## Experimental section

### Materials

All chemicals were purchased from Shanghai Macklin Biochemical Co., Ltd. and Guoyao Chemical Reagent Co. Ltd, Shanghai, which were used without further purification. CSC was synthesized according to the previous report. [S1]

### Characterizations

Proton nuclear magnetic resonance spectra were obtained using Bruker AM-400 spectrometer at room temperature with tetramethylsilane (TMS) as the reference. Transmission electron microscope (TEM) was measured on a JEM-100CX II electron microscope. The samples for TEM detection were dropped in the copper grid and air-dried. CD and CPL were collected with an Applied Photophysics ChirascanV100 model. Scanning electron microscope (SEM) images were recorded using a Zeiss scanning electron microscope.

The samples for SEM detection were dropped in the silicon pellet, sucking most of solvents by a filter paper and air dried, followed by gold spraying. UV-vis spectra were measured by Agilent ultraviolet visible near infrared spectrophotometer Cary5000. FL spectra were collected by a Shimadzu RF-6000.

### MD simulation of 3-CSC and 4-CSC

The native structures of 3-CSC and 4-CSC were built from the GaussView6.0 program. The obtained configurations of CSCs were initially optimized. All MD simulations were implemented with the GROMACS 2020 program. The water solvent simulated was the SPC216 model. The organic molecules and solvent were coupled to

temperature in 298K. The cut-off distance for non-bonded interactions was set to 1 nm. Energy minimization was conducted using the steepest descent algorithm before performing dynamic simulations. MD simulations for two systems were carried out for 20 ns with a time step of 0.002 ps per integration step under the ensemble conditions of T = 298 K. All S-6 simulations were visualized using VMD program.

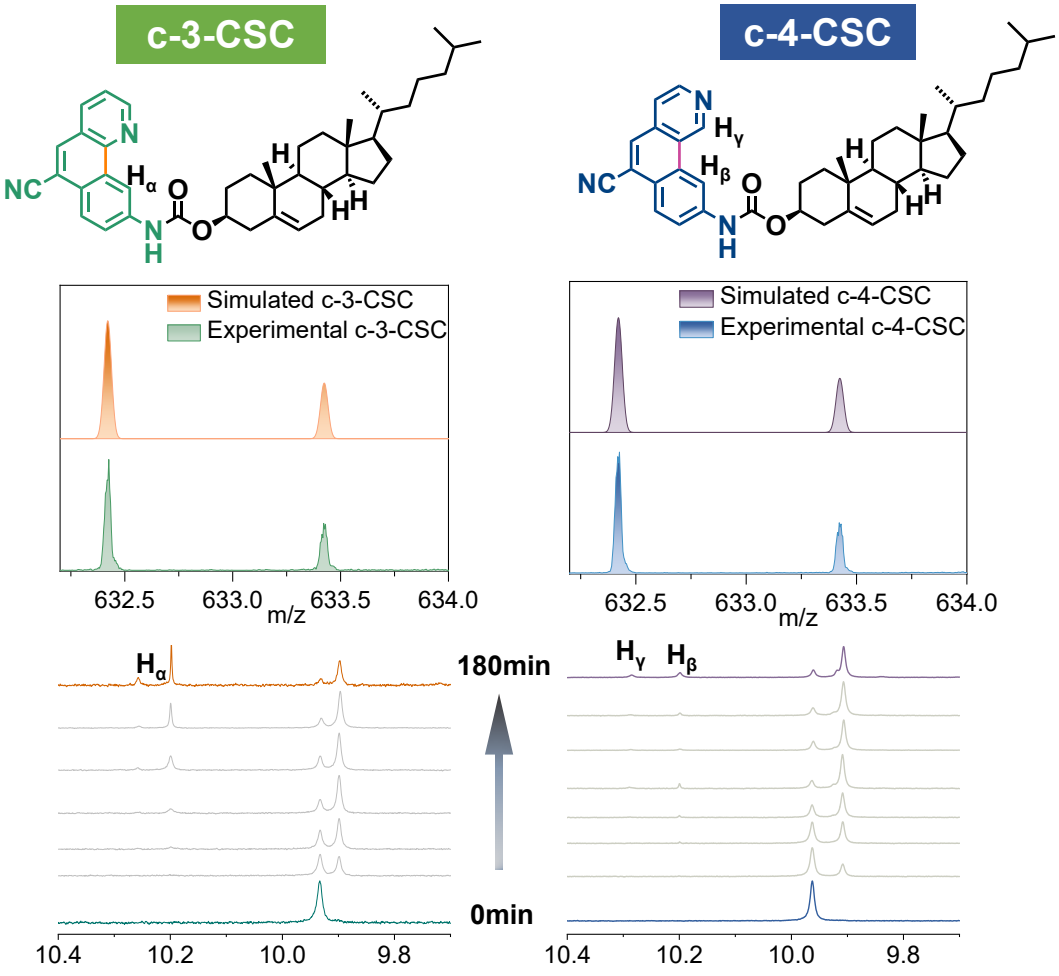


**Figure S1.** Molecular formula, mass spectrum (THF), $^{1}H$ NMR (DMSO-$d_6$) spectrum of photocyclization.

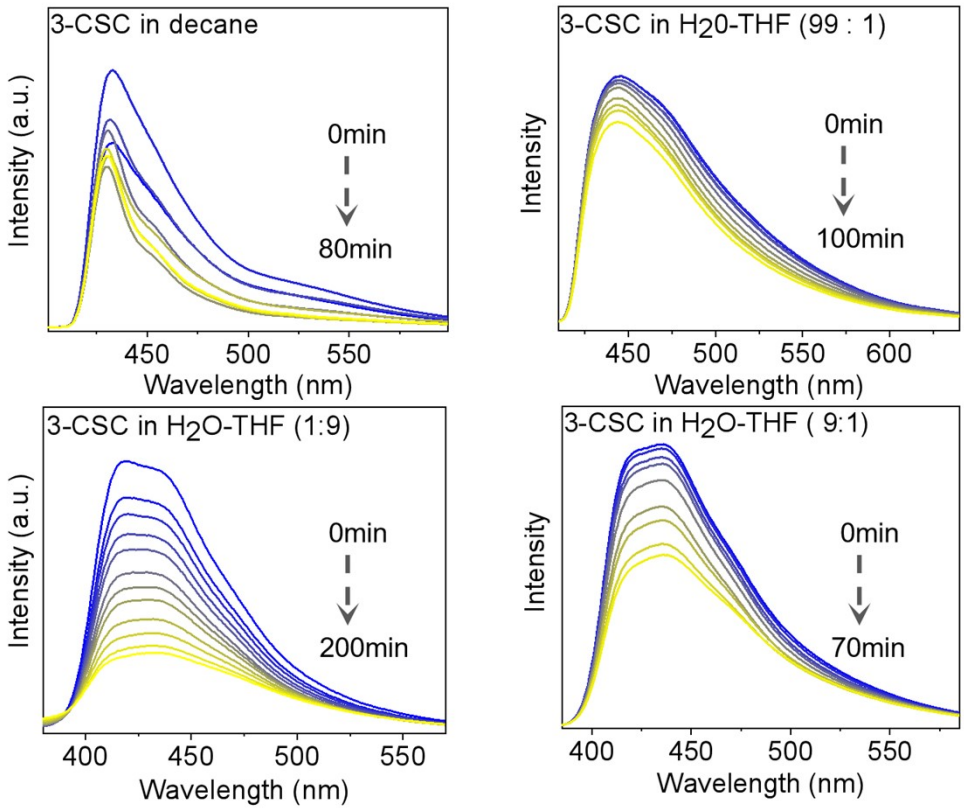


**Figure S2.** PL spectra of Z-3-CSC (0.05 mM) in different solution irradiated with 365 nm UV light from a hand-held UV lamp for different time.

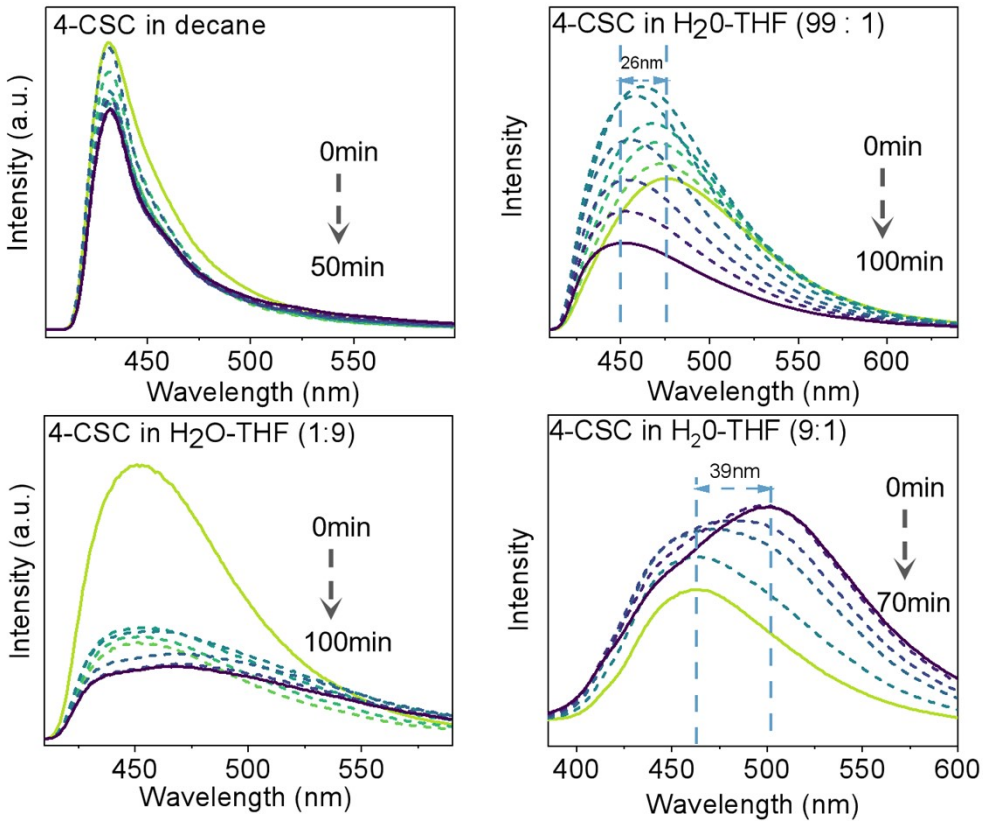


**Figure S3.** PL spectra of Z-4-CSC (0.05 mM) in different solution irradiated with 365 nm UV light from a hand-held UV lamp for different time.

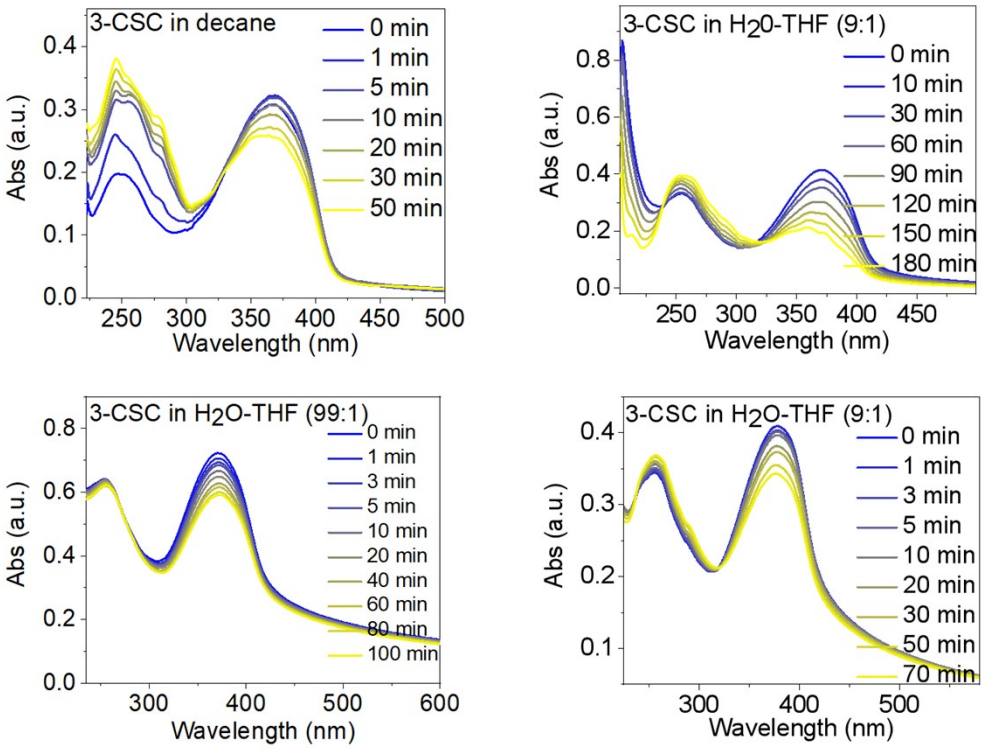


**Figure S4.** UV−vis spectra of Z-3-CSC(0.05 mM) before and after irradiation with 365 nm UV light.

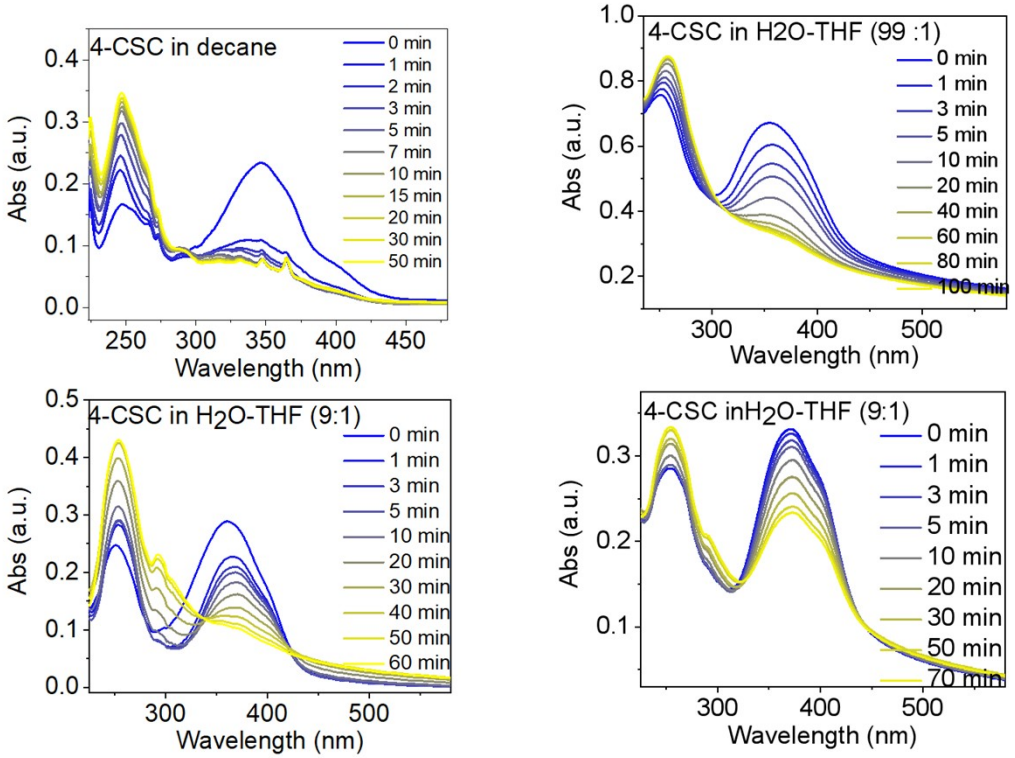


**Figure S5.** UV−vis spectra of Z-4-CSC(0.05 mM) before and after irradiation with 365 nm UV light.

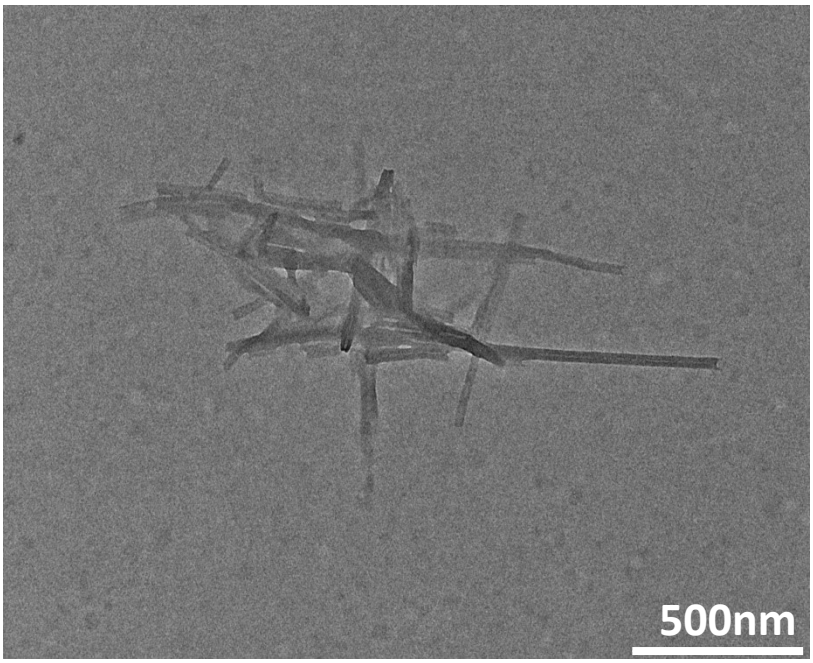


**Figure S6.** TEM image (0.1 mM) of 3-CSC in decane before 365 nm illumination.

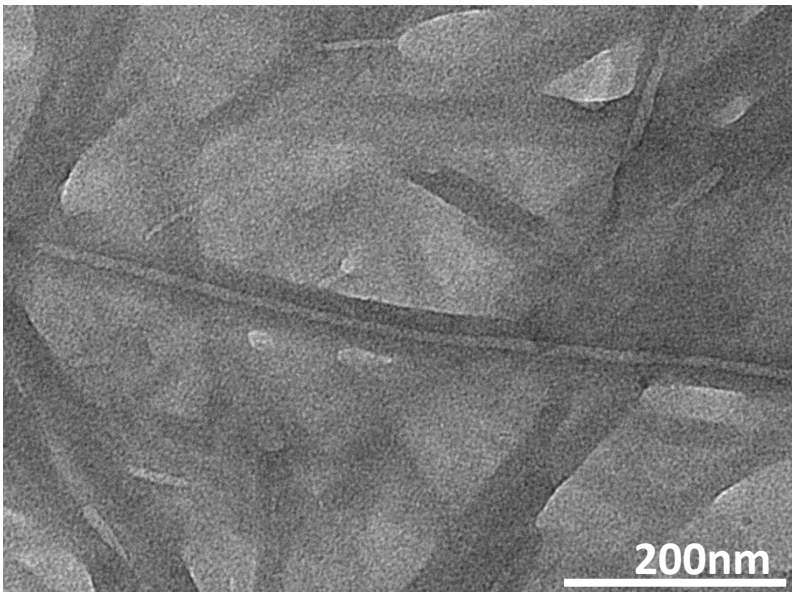


**Figure S7.** TEM image (0.1 mM) of 3-CSC in decane after 1 h of 365 nm illumination.

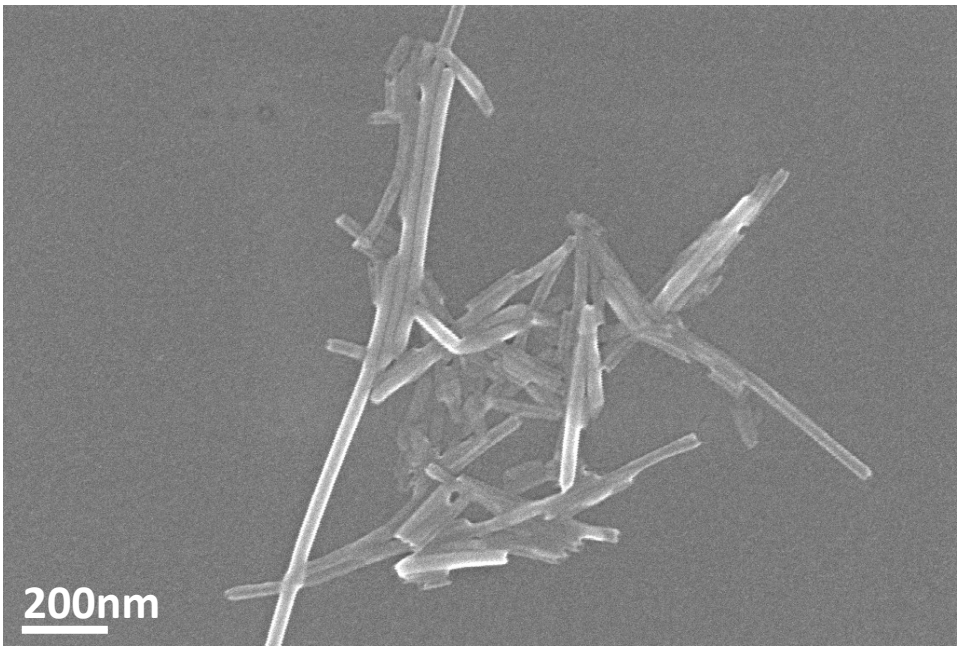


**Figure S8.** SEM image (0.1 mM) of 3-CSC in decane before 365 nm illumination.

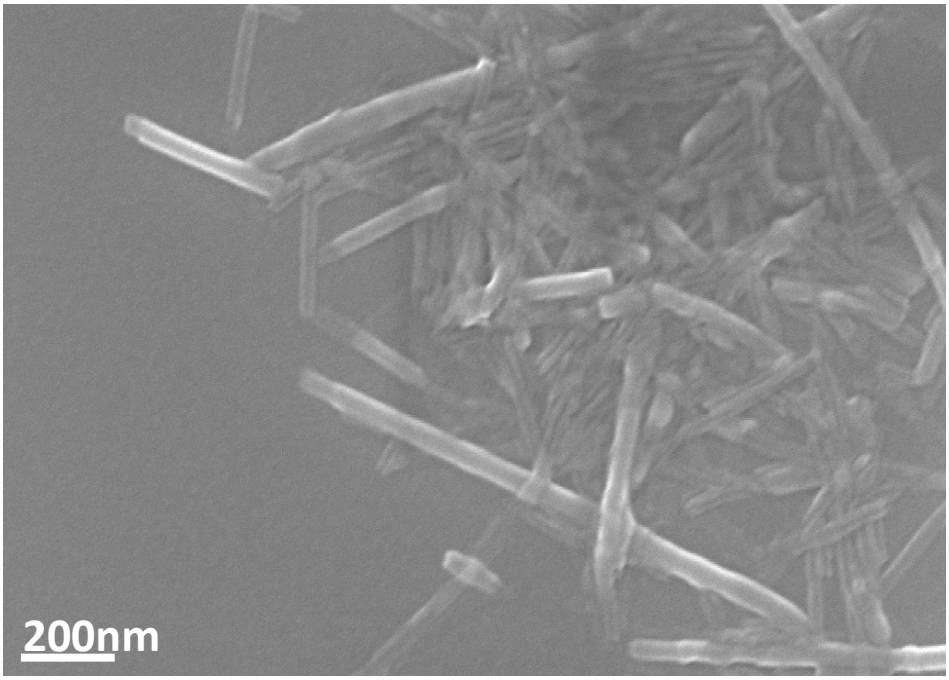


**Figure S9.** SEM image (0.1 mM) of 3-CSC in decane after 1 h of 365 nm illumination.

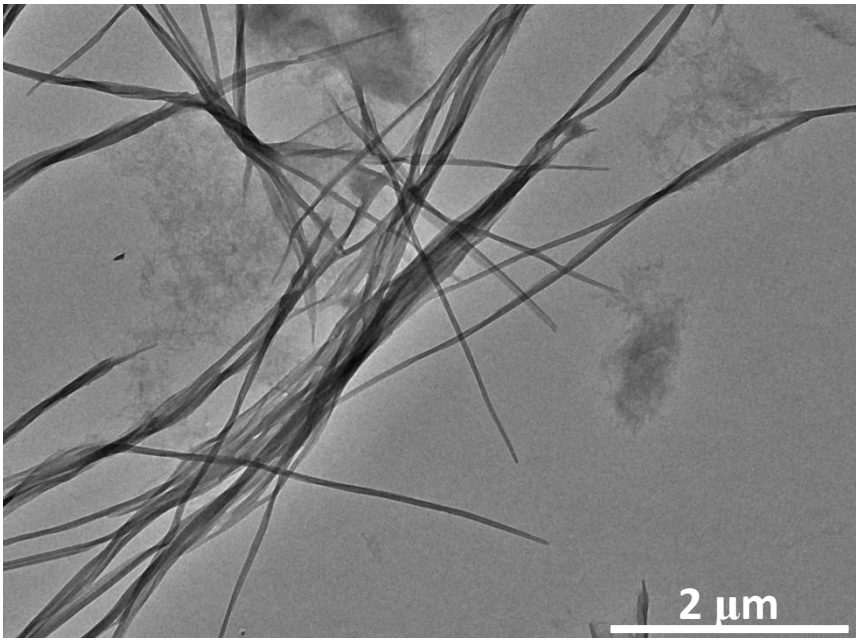


**Figure S10.** TEM image (0.1 mM) of 4-CSC in decane before 365 nm illumination.

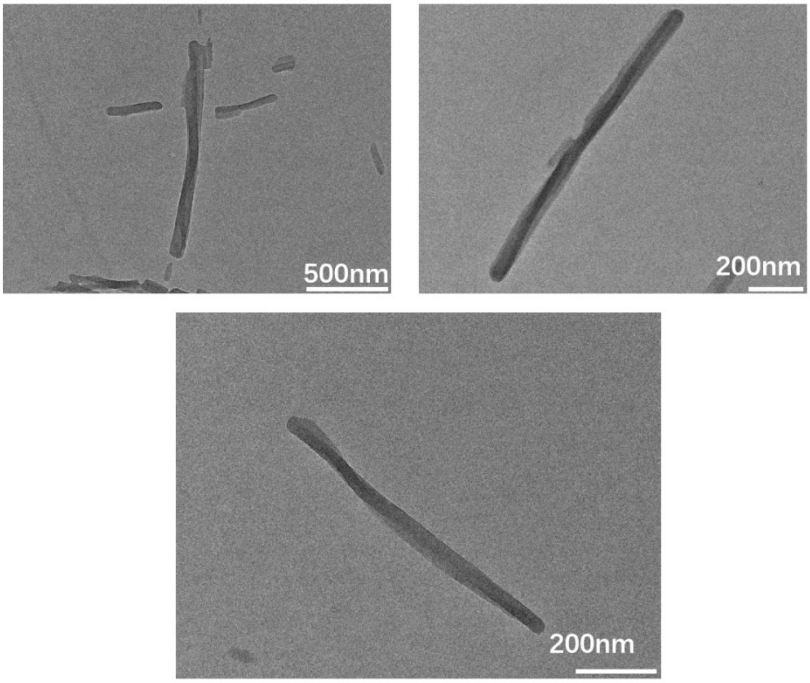


**Figure S11.** TEM image (0.1 mM) of 4-CSC in decane after 1 h of 365 nm illumination.

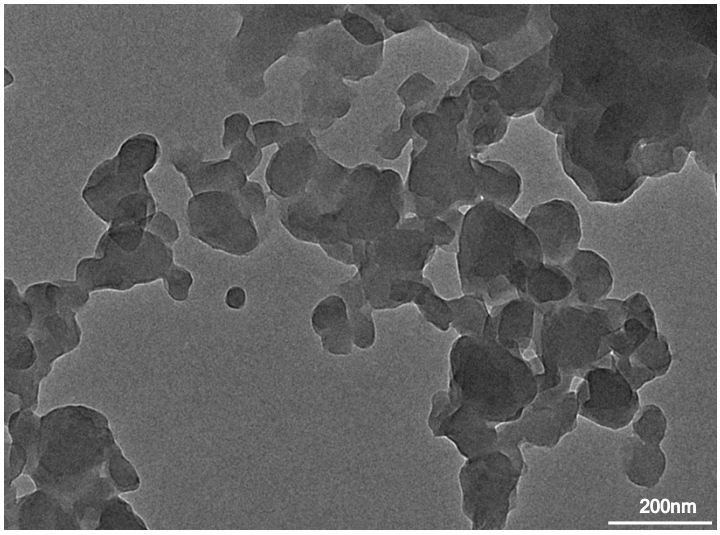


**Figure S12.** TEM image (0.05 mM) of 3-CSC in H20-THF(9:1) after 1 h of 365 nm illumination.

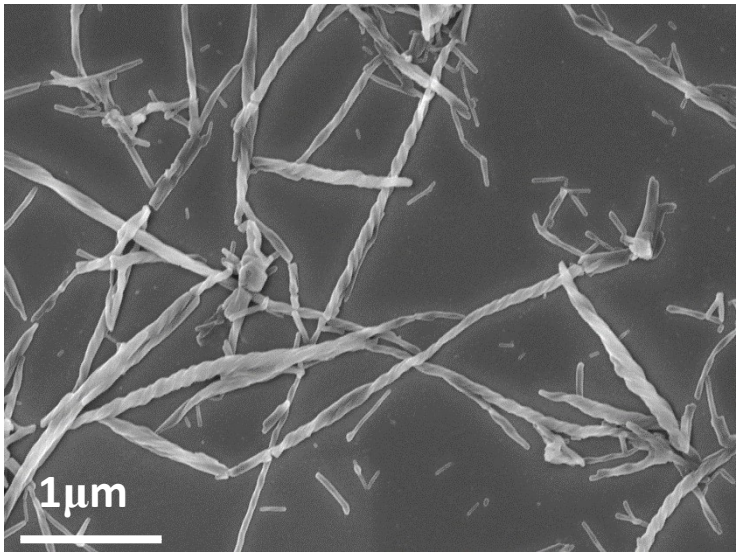


**Figure S13.** SEM image (0.05 mM) of 4-CSC in H20-THF(9:1) before 365 nm illumination.

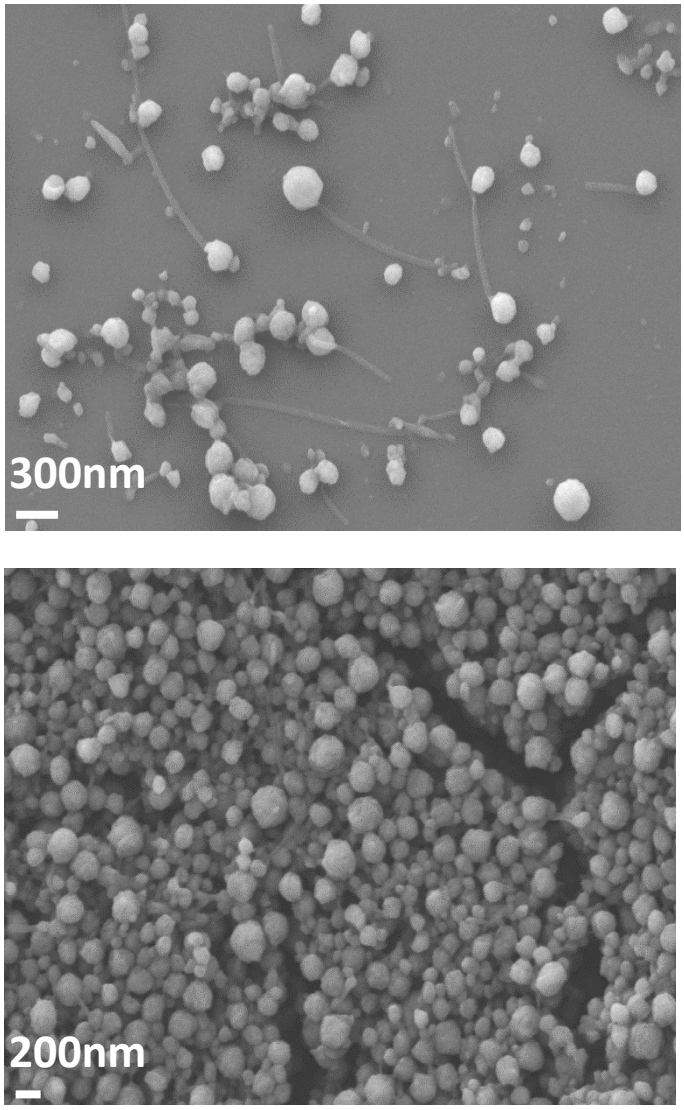


**Figure S14.** SEM image (0.1 mM) of 4-CSC in H20-THF(9:1) after 1 h of 365 nm illumination.

**References**

1. An, S.; Hao, A.; Xing, P. Halogen Bonding Mediated Hierarchical Supramolecular Chirality. *ACS Nano* **2021**, 15, 15306−15315.