Supporting Information

For

Novel Luminescent Hierarchical Porous Hydrogels with Three-dimensional Interconnected Network Structure from Feather Keratin Crosslinking Reaction

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Experimental section

Materials and methods

White duck feathers were gathered from the poultry market in Guangzhou, China. Seaweed powders (SP) were purchased from the Wu you fang in Anyang, China. Chitosan (CS) and rhodamine B (Rh B) were purchased from Macklin Biochemical Reagent Co., Ltd (Shanghai, China). All aqueous solutions required were prepared with ultrapure water. All chemicals used in this study were purchased and employed without further purification. All starting materials were used without further purification.

Surface morphology, and pore size of these hydrogels were characterized by FE-SEM (Zeiss Sigma 30) with a Bruker Quantax XFlash SDD 6130. Thermogravimetric analyses (TG) and derivative thermogravimetry (DTG) were performed on Mettler TGA/DSC3+T, the sample in nitrogen was heated at a rate of 5 K min^{-1} in nitrogen atmosphere. Differential scanning calorimetry (DSC) was performed on Mettler Mettler DSC1+, the sample in nitrogen was heated at a rate of 5 K min^{-1} in nitrogen atmosphere. Infrared spectra were recorded in the range of 400−4000 cm^{-1} on a Bruker TENSOR 27 spectrometer. The compression tests were carried out using a mechanical testing machine. Solid-state emission spectra of fluorescence were recorded on an FluoroLog-3 fluorescence spectrophotometer.

Preparation of FK-1, and FK-2 hydrogels and aerogels

White duck feathers were washed with surfactant three times and washed down for several times with ultrapure water to remove grease and solid impurities. Clean duck feathers were dried at 350 K in oven for over 15 h. The puffed feather meal (PFM), yellow granular (or powder) was extruded from duck feathers using feather puffing extruder machine at 433 K. PFM used in the experiment was obtained by flour fractionating through standard sieves of 100 mesh. Firstly, 5.0 g of PFM was suspended in 25 mL of ultrapure water, and then the resulting suspension was incubated in an orbital shaker (200 rpm) for 20 min. Secondly, the reaction suspension was sonicated for 10 min, followed with transferring this suspension into a pressure vapour sterilizer and kept at 394 K for 30 min under static conditions. The resulting porous honeycomb-like hydrogel was collected and washed properly with ultrapure water for removal of unreacted

reagents. The brown sample was designated as **FK-1** hydrogel (FK: feather keratin). The aerogel was obtained via freeze-drying.

The preparation process of the **FK-2** hydrogel is similar to that of **FK-1** except 50 mL ultrapure water was used for the incubation process of PFM. The aerogel was obtained via freeze-drying.

Preparation of FK-SP-1, FK-SP-2, FK-CS, and FK-Rh B hydrogels and aerogels

SP used in the experiment was obtained by flour fractionating through standard sieves of 100 mesh. Firstly, PFM (5.0 g) and SP (1.0 g) was suspended in 50 mL of ultrapure water, and then the resulting suspension was incubated in an orbital shaker (RT, 200 rpm) for 20 min. Secondly, the suspension was sonicated for 10 min, followed with transferring this suspension into a pressure vapour sterilizer and kept at 394 K for 30 min. The resulting spongy-like hydrogel was washed with ultrapure water for removal of unreacted raw materials. The dark brown sample was designated as **FK-SP-1** hydrogel. The aerogel was obtained via freeze-drying.

FK-SP-2 hydrogel was prepared using the same procedure except for substituting 1.0 g of SP with 2.0 g of SP. The aerogel was obtained via freeze-drying.

The coffee-coloured **FK-CS** hydrogel was perpared using the same procedure is similar to that of **FK-SP-1** except substituting SP with CS. The aerogels was obtained via freeze-drying.

The preparation process of the fuchsia **FK-Rh B** hydrogel is similar to that of **FK-1** except substituting pure PFM with the mixture of Rh B (1.0 g) and PFM (5.0 g). The aerogel was obtained via freeze-drying.

Water absorption was obtained by the gravimetric method, and the weights of hydrogels were measured for the calculation of water absorption. Before testing, the hydrogels were dried in a freezer oven until constant weight, and the weight of each dried sample was recorded as W_1 (g). Then, the samples were immersed in ultrapure water at RT for 12 h. After removing water droplets from the sponge surface using a filter paper, the swollen sample was quickly weighed and recorded as W_2 (g). Water absorption was calculated as follows:

$$swelling\ rate\ (\%) = \frac{W_2 - W_1}{W_1} \times 100\% \qquad (1)$$

TG/DTG and DSC curves of **FK-1**, **FK-2**, **FK-SP-1**, **FK-SP-2**, **FK-CS**, and **FK-Rh B** hydrogels are shown in Figure S1–S7. In the first stage, the weight loss percentage of these hydrogels was 56.45%, 65.45%, 64.77%, 62.14%, 62.25%, and 56.82% before 200 °C, respectively (TG). The DTG curves presented in Figs S1–S6 show, in all cases, the presence of the first peak at about 100 °C, and the DSC curves of these hydrogels has a large endothermic peak in the same temperature range, and the peak temperature is about 100 °C (Figure S7). It can be inferred that the weight losses between about 55–65% were due to the loss of free water and partially bound water in these wet hydrogels. In the second stage, the loss of weight (TG) and the second peak of DTG appearing in the temperature range 200–400 °C is assigned to the degradation, cross-linking and the decomposition of hydrogels.

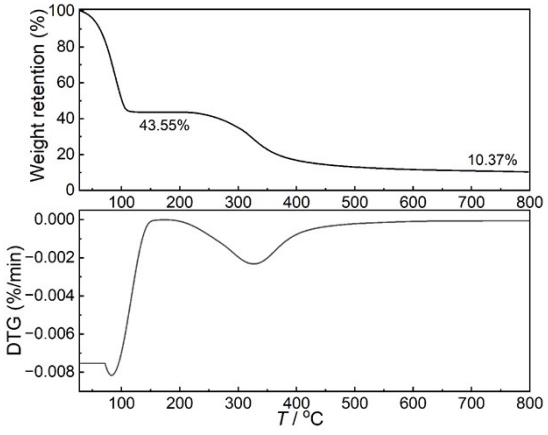

Figure S1 TG/DTG curves of **FK-1** hydrogel.

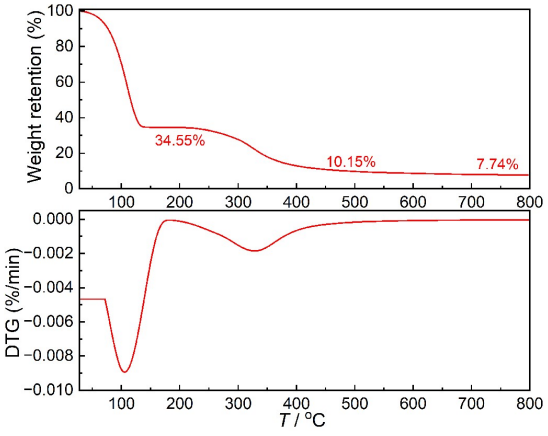

Figure S2 TG/DTG curves of **FK-2** hydrogel.

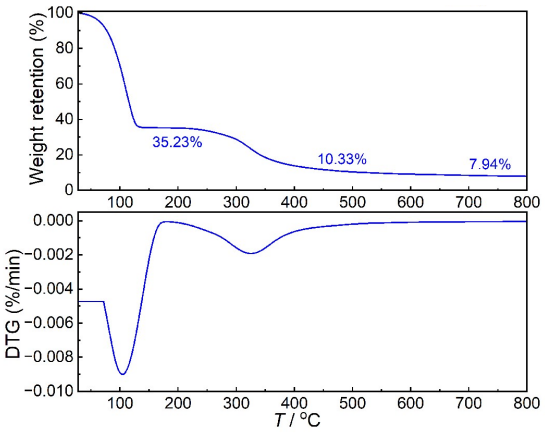

Figure S3 TG/DTG curves of **FK-SP-1** hydrogel.

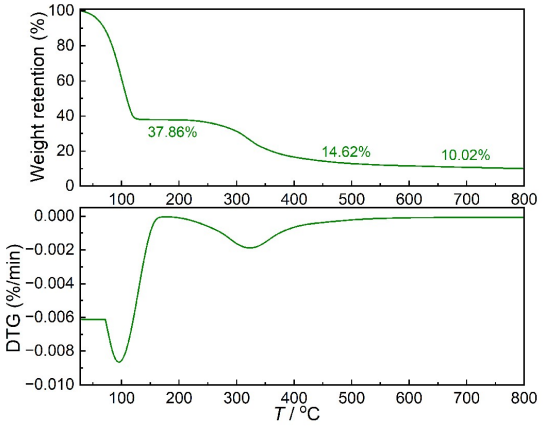

Figure S4 TG/DTG curves of **FK-SP-2** hydrogel.

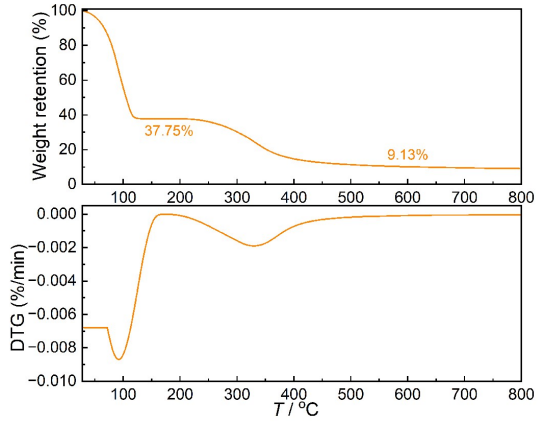

Figure S5 TG/DTG curves of **FK-CS** hydrogel.

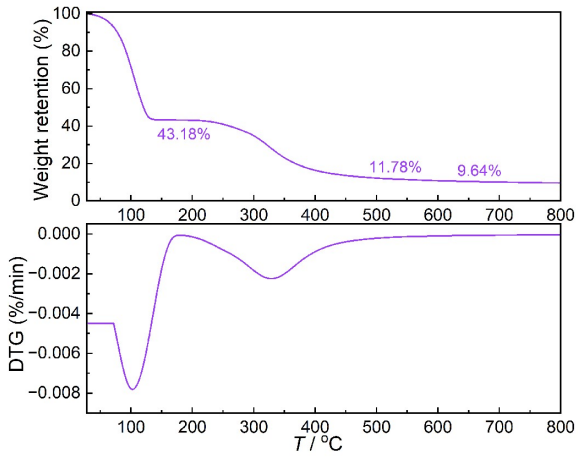

Figure S6 TG/DTG curves of **FK-Rh B** hydrogel.

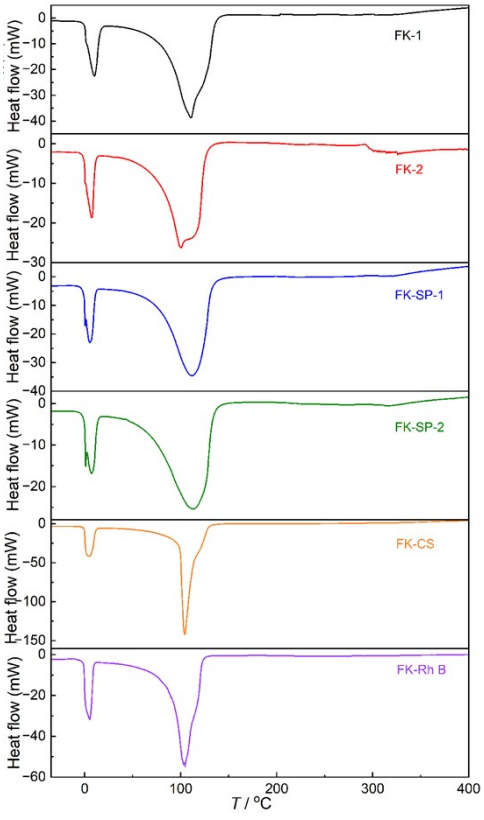

Figure S7 DSC curves of **FK-1**, **FK-2**, **FK-SP-1**, **FK-SP-2**, **FK-CS**, and **FK-Rh B** hydrogels.

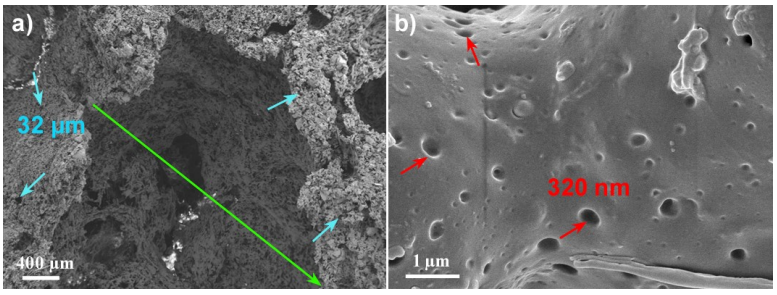

Figure S8 SEM images of (a) the FK-SP-2 aerogel (× 25), and (b) the FK-SP-2 aerogel (× 15,000). The primary hole structure, the secondary hole structure, and the tertiary pore structure are colored in green, blue, and red, respectively. The primary hole structure of FK-SP-2 sample is very large, and it is difficult to show the whole hole from SEM. Pore sizes of the secondary and tertiary hole structures ranged from 10 to 100 μm and 100 to 500 nm, respectively.

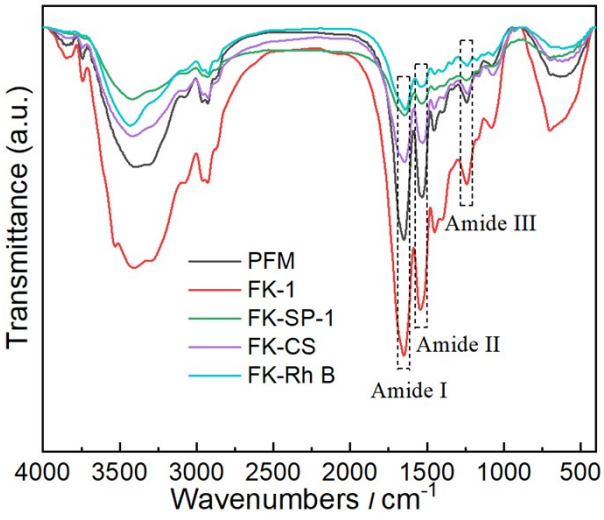

Figure S9. FT-IR spectra of PFM, **FK-1**, **FK-SP-1**, **FK-CS**, and **FK-Rh B**. Characteristic bands of protein were observed in the spectra of all samples. A band at 1650 cm^{-1} was observed in the spectra corresponding C=O deformation vibrations ($\nu_{C=O}$) from –CONH– of feather keratin (amide I). A band from 1549 cm^{-1} to 1513 cm^{-1} was assigned to the N-H bending vibrations (δ_{N-H}) (amide II). Additionally, the band at 1240 cm^{-1} can be assigned to stretching vibration of C–N (ν_{C-N}) (amide III). These bands imply that keratin molecules exist in all samples.

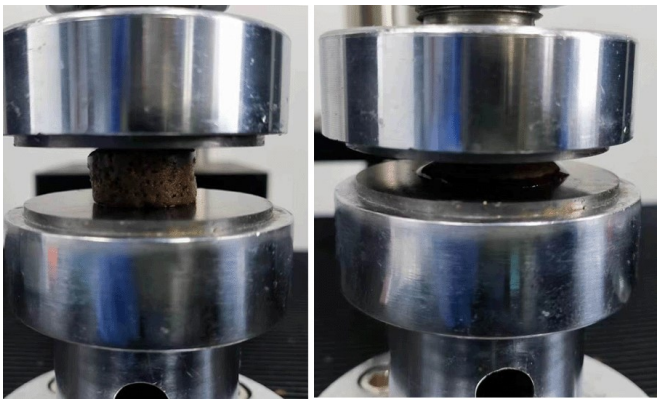

Figure S10 The diagram of compression process of **FK-1** hydrogel using a mechanical testing machine

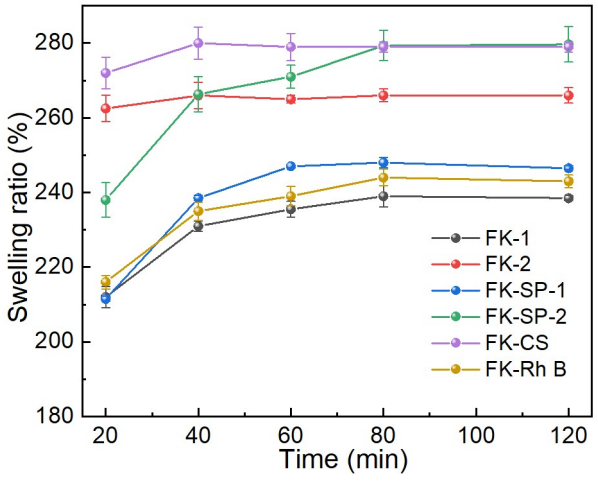

Figure S11 water adsorption capacity of **FK-1**, **FK-2**, **FK-SP-1**, **FK-SP-2**, **FK-CS**, and **FK-Rh B** at room temperature. It can be clearly observed that swelling rate increases sharply during first 20 min Swelling rate of **FK-1**, **FK-2**, **FK-SP-1**, **FK-SP-2**, **FK-CS**, and **FK-Rh B** reached 211%, 262% 211%, 238%, 272%, and 216%, respectively. The equilibrium water adsorption capacities for **FK-1**, **FK-2**, **FK-SP-1**, **FK-SP-2**, **FK-CS**, and **FK-Rh B** were around 239%, 266%, 248%, 279%, 279%, and 244% after 80 min, respectively. A higher value was observed for **FK-2**, **FK-SP-2**, and **FK-CS**, which might be ascribed to the exposed groups providing more hydrogen bond attraction between SP (or CS) and water molecules. Therefore the incorporation of SP or CS could increase water adsorption.

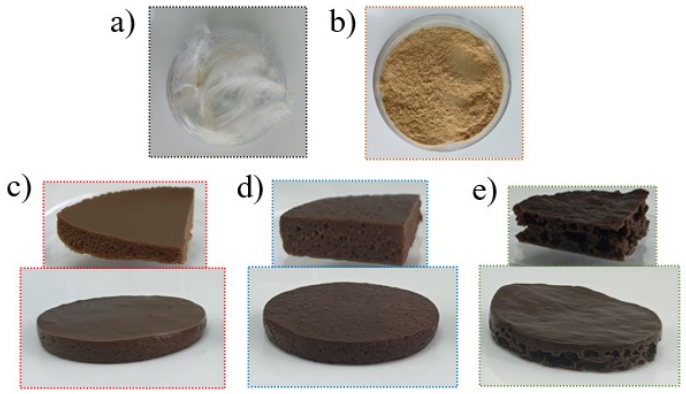

Figure S12 Specimens of raw material, PFM, the keratin hydrogel and hydrogel composites, (a) duck feathers, (b) PFM, (c) **FK-1** hydrogel, (d) **FK-SP-1** hydrogel, and(e) **FK-CS** hydrogel.