Supporting Information

Materials and general methods:

Chemicals: Fmoc-amino acids were obtained from GL Biochem (Shanghai, China). Phenothiazine was obtained from A&K (Beijing, China). Naphthalene acetic acid was obtained from Aladdin (Shanghai, China). 2-Cl-trityl chloride resin was obtained from Nankai University resin Co. Ltd. All the other starting materials were obtained from Alfa. Commercially available reagents were used without further purification, unless noted otherwise. All other chemicals were reagent grade or better.

General methods: The synthesized compounds were characterized by $^1$H NMR (Bruker ARX 300) using DMSO-d$_6$ as the solvent and ESI-MS spectrometric analyses were performed at the Thermo Finnigan LCQ AD System. HPLC was conducted at LUMTECH HPLC (Germany) system using a C$_{18}$ RP column with MeOH (0.1% of TFA) and water (0.1% of TFA) as the eluents, LC-MS was conducted at the LCMS-20AD (Shimadzu) system, and rheology was performed on an AR 2000ex (TA instrument) system using a parallel plates (40 mm) at the gap of 500 μm. TEM was done on a Tecnai G2 F20 system, operating at 200 kV. Confocal microscopy images were obtained on a Leica TCS SP5 system (Germany). Fourier transform infrared (FT-IR) spectra were scanned on a bio-rad FTS6000 FT-IR spectrophotometer (America). Circular dichroism (CD) data were collected on a Jasco J-715 CD spectrometer (Japan).

Syntheses and characterizations:

3-(Phenothiazin-10-yl)acetic acid synthesis: procedure was modified from the literature$^1$. Phenothiazine (1) (509 mg, 2.56 mmol), potassium hydroxide (171 mg, 2.61 mmol, 85%, freshly ground), and sodium iodide (39 mg, 0.26 mmol) were combined under argon in 10 mL dry DMF. After 5 min stirring at rt, the ethyl bromoacetate (7.7 mmol) was added dropwise. The reaction mixture was heated at 50 ºC for 48 h. After the reaction mixture had cooled at rt for 30 min, it was diluted with DCM (15 mL) and washed with water (3 × 10 mL). The combined organic layers were dried over magnesium sulfate, filtered, and rotary-evaporated to give a yellow liquid (1.97 g). The crude product was purified on a flash silica column with EtOAc/hexanes (4% to 8% to 15%) as eluent to give the pure compound of 2. The ester 2 was then hydrolyzed in EtOH/MeOH/4M-KOH (25:25:8, 116 mL) at 60 ºC for 30 min. After rotary-evaporating, the reaction mixture was poured into water (100 mL) and acidified by slow addition of 10 % citric acid until the salt was converted completely to the free acid. The resulting crystal was filtered and washed with cold water. The crude acid was re-crystallized from ethanol to afford the final product of 3. $^1$H NMR (300MHz, DMSO-d$_6$) δ 7.08-7.17 (m, 4H), 6.90-6.96 (t, 2H), 6.68-6.72 (d,J=8.12Hz, 2H), 4.55 (s, 2H).
**Scheme S-1.** Synthetic pathway for the preparation of 3-(Phenothiazin-10-yl)propionic acid

**Fig. S-1.** $^1$H-NMR of 3-(Phenothiazin-10-yl)acetic acid

**Fig. S-2.** LC-MS of 3-(Phenothiazin-10-yl)acetic acid
Peptide synthesis: The peptide derivative was prepared by solid phase peptide synthesis (SPPS) using 2-chlorotrityl chloride resin and the corresponding N-Fmoc protected amino acids with side chains properly protected by a tert-butyl group. The first amino acid was loaded on the resin at the C-terminal with the loading efficiency about 0.5 mmol/g. 20% piperidine in anhydrous N,N'-dimethylformamide (DMF) was used during deprotection of Fmoc group. Then the next Fmoc-protected amino acid was coupled to the free amino group using O-(Benzotriazol-1-yl)-N,N,N',N'-tetramethyluroniumhexafluorophosphate (HBTU) as the coupling reagent. The growth of the peptide chain was according to the established Fmoc SPPS protocol. At the final step, Naphthalene acetic acid, 3-(Phenothiazin-10-yl)propionic acid or benzyloxycarbony were used to attach on the peptide. After the last coupling step, excessive reagents were removed by a single DMF wash for 5 minutes (5 mL per gram of resin), followed by five steps of washing using DCM for 2 min (5 mL per gram of resin). The peptide derivative was cleaved using 95% of trifluoroacetic acid with 2.5% of TMS and 2.5% of H₂O for 30 minutes. After rotary-evaporate process, 20 mL per gram of resin of ice-cold diethylether was then added to cleavage reagent. Afterward the supernatant was decanted and the resulting solid was dissolved in DMSO for HPLC separation.

Preparation of Fmoc-GFFY: ¹H NMR (300MHz, DMSO-d₆) δ 9.21 (s, 1H), 8.15-8.27 (m, 2H), 7.84-7.95 (m, 3H), 7.64-7.71 (d, J=7.37Hz, 2H), 7.44-7.51 (t, 1H), 7.35-7.43 (t, 2H), 7.26-7.33 (t, 2H), 7.18-7.24 (d, 4H), 7.09-7.17 (m, 6H), 6.98-7.04 (d, J=8.40, 2H), 6.60-6.68 (d, J=8.42, 2H), 4.44-4.59 (m, 2H), 4.32-4.41 (m, 1H), 4.15-4.27 (m, 3H), 3.42-3.65 (m, 2H), 2.63-3.05 (m, 6H). MS: calc. M⁺ = 754.3, obsvd. (M+1)⁺ = 755.3. HR-MS: (M+1)⁺ = 755.3070.
Fig. S-3. $^1$H-NMR of Fmoc-GFFY

Fig. S-4. HR-MS spectrum of Fmoc-GFFY
Preparation of Nap-GFFY: $^1$H NMR (300MHz, DMSO-d$_6$) δ 9.22 (s, 1H), 8.14-8.28 (m, 3H), 7.97-8.04 (d, J=8.41Hz, 1H), 7.77-7.88 (m, 3H), 7.73 (s, 1H), 7.36-7.50 (m, 3H), 7.26-7.33 (t, 2H), 7.18-7.23 (d, 4H), 7.10-7.17 (m, 6H), 6.98-7.05 (d, J=8.45, 2H), 6.61-6.68 (d, J=8.42, 2H), 4.43-4.60 (m, 2H), 4.31-4.41 (m, 1H), 3.50-3.76 (m, 4H), 2.60-3.05 (m, 6H). MS: calc. M$^+$ = 700.3, obsvd. (M+1)$^+$ = 701.3. HR-MS: (M+1)$^+$ = 701.2978.

Fig. S-5. $^1$H-NMR of Nap-GFFY
Preparation of PTZ-GFFY: $^1$H NMR (300MHz, DMSO-d$_6$) $\delta$ 9.23 (s, 1H), 8.00-8.48 (m, 4H), 7.20-7.26 (d, J=4.19Hz, 4H), 7.11-7.19 (m, 6H), 6.99-7.09 (m, 6H), 6.83-6.92 (t, 2H), 6.60-6.73 (m, 4H), 4.48-4.63 (m, 2H), 4.30-4.46 (m, 3H), 3.73-3.87 (m, 1H), 3.57-3.70 (m, 1H), 2.90-3.07 (m, 3H), 2.62-2.89 (m, 3H). MS: calc. M$^+$ = 771.3, obsvd. (M+1)$^+$ = 772.3. HR-MS: (M+1)$^+$ = 772.2792.
Fig. S-7. $^1$H-NMR of PTZ-GFFY

Fig. S-8. HR-MS spectrum of PTZ-GFFY
**Preparation of Cbz-GFFY:** $^1$H NMR (300MHz, DMSO-d$_6$) $\delta$ 9.22 (s, 1H), 8.15-8.27 (m, 2H), 7.86-7.93 (d, J=8.26Hz, 1H), 7.35-7.41 (t, 1H), 7.28-7.35 (m, 4H), 7.19-7.25 (d, 4H), 7.09-7.18 (m, 6H), 6.98-7.04 (d, J=8.42, 2H), 6.61-6.67 (d, J=8.41, 2H), 4.99 (s, 2H), 4.43-4.60 (m, 2H), 4.32-4.41 (m, 1H), 3.47-3.64 (m, 2H), 2.63-3.05 (m, 6H). MS: calc. M$^+$ = 666.3, obsvd. (M+1)$^+$ = 667.3. HR-MS: (M+1)$^+$ = 667.2766.

**Fig. S-9.** $^1$H NMR of Cbz-GFFY
### Formation of the gels:

The gelators with one equiv. of Na₂CO₃ (to neutralize the terminal carboxylic acid on the peptide) were suspended in PBS buffer (pH = 7.4). The suspensions were then heated in a hot water bath to make the peptides totally dissolved. Hydrogels would form after cooling back to room temperature within 10 minutes.

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**Fig. S-10.** HR-MS of Cbz-GFFY

**Fig. S-11.** The rheological measurement with the mode of dynamic strain sweep for the gel of Fmoc-GFFY
**Fig. S-12.** The rheological measurement with the mode of dynamic strain sweep for the gel of Nap-GFFY

**Fig. S-13.** The rheological measurement with the mode of dynamic strain sweep for the gel of PTZ-GFFY
Fig. S-14. Rheological measurements in the dynamic frequency sweep mode for the gels containing 0.2 wt% of gelators at a strain of 0.5%
Fig. S-15. FT-IR of freeze-dried hydrogels