Supporting Information

Magnetic soft organogel supercapacitor for energy storage

Xinxian Ma, *‡ Jiuzhi Wei,‡ Yuehua Liang, Juan Zhang, Enke Feng, Zhenxing Fu, Xinning Han*

1. Materials and Experimental apparatus

a) Materials

Polyvinyl alcohol (PVA,polymerization degree 1799,98-99% alcoholysis), Iron acetyl acetone, dibenzyl ether, oleamine, N,N-Dimethylformamide(DMF) and dimethyl sulfoxide (DMSO) were provided by Shanghai Energy Chemical Co. Ethanol, 2,2'-Biimidazole, Ethyl chloroacetate, H_2SO_4, Na_2CO_3, KI, NaOH was purchased from Shanghai Titan Scientific Co.,Ltd. All reagents were not further purified unless otherwise specified

b) Experimental apparatus

NMR spectra were carried out on a Bruker 400 MHz spectrometer with internal standard tetramethylsilane (TMS) and solvent signals as internal references at room temperature(23℃), and the chemical shifts (δ) were showed in ppm. The FT-IR spectra of the synthesized samples and the organogel were measured on an IR spectrophotometer (Thermo Scientific Nicolet iS5) with wavenumbers from 500 to 4000 cm^{-1}. The morphologies of the organogel were characterized with a ZEISS Gemini 300 SEM, the acceleration voltage is 5kV and the magnification is 1500 times . The tensile test of the sample was performed with rheological test on a rheometer (Anton Paar, 302, GmbH) . The electrochemical test of supercapacitor was carried out by CHI660A.

2. Preparation of nano ferric oxide

Scheme 1

Iron acetyl acetone (0.706g) was dissolved in a mixture of dibenzyl ether (10ml) and oleamine (10ml), and the solution was placed in a three-mouth flask and heated to 100℃ under the protection of nitrogen for one hour, then heated to 300℃ under the protection of nitrogen for two hours, and then cooled to room temperature. After adding 40ml

ethanol, the solution was divided into two parts. After centrifugal washing at 3000rpm, extraction was done by freeze-drying.

3. Preparation of PS

Scheme 2

Weigh 5g 2,2' -biimidazole and dissolve it in 150ml N, N-dimethylformamide (DMF), add 18.272g ethyl chloroacetate and 7.901g Na_2CO_3, then add appropriate amount of KI to catalyze, react at 75℃ for 10 hours, and then add 5.964g sodium hydroxide solution for 5 hours. PS was obtained after rinsing with ethanol and drying (Figure I).

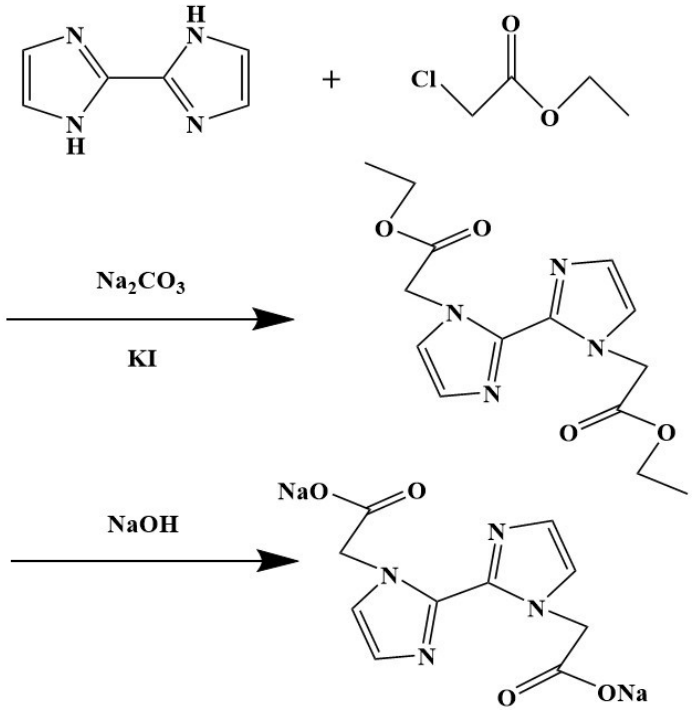

Figure I The synthesis path of PS

4. Solubility of PS

(1) Weigh the 0.045g PS powder and place it at the bottom of the bottle, adding DMSO, EG, Gly separately 2ml each to test its solubility in different solutions. After standing at room temperature (20℃) for 0.5h, we can see that the PS powder is completely dissolved in DMSO, while there is still some residue in other solvents (Figure II A).

(2) Weigh 0.03g, 0.045g, 0.06g of PS powder into the bottom of the bottle, and add 2ml DMSO into it, the solubility in DMSO at different masses was tested. After standing

at room temperature (20℃) for 0.5h, We can see that the PS powder of 0.03g and 0.045g can be completely dissolved, while the PS powder of 0.06g has some undissolved (Figure II B).

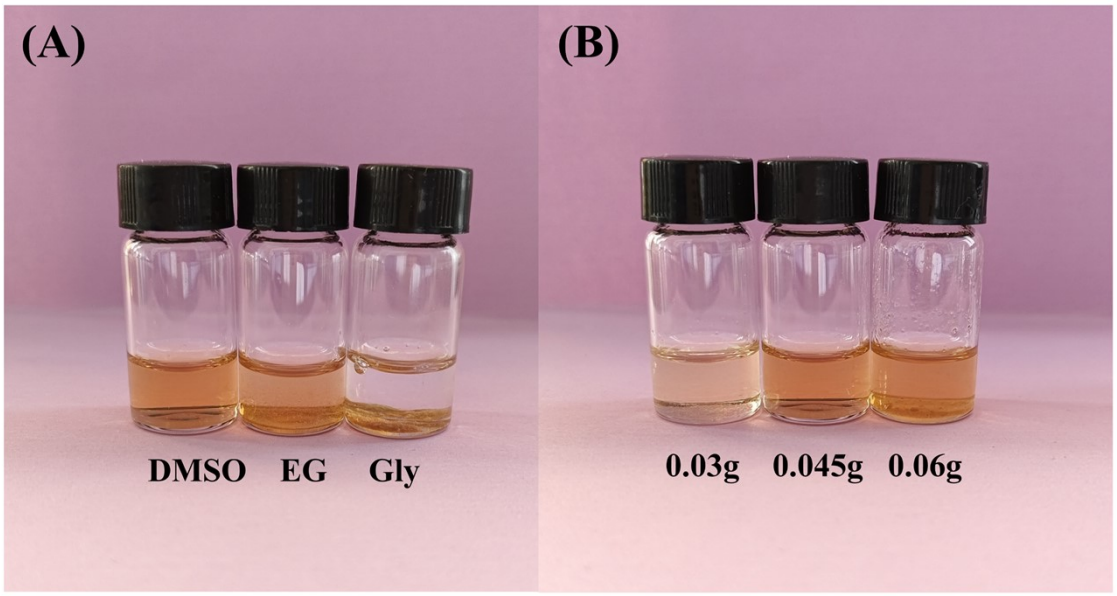

Figure II (A) PS powder is dissolved in different solvents, (B) different quality PS powder is dissolved in DMSO.

5. Preparation and of organogel

PS(0.245g, molar mass 294.18) and H_2SO_4 (80μL) were dissolved in DMSO(10ml) solution and stirred for 0.5h to obtain a homogeneous transparent solution(At 20℃, S (PS) =2.23g/100g DMSO). Then PVA (2.2g, molar mass 44.05) was added to the PS solution and stirred at 120℃ for 5h until completely dissolved. Add Fe_3O_4 (0.77g, molar mass 231.533) and hold for 2h under intense stirring. Next, the resulting mixture is poured into plastic petri dishes and left overnight at minus 20 degrees Celsius. Finally, thaw at room temperature (23℃) for one hour to obtain an organogel with high tensile strength (~700%), strong toughness (can lift 2500g heavy objects) and high magnetism.

6. Preparation and of Flexible Supercapacitor

a) Preparation of flexible supercapacitors

First, AC powder, acetylene black and PVDF adhesive (W/W=8:1:1:) are evenly mixed in NMP solution, and then they are evenly applied to 20 mm x 10 mm stainless steel mesh. After drying at 60 ℃ overnight, the AC electrode was obtained. The weight of the active material for each electrode increases between about 5 - 5.5 mg. Finally, a

sample of organogel (20 mm × 10 mm × 1 mm) is sandwiched between two identical electrodes to make a flexible supercapacitor.

b) Specific capacitance and ionic conductivity calculation

The specific capacitance for a single electrode (Cs, F g^{-1}) was estimated from the GCD curves by the equation was calculated as follow :

$$\mathbf{Cs = 4 \times I \times \Delta t / (M \times \Delta V).}$$

Where I (A) is the discharge current, Δt (s) is the discharge time, M (g) is the total mass of the active material in both electrodes, and ΔV (V) is the actual voltage excluding the IR drop of the discharge process.

The ionic conductivity of the organogel electrolytes are evaluated by the equation was calculated as follow :

$$\mathbf{\sigma = L / (R \times S)}$$

Where σ (mS cm^{-1}) is the ionic conductivity, L (cm) is the distance between the two electrodes, R (Ω) is the bulk electrolyte resistance obtained from EIS, and S (cm^2) is the geometric area of the electrode/electrolyte interface.

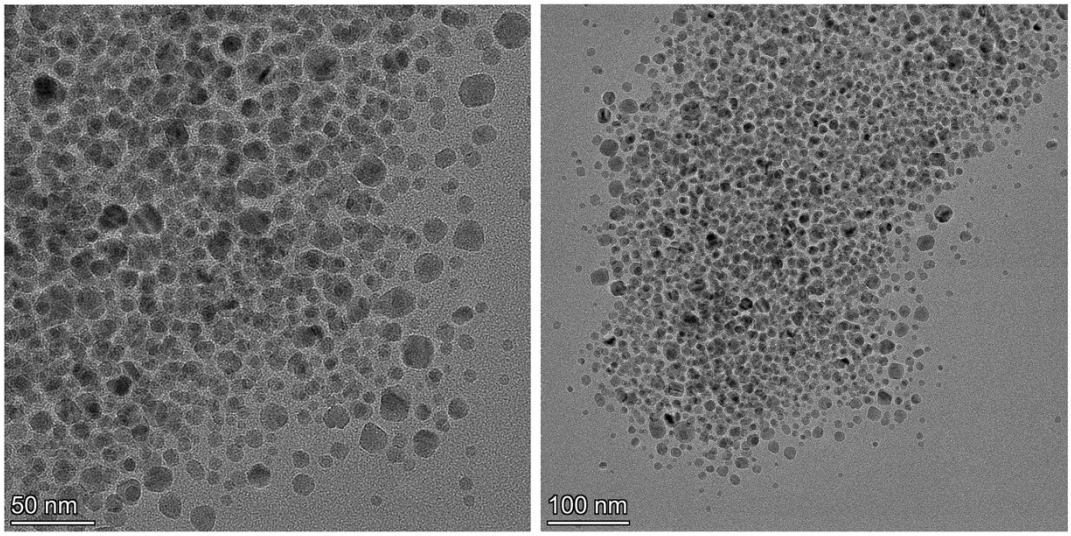

FIG S1 TEM micrographs of Fe_3O_4.

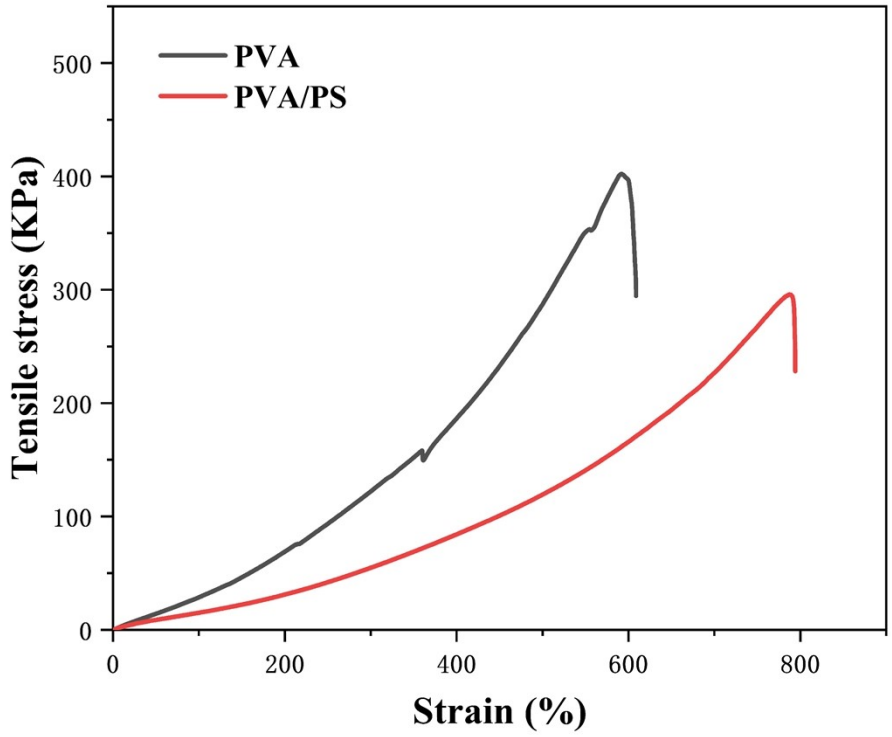

FIG S2 Tensile stress-strain curves of PVA organogel and PVA/PS organogel.

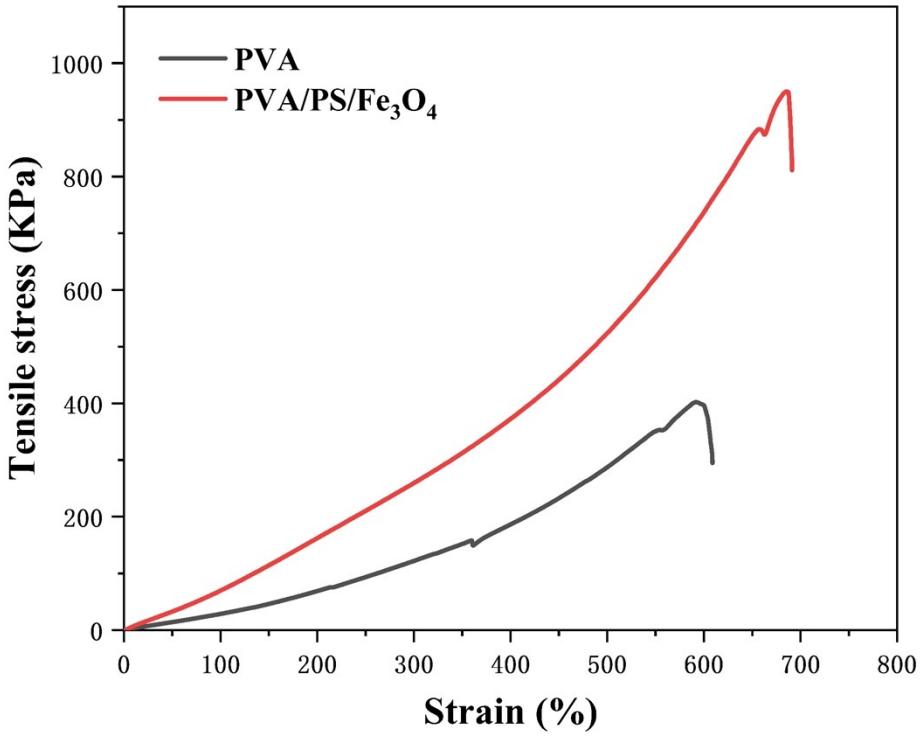

FIG S3 Tensile stress-strain curves of PVA organogel and PVA/PS/Fe_3O_4 organogel.

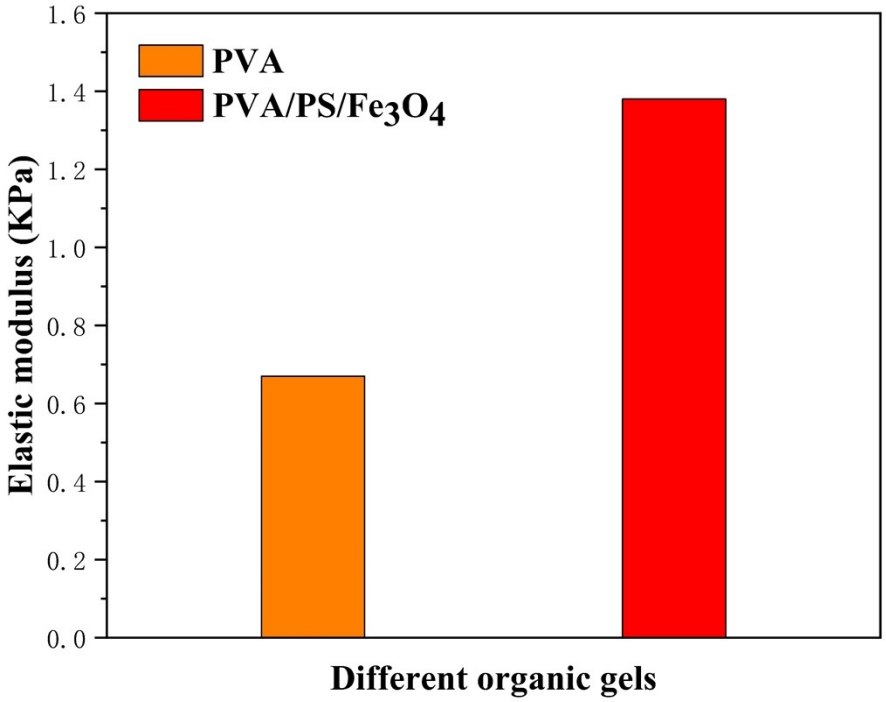

FIG S4 Elastic modulus of PVA organogel and PVA/PS/Fe_3O_4 organogel.

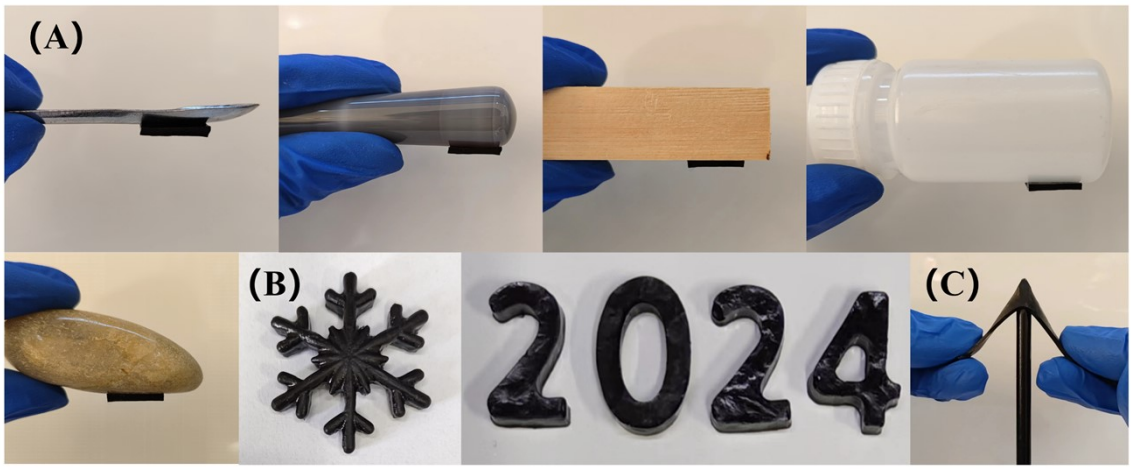

FIG S5 (**A**) PVA/PS/Fe_3O_4 organogel can be attached to various materials, such as iron, agate , wood, plastic and stone. (B) Organogels are made into various shapes.(C) Demonstration of puncture resistance of organogel.

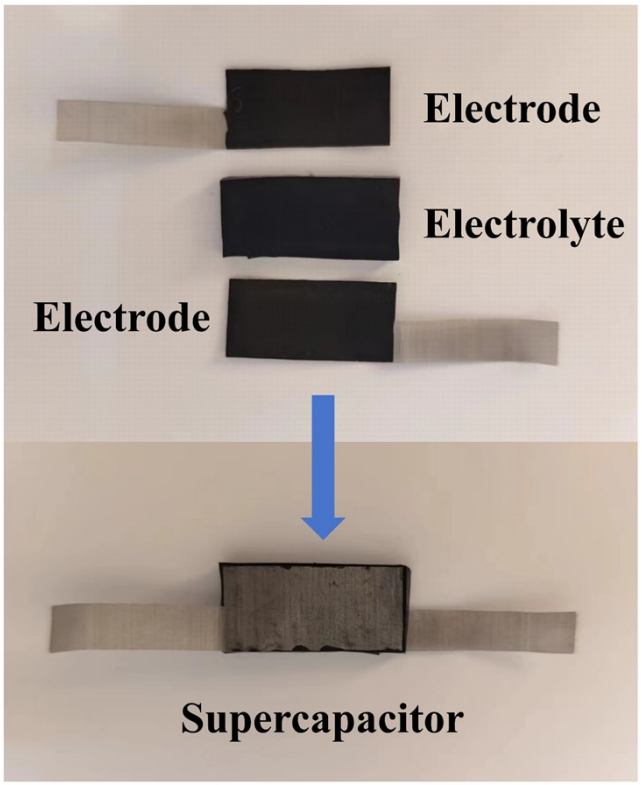

FIG S6 Schematic diagram of the prepared supercapacitor.

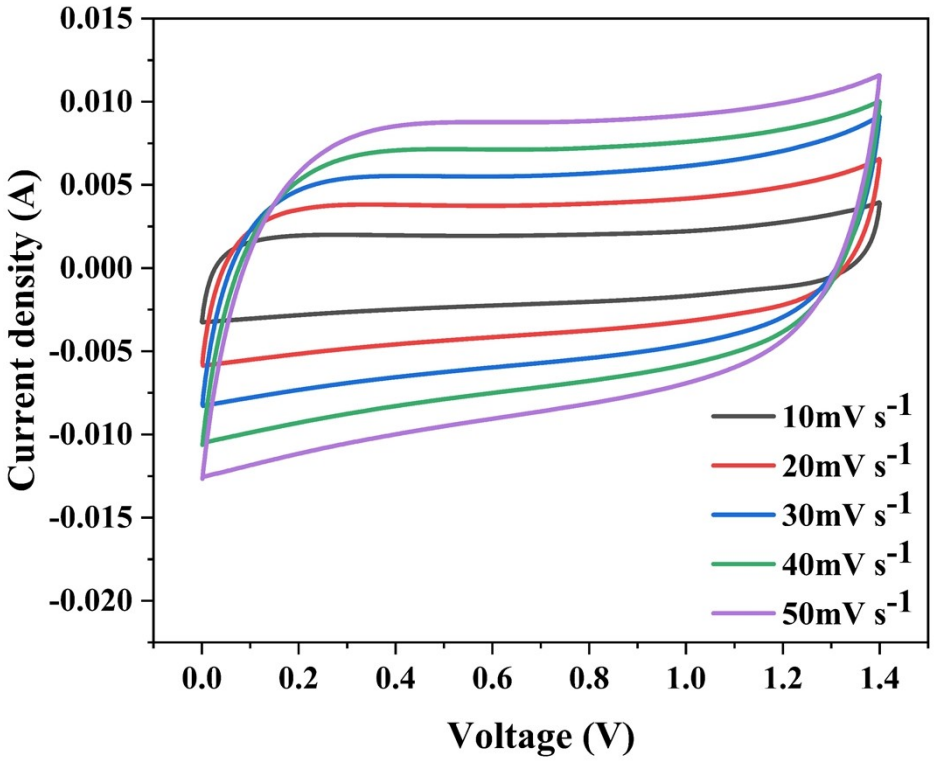

FIG S7 CV curve of PVA/PS organogel.

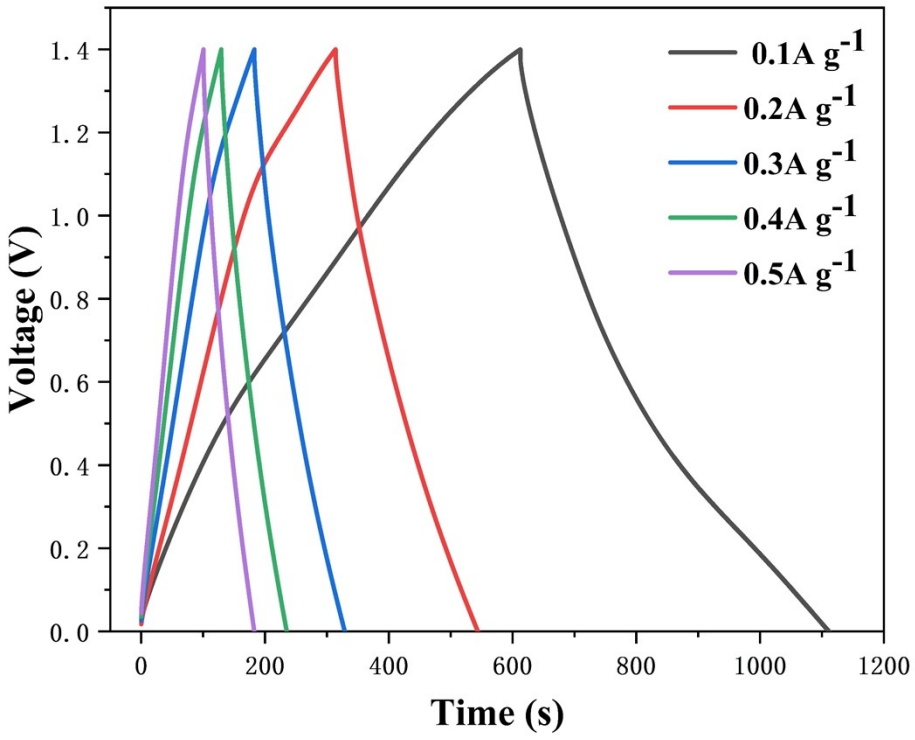

FIG S8 GCD curve of PVA/PS organogel.

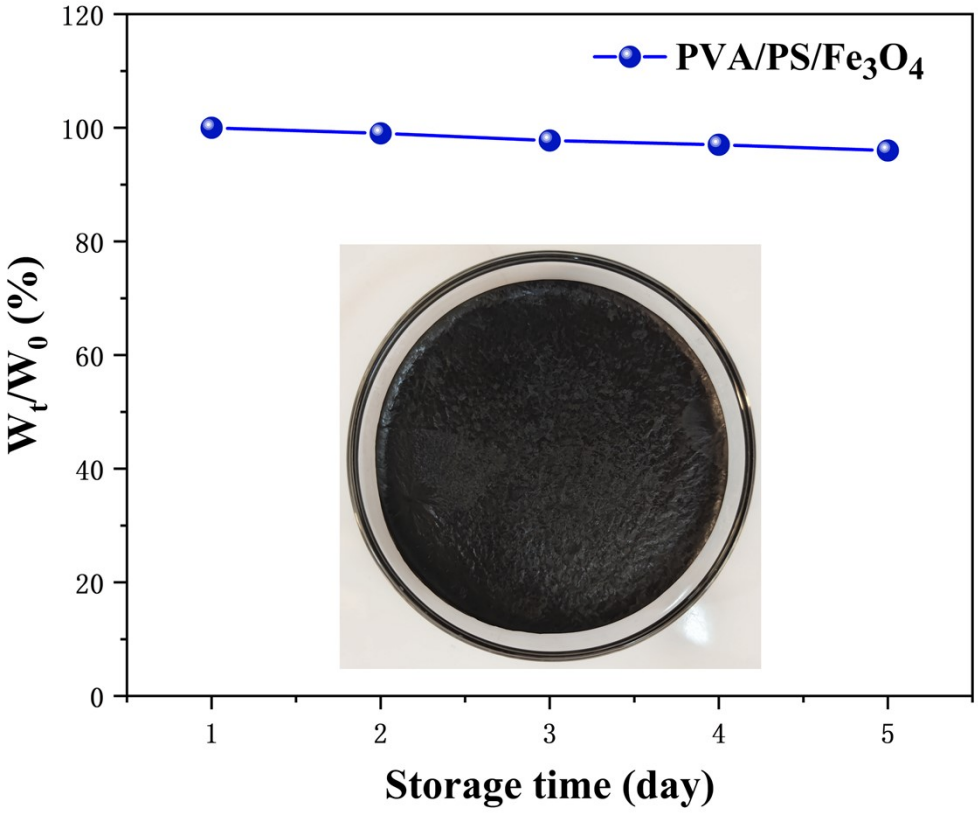

FIG S9 Weight loss ratio of organogel at 23℃ for five days.

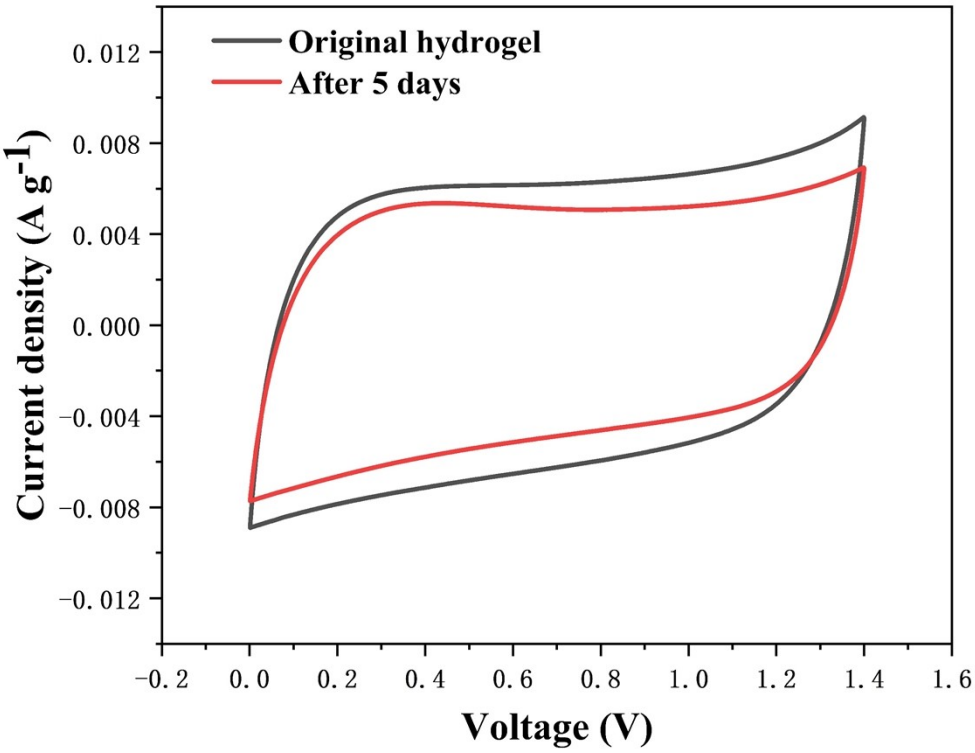

FIG S10 CV curve after five days of PVA/PS/Fe_3O_4 organogel placement.

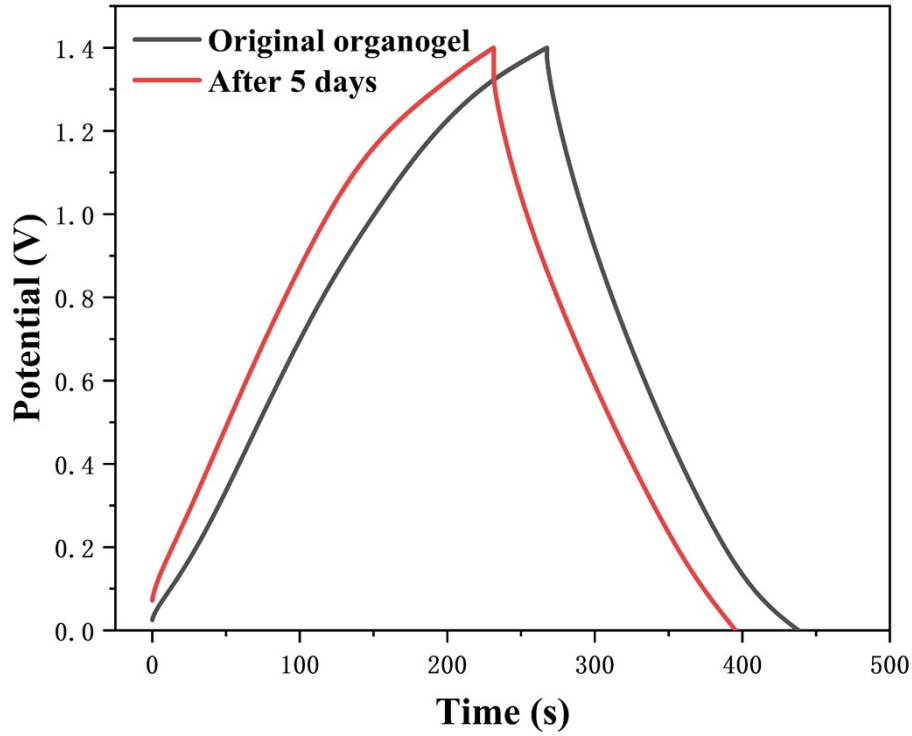

FIG S11 GCD curve after five days of PVA/PS/Fe_3O_4 organogel placement.