Three New Organic Scintillators with Large Stokes Shifts

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Three new organic scintillators (PMP-410, PMP470, and PMP480) were synthesized on the basis of 1-phenyl-3-mesityl-2-pyrazoline (PMP). Their synthesis and their polymerization in polystyrene are described. We measured their Stokes shifts in polystyrene (~ 1.3 eV) between their absorption maxima (PMP-410: 287 nm; PMP-470: 315 nm; PMP-480: 314 nm) and their emission maxima (PMP-410: 408 nm; PMP-470: 480 nm; PMP-480: 479 nm). Absorption and emission maxima, Stokes shifts, and light yields of all eight pyrazoline scintillators so far synthesized are listed and are compared with results for 3-hydroxyflavone (3-HF). The advantages of these one-component dopants are discussed with respect to the usually applied two-component (scintillator plus wavelength shifter) dopants.

Index Heading: **Fluorescence.**

INTRODUCTION

Small quantities of organic scintillators are added to organic solvents (e.g., toluene, benzene, etc.) or to solid polymers (e.g., polystyrene, polyvinyl toluene) to enhance the poor fluorescence yield of these solvents for the detection of ionizing radiation. Intramolecular energy transfer between the respective quantum levels requires that the ultraviolet emission band of the solvent or polymer overlap the scintillator's absorption band. If the added scintillator emits in the opaque (UV) region of the solvent or polymer (e.g., *p*-terphenyl at 340 nm) a second organic scintillator (wavelength shifter) is needed to shift the emitted light into the visible (>400 nm) region (two-component system). If, however, one scintillator spans the large energy difference (Stokes shift) between the emission band of its solvent or polymer in the UV region and its fluorescence emission in the visible (>400 nm) region, a wavelength shifter can be avoided (one-component system).

The advantage of the one-component system becomes obvious if we compare its self-absorption, caused by the overlapping of the scintillator's absorption and emission bands, with that of the two-component system. Here, the two scintillators bridge energy gaps of about 0.8 eV (primary scintillator) and 0.5 eV (wavelength shifter) with their respective bandwidths [full width at half-maximum (FWHM)] of some 0.4 eV. This factor necessarily results in a large overlap of absorption and emission of the wavelength shifter and therefore causes strong self-absorption. For reduction of this self-absorption, the concentration of the wavelength shifter must be heavily reduced to finally achieve reasonably low attenuations of its own light emission.

The energy transfer from the solvent or polymer to the added scintillator, or from the scintillator to its wavelength shifter, can be either nonradiative (Förster† transitions[1]) or radiative via light emission and its reabsorption, depending on the concentrations of the scintillators added. Sufficiently high scintillator concentrations (>0.02 molar fraction) provide mainly nonradiative and therefore local (transfer distances of less than 5 nm) energy transfer. Such concentrations can be achieved with one-component scintillators of large Stokes shift (>1.0 eV) since their self-absorption due to overlapping of absorption and fluorescence bands is reasonably small if not zero. In a two-component system, the concentration of the wavelength shifter must be kept between one and two orders of magnitude smaller to reduce its self-absorption, and the energy transfer is therefore purely radiative, with an average reabsorption length of about 150 μm between the primary light emission and the reabsorption of its wavelength shifter. This pattern results in nonlocal light emissions, which account for cross-talk in small-diameter fibers or mask the decay length of short-lived (≤1 ps) particles in scintillating liquids.

These were our motivations to search for scintillators with large Stokes shifts in order to replace by one-component systems the two-component systems previously used. Their main applications would be large-volume liquid scintillators for neutrino detection and small-diameter scintillating fibers for particle tracking at electron or hadron colliders. Apart from longer light absorption lengths, these one-component systems provide local energy transfer and therefore avoid unwanted cross-talk (~ 150 μm) to neighboring detector regions.

A systematic search for scintillators with large Stokes shifts was already started in 1977 by Güsten et al.,[3] who synthesized a series of methyl and/or methoxy substituted 1,3-diphenyl-2-pyrazolines as scintillators for liquid scintillation counting systems. With the recent advent of scintillating fibers,[4] one of these substances, 1-phenyl-3-mesityl-2-pyrazoline (PMP), has been successfully used as a one-component scintillator in small-diameter (60 μm) fibers[5] and in large-volume scintillators.[6] Encouraged by this experience, we systematically synthesized during the past years other pyrazoline-based compounds and eval-

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† Förster has shown that radiationless energy transitions can occur between donor molecules (e.g., polystyrene) and acceptor (scintillator) molecules. They require distances <0.5 nm between donors and acceptors, which calls for scintillator concentrations at which, on the average, one scintillator molecule is inside a sphere with a radius corresponding to these distances. An extensive list of computed critical distances and molar concentrations can be found in Ref. 2.

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uated their properties with respect to Stokes shift, self-absorption, and light yield.

BASIC CONSIDERATIONS

Molecular Mechanisms of Large Stokes Shifts. More than a century ago, Stokes[7] pointed out that molecules generally fluoresce at longer wavelengths than the exciting one. In aromatic molecules, the light absorption process causes rapid transitions (time $< 10^{-15}$ s) of π electrons between different energy levels in which the nuclear configuration of the molecule's ground state is retained (Franck–Condon state). The corresponding fluorescence emission is generally delayed by some 10^{-7} to 10^{-9} s with respect to the absorption process. During this delay time, the molecule has time enough to rearrange its energy levels from the Franck–Condon state to a more stable configuration of lower potential energy (singlet state) from which fluorescence is emitted. The loss in energy is termed Stokes shift or Stokes loss. The light yield of a fluorescing molecule depends on the efficiency of other competing processes like vibrational deactivation or photochemical processes in the excited singlet state.

The Stokes shift can be correlated with molecular features such as sterical configuration and intramolecular electron transfer processes. The more the configuration of the molecule differs in the electronic states between which transitions take place, the wider will be the separation between absorption and fluorescence spectra; i.e., the Stokes shift will be larger. In particular, the medium (solvent, polymer) has an effect on intramolecular electron transfer. The polarity or solvation properties of the solvent may affect the energy levels of the electronically excited singlet state. Polar solvents tend to increase the Stokes shift of a fluorescing molecule. In more viscous solvents or polymers, the orientational relaxation of the solvent molecules may not occur within the limited lifetime of the excited singlet state, and fluorescence may come from excited singlet states that, in terms of energy and configuration, are closer to the Franck-Condon state than to the relaxed excited singlet state. This pattern would result in a smaller Stokes shift.

The molecular requirements and mechanisms for generating large Stokes shifts have been discussed over the past 25 years.[8–11] In principle, a purely photophysical mechanism and one in which an adiabatic photochemical reaction leads to a new fluorescing product are conceivable to produce large Stokes shifts.[10] Unfortunately, all these photochemical mechanisms leading to fluorescing molecules or species with large Stokes shifts generally show poor fluorescence quantum yields. Even the scintillator known as 3-HF (3-hydroxyflavone)[12] with an extremely large Stokes shift has a poor fluorescence quantum yield.[13] For the design of novel organic scintillators with large Stokes shifts, twisted intramolecular charge transfer[9,10] cannot be used because the scintillation process is restricted to unpolar aromatic solvents (e.g., toluene) or polymers (e.g., polyvinyl toluene, polystyrene) with high π electron densities. Thus, what generally remain are organic molecules in which, during electronic excitation, a change of physical properties occurs, which, in turn, gives rise to large Stokes shifts.

The class of sterically hindered pyrazoline compounds proved to possess both properties: large Stokes shifts and excellent fluorescence quantum yields.[3,13–15] A combination of two photophysical effects—namely, the internal rotation of a moiety to ease sterical hindrance[3,14] and the intramolecular charge transfer in the excited singlet state[16]—gives rise to these favorable scintillation properties. The amount of the structural change in the excited singlet state and the degree of electron transfer are determined by the polarity and viscosity of the solvent,[16] by the different substituents,[17] as well as by the size and electron-donating ability of the rotatable moiety in the pyrazoline compound.[14] The concept of different degrees of sterical hindrance in the ground and in the electronically excited singlet states proved to be valid for larger Stokes shifts in another class of organic fluorescers.[18] Furthermore, unlike in other organic scintillators, these structural features lead to a unique self-quenching behavior of the sterically hindered pyrazoline compounds in the excited singlet state.[15] This characteristic allows concentration values of the solute in large-volume scintillators or scintillating fibers at which only nonradiative energy transfer is effective.[15] The parent compound of these sterically hindered novel scintillators, 1-phenyl-3-mesityl-2-pyrazoline, known as PMP, has been successfully altered in order to obtain scintillators with even larger Stokes shifts and satisfactory fluorescence quantum yields.[13]

Comparison of Scintillator Self-Absorption with Other Light Losses in Scintillation Detectors. Light attenuation caused by self-absorption due to overlapping absorption and emission bands of the added scintillator must be compared with other light losses due to scattering and reflections in order to make a reasonable estimation of transmission performance.

The absorption length Λ_A depends on the decadic molar extinction coefficient ϵ_λ (l mole^{-1} m^{-1}) of the scintillator or wavelength shifter. It is defined[19] as the value of the light path Λ_A (m) after which the transmitted light intensity Y drops to $1/e$ of its initial value $Y(0)$:

$$\frac{Y_0}{e} = Y(\Lambda_A) = \int_{\mathrm{em}} I_\lambda^{\mathrm{em}} 10^{-\varepsilon_\lambda C \Lambda_A} \frac{\mathrm{d}\lambda}{\lambda^2}. \qquad (1)$$

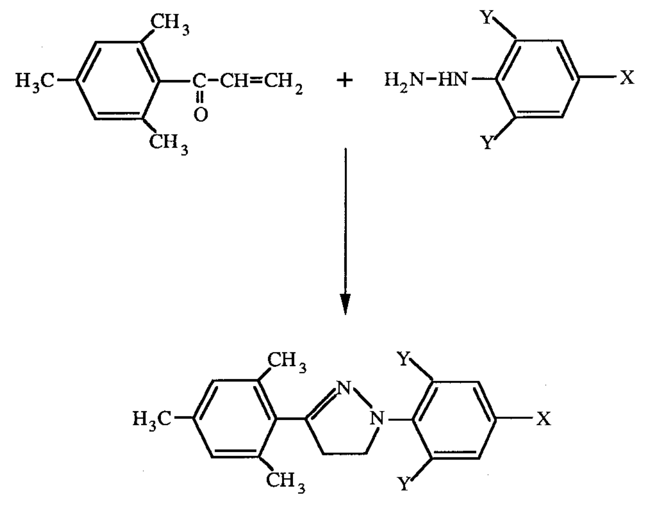
The emission intensity I_λ^{em} (normalized to unity) and ϵ_λ are integrated over the scintillators or wavelength shifters (concentration C [mole l^{-1}]) emission band.

Light scattering mostly occurs on statistical density variations and is attributed mainly to Rayleigh scattering, which varies inversely to the fourth power of the wavelength. The scattering length Λ_{SC} after which the initial light intensity Y_0 drops to $1/e$ is defined as

$$\frac{Y_0}{e} = Y(\Lambda_{SC}) = \int_{\mathrm{em}} I_\lambda^{\mathrm{em}} 10^{-(kC_d/\lambda^4)\Lambda_{SC}} \frac{\mathrm{d}\lambda}{\lambda^2} \qquad (2)$$

where C_d represents the concentration of density variations.

Reflection losses occur at imperfections at the core/cladding interface of the fibers. They cause small reductions from unity for the coefficient q of total reflections, and the transmitted light intensity I decreases exponentially with the number $N = tg\vartheta_d^L$ of reflections along a



PMP-420 : X = H, Y = H
PMP-430 : X = CH_3, Y = H
PMP-450 : X = OCH_3, Y = H
PMP-410 : X = CH_3, Y = CH_3

FIG. 1. Scheme for synthesis of PMP-420, PMP-430, PMP-450, and PMP-410.

fiber length L with the diameter d, for a given reflection angle ϑ:‡

$$\frac{I}{I_0} = q^N. \tag{3}$$

By integrating over the polar angle, we obtain as an approximation[20] the reflection length Λ_R of a fiber, after which the injected light intensity drops to $1/e$:

$$\Lambda_R = \frac{1.5}{(1 - q)_\lambda} \frac{n_1}{(n_1^2 - n_2^2)^{1/2}} d. \tag{4}$$

In a first approximation, the scattering length Λ_{SC} (from relation 2) and absorption length Λ_A (from relation 1) add up in scintillation liquids (relation 5), together with the reflection length Λ_R (relation 4) in scintillating fibers (relation 6) to become the total light attenuation length:

$$(\Lambda_{tot}^{liquid})^{-1} = \Lambda_{SC}^{-1} + \Lambda_A^{-1} \tag{5}$$

$$(\Lambda_{tot}^{fiber})^{-1} = \Lambda_{SC}^{-1} + \Lambda_A^{-1} + \Lambda_R^{-1}. \tag{6}$$

Relations 1, 2, and 4 show that Λ_{tot} is not really the attenuation length of an exponential but represents the approximation of many exponentials varying with the light path length due to the broad emission band of the scintillator.

SYNTHESES OF NEW PYRAZOLINE COMPOUNDS WITH LARGE STOKES SHIFTS

PMP-420,§ PMP-430, PMP-450, and PMP-410 were synthesized by condensation of mesityl vinyl ketone with

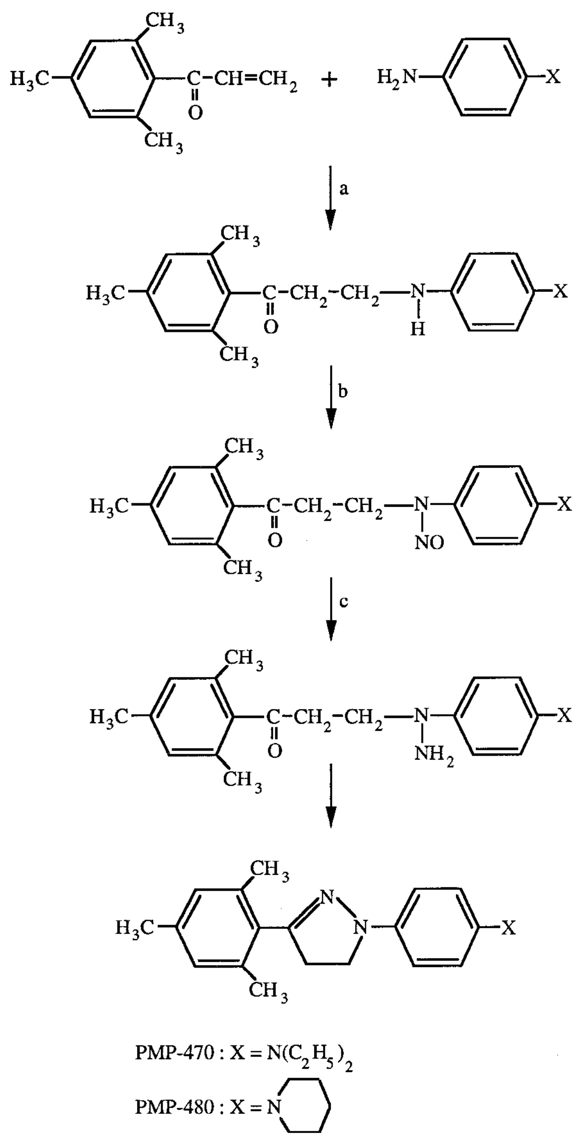


FIG. 2. Synthesis of PMP-470 and PMP-480. The amino group of a p-substituted aniline reacts with the carbon double bond of mesityl vinyl ketone. The secondary amine is nitrosated. Finally, the nitroso group is reduced with zinc and acetic acid to close the 2-pyrazoline ring via an intermediate hydrazine.

phenylhydrazine, p-tolylhydrazine, p-anisylhydrazine, and mesitylhydrazine, respectively[21] (Fig. 1) and PMP-440 and PMP-460 by condensation of the appropriate Mannich base of 2,6-dimethoxyphenylacetophenone, with p-tolylhydrazine and p-anisylhydrazine respectively.[21] PMP-470 and PMP-480 were prepared by the method that Mannich and Heilner[22] used for the synthesis of 1-methyl-3-phenyl-2-pyrazoline: the amino group of an appropriately p-substituted aniline reacts with the carbon–carbon double bond of mesityl vinyl ketone, the secondary amine is nitrosated, and, finally, the nitroso group is reduced with zinc and acetic acid to close, in one step, the 2-pyrazoline ring, via hydrazine intermediate (Fig. 2). The procedure of this synthesis is a reversal of previous methods, i.e., the intermolecular condensation of a carbonyl compound with a hydrazine derivative.

The N,N-diethylamino and the piperidino groups are strong electron-donating substituents and decrease the

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‡ Since some of the imperfections at the core/cladding interface cause Rayleigh-scattering, the $(1 - q)$ values in relation 3 show also a wavelength dependence.

§ From now on we will call the basic PMP compound (1-phenyl-3-mesityl-2-pyrazoline[3]) PMP-420. In the same way, we label similarly structured PMP derivatives as 430, 440, 450, and 460[13] as well as the newly synthesized ones as 470, 480, and 410. These labels indicate approximately the wavelengths of their emission maxima.

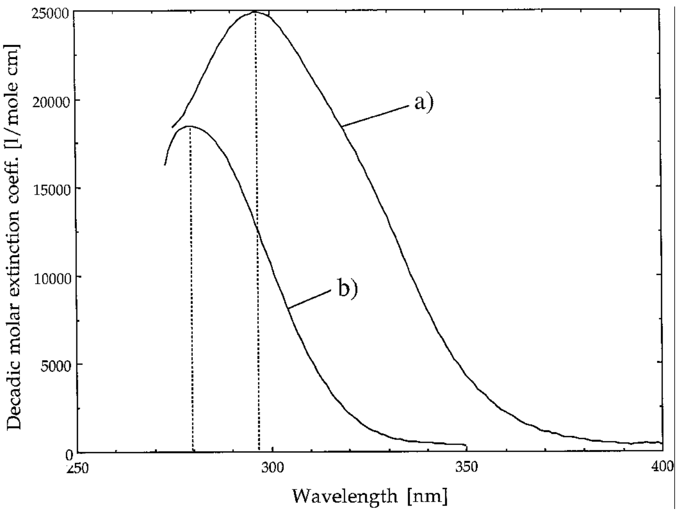

FIG. 3. Absorption spectra of scintillators measured with 10-μm-thick sample layers: (*a*) 0.01 molar PMP-420 solution in toluene; (*b*) 0.025 molar PMP-410 solution in toluene.

stability of hydrazine derivatives. Neither the 4-*N,N*-diethylamino nor the 4-piperidinophenyl hydrazine is commercially available, and their preparation by the reduction of diazonium salt of the corresponding aryl amines requires two steps as well as laborious handling precautions.[23] Again, the instability of these electron-rich intermediates increases the probability of competitive side transformations during the condensation reaction. On the other hand, the subsequent reaction of arylamine to mesityl vinyl ketone and the following nitrozation are performed very easily. The preformation of a carbon–nitrogen bond allows an immediate intramolecular reaction of the ketonic carbonyl group to take place with the amino functionality of nascent hydrazine during the reduction step. Keeping in mind that the commercially available starting products are the same under both methods, the reaction pathway used for the synthesis of PMP-470 and PMP-480, depicted in Fig. 2, appeared more interesting than the general method (Fig. 1) because of its higher selectivity and final yield.

All the PMP derivatives were purified by flash chromatography and crystallization techniques. They furnished a correct elemental analysis and were characterized by absorption and fluorescence spectroscopy, ^{1}H and ^{13}C NMR, and mass spectroscopy.[24,25]

PREPARATION OF POLYSTYRENE DOPED WITH NEW PYRAZOLINE COMPOUNDS

The preparation of scintillating plastics has already been described elsewhere.[26] Styrene was purchased from Aldrich.[‖] To eliminate inhibitors and contaminants, it was first filtered through basic aluminum oxide[¶] and silica gel,[**] and then distilled in vacuum at only 40 °C, to avoid polymerization to start during this process.

A glass tube (40 cm in length and 3 cm in diameter) was treated for 10 min with a 3% solution of dimethyldichlorosilane in chloroform to build up a hydrophobic monolayer of CH_3 groups. This coating prevented the sample from sticking to the walls after polymerization.

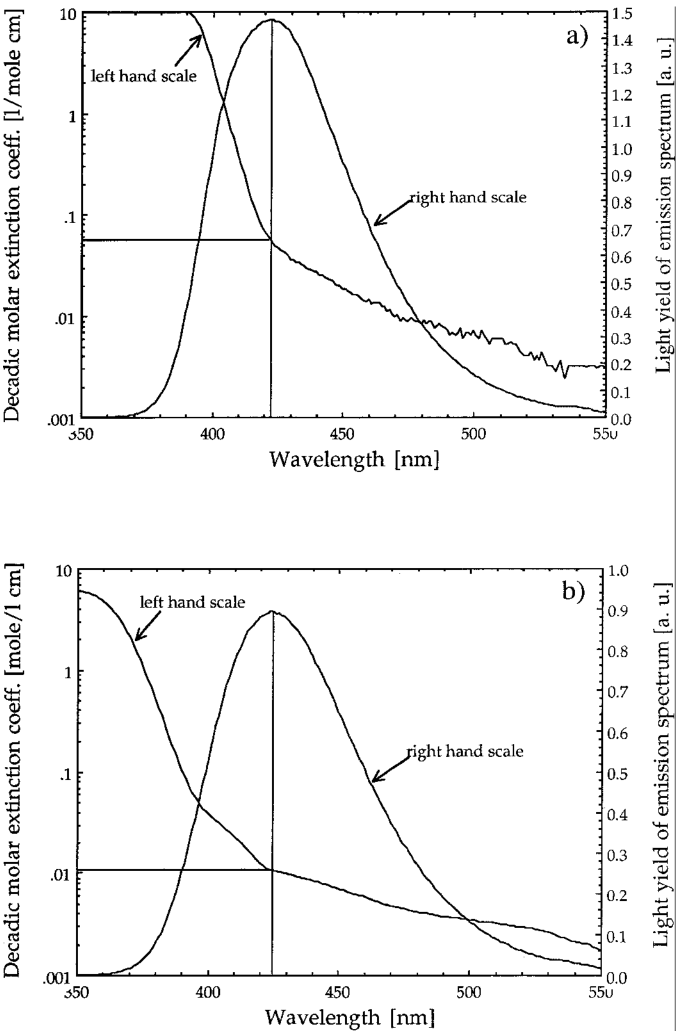

FIG. 4. Surface emission spectra (right-hand scales) and molar extinctions (left-hand scales) of the long wavelength absorption tails overlapping the emission bands: (*a*) 0.025 molar PMP-420 solution in toluene; (*b*) 0.045 molar PMP-410 solution in toluene.

A specified amount of the scintillator was introduced into the tube. Antioxidants were added, and the freshly distilled styrene was poured into the ampoule. The mixture was degassed in six successive freezing and thawing cycles, and the tube then was sealed under vacuum and placed for 5 days into an oil bath maintained at 130 ± 0.1 °C.

Finally a motor moved the sample downwards out of an oven (set to 130 °C) at a speed of 20 cm h^{-1}. In this way, it was progressively cooled down to room temperature, which prevented the appearance of bubbles in the bulk of the polymer. The glass tube was sawed across its upper part, and the polymer was easily extracted. Samples of different lengths were cut from the rod.

ABSORPTION AND EMISSION PROPERTIES OF THE NEW PYRAZOLINE COMPOUNDS DISSOLVED IN TOLUENE OR POLYSTYRENE

Spectral light absorption was determined with a double-beam spectrophotometer[‡‡] as described in Ref. 13.

[‖] Aldrich S497-2.

[¶] Aldrich 19,944-3.

[**] Merck index 11,8437.

[††] Lambda 6, Perkin-Elmer, Norwalk, CT, USA.

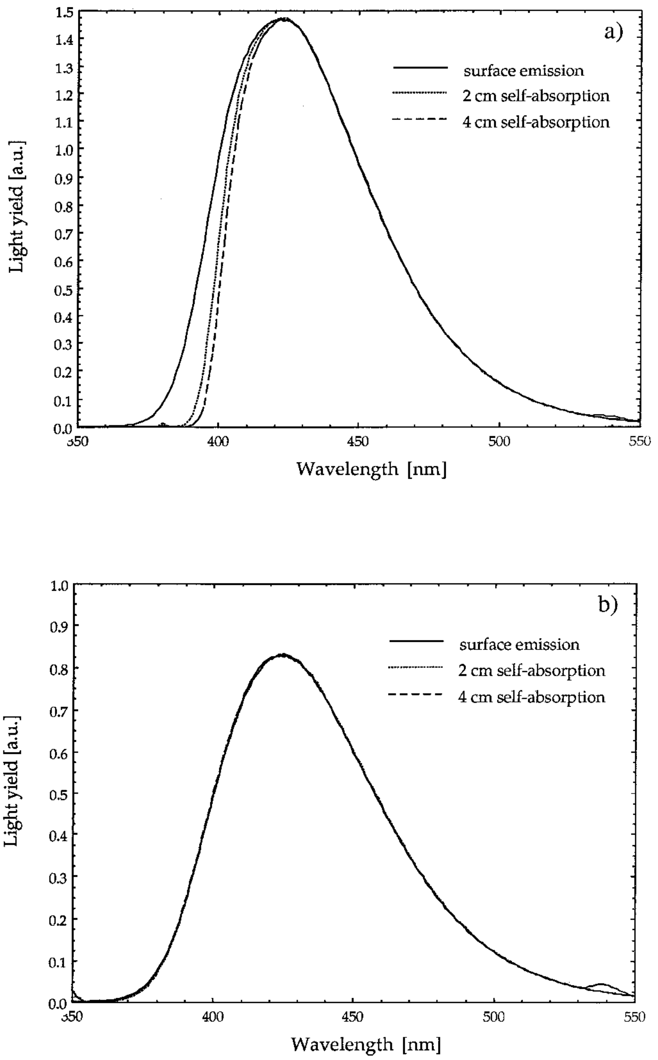

FIG. 5. Self-absorptions measured with zero, 20-mm, and 40-mm absorption samples (Fig. 1) in the light path of "surface emissions". Both emission curves are normalized at 440-nm wavelength: (*a*) 0.025 molar PMP-420 solution in toluene; (*b*) 0.025 molar PMP-410 solution in toluene.

The toluene solutions were contained in quartz cuvettes[‡‡] and the bulk polystyrene samples had polished end faces. Their optical pathlength d ranged from 0.01 to 100 mm, depending on the measured wavelength regions of the scintillators added.

The measured absorption maxima of 0.01 and 0.025 molar PMP-420 and PMP-410 solutions in toluene are shown in Fig. 3. Because of the large molar extinction coefficient, the sample thickness had to be reduced to 10 μm in order to obtain reasonable absorption ratios I/I_0 in the spectrophotometer. Below 275 nm wavelength, the toluene absorption band begins to mask the scintillator absorption.

Surface emission spectra measured with the spectrofluorimeter[§§] (see Ref. 13) are shown in Figs. 4a and 4b for PMP-420 and PMP-410 solutions in toluene. These figures also display the molar extinction coefficients for the overlapping parts of the PMP-420 and PMP-410 absorption bands. The measured self-absorptions for both scintillators dissolved in toluene are shown in Figs. 5a and 5b for 20 and 40 mm lengths of the absorption samples.[13] Whereas PMP-420 clearly shows a reduced emission band, the absorption of PMP-410 shows a zero effect on its emission (or fluorescence) yield. Self-absorption for the same scintillators, but this time added at 0.025 molar concentration to polystyrene, is plotted in Figs. 6a and 6b. Again, the PMP-410 emission band is hardly affected by a 1-mm absorption length, which normally reduces the largest fraction of the overlapping emission band.

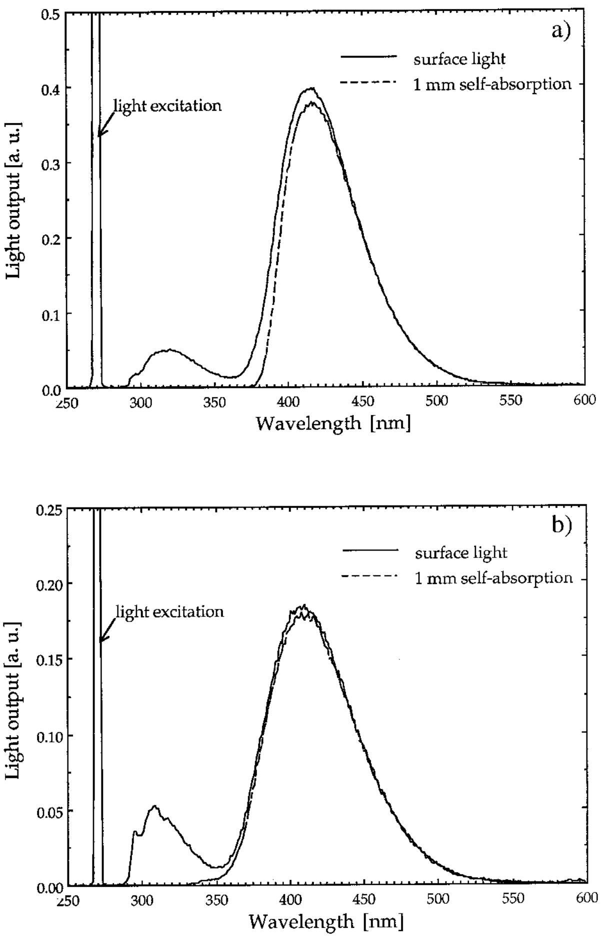

FIG. 6. Self-absorptions measured with zero and 1-mm absorption samples in the light path of "surface emissions". Both emission curves are normalized at 450 nm. The emission bands peaking at 320 nm are from direct excitations (at 270 nm) of the polystyrene.[19] They disappear with the absorption sample because of large polystyrene extinction coefficients: (*a*) 0.025 molar PMP-420 solution in polystyrene; (*b*) 0.025 molar PMP-410 solution in polystyrene. (Here, both emission curves coincide within their error margin.)

RESULTS AND DISCUSSION

The main results of our one-component scintillators so far synthesized and investigated are listed in Table I. Relevant values for the first five scintillators were already published in two earlier papers.[13,19] The last row shows for comparison the scintillator known as 3-HF[12] with Stokes shift and scintillation yield measured by us in the same way as for our compounds. Figure 7 shows the transmitted light intensities of the three newly synthe-

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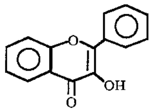
[‡‡] Hellma, Müllheim, Germany.

[§§] Model 8000, SLM Instruments Inc., Urbana, IL, U.S.A.

TABLE I. Properties of one-component scintillators dissolved in polystyrene (upper lines) and toluene (lower lines). The light yield is normalized to PMP-420. The structure formulae for the PMP-compounds are listed in Figs. 1, 2, and 3. Relevant values for the first five scintillators were already published.[13,19]

			Maxima		Stokes shift		
Compound	Molar mass g mole^{-1}	Melting point °C	Absorption nm	Emission nm	eV	cm^{-1}	Light yield
PMP-420	264	96	302	414	1.11	8960	100
			295	422	1.27	10,200	100
PMP-430	278	105–106	302	420	1.15	9300	102
			295	432	1.33	10,750	90
PMP-440	296	182–183	307	428	1.14	9210	96
			305	440	1.25	10,060	85
PMP-450	294	88–89	304	436	1.24	9960	83
			300	448	1.37	11,012	70
PMP-460	312	187–188	311	440	1.17	9430	79
			309	458	1.31	10,528	60
PMP-470	335	87	315	480	1.36	10,970	65
			310	495	1.50	12,140	33
PMP-480	347	133	314	479	1.36	10,970	50
			310	497	1.50	12,140	44
PMP-410	306	85	287	408	1.29	10,377	50
			280	423	1.50	12,074	60
3-HF[a]	238		343	528	1.27	10,215	27.2
			338	528	1.32	10,650	20

[a] 3-Hydroxyflavone



sized compounds and of PMP-420 and 3-HF, all dissolved in toluene. They were calculated according to Eq. 2 by taking the measured molar extinction coefficients $\epsilon(\lambda)$ for the different compounds and their concentrations C.

The light yields listed in Table I are all normalized to PMP-420 dissolved in toluene and in polystyrene, respectively. They differ considerably without any correlation to the Stokes shift or absorption length of the compound. The highest yields were obtained with PMP-420 and PMP-430. Unfortunately, PMP-410 and PMP-480 with their large Stokes shifts and absorption lengths yield only some 50% of the PMP-420 light emission.[||||] In comparison with the relatively small light yield of 3-HF, PMP-410 and PMP-480 compounds perform much better for applications with long scintillating fibers.

The PMPs combine very good photophysical properties such as high light yields and large Stokes shifts (see Table I) with excellent solubilities in toluene and polystyrene. Due to the sterical hindrance in the ground state of PMP-420, its solubility in toluene is nearly 1.3 molar.[15] Furthermore, the steric configuration of PMP is responsible for a unique self-quenching behavior. Compared to the standard scintillator PPO at 1 molar concentration in toluene where its light output dropped below 20%, there is a slight reduction of the scintillation yield of PMP-420 only at 1 molar concentration.[15] As has been discussed above, the sterical configuration of PMPs in the ground and electronic excited states gives rise to their large Stokes shifts.

In polystyrene, the Stokes shifts of the PMPs are generally smaller by 10–15% compared with those for toluene solutions; the light yields, however, are larger[||||] by 10–25% (in the case of PMP-470 even by 50%). It has been observed previously[27,28] that, when going from fluid solutions to polymeric matrices, an increase in fluorescence quantum yield of fluorophors occurs. Thus, giving a figure of merit for the use of PMPs in polymeric fibers, the smaller Stokes shift and the slightly longer absorption wavelength in polystyrene (see Table I) are compensated by a higher light output.

Even longer absorption lengths may be obtained by a more advanced polymerization technique. In fact, the produced polystyrene samples show some opaqueness, which may be caused by microscopic irregularities in the

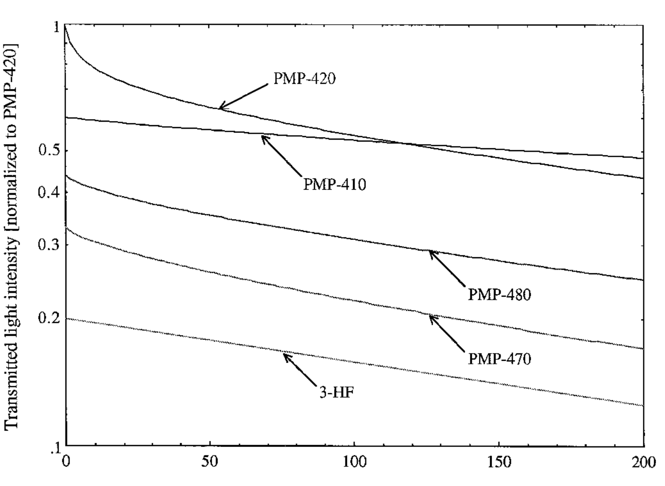


FIG. 7. Estimation of the transmitted light intensities for the newly synthesised dopants and for 3-HF dissolved in toluene. The curves are calculated from relation 1.

[||||] PMP-410 is the only compound on this list that does not increase its light yield in polystyrene compared to that in toluene. Probably some dissociation occurred during the polymerization process.

polymeric matrices. The light scattering at these irregular centers decreases the attenuation length.

CONCLUSION

Three new organic scintillators were synthesized on the basis of 1-phenyl-3-mesityl-2-pyrazoline (PMP). Two of them emit in the green region of wavelength (PMP-470 and PMP-480) and one in the blue region (PMP-410). Characteristics of all three scintillators are the very large Stokes shifts (larger than 3-HF) and consequently the quasi-absence of self-absorption. Their light yield amounts to 35–65% of that measured for PMP-420. They constitute an excellent alternative to 3-HF in the green region of emission (higher light yields) and an alternative to PMP-420 in applications where attenuation lengths longer that 1 m are required (large scintillator volumes or trackers).

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