

Tetraphenylpyrene-Bridged Periodic Mesostructured Organosilica Films with Efficient Visible-Light Emission

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Mesostructured organosilica films with strong blue fluorescence emission were synthesized by surfactant-templated sol-gel polycondensation using a 1,3,6,8-tetraphenylpyrene (TPPy)-containing organosilane precursor. The TPPy precursor, which contained four polymerizable silyl groups, was suitable for the preparation of mesostructured films with high TPPy content in the framework. The fluorescence quantum yields of the TPPy-bridged mesostructured organosilica films reached more than 0.7, despite the dense accumulation of TPPy units within the framework. Doping of the mesostructured films with fluorescence dyes enabled fine-tuning of the emission colors over a wide range of the visible spectrum. Such mesostructured organosilica films, in which different chromophores can be distributed into the framework and mesopores, have significant potential for luminescence applications.

Introduction

Periodic mesoporous organosilica (PMO) is a versatile inorganic/organic hybrid with mesoscale porous structures and molecular-scale functionalities.¹⁻⁴ PMOs are synthesized by surfactant-templated sol-gel polycondensation of organic-bridged alkoxysilane precursor compounds, generally represented as $R[Si(OR')_3]_n$ $(n \ge 2)$. Various organic species ranging from hydrocarbons and heteroaromatics to metal complexes have been introduced

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into the pore walls of PMOs as the R bridging group.^{5–18} The density, distribution, and molecular-scale ordering of the bridging groups within pore walls can be controlled by the molecular design of the precursors, optimization of the synthesis conditions, and appropriate selection of the co-condensation technique. $^{19-30}$ Expansion of the variation of organic bridges has broadened the potential applications of PMOs to optical materials, solid catalysts, sensing systems, and electronic devices.^{31–35}

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Article

Among these applications, organosilica hybrids with periodic mesostructures have significant potential for luminescence applications.^{36,37} Acidic sol-gel polycondensation of organosilane precursors in the presence of organic solvents yields highly transparent films favorable for optical materials.^{36–38} Different fluorescent chromophores can then be distributed into both the framework (as a bridging group) and mesopores (as a dye dopant) of PMO.^{13,14,39} In addition, PMOs have been found to exhibit light-harvesting antenna properties, i.e., the funneling of light energy absorbed by the framework organics into dyes placed in the mesochannels through efficient nonradiative energy transfer; therefore, flexible tuning of the fluorescence emission intensities of the framework and the dyes can be realized with almost no energy transfer loss.³⁹ These advantages of mesostructured organosilicas can lead the way to high-performance luminescence applications.

Recently, we reported the construction of highly fluorescent periodic mesostructured organosilica films.^{23,40} We focused on alkoxy-substituted oligo(phenylenevinylene) (OPV) precursors²³ as visible-light-absorptive chromophores and achieved efficient visible light emission in dye-doped mesostructured OPV-silica hybrid films.⁴⁰ The emission colors were successfully tuned from blue to white and yellow by combining the blue emission of the organosilica framework and the yellow emission of a doped dye, which was excited by fluorescence resonance energy transfer from the organosilica framework (Figure 1). However, the preparation of mesostructured films from 100% OPV precursors could not be achieved, probably because of weak interactions between the template surfactant micelles and the hydrolyzed precursors with only two silvl groups on both ends of the large OPV precursors. In addition, organosilica films with high OPV contents exhibited relatively low fluorescence quantum yields, because the hydrophobic OPV moieties tended to form nonemissive side-by-side aggregates in the pore walls.²³ Therefore, the addition of a large amount of pure silica precursors (67-80 wt % of tetraethyl orthosilicate (TEOS)) was required to form periodic mesostructures and achieve high fluorescence quantum yields, even though the strong absorption of excitation light with a high chromophore content is desirable for photoluminescent materials.^{36,37} Furthermore, vinylenes and acenes used as highly fluorescent components in our previous works^{22,23,40} are known to be photochemically reactive

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Figure 1. Schematic illustration of color-tunable fluorescence emission from dye-doped mesostructured organosilica film and photograph of a transparent organosilica film spin-coated on a glass substrate.

and unstable,^{41–45} which is problematic in practical applications. For the preparation of luminescent PMOs, it is desirable to introduce a high density of visible-light-absorptive, highly fluorescent, and photochemically stable chromophores into the mesostructured framework without strong quenching of fluorescence.

The aim of the present study was to introduce 1,3,6,8tetraphenylpyrene (TPPy) as a fluorescent bridging group in a mesostructured organosilica framework. The use of TPPy is suitable for the preparation of mesostructured and highly fluorescent organosilica films for the following reasons: (i) TPPy has an absorption in the near-UV to visible-light wavelength region and exhibits strong blue fluorescence;⁴⁶ (ii) a vacuum-deposited solid film consisting of 100% TPPy is reported to show a high fluorescence quantum yield (ca. 0.68),⁴⁷ which suggests that dense accumulation of TPPy units into pore walls is possible without strong fluorescence quenching; (iii) a four-armed organosilane precursor can be designed by the introduction of polymerizable silyl groups onto the phenyl substituents, which facilitates periodic mesostructure formation from a precursor having a large TPPy chromophore through interactions between the template surfactant micelles and the four hydrophilic silyl groups; and (iv) high photochemical stability can be expected for TPPy chromophores that contain neither vinylenes nor acenes.

In this article, we report the preparation of highly fluorescent mesostructured organosilica films from

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Figure 2. Chemical structure of the TPPy-Si organosilane precursor.

TPPy-Si containing TPPy as a bridging unit (Figure 2). The organosilica films exhibit efficient blue light emission with quantum yields of over 0.7. The emission colors can also be finely tuned over a wide range of the visible spectrum by the doping of fluorescent dyes into the mesostructured films.

Experimental Section

Materials and Methods. All reagents and solvents were purchased from Aldrich and Tokyo Chemical Industry and used without further purification. A poly(ethylene oxide)-block-poly-(propylene oxide)-block-poly(ethylene oxide) (PEO-PPO-PEO) triblock copolymer (EO20-PO70-EO20, P123, Aldrich) was used as a nonionic template surfactant. ¹H and ¹³C NMR spectra were measured using a Jeol JNM-ECX400P spectrometer. Mass spectra were recorded on a Bruker Daltonics Autoflex mass spectrometer with matrix-assisted laser desorption/ionization (MALDI). Infrared (IR) absorption spectroscopy (Thermo Nicolet Avatar 360 FT-IR) was conducted using an attenuated total reflection (ATR) attachment. X-ray diffraction (XRD) measurements were performed using $Cu-K\alpha$ radiation (Rigaku RINT-TTR, 50 kV, 300 mA). Transmission electron microscopy (TEM) of the organosilica hybrid films was performed using a Jeol JEM 2100F with an accelerating voltage of 200 kV. In order to obtain high-contrast images, the organosilica samples were annealed at 200 °C for 6 h and then immersed in ethanol for 6 h to extract P123. UV-vis absorption spectra were measured using a Jasco V-670 spectrometer. Fluorescence spectra were obtained with a Jasco FP-6500 spectrometer. Fluorescence quantum yields (within an error margin of \pm 3%) were determined using a photoluminescence quantum yield measurement system equipped with a calibrated integrating sphere (Hamamatsu Photonics, C9920-02). The Commission Internationale de l'Eclairage (CIE) 1931 chromaticity coordinates were calculated using a U6039 software (Hamamatsu Photonics).

Synthesis of TPPy-Bridged Precursor, 1,3,6,8-Tetrakis[4-(3-triethoxysilylpropylaminocarbonyloxy)phenyl]pyrene (TPPy-Si). 3-Triethoxysilylpropyl isocyanate (1.98 g, 8.00 mmol) was added to a mixture of 1,3,6,8-tetrakis(4-hydroxyphenyl)pyrene⁴⁶ (0.80 g, 1.40 mmol), dry dichloromethane (80 mL), and distilled triethylamine (10 mL). The reaction mixture was stirred at room temperature for 24 h and then refluxed for 2 h. After removal of the solvents using a rotary evaporator, the residue was dissolved in toluene and filtered to remove insoluble materials. Reprecipitation in toluene/hexane was repeated twice, and the precipitate was collected by suction filtration

Table 1. Composition and Structural Properties of Organosilica Films

sample	P123/ TPPy-Si / TEOS ^a (mg)	mesoscale periodicity ^b (nm)	
F-0a	40/60/0	8.8 (broad)	
F-0b	60/60/0	9.1 (broad)	
F-0c	80/60/0	9.7 (broad)	
F-1a	40/30/30	8.7	
F-1b	60/30/30	10.0	
F-1c	80/30/30	10.5	
F-2a	40/20/40	9.0	
F-4a	40/12/48	10.0	

^{*a*}Weights of the components dissolved in 2.0 mL of solvent. ^{*b*}The largest *d*-spacing value obtained from XRD measurements.

and dried under reduced pressure to afford a light yellow solid (1.62 g, 74%). ¹H NMR (CDCl₃) δ 8.16 (s, 4H), 7.97 (s, 2H), 7.63 (d, J = 8.3 Hz, 8H), 7.31 (d, J = 8.3 Hz, 8H), 5.59 (t, J = 5.9 Hz, 4H), 3.87 (q, J = 7.0 Hz, 24H), 3.33 (m, 8H), 1.76 (m, 8H), 1.27 (t, J = 7.0 Hz, 36H), and 0.73 (t, J = 8.0 Hz, 8H) ppm; ¹³C NMR (CDCl₃) δ 154.5, 150.5, 137.7, 136.4, 131.3, 129.6, 128.0, 125.8, 125.2, 121.4, 58.4, 43.5, 23.0, 18.2, and 7.6 ppm; IR (ATR, cm⁻¹) 3329 (NH st), 3039, 2974, 2928, 2885, 1716 (C=O st), 1533, 1489, 1203, 1165, 1070; MS (MALDI, m/z) calcd for 1558.6790 [M]⁺, found 1558.6809.

Preparation of Organosilica Films. The TPPy-based precursor TPPy-Si, TEOS, and P123 with the compositions given in Table 1 were dissolved in 2.0 mL of a 1:1 (w/w) mixture of tetrahydrofuran (THF) and ethanol, followed by the addition of 2 M hydrochloric acid (8 μ L) and deionized water (40 μ L). The sol mixtures were stirred at room temperature for 24 h. Organosilica thin films were prepared by spin-coating of the solutions onto quartz substrates (4,000 rpm, 30 s) and drying under reduced pressure at room temperature for 24 h. The organosilica films were stored in a dark place prior to optical measurements. For the preparation of dye-doped mesostructured films, a 5-fold scale sol mixture was prepared and the total amount of the solution was adjusted to 10.0 mL using a volumetric flask $([\mathbf{TPPy}-\mathbf{Si}] = 9.6 \text{ mM})$ in order to accurately weigh the amounts of chromophores. Appropriate amounts of dyes (0-5.0 mol % ratios to the amount of TPPy units) were added to the sol solution using a 9.6 mM solution of dyes (solvent: THF for Bodipy, ethanol for Rhd6G). The dye-doped organosilica films were prepared in the same manner as the nondoped films.

Results and Discussion

Preparation of TPPy-Bridged Mesostructured Films. The precursor compound, **TPPy-Si**, was easily synthesized by the reaction of 1,3,6,8-tetrakis(4-hydroxyphenyl)pyrene⁴⁶ with 3-triethoxysilylpropyl isocyanate and purified by repetitive reprecipitation in toluene/hexane. Figure 3 shows UV–vis absorption and fluorescence emission spectra of **TPPy-Si** in a 2-propanol solution $(1.0 \times 10^{-5} \text{ M})$. The absorption maximum (λ_{max}) was 384 nm and the absorption edge reached into the visible light region (ca. 420 nm). The emission wavelength (λ_{em}) and fluorescence quantum yield (Φ_F) excited at $\lambda = 400$ nm were 423 nm and 0.92, respectively; the TPPy-bridged precursor is favorable as a blue-light-emitting component of mesostructured organosilica films.

Mesostructured organosilica films with a TPPy bridging group were obtained by acidic sol-gel polycondensation of **TPPy-Si** via an evaporation-induced



Figure 3. UV-vis absorption (gray) and fluorescence emission (black, excited at $\lambda = 400$ nm) spectra of **TPPy-Si** in 2-propanol solution.



Figure 4. XRD patterns of (a) F-na and (b) F-1a-c. (c) TEM images of the mesostructured films F-1a and F-4a.

self-assembly process. Table 1 gives the composition of template surfactant P123, TPPy-Si, and TEOS in the sol mixtures and the main d-spacing values (mesoscale periodicity) obtained from XRD measurements. The XRD patterns of all the films prepared in this study (denoted as F-na, F-nb, and F-nc; n = weight ratio of TEOS to TPPy-Si in the sol mixtures; a, b, and c indicate amounts of P123 (a, 40 mg, b, 60 mg, c, 80 mg)) indicated mesoscale periodicity of 8.7-10.5 nm. Figure 4a shows XRD patterns of the organosilica films prepared using the same amount of P123 (F-na). A fairly broad diffraction pattern is observed for F-0a, which indicates the formation of a less-ordered mesostructure. With an increase in the amount of TEOS, the diffraction peaks become conspicuous and the dspacing of the periodic mesostructure increases. Addition of TEOS is effective for the enhancement of mesoscale ordering, as reported for organosilica films containing large hydrophobic bridging units.¹⁹⁻²³ Increasing the amount of P123 in F-1 films (F-1a-c) also improves the ordering of the mesostructures, accompanied with an

Table 2. Optical Properties of TPPy-Si and TPPy-Silica Hybrid Films

sample	λ_{\max} (nm)	$\lambda_{\rm em} ({\rm nm})$	Φ_{F}	light transmittance ^b at 500 nm (%)
TPPv-Si ^a	384	423	0.92	
F-0a	387	458	0.70	96
F-0b	387	458	0.71	97
F-0c	387	458	0.71	97
F-1a	386	457	0.75	> 99
F-1b	387	457	0.74	> 99
F-1c	387	457	0.74	> 99
F-2a	387	445	0.76	> 99
F-4a	386	443	0.79	> 99

^{*a*} Measured in 2-propanol solution (1.0×10^{-5} M). ^{*b*} A quartz substrate was measured as the background.

increase in the *d*-spacing values (Table 1, Figure 4b). The formation of mesoscale porous structures was further confirmed by TEM observation of the thermally annealed and ethanol-washed samples (Figure 4c). For organosilica film F-1a, wormholelike disordered mesopores are observed. For F-4a prepared by addition of a large amount of TEOS, a mesochannel array with a periodicity of ca. 7 nm is formed in the film. The mesoscale periodicity of F-4a observed by TEM is smaller than that given in Table 1 because of contraction of the mesostructures by thermal annealing and template extraction (see the Supporting Information). Here, it should be noted that large amounts of the hydrophobic and bulky TPPy units were successfully embedded within the framework with the aid of enhanced interaction between the hydrophilic surface of the micelles and the TPPy precursor with the adopted silvl-terminated, four-armed molecular design.

Optical Properties of TPPy-Silica Mesostructured Films. The TPPy-bridged mesostructured films exhibited efficient blue light emission upon excitation by near-UV to visible light (Table 2). Figure 5 shows UV-vis absorption and fluorescence emission spectra of the F-na mesostructured organosilica films. The organosilica films exhibited λ_{max} of ca. 387 nm with strong absorption of visible light around $\lambda = 400$ nm (Figure 5a). Although the absorption intensities decreased with the decrease in the TPPy content, the spectral shape was unchanged and similar to that of the precursor solution (Figure 3), which indicated that the weak interaction between TPPy units was preserved even at high TPPy concentration. With respect to the emission spectra, all of the TPPy-containing mesostructured films exhibited strong blue emission around $\lambda = 460$ nm (Figure 5b). The Φ_F of **F-0a** upon excitation at $\lambda = 400$ nm was 0.70 (Table 2). It is remarkable that such a high fluorescence quantum yield is achieved for a mesostructured organosilica prepared from 100% organosilane precursor TPPy-Si. The TPPy unit in the framework is not likely to form strong quenching sites, even in the densely accumulated state.⁴⁶ The $\Phi_{\rm F}$ values of the organosilica films were increased up to 0.79 by the addition of TEOS (Table 2). The emission spectra of **F**-na gradually shifted to the shorter wavelength region with an increase in the amount of added TEOS, as shown in Figure 5b, which indicates that highly fluorescent monomer-like TPPy species increased in the organosilica



Figure 5. (a) UV-vis absorption and (b) fluorescence emission spectra (normalized at 455 nm) for F-na. The excitation wavelength is 400 nm.

framework by further isolation of TPPy within the pure silica framework. Therefore, the dilution of TPPy units in the organosilica framework is effective for enhancing Φ_F , although the absorption intensity of the excitation light decreases.

On the other hand, the spin-coated films of F-0a-cprepared without TEOS showed a slight haze and 3-4% decrease in light transmittance at $\lambda = 500$ nm, whereas the films prepared with TEOS exhibited high transparency (Table 2). The baseline of the UV-vis spectrum of **F-0a** is shifted upward, due to partial scattering of the incident light, as shown in Figure 5a. Microscopic observation of the organosilica films revealed that the partial light scattering results from micrometer-scale wrinkles formed on the surface of the F-0 films (see the Supporting Information). The wrinkle formation is thought to be brought about by heterogeneous shrinkage during solvent evaporation, especially for the F-0 films made from 100% TPPy-Si. The TPPy-Si precursor is able to form hydrogen bonds through urethane moieties; therefore, the concentrated precursors can form a dense hydrogenbonded network that affects the reactivity of the silyl groups and the rate of evaporation of the solvents. The addition of TEOS possibly weakens the hydrogenbonded aggregation of the organic moieties during the evaporation-induced self-assembly process.48

To examine the hydrogen-bonded states and distribution of the TPPy component in the framework, infrared absorption spectra of the **F-na** organosilica



Figure 6. IR absorption bands of mesostructured films **F-na** corresponding to the C=O stretching mode of the urethane bond.

mesostructured films were measured. Figure 6 shows infrared absorption bands of the films in the range of $1680-1780 \text{ cm}^{-1}$, which corresponds to the C=O stretching mode of the urethane moieties of the organic bridge. The TPPy-silica hybrid films had two absorption bands at 1716 and 1742 cm⁻¹. The absorption band at 1742 cm⁻¹ is attributed to non-hydrogen-bonded C=O (see the Supporting Information). The main absorption band observed at 1716 cm⁻¹ indicates that most of the urethane bonds are in the weakly hydrogen-bonded state, because the strongly hydrogen-bonded C=O of urethane linkage is reported to exhibit an infrared absorption band at $1690-1700 \text{ cm}^{-1.49}$. Comparing the infrared spectra of the F-na films, the relative absorption intensity of the hydrogen-bond-free C=O at 1742 cm^{-1} increased with increasing amount of TEOS. These results suggest that the co-condensation of TPPy-Si with TEOS partially suppresses the hydrogenbonded aggregation of TPPy units in the organosilica framework, which may result in blue shifts of the fluorescence wavelengths, enhancement of the fluorescence quantum yields, and high transparency of the films by hindering the wrinkle formation during solvent evaporation.

Tuning of Emission Colors by Dye Doping. Doping of fluorescent dyes into the TPPy-bridged mesostructured films enabled flexible tuning of the fluorescence emission colors of the films within a wide range of the visible spectrum. As a host mesostructured film, F-1a was selected due to the high content of TPPy units in the film, good transparency, and a sufficiently high fluorescence quantum yield. Two dyes, green-light-emitting **Bodipy**⁵⁰ $(\Phi_F = 0.89 \text{ in 2-propanol})$ and yellow-light-emitting rhodamine $6G^{51}$ (**Rhd6G**, $\Phi_{\rm F} = 0.95$ in 2-propanol), were doped in the mesostructured film at concentrations of 0-5.0 mol % to the TPPy units (Figure 7). The broad fluorescence spectrum of **F-1a** ranges from $\lambda = 420$ to 550 nm and sufficiently overlaps with the absorption spectra of the fluorescent dyes; therefore, efficient excitation energy transfer from the TPPy-silica framework to the guest dyes can be expected. Figure 8 shows the emission spectra of the dye-doped F-1a films upon excitation at $\lambda = 400$ nm. The blue light emission of **F-1a** at

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⁽⁴⁸⁾ In the sol mixtures of F-0a-c containing 100% TPPy-Si as a silica source, precipitation of the TPPy derivatives and macroscopic gelation of the mixtures occur in a few days after the preparation of the sol mixtures. On the other hand, sol mixtures containing TEOS remain as homogeneous solutions for at least one month. These results indicate that the aggregation of TPPy-Si is suppressed by the presence of TEOS even in the solution states.

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Figure 7. Chemical structures of fluorescent dye dopants and UV–vis absorption and fluorescence emission spectra of the dyes in 2-propanol solutions.



Figure 8. Fluorescence emission spectra of the (a) F-1a/Bodipy(0-5.0 mol %) and (b) F-1a/Rhd6G(0-5.0 mol %) films. The excitation wavelength is 400 nm.

around $\lambda = 460$ nm was quenched with an increase in the dye concentration, with the subsequent appearance of emission bands attributed to the fluorescence of the doped dyes in the longer wavelength region. The two dyes do not exhibit absorption bands at $\lambda = 400$ nm (Figure 7); therefore, the fluorescence emission from the dyes is caused by excitation energy transfer from TPPy within the pore walls. The colors of the emissions from dye-doped **F-1a** were plotted in the CIE 1931 chromaticity diagram (Figure 9a). The emission colors of the **F-1a**/**Bodipy** films changed from blue with CIE coordinates of (0.15, 0.13) to green (0.22, 0.59). The **F-1a/Rhd6G** films



Figure 9. (a) Emission colors in the CIE 1931 chromaticity diagram calculated from the fluorescence emission spectra of the dye-doped F-1a films. (b) Photographs of the LED lamp (emission wavelength: 390 nm) coated with a F-1a/Rhd6G(1.0 mol %) film.

exhibited fluorescence emissions between (0.15, 0.13) and (0.41, 0.38). In particular, the F-1a/Rhd6G film containing 1.0 mol % Rhd6G exhibited a white light emission with CIE coordinates of (0.32, 0.34). The dye-doped F-1a fluorescent films were applied to the development of a white light-emitting diode (LED). Light scattering loss is generally inevitable in conventional white LED lamps consisting of near-UV (e.g., $\lambda = 400$ nm) LED chips and micrometer-sized phosphor powders; however, the organosilica films, which are transparent and flexible in shape, can suppress light scattering loss, leading to an improvement in the external quantum efficiency of the light-emitting system. Moreover, efficient light absorption and strong visible-light emission can be achieved because of the high content of fluorescent organic chromophores in the transparent films. The dye-doped TPPy-silica films were used as phosphors and LEDs as an excitation light source. Figure 9b shows that the near-UV emission of a commercial LED at $\lambda = 390$ nm was successfully converted to a white light emission by simply coating the transparent F-1a/Rhd6G(1.0 mol %) film (ca. 10 μ m thickness) at the curved surface of the LED.

The fluorescence quantum yields of the dye-doped films were significantly dependent on the guest dyes used. The $\Phi_{\rm F}$ of the **F-1a/Bodipy**(1.0 mol %) film upon excitation at $\lambda = 400$ nm was 0.74, which is comparable to that for the highly fluorescent nondoped **F-1a**. On the other hand, the $\Phi_{\rm F}$ of the **F-1a/Rhd6G**(1.0 mol %) film decreased to 0.43. The $\Phi_{\rm F}$ of the dyes doped in the

mesostructured films were also measured by direct excitation of the dyes. Although the $\Phi_{\rm F}$ value of **Bodipy** in the 1.0 mol % doped film was ca. 0.7-0.8 upon direct excitation at $\lambda = 485$ nm, that of **Rhd6G** excited at $\lambda =$ 530 nm was ca. 0.2–0.3, which is much lower than the $\Phi_{\rm F}$ in dilute 2-propanol solution. Decreased intensity and slight red shift of the fluorescence emission from Rhd6G at 2.0-5.0 mol % doping are also observed in Figure 8b, which indicates that the fluorescence emission of Rhd6G is partially quenched by dye aggregation in the mesostructured film. Hydrophobic dyes such as Bodipy can be well-dispersed in the hydrophobic cores of micellar aggregates of the template surfactants.⁴⁰ In contrast, hydrophilic dyes such as Rhd6G are located and concentrated in the proximity of hydrophilic pore walls, which might promote partial aggregation of the dye molecules.

Conclusions

The four-armed TPPy unit was shown to be suitable as a highly fluorescent bridging group for the formation of periodic mesostructured organosilicas. The TPPy unit was densely embedded within the framework of the mesostructured films without strong fluorescence quenching and the films exhibited efficient blue light emission with quantum yields of over 0.7. Fluorescent dye doping of the mesostructured films enabled color-tunable photoluminescence over a wide range of the visible spectrum, including a white light emission with CIE coordinates of (0.32, 0.34). The transparent and highly fluorescent organosilica hybrids with mesostructures have significant potential for various luminescence applications including LEDs and fluorescence sensing.

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Supporting Information Available: Optical photomicrographs of **F-0a** and **F-1a**, additional infrared absorption spectra, and XRD patterns of the films after extraction of the template surfactant (PDF). This material is available free of charge via the Internet at http://pubs.acs.org.