Electronic Supplementary Information

## Fast and Controlled Thermoresponse in Photoluminescence of Well-Designed Hydrogels of Two Separate Nanodomains with Solvatochromic Dyes

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Fig. S1 SEC curve of PDMAAm macro-CTA. Reaction conditions: [DMAAm] = 3000 mM, [CTA]= 10 mM, [AIBN] = 1.0 mM in 1,4-dioxane at 60 °C for 24 h (monomer conversion: 81 %).



**Fig. S2** <sup>1</sup>H NMR spectrum of PDMAAm macro-CTA. The DP<sub>n</sub> and  $M_{n, NMR}$  were calculated from the ratio of integral values between the signals *a* and *d*. Reaction conditions: see the caption of Fig. S1.



Fig. S3 Appearances of an aqueous dispersion of Nile Red (30  $\mu$ g/mL) under (a) visible light and (b) irradiation of UV light (wavelength: 365 nm).



**Fig. S4** Appearances under (a) visible light and (c) irradiation of UV light (wavelength: 365 nm), (b) UV-vis spectra and (d) photoluminescent spectra (dashed line: excitation, solid line: emission) of the reaction solutions for hydrogels with thermoresponsive CDs containing Nile Red under various concentration ratios ([NIPAAm] = 500, 750 and 1000 mM, [NIPAAm] + [DMAAm unit] = 2000 mM, [BIS] = 20 mM, Nile Red: 30 µg/mL in water). In the panel (c), the reaction solution under [NIPAAm] = [DMAAm] = 1000 mM is shown as the representative. The spectra of Nile Red dispersion are included in (b) and (d).



**Figure S5**. Effect of the composition of CD gels containing Nile Red on the absorption and photoluminescent properties in response to temperature change. UV-vis spectra of (a) NG<sub>500</sub> and (b) NG<sub>750</sub>, and photoluminescent spectra (dashed lines: excitation, solid lines: emission) of (c) NG<sub>500</sub> and (d) NG<sub>750</sub> at room temperature (blue lines) and after heating at 40 °C for 20 minutes (red lines).



**Figure S6**. Photoluminescent spectra (dashed lines: excitation, solid lines: emission) of NG<sub>1000BIS80</sub> at room temperature (blue lines) and after heating at 40 °C for 20 minutes (red lines).



Figure S7. (a) Appearances of  $FG_{1000}$  under irradiation of UV light (wavelength: 365 nm) at room temperature and upon heating at 40 °C for 20 minutes. (b) Photoluminescent spectra (dashed lines: excitation, solid lines: emission) of  $FG_{1000}$  at room temperature (blue lines) and after heating at 40 °C for 20 minutes (red lines). (c) Appearance of  $FG_{1000}$  after heating at 40 °C for 16 h. Noticeable syneresis was observed and the weight loss was ca. 50%.



Fig. S8 Time dependence of (a) UV-vis spectra, (b) the maximum absorption wavelength, and (c) the maximum absorbance of  $NG_{1000}$  during heating from 20 °C to 40 °C.



**Fig. S9** Time dependence of (a) UV-vis spectra, (b) the maximum absorption wavelength, and (c) the maximum absorbance of **NG**<sub>500</sub> during cooling from 40 °C to 20 °C. The sample was heated at 40 °C for 60 minutes before the measurement. The spectrum before heating is also shown in the panel (a).