Supporting Information

Interference of Solvatochromic Twist in Amyloid Nanostructure for Light-Driven Biocatalysis

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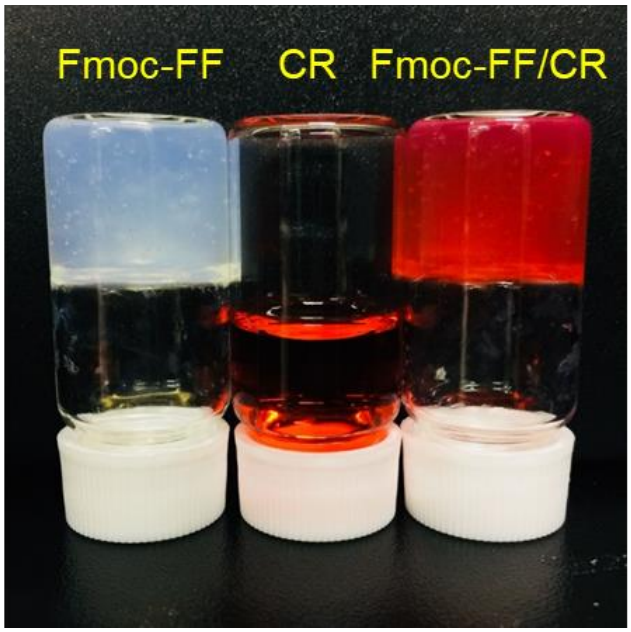

Figure S1. Photographs of CR solution, Fmoc-FF, and Fmoc-FF/CR hydrogels. The Fmoc-FF hydrogel was white, while the Fmoc-FF/CR hydrogel was red. The concentrations of Fmoc-FF and CR were 10 mg ml^{-1} and 200 μM, respectively.

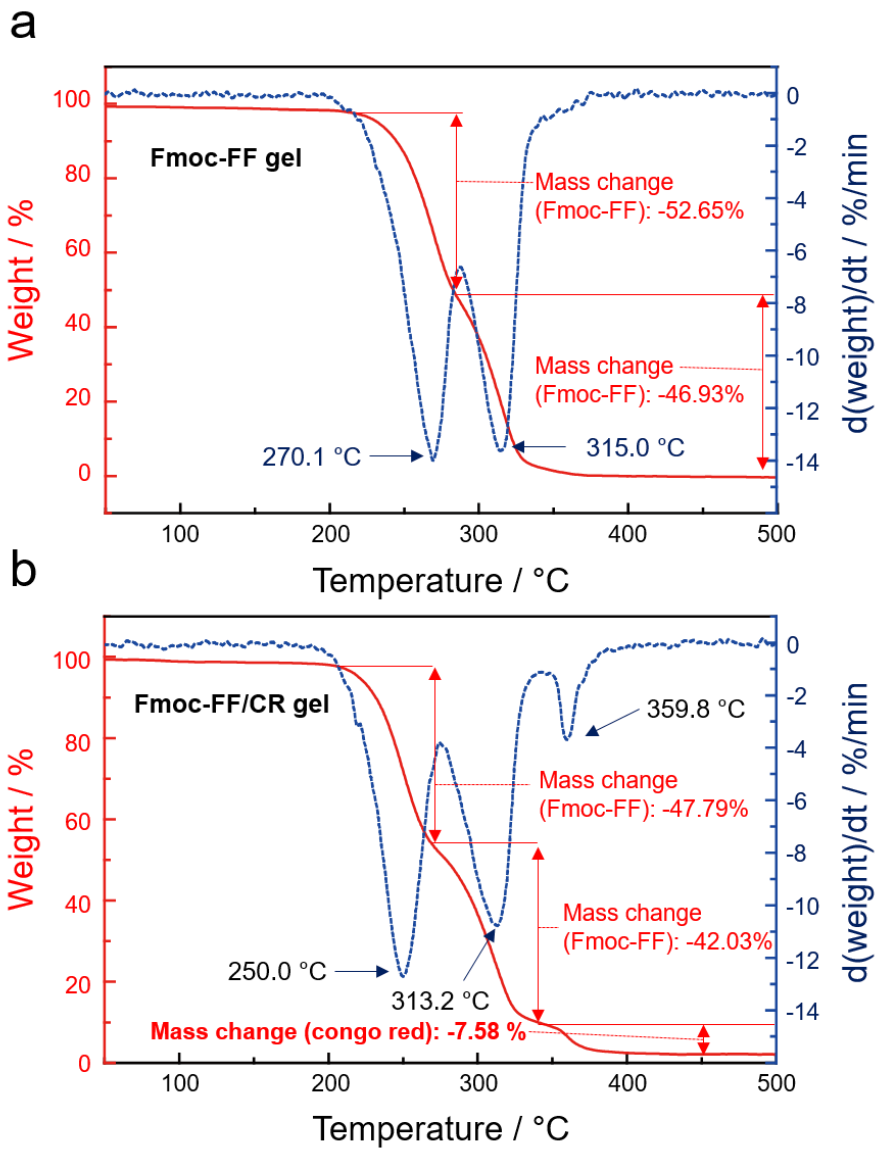

Figure S2. Thermogravimetric analysis of (a) Fmoc-FF and (b) Fmoc-FF/CR hydrogels. These two hydrogels were lyophilized before the analysis. The weight loss of the pristine Fmoc-FF hydrogel occurred from 200 to 350 °C. This thermal degradation range of the Fmoc-FF was similar in the Fmoc-FF/CR hydrogel; the CR dyes were thermally decomposed after 350 °C.

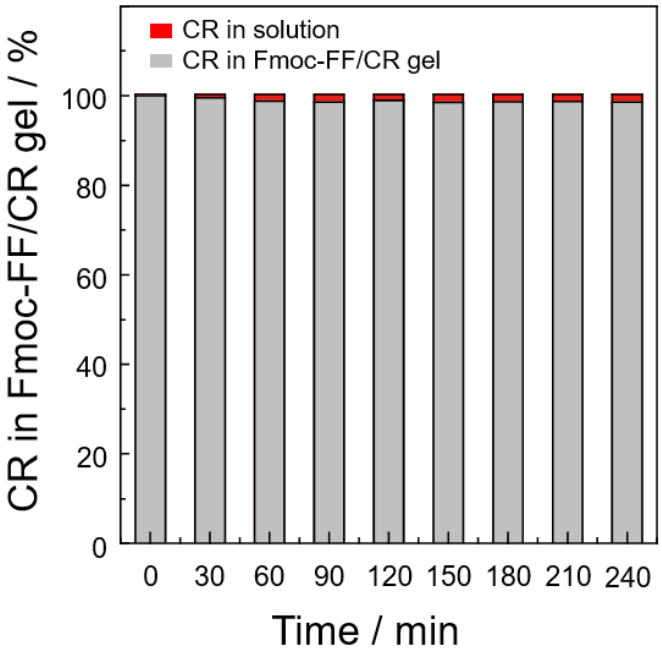

Figure S3. Time profile of the relative amount of CR in the supernatant and Fmoc-FF/CR hydrogel. Note that we injected deionized water above the Fmoc-FF/CR hydrogel and obtained the absorbance spectra of the supernatant to measure the concentration of free CR. Then, we estimated a relative amount of the CR in the Fmoc-FF/CR hydrogel by subtracting the amount of free CR in the solution from the initial amount of CR in Fmoc-FF/CR hydrogel.

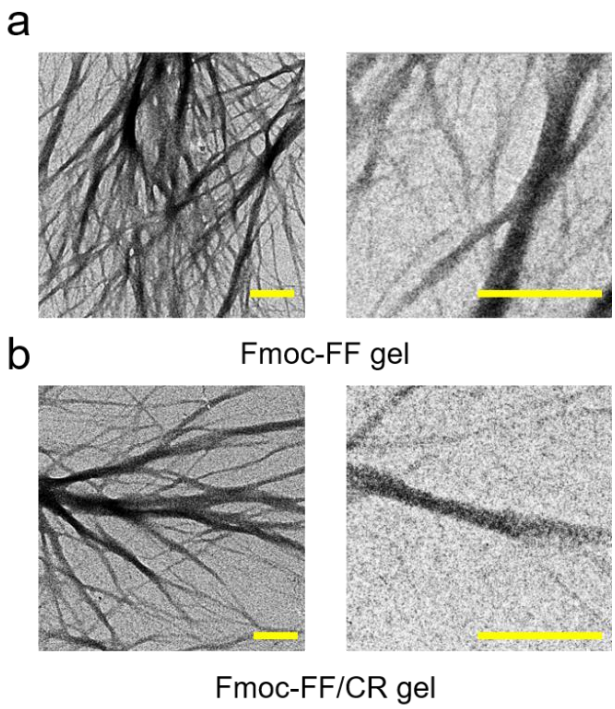

Figure S4. Transmission electron microscopy images of (a) Fmoc-FF and (b) Fmoc-FF/CR hydrogels. Scale bar: 500 nm.

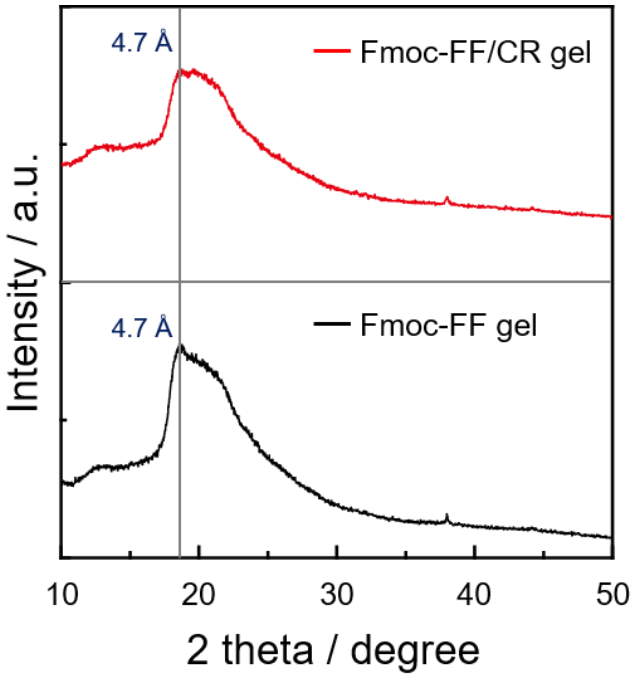

Figure S5. X-ray diffraction patterns of Fmoc-FF and Fmoc-FF/CR hydrogels. Cu Kα1 radiation wavelength: 1.5406 Å. The characteristic peak at 18.65º (d: 4.7 Å) stemmed from the interstrand distance of Fmoc-FF nanofibers (*Biomacromolecules* **2017**, *18*, 3551-3556).

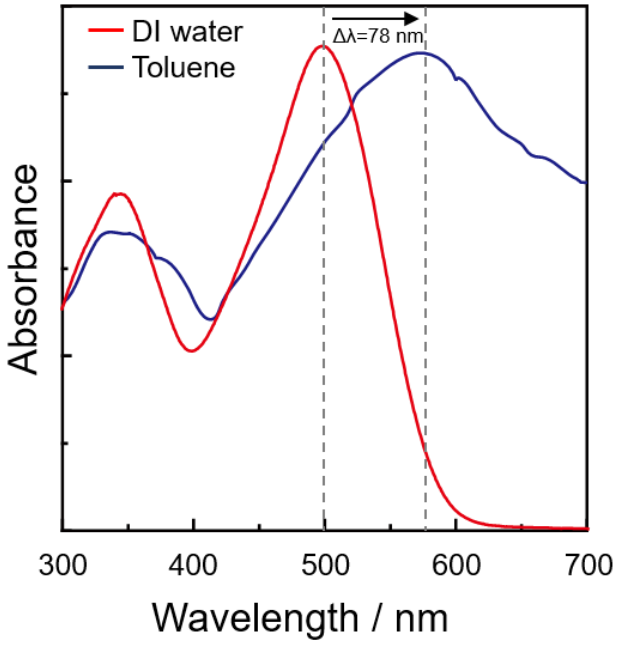

Figure S6. Absorbance spectra of the CR in deionized water and toluene to investigate the solvatochromic effect of free CR. The concentration of CR was 100 μM.

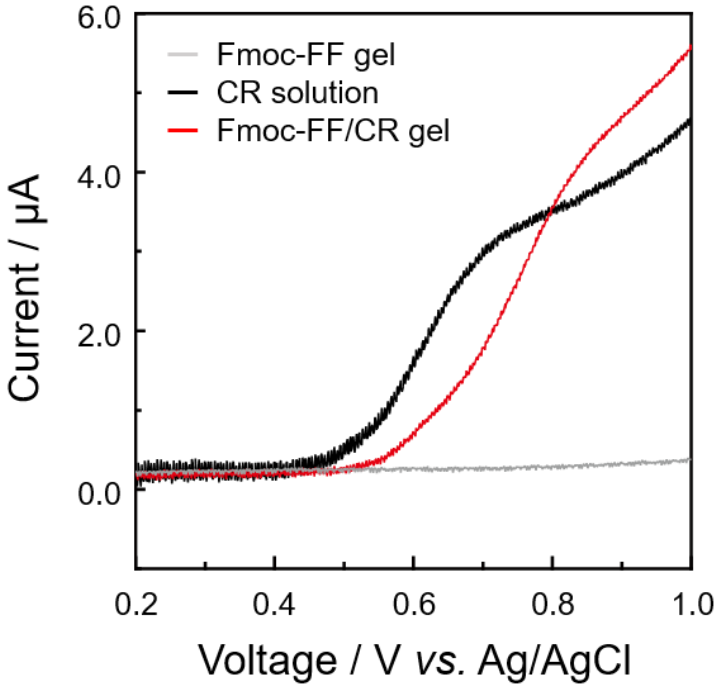

Figure S7. Linear sweep voltammograms of CR solution, Fmoc-FF and Fmoc-FF/CR hydrogels. Scan rate: 50 mV s^{-1}. The working electrodes of Fmoc-FF and Fmoc-FF/CR hydrogels were Fmoc-FF-coated ITO and Fmoc-FF/CR-coated ITO, respectively.

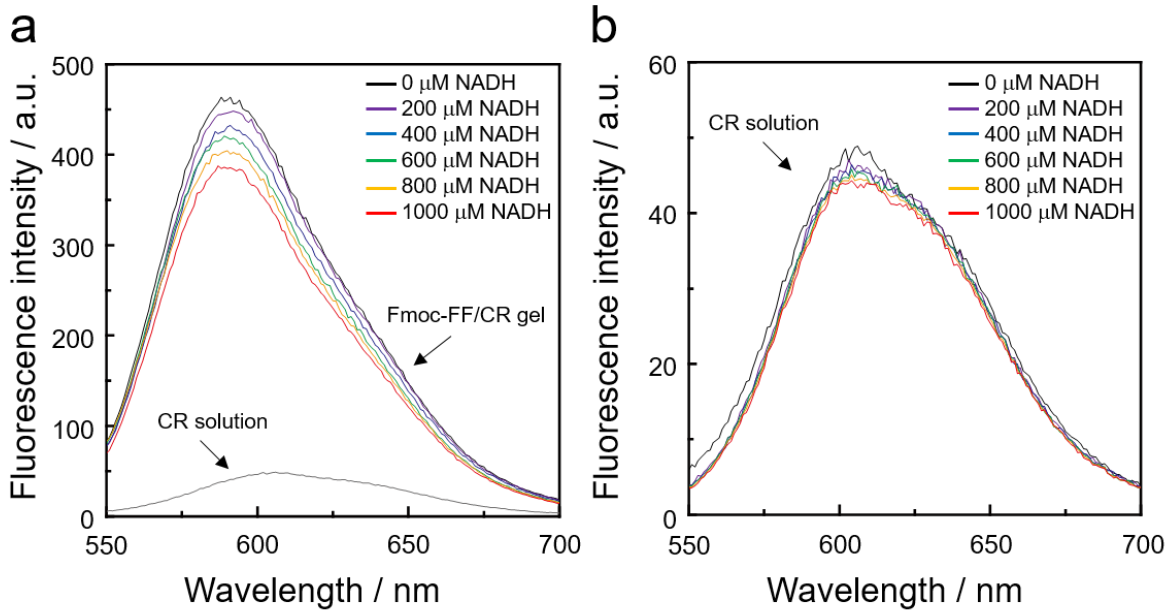

Figure S8. Fluorescence spectra of (a) Fmoc-FF/CR hydrogel and (b) CR solution at different concentrations of NADH. Excitation wavelength: 500 nm.

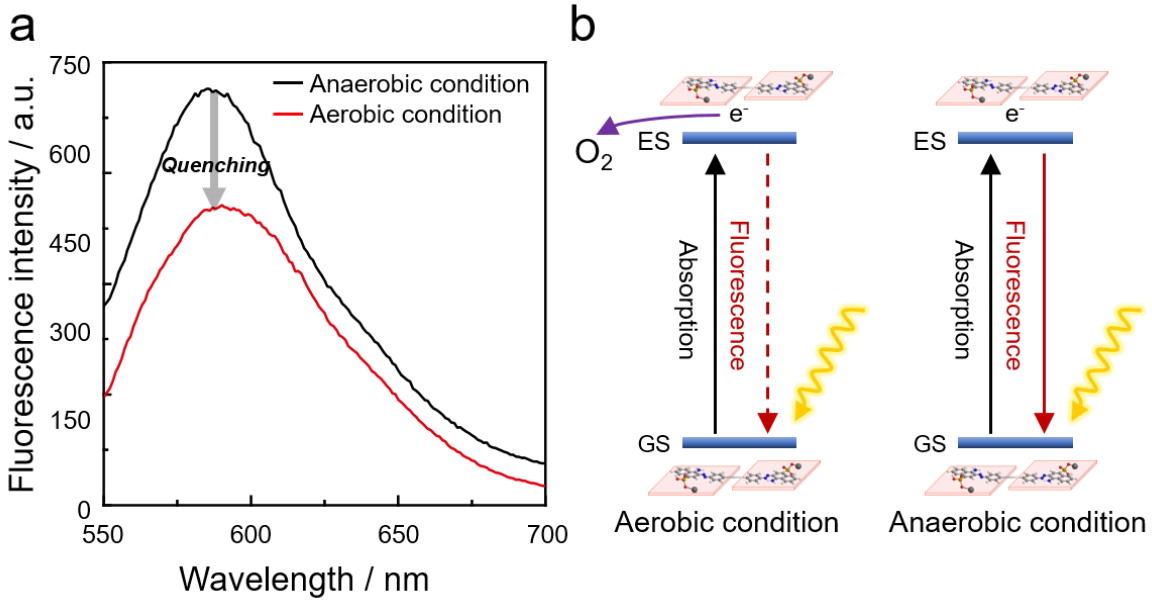

Figure S9. (a) Fluorescence spectra of Fmoc-FF/CR hydrogel under anaerobic or aerobic conditions. Excitation wavelength: 500 nm. The anaerobic solution was prepared by purging with Ar gas. (b) Illustration of an oxidative quenching of Fmoc-FF/CR hydrogel by O_2 molecules under light irradiation.

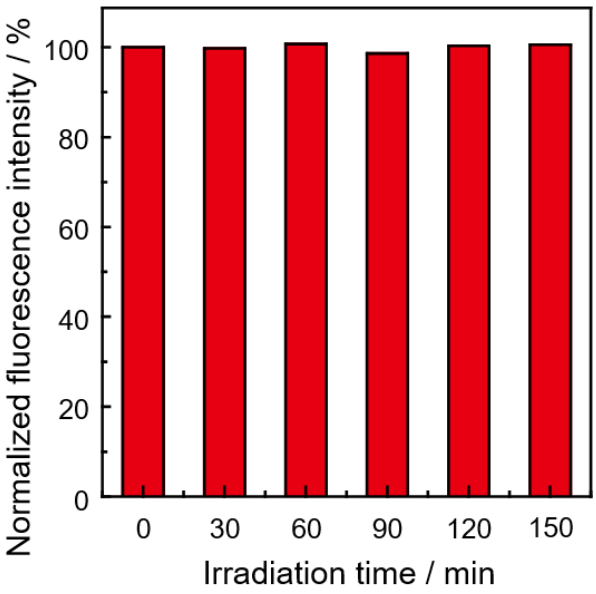

Figure S10. Time profile of normalized fluorescence intensity (I/I_0) of Fmoc-FF/CR hydrogel during photobiocatalytic oxidation. Excitation wavelength: 500 nm. Emission wavelength: 600 nm. Reaction condition: 10 mg ml^{-1} Fmoc-FF, 200 μM CR, 5 U ml^{-1} *Sc*ADH, 2 mM NADH, and 10 mM ethanol in a sodium phosphate buffer (25 mM, pH 8.8).

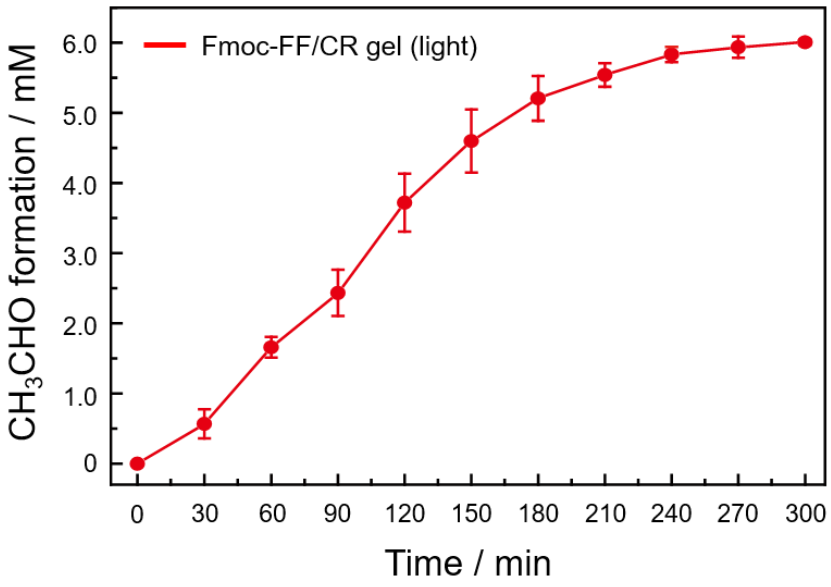

Figure S11. Time profile of photoenzymatic production by Fmoc-FF/CR hydrogel with *Sc*ADH. Reaction condition: 10 mg ml^{-1} Fmoc-FF, 200 μM CR, 5 U ml^{-1} *Sc*ADH, 2 mM NADH, and 10 mM ethanol in a sodium phosphate buffer (25 mM, pH 8.8).