

Electronic Supplementary Material (ESI) for Soft Matter.

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## Supporting Information

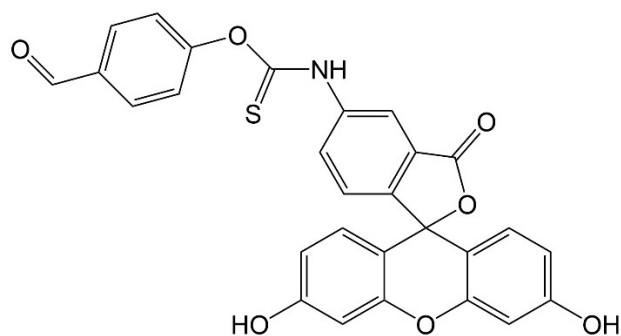
### **Localized self-assembly of macroscopically structured supramolecular hydrogels through reaction-diffusion**

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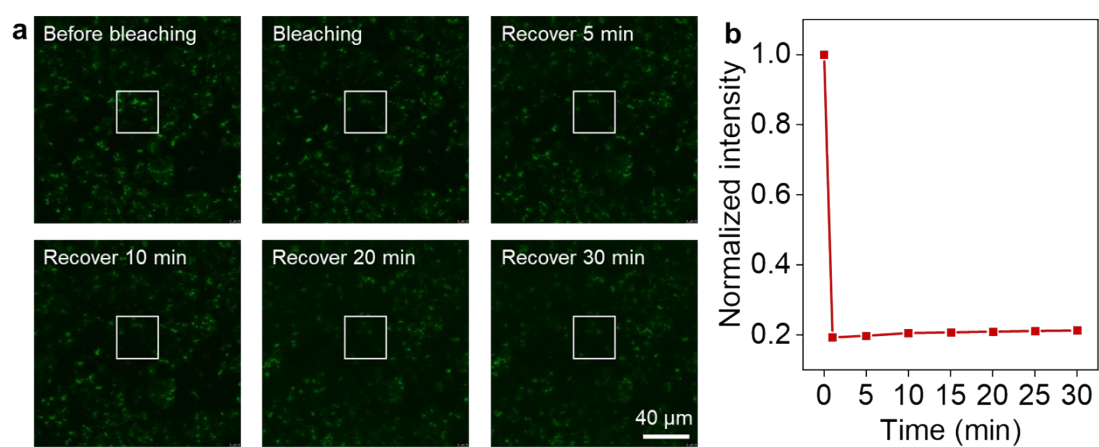
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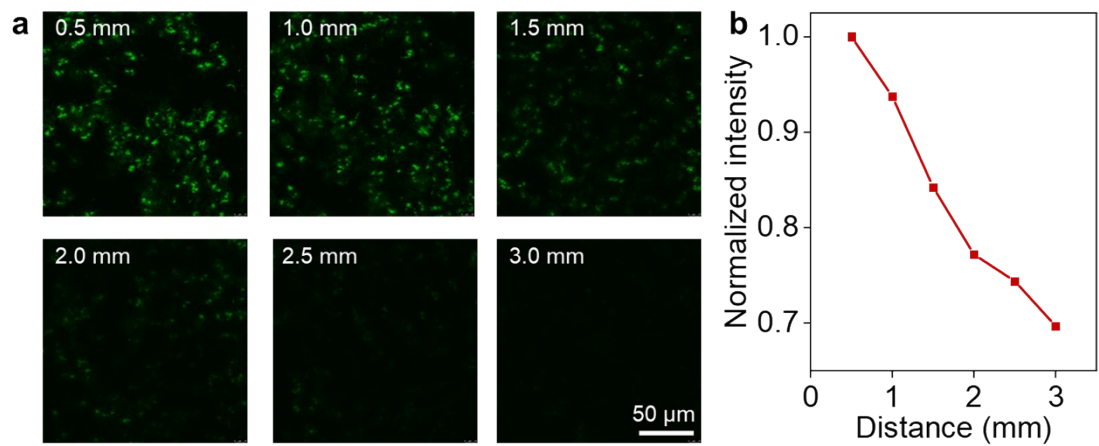
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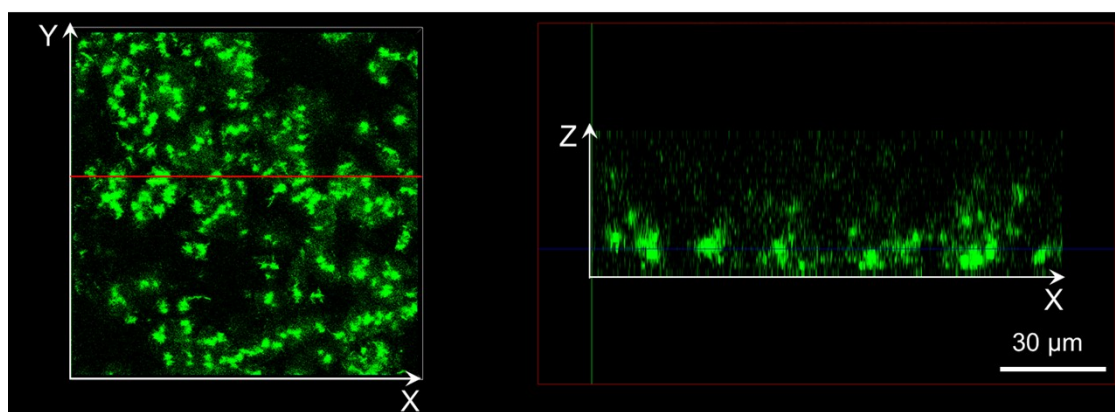
**Fig. S1.** Chemical structure of **A-FL**.



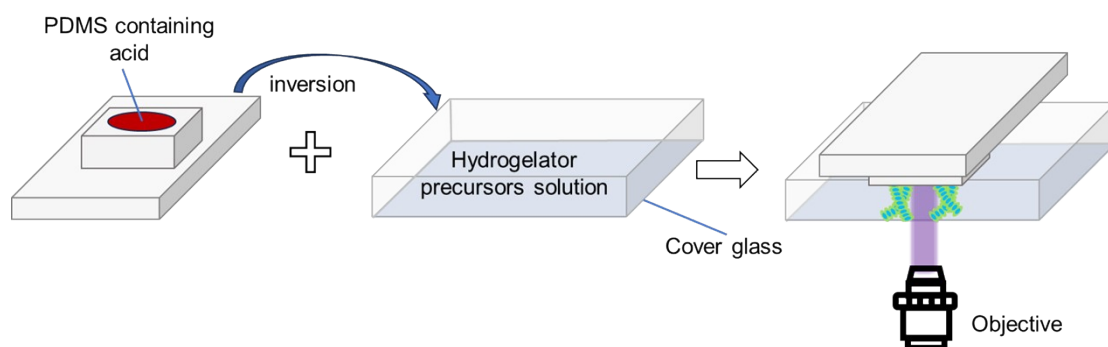
**Fig. S2.** Fluorescence recovery after photobleaching (FRAP) experiments demonstrated the formation of solid hydrogels. a) CLSM images and b) fluorescence recover curve at the bleached area of hydrogel fibers. The photobleached area is delineated by the white rectangle. Sample:  $[H] = 20 \text{ mM}$ ,  $[A] = 120 \text{ mM}$ ,  $[A\text{-FL}] = 30 \text{ }\mu\text{M}$ ,  $[HCl] = 12 \text{ M}$ .



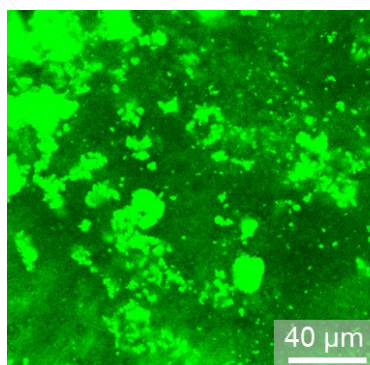
**Fig. S3.** The hydrogel network as a function of the distance relative to the surface of PDMS. a) CLSM images and b) evolution of the average fluorescence intensity against the distance. Sample:  $[\text{H}] = 20 \text{ mM}$ ,  $[\text{A}] = 120 \text{ mM}$ ,  $[\text{A-FL}] = 30 \mu\text{M}$ ,  $[\text{HCl}] = 12 \text{ M}$ .



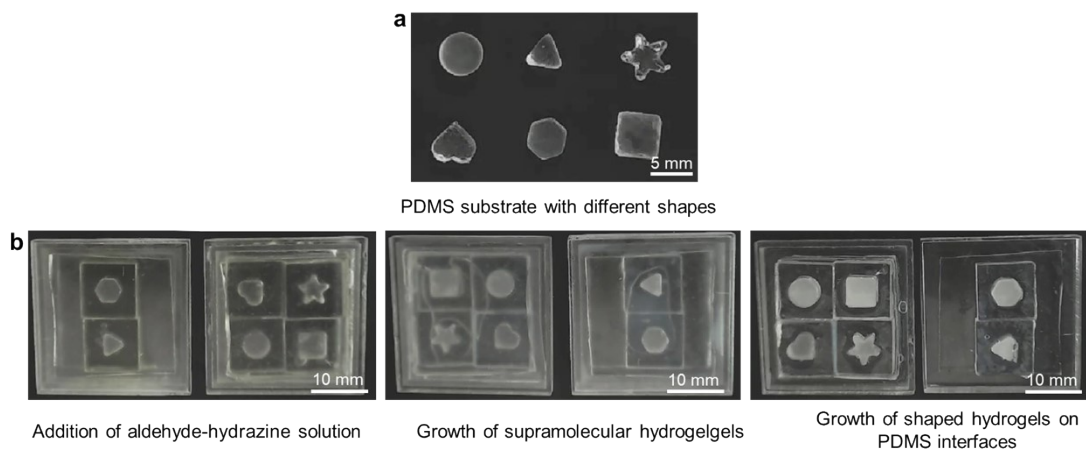
**Fig. S4.** Network density of the supramolecular hydrogel fibres obtained from CLSM 3D scanning experiment. Sample:  $[\text{H}] = 20 \text{ mM}$ ,  $[\text{A}] = 120 \text{ mM}$ ,  $[\text{A-FL}] = 30 \mu\text{M}$ ,  $[\text{HCl}] = 12 \text{ M}$ .



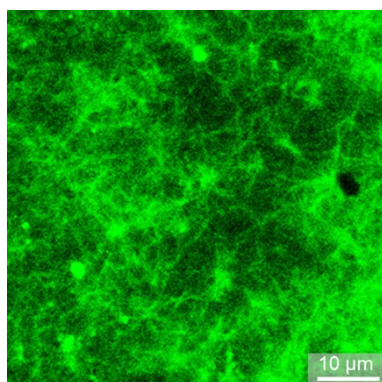
**Fig. S5.** Illustration of the setup for the CLSM monitors interfacial assembly of supramolecular hydrogels. To observe the dynamic assembly of hydrogel fibers, we used a 3D printed mold to fix 5 mm<sup>3</sup> cubic PDMS containing acid. The mold was integrated with a container containing 1 mL of an aqueous solution containing 10 mM hydrogelator precursors at pH 7.0. The container was equipped with a coverslip at the bottom side, which allows for CLSM observation.



**Fig. S6.** Magnified CLSM image showing the hydrogel network of the supramolecular hydrogels grown at the surface of the PDMS substrate. Sample:  $[H] = 10 \text{ mM}$ ,  $[A] = 60 \text{ mM}$ ,  $[A\text{-FL}] = 30 \text{ }\mu\text{M}$ .



**Fig. S7.** Preparation of macroscopically patterned supramolecular hydrogels. a) the PDMS substrates used for the fabrication of macroscopically patterned supramolecular hydrogels; b) the growth process of the patterned supramolecular hydrogels.



**Fig. S8.** CLSM image showing the fibrous network of the patterned supramolecular hydrogels. Sample:  $[H] = 10$  mM,  $[A] = 60$  mM,  $[A-FL] = 30$   $\mu$ M.