Sigma-Aldrich, USA) were used as traditional wavelength shifters. Furthermore, 2,5-Bis(5-tert-butyl-benzoxazol-2-yl)thiophene, hence called BTE, 7-Amino-4-methylcoumarin, hence called AMC and 7-Amino-1,3-naphthalenedisulfonic acid monopotassium salt monohydrate, hence called ASA, were investigated (all Sigma-Aldrich, USA) as a possible secondary fluor.

Toluene, cyclohexane (both Chromservis, s. r.o., Czechia), 1-Methylnaphthalene (Sigma-Aldrich, USA) di-isopropylnaphthalene (DIPN) (Fisher Scientific, United Kingdom) were used as solvents.

Triton™ X-100 (T-100), Triton™ X-114 (T-114) (both Sigma-Aldrich, USA) and butyldiglycol (Carl Roth GmbH, Germany) were used as surfactants to ensure water miscibility. Water was obtained from a DEMIWA 5 ROSA™ (Watek) water purification system. All other materials were ACS reagent grade and were used as received.

Radiochemical isotope tracers Am-241 ( $1.67 \text{ Bq } \mu\text{l}^{-1}$ ) and Sr-90 ( $0.55 \text{ Bq } \mu\text{l}^{-1}$ ) were obtained from CMI (the Czech Metrological Institute).

### 2.3. Procedures

#### 2.3.1. Luminescence properties of investigated fluorophores

The fluorescence spectra were obtained using UV-VIS luminescent spectrofluorometer 199 S, Edinburgh Instrument. The excitation was performed using X-ray source Seifert at a voltage of 40 kV and a current of 40 mA.

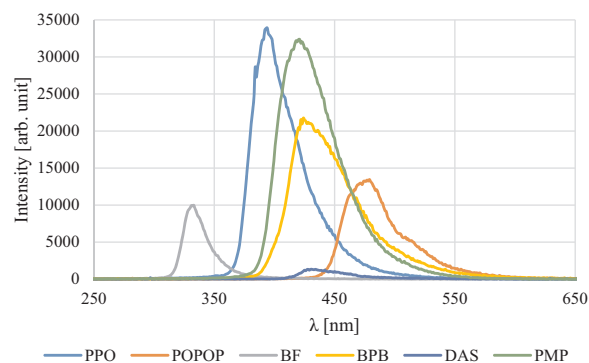


Fig. 1. Fluorescence spectra of PPO, POPOP, BF, BPB, DAS and PMP at room temperature.

#### 2.3.2. Solvent and fluor concentration influence

Every mentioned fluor was separately dissolved in a mixture of 9 ml of DIPN and 1 ml of T-100 in the concentration of 0.01, 0.1 and  $1 \text{ g l}^{-1}$ . Each mixture was prepared six times; three samples were spiked with  $30 \mu\text{l}$  of Am-241 and three samples were spiked with  $50 \mu\text{l}$  of Sr/Y-90. This procedure was repeated for all remaining solvents, such as toluene, cyclohexane and 1-methylnaphthalene. The measurement time was 600 s, PLI was zero and YGain was 20. The range of alpha and beta window was from 0 up to 1023 channels. Other parameters were the same as in original program “RN222”.

#### 2.3.3. Secondary fluorophores influence on detection efficiency

The ratio of solvent and emulsifier and measurement settings were the same as stated in “Solvent and fluor concentration influence”. The mass concentration of the fluor was  $1 \text{ g l}^{-1}$  and the mass of secondary fluor was  $0.1 \text{ g l}^{-1}$ . Each mixture containing the same primary and secondary fluor was prepared six times; one-half was spiked with  $30 \mu\text{l}$  of Am-241 and the other half was spiked with  $50 \mu\text{l}$  of Sr/Y-90. This procedure was repeated for all combinations of the remaining primary and secondary fluors.

#### 2.3.4. Alpha/beta separation

The ratio of solvent, primary fluor and emulsifier, as well as the measurement settings were the same as stated in the previous paragraph. Each mixture was prepared two; one sample was spiked with  $30 \mu\text{l}$  of Am-241 and the other sample was spiked with  $50 \mu\text{l}$  of Sr/Y-90.

#### 2.3.5. Afterglow properties of liquid scintillator components

The measurement was performed for used solvents, namely DIPN, toluene and cyclohexane, then for 3.3 ml of T-100 and T-114 and their mixtures with DIPN (6:3) and finally for all investigated fluors dissolved in the mixture of DIPN and T-100 (6:3). The phosphorescence dependence on the fluor concentration was also measured. The vial with proper mixture without any artificial nuclide was exposed for one minute to the UV light with  $\lambda = 254 \text{ nm}$  and then measured. The mode of measurement was switched to the “Lumi” mode. The time of the measurement was 5 s and the number of repetition was 250.

## 3. Results and discussion

### 3.1. Luminescence properties of investigated fluorophores

Fluorophores are added into the liquid scintillator for shifting the maximum emission wavelength of scintillation cocktail to the wavelength during which the photomultiplier tube has a maximum quantum efficiency. Usually, this a maximum quantum efficiency of the most used photomultipliers lying in the range of 370–500 nm [25]. The frequently used fluorophore PPO has the emission peaks at 385 nm, which is in the range of UV light. Because the emission curve does not ideally

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