Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

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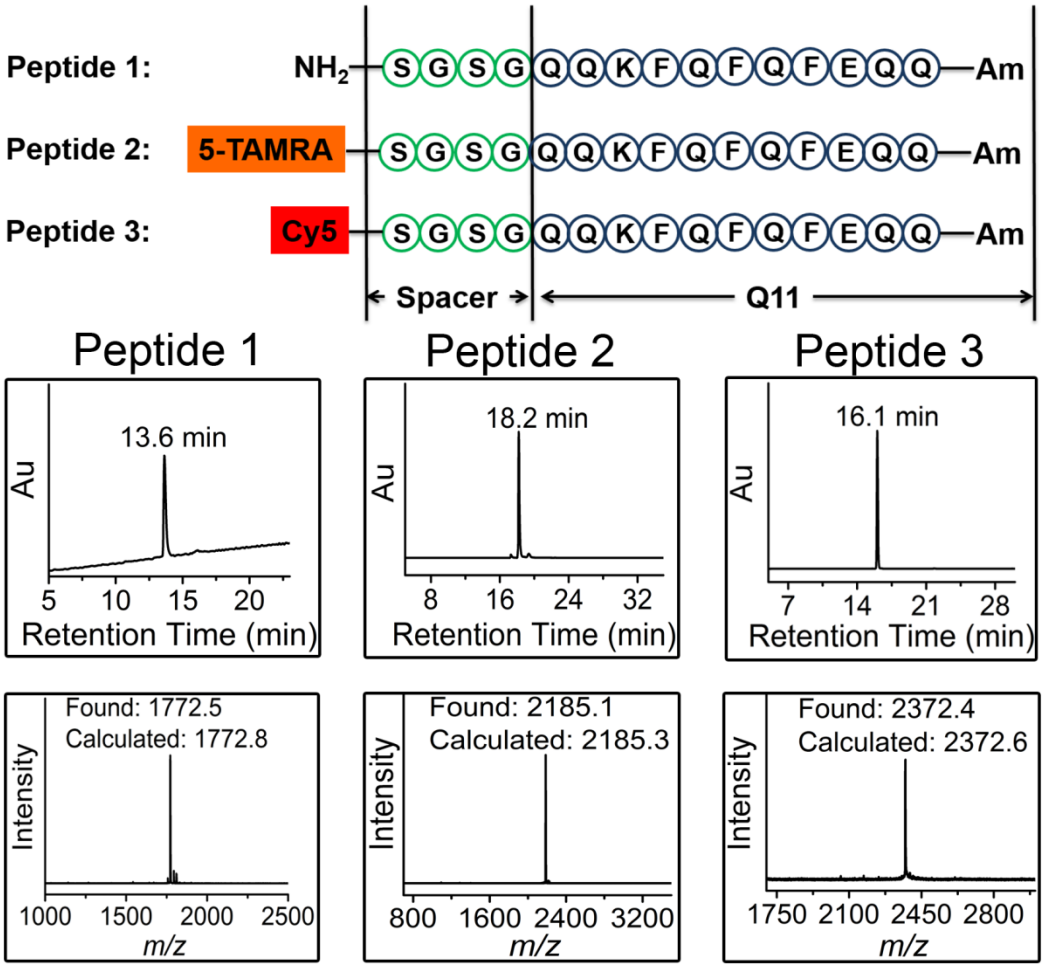

Figure S1. Primary sequences, analytical HPLC and MALDI-TOF mass spectra of synthesized SGSG-Q11 peptides. For analytical reverse phase HPLC, we chose a Waters Xbridge C18 column (5 μm, 4.6 mm × 150 mm). Solvent A was Milli-Q water (0.1% TFA) and solvent B was Acetonitrile (0.1% TFA). The elution gradient was set from 5% B to 65% B in 30 min. Fractions containing the obtained peptides were collected and mixed with matrix α-cyano-4-hydroxycinnamic acid (CHCA), then 1 μL of each fraction mixture was added onto the surface of a Bruker 96 sample plate and dried at room temperature. After that, samples were investigated by Bruker Microflex MALDI-TOF and analyzed by FlexAnalysis software.

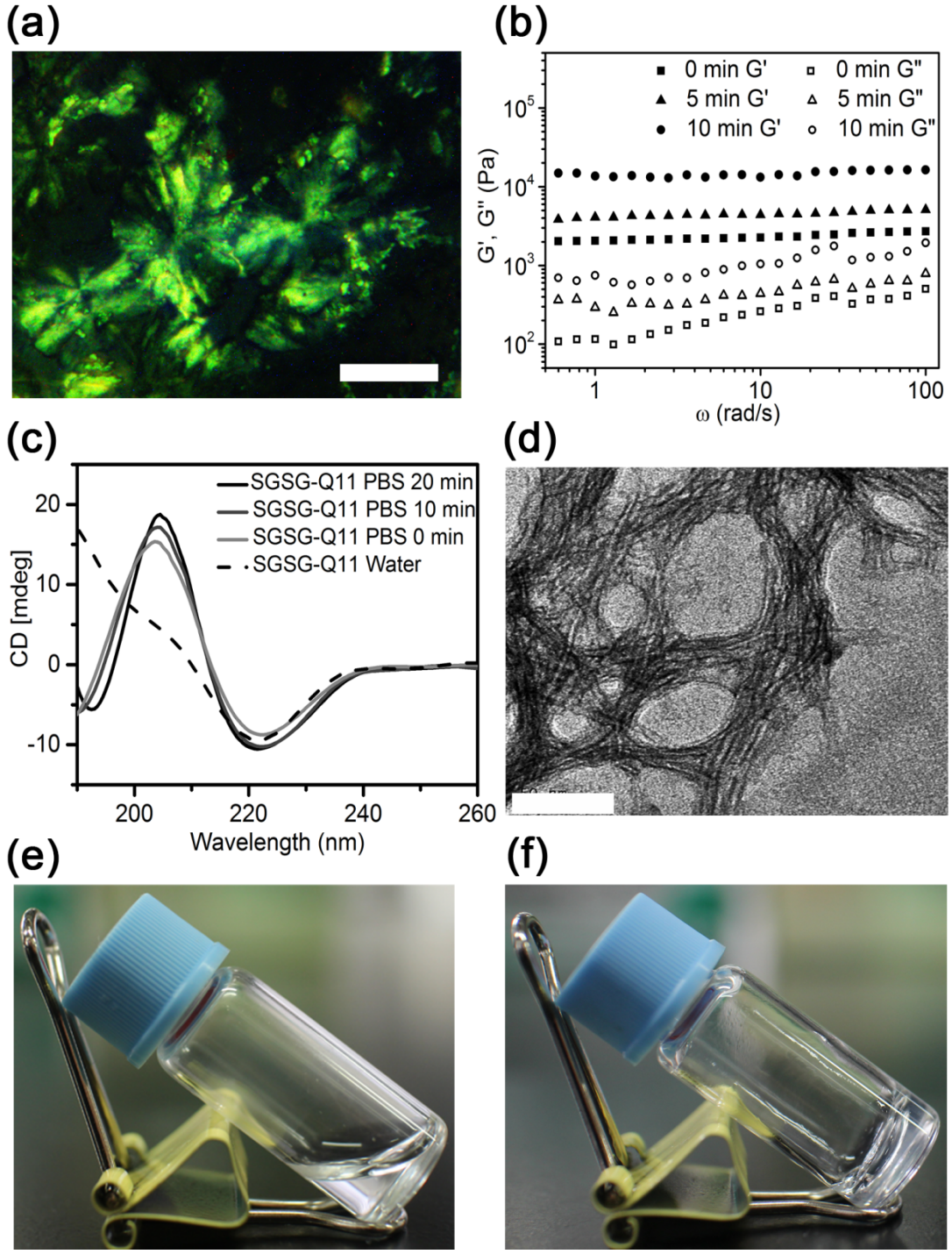

Figure S2. Characterization of the SGSG-Q11 hydrogel. (a) Hydrogel imaging by a polarizing microscope after ThT staining, showing the Maltese Cross pattern. Scale bar = 30 μm; (b) Dynamic oscillatory frequency of the SGSG-Q11 peptide hydrogel before and after PBS treatment. G' was storage modulus, and G" was loss modulus. The increasing G' and G" indicated enhanced intermolecular interactions of SGSG-Q11 after PBS

treatment. (c) CD spectrum of SGSG-Q11 before and after PBS treatment. The peaks at 205 nm (+) and 220-230 nm (-) indicate a structural change from 50% β-sheet to 100% parallel β-sheet. (d) TEM image of SGSG-Q11 fibrils negatively stained with 2% uranyl acetate for 30 s; scale bar = 50 nm. (e) 200 μL 3 mg/mL SGSG-Q11 solution before 1X PBS treatment. (f) 200 μL 3 mg/mL SGSG-Q11 solution after 30 min treatment with 1X PBS.

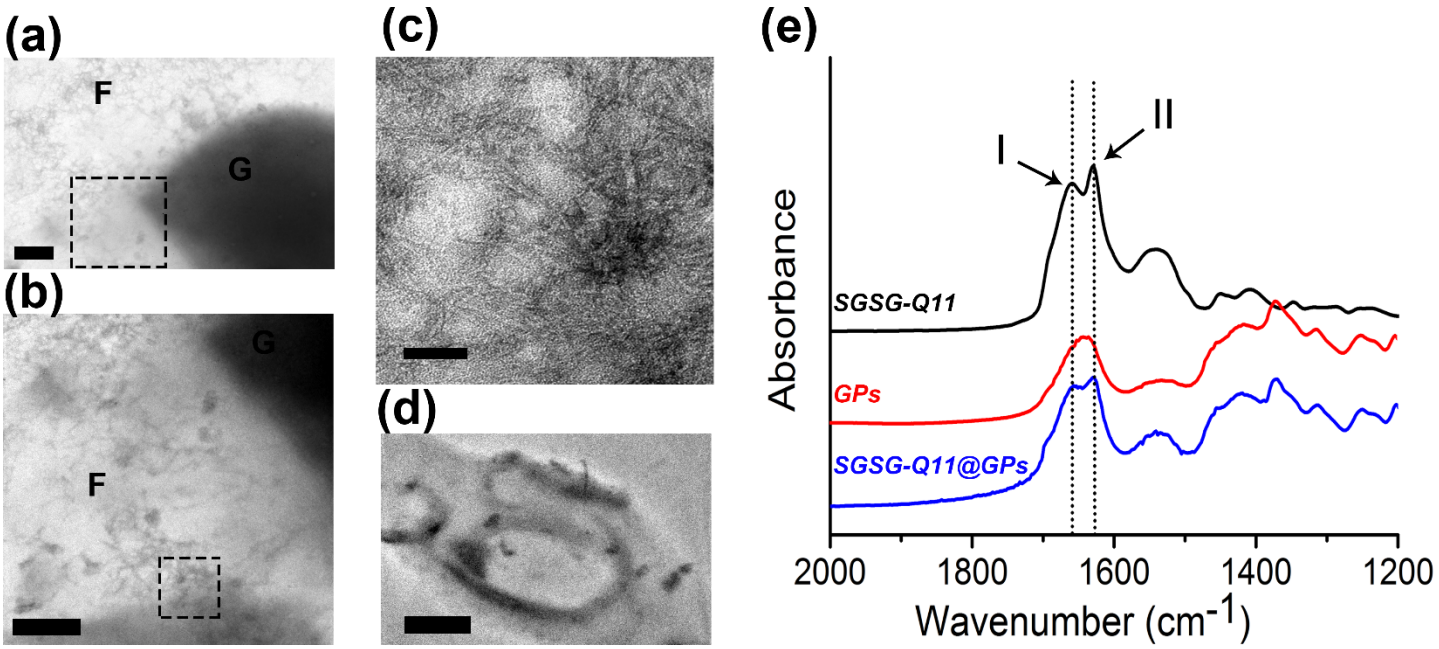

Figure S3. SGSG-Q11 peptides were microencapsulated into GPs, then grinded in liquid nitrogen. The disrupted SGSG-Q11@GPs capsules were imaged by TEM to investigate the self-assembled structures of SGSG-Q11 peptide. (a), TEM image showing the fibrous structure of assembled SGSG-Q11 peptides (F) and the disrupted GPs (G); (b), enlarged ROI in (a); (c), enlarged ROI in (b), demonstrating the assembled SGSG-Q11 fibers; (d), disrupted empty GPs; (e), FTIR spectra of assembled SGSG-Q11, empty GPs and SGSG-Q11@GPs capsules, showing absorbance peak at 1675 cm^{-1} (Ⅰ) and 1626 cm^{-1} (Ⅱ), indicative of antiparallel β-sheet structure in assembled SGSG-Q11 peptides and SGSG-Q11@GPs capsules. Note: (a) to (d) were stained with 2% uranyl acetate for 3 min. Scale bar: (a), (b) and (d), 500 nm; (c), 50 nm.

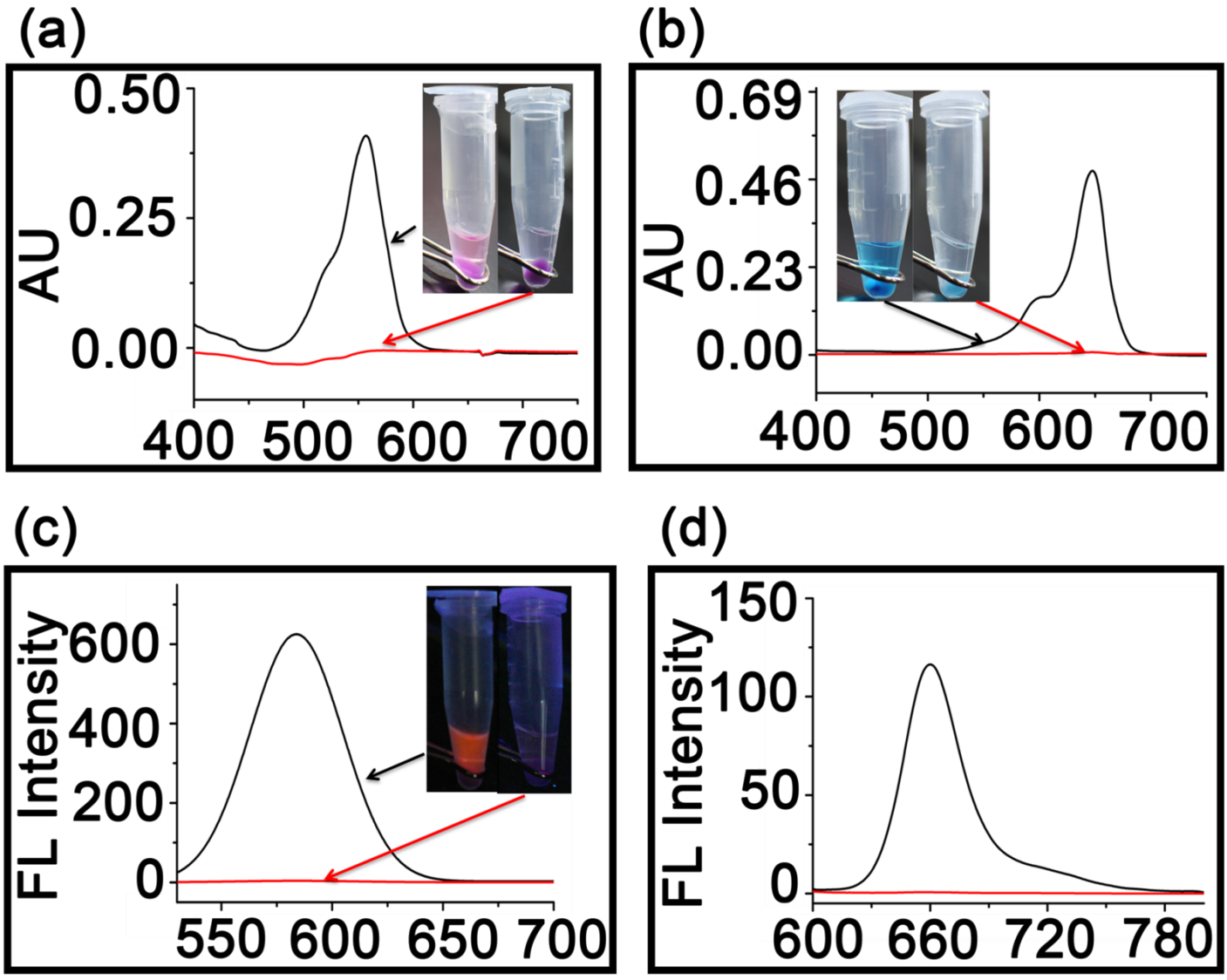

Figure S4. UV/Vis absorption spectra of supernatants during the encapsulation process and after complete encapsulation of (a) SGSG-Q11-5-TAMRA and (b) SGSG-Q11-Cy5 into GPs. Fluorescence emission spectra of supernatants during the encapsulation process and after complete loading of (c) SGSG-Q11-5-TAMRA and (d) SGSG-Q11-Cy5**.** The photos of 1.5 mL microfuge tubes demonstrate the presence of visible and fluorescent dye in the supernatants, indicating incomplete or ongoing encapsulation, while the clear supernatants indicate complete encapsulation of Q11-dye molecules into GPs.

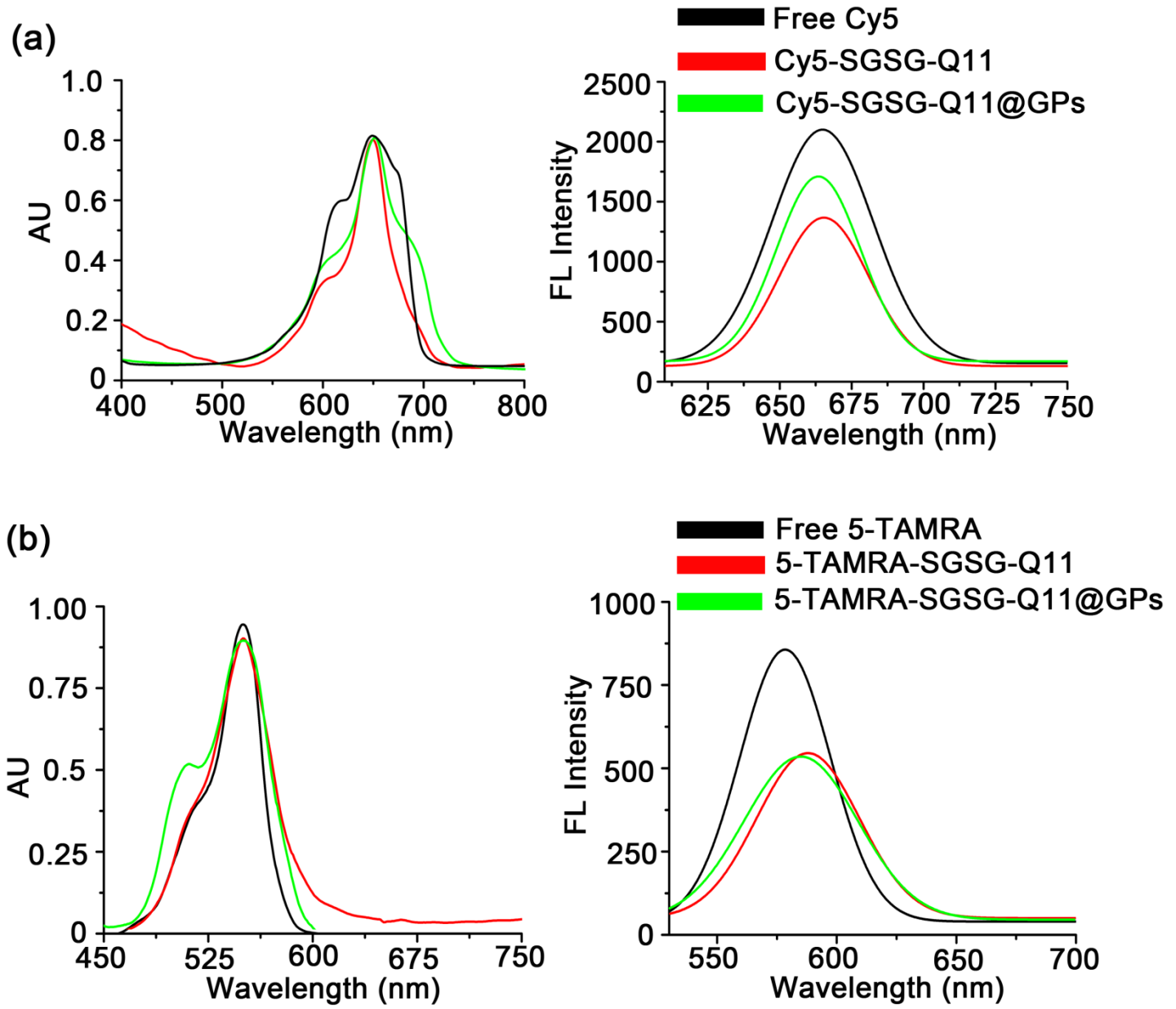

Figure S5. UV/Vis absorption spectra and fluorescence emission spectra of (a) 1 mL free Cy5, Cy5-SGAG-Q11, 3-fold diluted Cy5-SGAG-Q11@GPs, and (b) 1 mL free 5-TAMRA, 5-TAMRA-SGSG-Q11, 5-TAMRA-SGSG-Q11@GPs. According to the Lambert-Beer law,

$$A_\lambda = \varepsilon\, c\, L \tag{1}$$

where "A_λ" represents the UV/Vis absorbance at a certain wavelength, L stands for the length of light path, c is the molar concentration, and ε is the molar extinction coefficient. Then we get:

$$c = A_\lambda \varepsilon^{-1} L^{-1} \quad (2)$$

In this figure, the 1 mL solution of GP capsules was directly suspended from prepared tubes as described in the experimental section. For Cy5, A_{650nm}=0.8012 (ε=270000 $M^{-1}cm^{-1}$, L=1 cm). For 5-TAMRA, A_{548nm}=0.9213 (ε=85000 $M^{-1}cm^{-1}$, L=1 cm). We calculated the molar concentration of each encapsulated peptide as follows:

$C_{Cy5\text{-}SGSG\text{-}Q11}=0.9213\times(270000\ M^{-1}cm^{-1}\times 1\ cm)^{-1}=2.96\times10^{-6}$ M,

$C_{5\text{-}TAMRA\text{-}SGSG\text{-}Q11}=0.8012\times(85000\ M^{-1}cm^{-1}\times 1\ cm)^{-1}=1.06\times10^{-5}$ M.

The molecular weight of Cy5-SGSG-Q11 is 2372, and that of 5-TAMRA-SGSG-Q11 is 2185. Then we calculated the mass/volume concentration of each encapsulated peptide:

$C_{Cy5\text{-}SGSG\text{-}Q11}= (2.96\times10^{-6}\ M\times 2372\ g/mol)\times3=21\ \mu g/mL$,

$C_{5\text{-}TAMRA\text{-}SGSG\text{-}Q11}=1.06\times10^{-5}\ M\times 2185\ g/mol=23\ \mu g/mL$

The total volume was 1 mL. Therefore, the amount of encapsulated peptide was 21 μg and 23 μg, respectively; and the encapsulation yields were:

$Y_{Cy5\text{-}SGSG\text{-}Q11}=21/24=88\%$, and $Y_{5\text{-}TAMRA\text{-}SGSG\text{-}Q11}=23/24=97\%$, respectively.

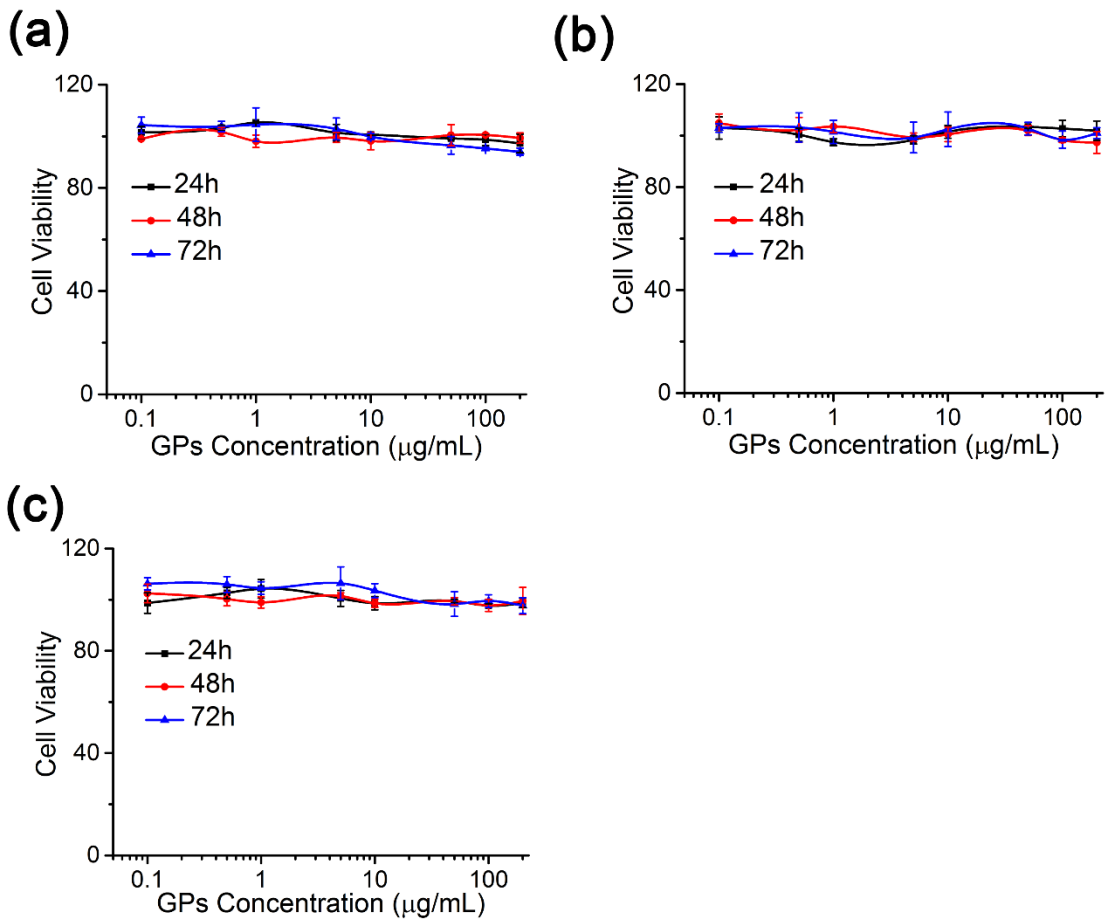

Figure S6. Superb biocompatibility of empty GPs and Q11-dye hydrogel-loaded GPs. Macrophages were incubated with (a) empty GPs, (b) 5-TAMRA –SGSG-Q11@GPs and (c) Cy5-SGSG-Q11@GPs for 24 h, 48 h and 72 h, then MTT assays were carried out to evaluate the concentration-dependent cell viability.

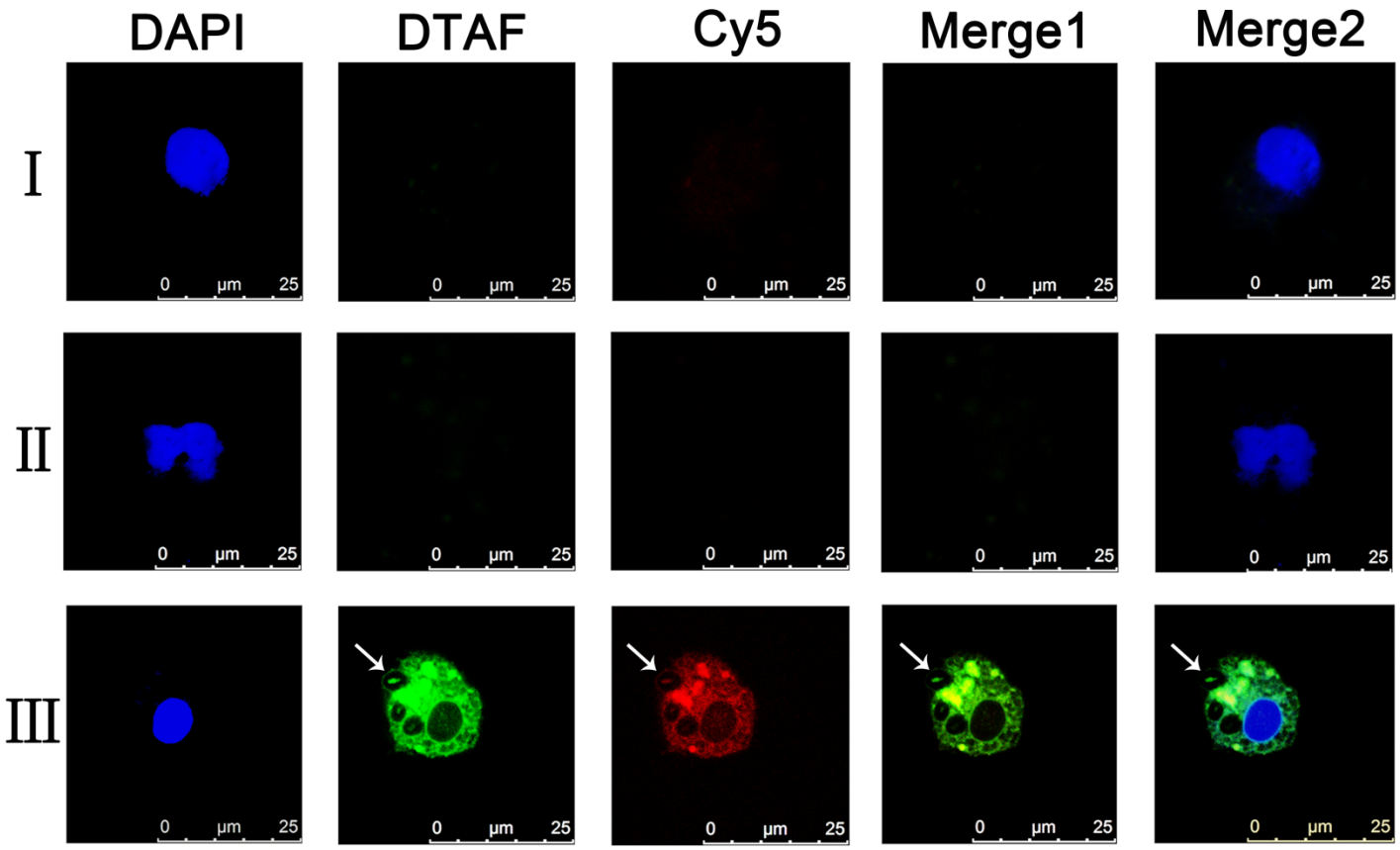

Figure S7. Confocal images of extracted peritoneal macrophages. White arrows indicate the GPs within a macrophage. Note: Ⅰ: Control, saline gavage mice; Ⅱ: Cy5-SGSG-Q11 gavage mice; Ⅲ: Cy5-SGSG-Q11@GPs gavage mice.

Table S1. MALDI-TOF-MS identification of SIF-digested Cy5-SGSG-Q11 hydrogel fragments

m/z	Compositions	Retention Time (min)
187.5	$[F+Na]^+$	6.5
162.9	$[GS+H]^+$	3.8
202.9	$[QG+H]^+$	6.5
250.6	$[SGS+H]^+$	2.3

532.3	$[EQQ+H]^+$	3.8
606.3	$[FQFQ+K]^+$	3.8
699.8	$[QFEQQ+Na]^+$ or $[QQKFQ+Na]^+$	3.8
947.1	$[Cy5\text{-}GS+Na]^+$	16.5