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# **Supporting information**

## For

## **Responsive and self-healing supramolecular photonic crystal hydrogel based on host-guest interactions**

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#### Materials

Ferrocene (purity: 98%), hydrogen peroxide (30 % aqueous solution), acetone (purity: 99%), ethanol (purity: 99%) were purchased from Sinopharm Chemical Reagent Co., Ltd., China. Acrylamide (AM, purity: 99.9%), 2-hydroxy-1-(4-(2-hydroxyethoxy) phenyl)-2-methylpropan-1-one (Photoinitiator 2959) (purity: 98%), 3,5-dimethyl-1-adamantanamine hydrochloride (purity: 98%) and N, N'-methylenebisacrylamide (MBAAm, purity: 98%) were purchased from Shanghai Macklin Biochemical Co., Ltd., China. 1-benzyl-3vinylimidazolium (G) and Cucurbit[8]uril (CB[8]) were prepared as our previous work.<sup>[1]</sup> Deionized water was used for all experiments.

### Characterization

The morphology of the Fe<sub>3</sub>O<sub>4</sub>@C MNPs and SPCH was characterized by scanning electron microscope (SEM, FEI Nova 400 Nano) and transmission electron microscope (TEM, JEOL JEM-2100). The crystal structure of photonic crystals was analyzed by X-ray diffractometer (XRD, PANalytical XPert PRO MPD). The particle size of nanoparticles was measured by dynamic light scattering (DLS, Nicomp 380 Z3000). The functional groups on the surface of nanoparticles were analyzed by Fourier transform infrared spectrum (FTIR, VERTEX70). The reflectance spectra of SPCH were measured by fiber optical spectrometer. The viscoelastic response of SPCH was analyzed by rheometer (TA DHR-2). **Synthesis of Fe<sub>3</sub>O<sub>4</sub>@C MNPs** 

Monodispersed Fe<sub>3</sub>O<sub>4</sub>@C MNPs were synthesized by a solvothermal method. For example, to prepare Fe<sub>3</sub>O<sub>4</sub>@C NPs with a diameter about 177 nm, 0.65 g ferrocene was first dissolved in 65.0 mL acetone under vigorous stirring to obtain a homogeneous solution. Then, 5.0 g hydrogen peroxide solution (30 %) was added dropwise to the above solution and the mixture was stirred for 90 min. The mixture was then transferred into a PPL autoclave and heated at 190 °C for 72 h. The resultant Fe<sub>3</sub>O<sub>4</sub>@C MNPs were separated with a magnet and washed with acetone three times.

#### Synthesis of photonic crystal hydrogels cross-linked by covalent bond

The Fe<sub>3</sub>O<sub>4</sub>@C MNPs (5 mg) were well dispersed in a aqueous solution (0.5 mL) consisting of acrylamide (AM, 0.1 g), *N*, *N'*-methylenebisacrylamide (MBAAm, 7 mg) and photoinitiator 2959 (5 mg) by a subsequently intense ultrasound treatment for 10 min. Then the mixture solution was injected into a glass plate to form sandwich structure and cross-linked with UV light under a magnetic field for 30 min at room temperature. Finally, the as-obtained photonic crystal hydrogels film was peeled off from the glass plate.



Fig. S1 DLS of Fe<sub>3</sub>O<sub>4</sub>@C MNPs (177 nm, PDI= 0.159).



Fig. S2 (a) SEM image, (b) TEM image of Fe<sub>3</sub>O<sub>4</sub>@C MNPs.

The XRD of the Fe<sub>3</sub>O<sub>4</sub>@C MNPs in Fig. S3a showed a typical broad diffraction peak characteristic of nanoscale samples. The peaks were corresponding to the crystal face of cubic crystal Fe<sub>3</sub>O<sub>4</sub> with spinel structure (JCPDS No 19-0629).<sup>[2]</sup> In addition, there was no peak of carbon in the spectrum, indicating that the encapsulated carbon layer exists in amorphous form. The FTIR of Fe<sub>3</sub>O<sub>4</sub>@C MNPs (Fig. S2b) showed the strong absorption peaks at 1630 cm<sup>-1</sup> and 3435 cm<sup>-1</sup>, which indicated the presents of hydrophilic carboxyl groups in the samples. And the Fe-O stretching vibration of the oxygen tetrahedron and the oxygen octahedron of Fe<sub>3</sub>O<sub>4</sub> caused the vibration peak at 586 cm<sup>-1</sup>. Therefore, the FTIR data could demonstrate that these negative charges mainly come from hydrophilic carboxyl and hydroxyl groups on the surface of nanoparticles.<sup>[3]</sup>



Fig. S3 (a) XRD, (b) FTIR spectra of Fe<sub>3</sub>O<sub>4</sub>@C MNPs.



Fig. S4 Reflection spectrum of the hydrogel without Fe<sub>3</sub>O<sub>4</sub>@C MNPs.



Fig. S5 Reflection spectrum and optical photographs (inset) of the hydrogel under 0% water content. Scale bar: 0.5 cm.



Fig. S6 (a) Reflection spectra, (b) optical photographs of the original,  $H_2O$  and EtOH stimulated photonic crystal hydrogels. Scale bar: 0.5 cm.



Fig. S7 Reflection spectra of the original and self-healed SPCH.

### References

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