Supplementary Information

Effect of the crosslinking agent on a supramolecular gel to control lost circulation

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Characterization of GP-A

Elemental analysis. A Vario EL - III elemental analyzer (Elementar, Germany) was used to analyze the quantity of the C, H, O, N, and S elements of the polymer GP-A.

FT-IR spectra analysis. FT-IR was conducted on a WQF-520A infrared spectrometer (BFRL, China) in the range of 4000 cm^{-1}-400 cm^{-1}.

The critical association concentration (CAC). The viscosity of GP-A and HPAM at different concentrations was measured by a HAAKE MARS III (Thermo Scientific) at a temperature of 25℃ and a shear rate of 7.34 s^{-1}. When the viscosity of the liquid increases suddenly, the concentration is the critical association concentration (CAC).

Rheology

The viscosity (η), shear stress (τ), storage modulus (G'), and loss modulus (G'') of

the GP-A were measured by a HAKKE MARS III, and the complex modulus G^* was calculated by *Eq. 1*. The results are shown in Fig. 1.

$$|G^*| = \sqrt{(G')^2 + (G'')^2} \quad \text{(Eq. 1)}$$

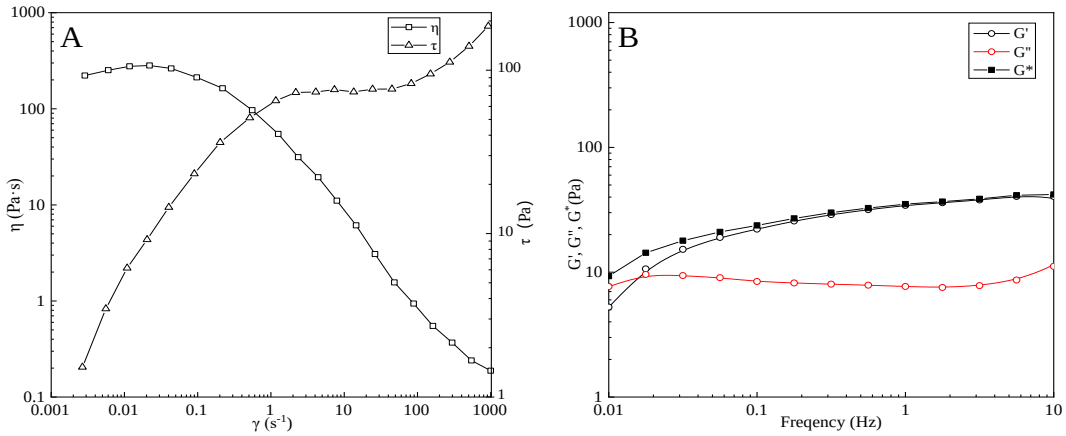

Figure S1. The rheological test results of 1.2 wt % GP-A. (A) Viscosity (η) and shear stress (τ) vary with the shear rate. (B) The modulus (G', G'', G^*) varies with frequency.

Scanning electronic microscopy (SEM)

For all measurements, the gel samples were prepared by 1.2% (w/v) gel powder dissolved in water, and stirring was stopped when the gel was completely dissolved and there were no bubbles in the gel solution. The scanning electron microscope (SEM, Hitachi S-4800, Japan) test steps are as follows:

(1) Put the copper block into a beaker, and pour liquid nitrogen into the beaker to drown the copper block so that the copper block can be cooled for more than 15 min;

(2) Take out the copper block, quickly cut a small piece of gel sample with scissors and place it on the copper block;

(3) The vacuum degree of the electron microscope was set as 267 Pa, and the copper block containing the sample was quickly put into the sample chamber for

vacuum pumping. After approximately 2 min, the vacuum reached the set value.

(4) After the sample was placed into an electron microscope for approximately 45 min, the water was condensed into ice crystals and sublimed. The microscopic morphology and structure of the gel were gradually revealed for observation

Rheology−cross linking agent Measurements

All the samples were measured by a HAAKE MARS III from Thermo Scientific. The type and concentration of the crosslinking agent in the gel system were changed, and the test included viscosity, shear stress, and modulus (G' & G'').

Rheology −Temperature& pH Measurements

The samples were measured by a HAAKE MARS III, and the rheological properties of the gel were tested by changing the temperature and pH value.

Microscopic Mechanism of GP-A for combating lost circulation

Atomic force microscope (AFM)

The gel polymer was tested using atomic force microscopy with a Bruker FastScan. The prepared gel sample is dropped onto the silicon wafer and cooled with liquid nitrogen. After water sublimation, the silicon wafer was placed under an atomic force microscope (AFM) for testing.

Transmission electron microscope (TEM)

Transmission electron microscopy (TEM, JEM-2100F Japan) is an important method for studying polymer morphology and structure. Detailed sample preparation and experimental steps are as follows:

(1) The carrying net is placed in a temperature/humidity-controlled environment

bin. The temperature can be adjusted between 5.85℃-61.85℃, and the humidity can be adjusted to 99% in the atmosphere.

(2) After dropping the sample suspension (usually 3 μL) on the electron microscope, some samples were absorbed through the automatic adsorption process of filter paper, and then the net was rapidly put into liquid ethane for freezing. After freezing, the carrier net was quickly transferred to the sample box and stored in liquid nitrogen for subsequent observation.

(3) The obtained frozen transmission samples were transferred to the cryogenic sample loading device in liquid nitrogen and placed on the frozen sample rod. After the sample was fixed, the sample rod was quickly pulled out and transferred to the transmission electron microscope. The samples were kept at low temperatures (usually below -173.15℃) during sample transfer and observation.

(4) Test sample transmission electron microscopy began.

RESULTS AND DISCUSSION

Characterization of GP-A

Elemental analysis

The elemental analysis of GP-A is shown in Table 1.

Table S1. Results of the elemental analysis and molecular weight of GP-A

Element	C	N	S	O	H	Na
Weight%	48.89	4.47	8.31	27.53	2.29	8.51
Atomic%	54.37	3.81	4.74	25.52	8.67	2.89

Table 1 lists the results of the elemental analysis for GP-A. The molecular structure

contains C (48.89%), N (4.47%), S (8.31%), O (27.53%), H (2.29%), and Na (8.51%).

Influence of the cross-linking agent on GP-A gel rheology

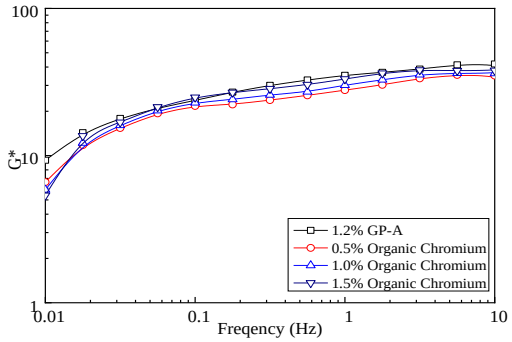

Figure S2. Strength analysis of the GP-A gel under different concentration crosslinking agents.

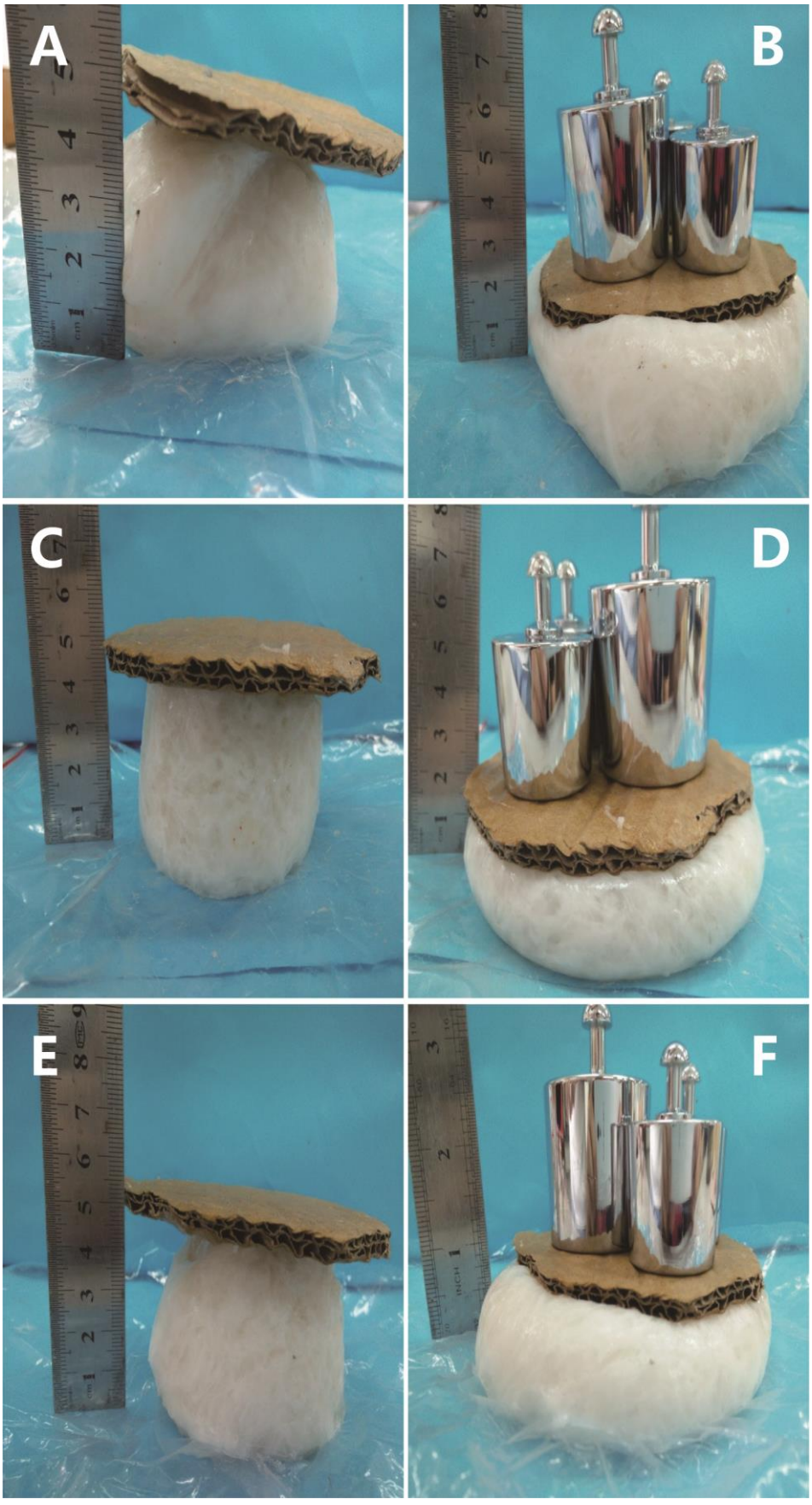
A
B
C
D
E
F

Figure S3 The gelatinous state of the gel after immersion in diesel oil.(A, B) Soak for 30min; (C, D) Soak for 1h; (E, F) Soak for 24h.

SEM analysis

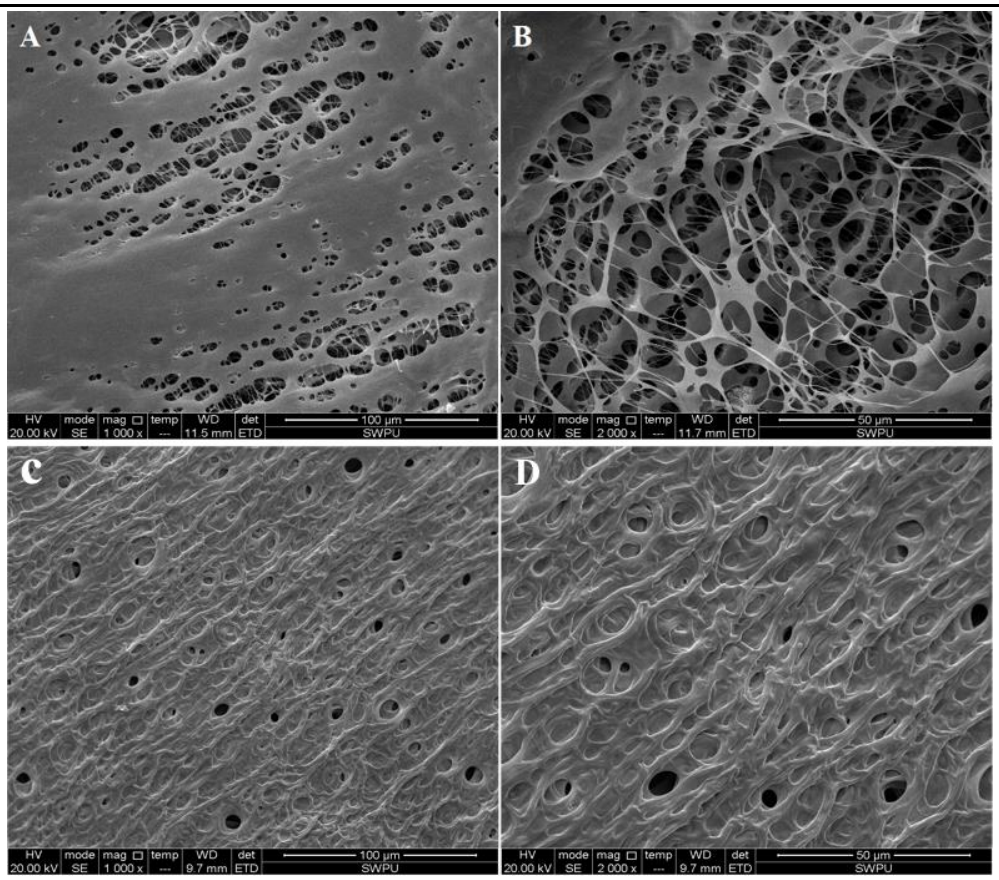

Figure S4. SEM of different samples cross - linked with organic chromium. 1.2 wt% GP-A contains 1.0% wt% organic chromium, scale bar 100 μm (A) and 50 μm (B); 1.2 wt% HPAM contains 1.0% wt% organic chromium, scale bar 100 μm (C) and 50 μm (D).

Atomic Force Microscope (AFM) Analysis

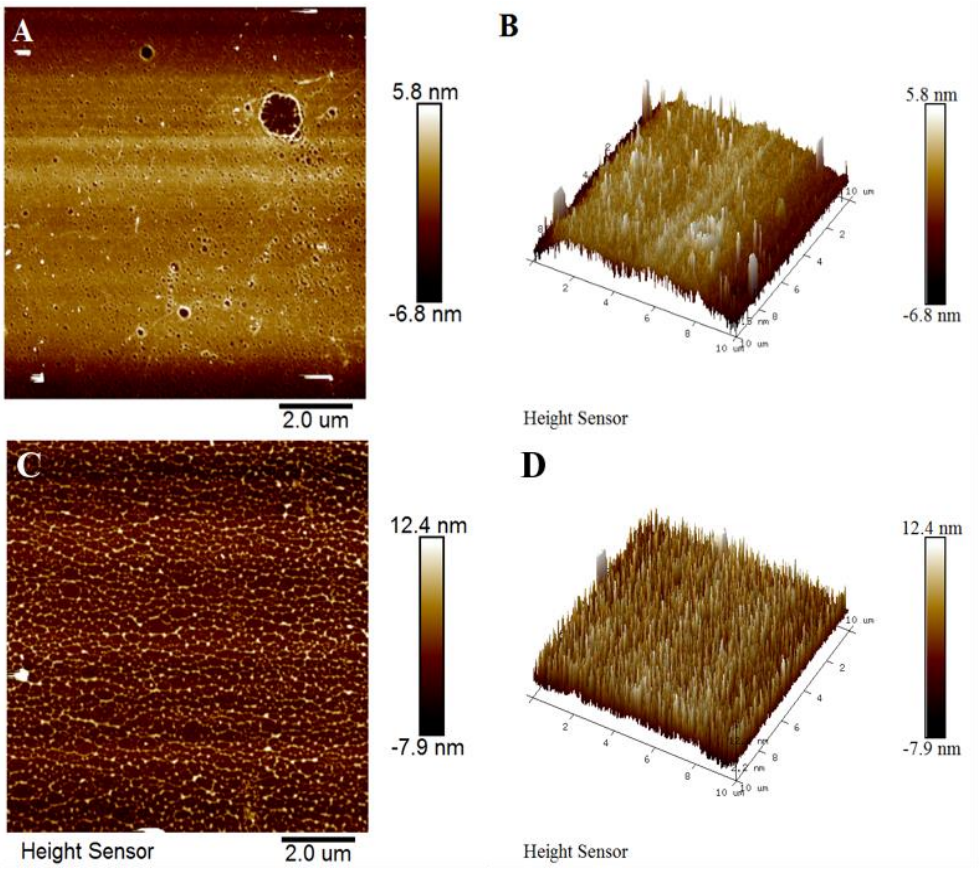

Figure S5. AFM of different samples cross - linked with organic chromium. 1.2 wt% GP-A contains1.0% wt% organic chromium, scale bar 2 μm, 2D diagram (A) and 3D diagram (B); 1.2 wt% HPAM contains 1.0% wt% organic chromium, scale bar 2 μm, 2D diagram (C) and 3D diagram (D).

TEM analysis

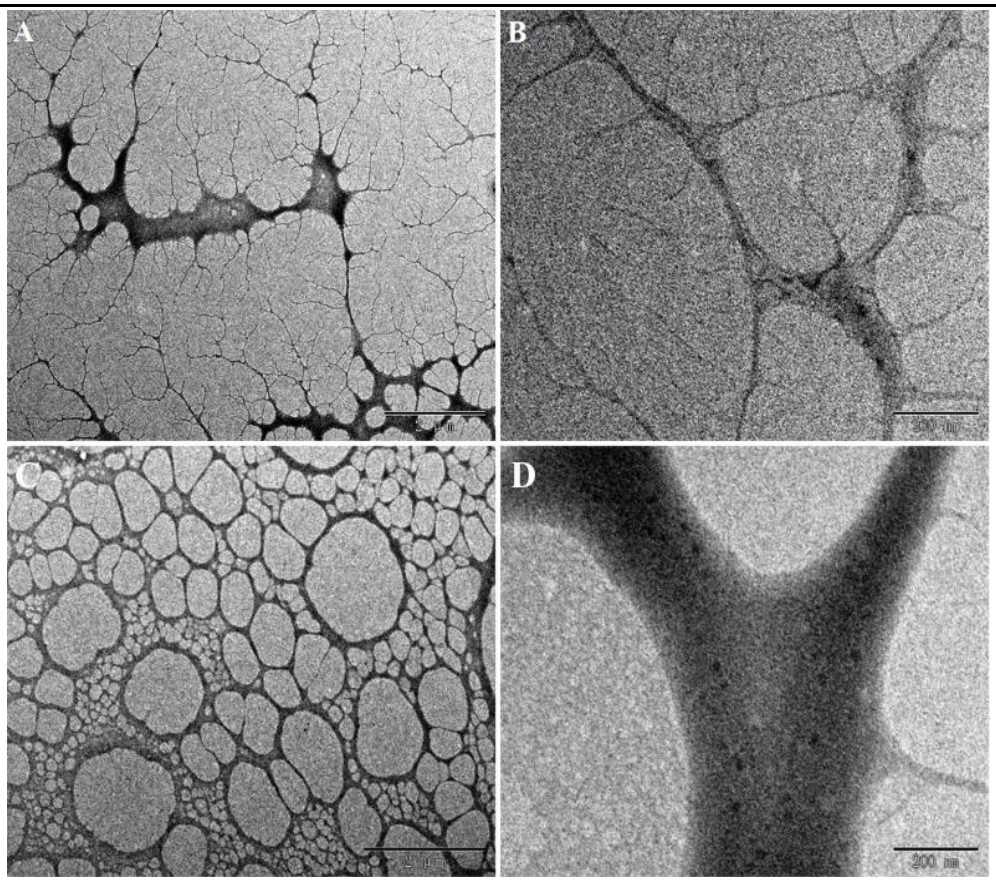

Figure S6. TEM of different samples cross - linked with organic chromium. 1.2 wt% GP-A contains 1.0% wt% organic chromium, scale bar 2 μm (A) and 200nm (B); 1.2 wt% HPAM contains 1.0% wt% organic chromium, scale bar 2 μm (C) and 200nm (D).