

**Electronic Supplementary Information (ESI)**

**Molecular Co-Assembly as a Strategy for the Synergistic  
Improvement of Hydrogels' Mechanical Properties**

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**Peptides and molecules.** Fmoc-Phe-Phe-OH and Fmoc-F<sub>5</sub>-Phe-OH were purchased as lyophilized peptides from Bachem (Budendorf, Switzerland) and Sigma Aldrich (Rehovot, Israel), respectively, and were used without further purification .

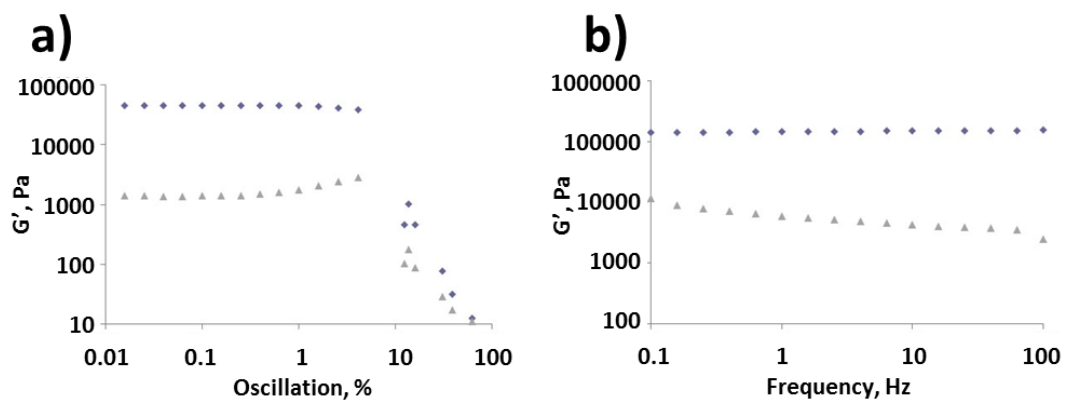
**Hydrogel preparation.** The Fmoc-F<sub>5</sub>-Phe stock solution was prepared by dissolving the lyophilized peptide powder in ethanol to a concentration of 10 mg/ml. The Fmoc-FF stock solution was prepared by dissolving the lyophilized peptide powder in dimethyl sulfoxide (DMSO) to a concentration of 100 mg/ml. Fmoc-F<sub>5</sub>-Phe and Fmoc-FF single peptides hydrogels were prepared by adding 500  $\mu$ L and 50  $\mu$ L of the stock solutions to 500  $\mu$ L and 950  $\mu$ L double distilled water (DDW), respectively, over vortex. Fmoc-F<sub>5</sub>-Phe:Fmoc-FF co-assembled hydrogels at a final concentration of 5 mg/ml were prepared by combining the two peptides stock solutions in ethanol and DMSO, respectively, at the desired molar ratios of 3:1, 1:1, 1:3 and then diluted in DDW: 3:1 (3.75 mg/ml Fmoc-F<sub>5</sub>-Phe, 1.25 mg/ml Fmoc-FF, 375  $\mu$ L ethanol, 12.5  $\mu$ L DMSO, 612.5  $\mu$ L DDW), 1:1 (2.5 mg/ml Fmoc-F<sub>5</sub>-Phe, 2.5 mg/ml Fmoc-FF, 250  $\mu$ L ethanol, 25  $\mu$ L DMSO, 725  $\mu$ L DDW) and 1:3 (1.25 mg/ml Fmoc-F<sub>5</sub>-Phe, 3.75 mg/ml Fmoc-FF, 125  $\mu$ L ethanol, 37.5  $\mu$ L DMSO, 837.5  $\mu$ L DDW).

**Rheological analysis.** The in-situ kinetics of hydrogel formation and mechanical properties were characterized by an AR-G2 rheometer (TA Instruments, USA). Time-sweep oscillatory tests in 20 mm parallel-plate geometry were conducted on 230  $\mu$ l of fresh solution (resulting in a gap size of 0.6 mm) at room temperature. In order to determine the linear viscoelastic region, at which the time sweep oscillatory tests were performed, oscillatory strain (0.01-100%) and frequency sweeps (0.01-100 Hz) were conducted 1 hour after attaching the sample to the rheometer (soak time). G' and G'', the storage and loss moduli, respectively, were obtained at 5 Hz oscillation and 0.5% strain deformation for each sample. All rheological measurements were conducted at a final peptide concentration of 5 mg/ml.

**TEM analysis.** The samples were prepared for TEM analysis by applying 10  $\mu$ l samples to a 400-mesh copper grids (Electron Microscopy Sciences LTD). The excess liquid was removed 2 minutes later. Samples were examined using a JEOL 1200EX electron microscope (JEOL), operating at 80 kV .

**Fluorescence spectroscopy.** The emission spectra of the gels were recorded using a Horiba JobinYvon FL3-11 fluorimeter (Horiba JobinYvon, NJ, USA). A quartz

cuvette with an optical path length of 1 cm was used. The gels were assembled within the cuvette and the spectra were monitored a few minutes following dilution. The experiments were carried out using an excitation wavelength of 280 nm and 5 nm excitation and emission slits.



**Fig.S1.** Rheological analysis of 1:1 hybrid hydrogel at 25°C: Strain Sweep (a) and Frequency Sweep (b) oscillatory measurements.