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Rapid gel-to-sol transition triggered by a photoacid generator under low-power light

Pengchong Xue,[a,b]* Jipeng Ding,[b] Jin Ming,[c] Ran Lu[b]

[a] Tianjin Key Laboratory of Structure and Performance for Functional Molecules; Key Laboratory of Inorganic-Organic Hybrid Functional Material Chemistry, Ministry of Education, College of Chemistry, Tianjin Normal University, Tianjin, 300387, P.R. China.

[b] College of Chemistry, Jilin University, Changchun, 130012, P. R. China.

[c] School of Materials Science and Engineering, Tongji University, Shanghai, 201804, P. R. China.

Experimental section

General information. All the raw materials were used without further purification. All the solvents as analytical reagent were purchased from Beijing Chemical Works (Beijing, China), and were used without further purification. Water used throughout all experiments was purified with the Millipore system. The UV-vis absorption spectra were obtained using a Mapada UV-1800pc spectrophotometer. Photoluminescence measurements were taken on a Shimadzu RF-5301 Luminescence Spectrometer. Transmission electron microscopy (TEM) images were observed with a Hitachi H-8100 apparatus by wiping the samples onto a 200-mesh carbon coated copper grid followed by naturally evaporating the solvent.

Gelation Test of Organic Fluids: The solution containing certain weighed hydrogen-bonded complexes in organic solvent was heated in a sealed test tube with 1 cm diameter in an oil bath until the solid was dissolved. After the solution was allowed to stand at room temperature for 6 h, the state of the mixture was evaluated by the "stable to inversion of a test tube" method.

Photolytic reactions of M-MePF6 in the dilute solution and the two-component gel were performed by irradiating the systems with a portable UV lamp (365 nm, 6w). Light intensity was measured using a light meter. Photoacid generation quantum yield was measured by using rhodamine B base as an acid indicator.

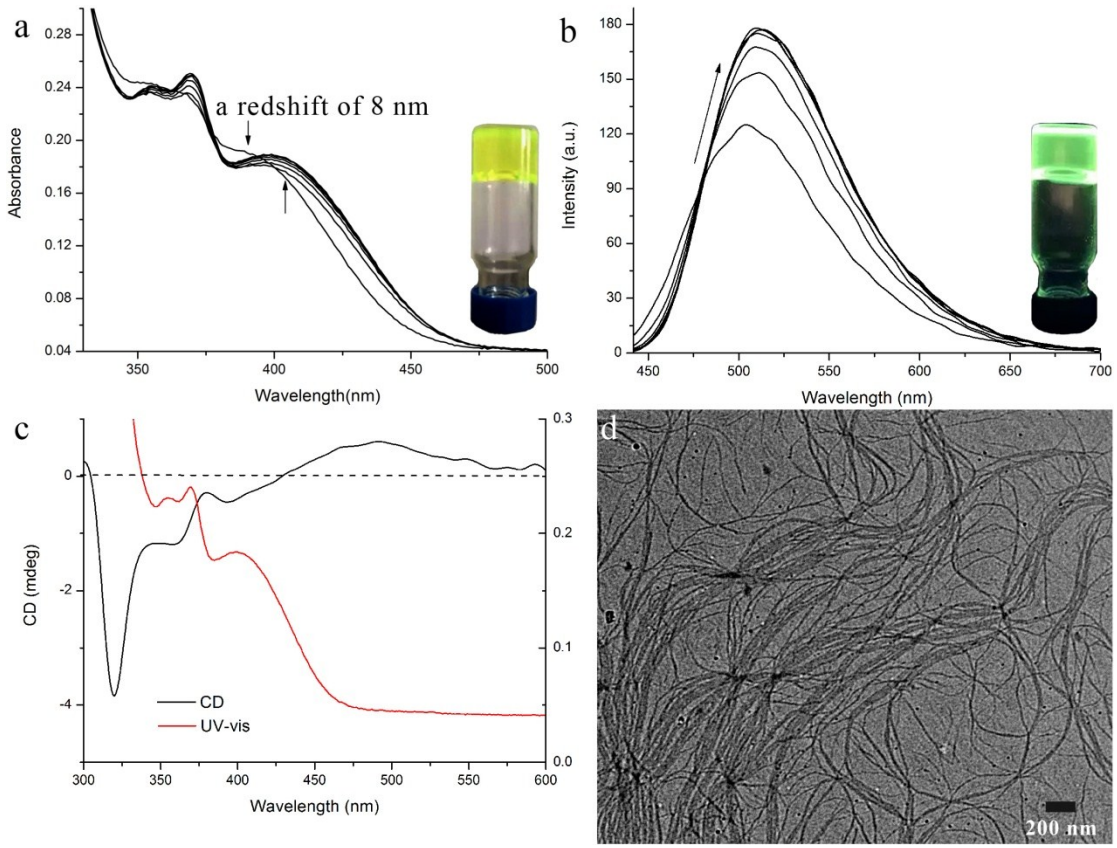

Fig. S1 Absorption (a) and fluorescence spectral (b) changes of **DCQ** in ODCB during gelation (from 120 to 20 °C). Concentration is 1.0 mg mL^{-1}, λ_{ex}= 400 nm. (c) Absorption and CD spectra of the gel and (d) TEM image of ODCB gel.

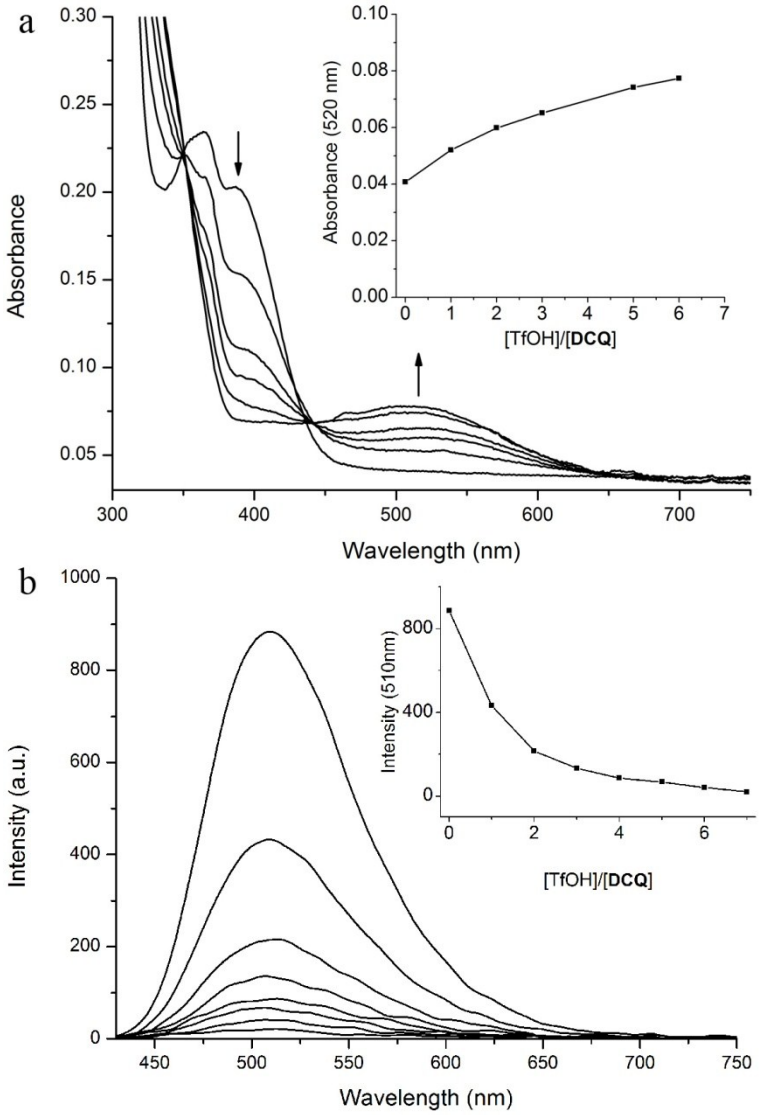

Fig. S2 (a) Absorption and (b) fluorescence spectral changes of **DCQ** in $CHCl_3$ solution (5×10^{-5} M) upon the addition of TfOH. λ_{ex} = 350 nm. Insets are the changes of (a) absorbance at 520 nm and (b) fluorescence intensity at 510 nm.

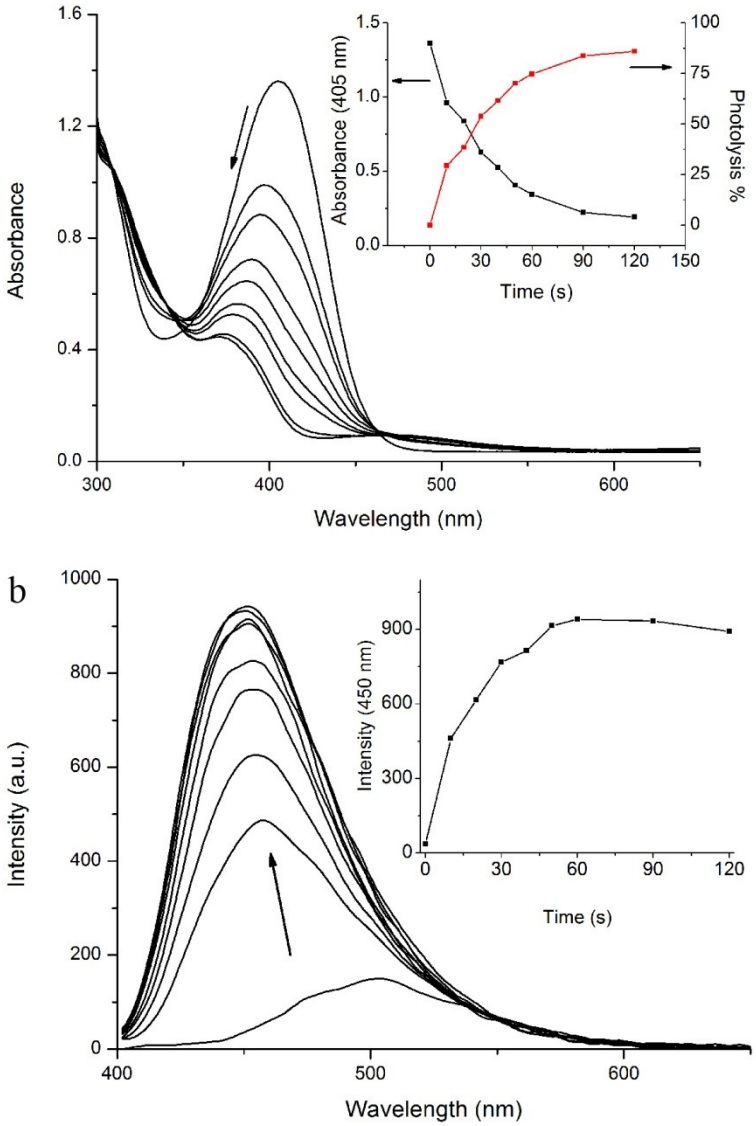

Fig. S3 Evolution of (a) absorption and (b) fluorescence spectra of **M-MePF6** in ODCB (5×10^{-5} M) upon irradiation at 365 nm (0.1 mW/cm^2). Insets are the changes of (a) absorbance at 405 nm and photolysis yield, and (b) fluorescence intensity at 450 nm.

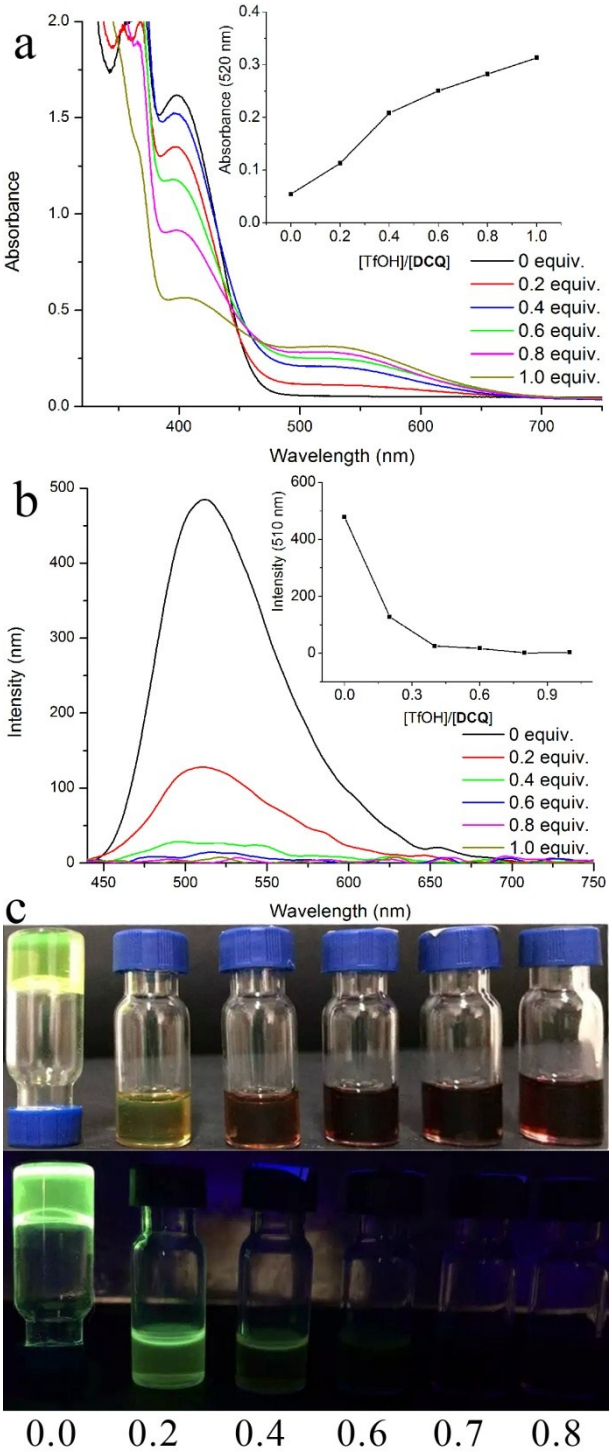

Fig. S4 (a) Absorption and (b) fluorescence spectral changes of **DCQ** in ODCB (1.0 mg/mL) upon the addition of TfOH. Insets are the changes of (a) absorbance at 520 nm and (b) fluorescence intensity at 510 nm. (c) Photos of **DCQ** in ODCB with different amounts of TfOH under (top) natural light and (bottom) 365 nm UV light.

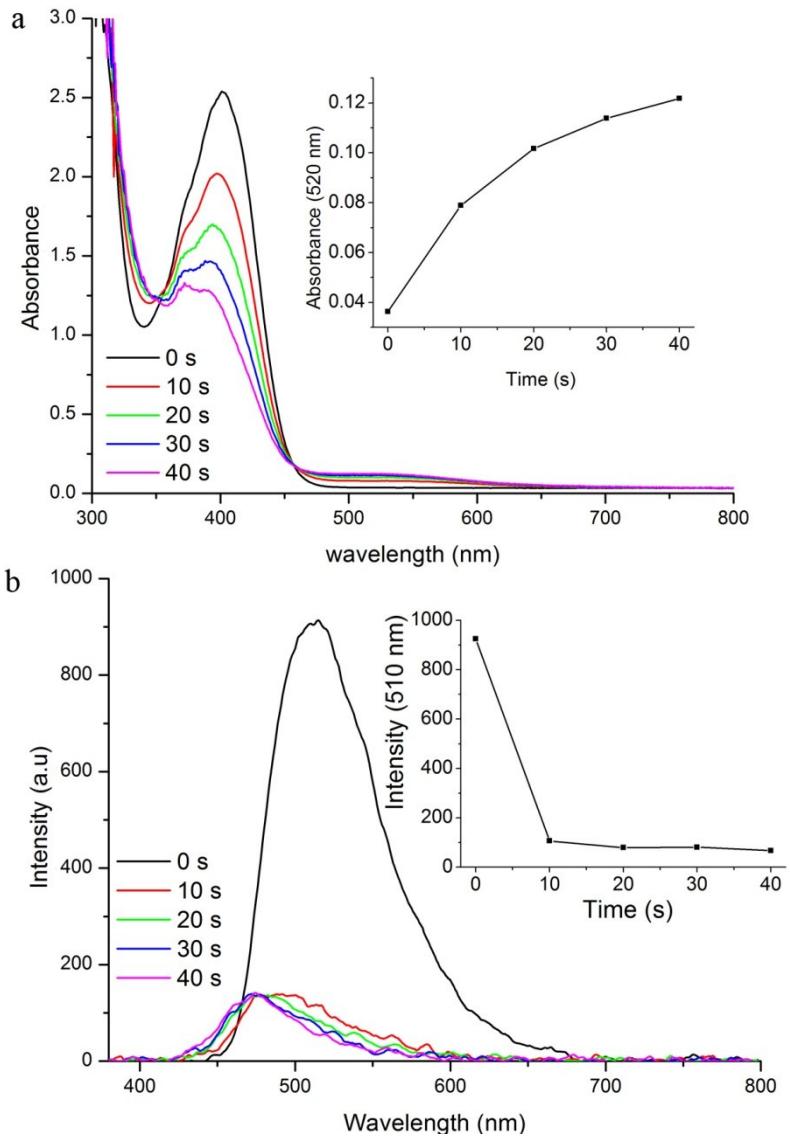

Fig. S5 Evolution of (a) absorption and (b) fluorescence spectra of the mixture of **DCQ**/**M-MePF6** (molar ratio = 1/3, [**DCQ**] = 1.27 ×10 $^{-3}$ M) in ODCB upon irradiation at 365 nm (0.1 mW/cm^2). Insets are the changes of (a) absorbance at 520 nm, and (b) fluorescence intensity at 510 nm.

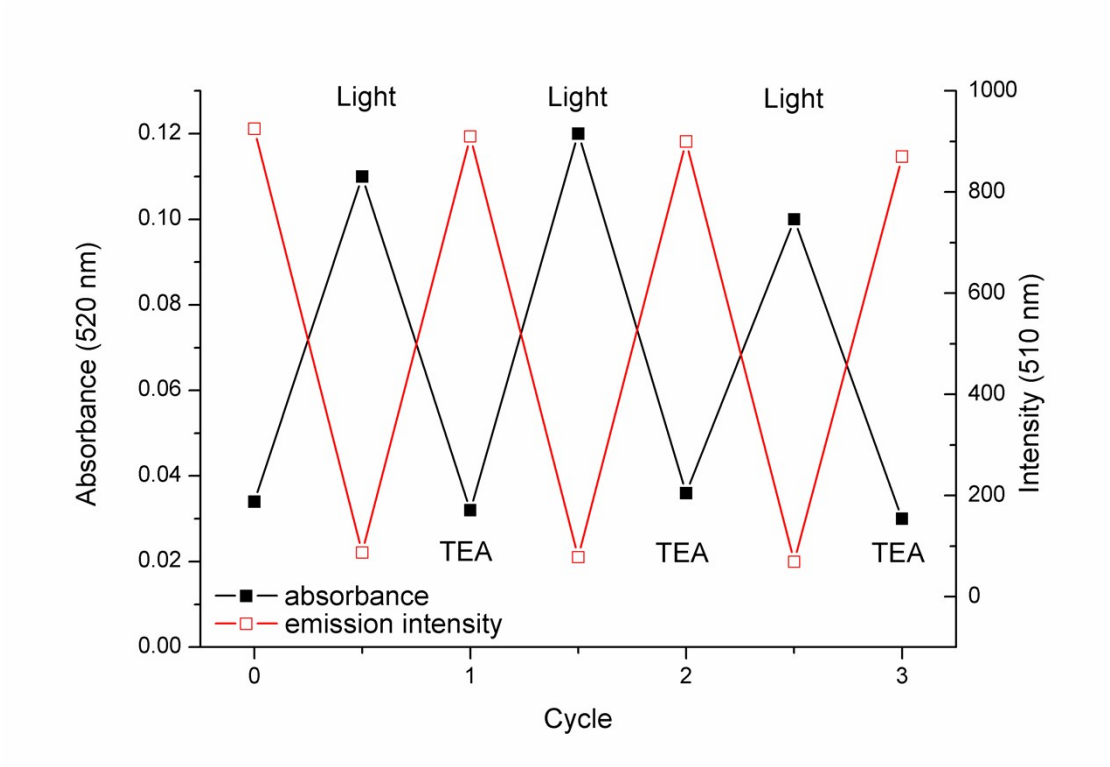

Fig. S6 Cycles of the gel-sol phase transition of the mixture of **DCQ**/**M-MePF6** (molar ratio = 1/3, [**DCQ**] = 1.27 $\times 10^{-3}$ M) upon irradiation at 365 nm (0.1 mW/cm^2) for 20 s and then addition of 0.2 equiv. triethylamine (TEA). The absorbance and the emission intensity were measured at 520 nm and 510 nm, respectively.