Supporting Information

Ultrathin redox active hydrogel electrolytes for high performance flexible supercapacitors

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S1 Materials

Polyvinyl alcohol (PVA) was purchased from Aladdin Biochemical Technology Co., Ltd. carboxymethyl chitosan (CMCS), 1-vinyl-3-ethylimidazolium bromide ([ViEtIm][Br]), activated carbon (AC, YP-80) with the specific surface area of 2100 m^2 g^{-1} was purchased from Kuraray Japan, and sulfuric acid (H_2SO_4) were purchased from Shanghai Macklin Biochemical Co., Ltd. All the reagents are of analytical grade and used without further purification.

S2 Fabrication of the supercapacitor

The 16 mg of activated carbon (AC, YP-80), 2 mg of acetylene black and 2 mg of polyvinylidene fluoride (PVDF) were dispersed in N-methyl-2-pyrrolidone (NMP) to form uniform slurry. Then, the slurry was coated on stainless steel meshes (2 cm × 1.5 cm) with the average mass of 8 mg, and dried at 60 ℃ for 8 h. Finally, a symmetric supercapacitor with sandwich structure (AC/hydrogel electrolyte/AC) was fabricated based on AC as electrode materials and PVA/CMCS and PVA/CMCS-[ViEtIm][Br] hydrogel as electrolyte.

S3. Measurement of ionic conductivity

The conductivity of the hydrogel electrolytes was measured by the impedance spectrum over a frequency range from 0.1 Hz to 10^5 Hz. Typically, a piece of the hydrogel electrolyte was sandwiched between two stainless steel meshes. The area (S) of the electrolyte was 3 cm^2 and the distance (L) between the two stainless steel meshes was 0.2 cm. Bulk resistance R (Ω) was determined by the intercept with the real axis. Ionic conductivity σ (mS cm^{-1}) was calculated by the following formula:

$$\sigma = L / (R \times S) \qquad (1)$$

S4. Characterizations

Fourier transform infrared (FT-IR) spectra was recorded on a Spectrum One spectrometer

(PerkinElmer, U.S.A). The morphology of electrolytes was examined by using FE-SEM (Ultra Plus, Carl Zeiss) at an acceleration voltage of 5 kV.

S5. Electrochemical measurements

The electrochemical performances of the two-electrode supercapacitor devices were evaluated by using a CHI660D electrochemical workstation (Chenghua, Shanghai, China). Electrochemical impedance spectroscopy (EIS) measurements were conducted at open circuit potential in the frequency range of 100000 Hz to 0.1 Hz with an alternate voltage amplitude of 5 mV.

The specific capacitance of the cell (C_{cell}) and specific capacitance of the electrode (C_s) were calculated according to follow formulas:

$$C_{cell} = I\,\Delta t / m\,\Delta V \qquad (2)$$

$$C_s = 4\,C_{cell} \qquad (3)$$

$$C_v = 2\,C_s / h \qquad (4)$$

where I, Δt, m, ΔV and h are the discharge current, discharge time, the total mass of two pieces of active electrodes, the voltage change upon discharging (excluding the IR drop) and h is the thickness of the device including the two working electrodes and the active material, respectively. Similarly, the energy density (E_{cell}, μWh cm^{-2}) and power density (P_{cell}, μW cm^{-2}) were estimated using the follow formulas:

$$E_{cell} = C_s\,\Delta V^2 / 8 \qquad (5)$$

$$P = E_{cell} / \Delta t \qquad (6)$$

Figure S1 The diagram of PVA/CMCS-[ViEtIm][Br] gel film.

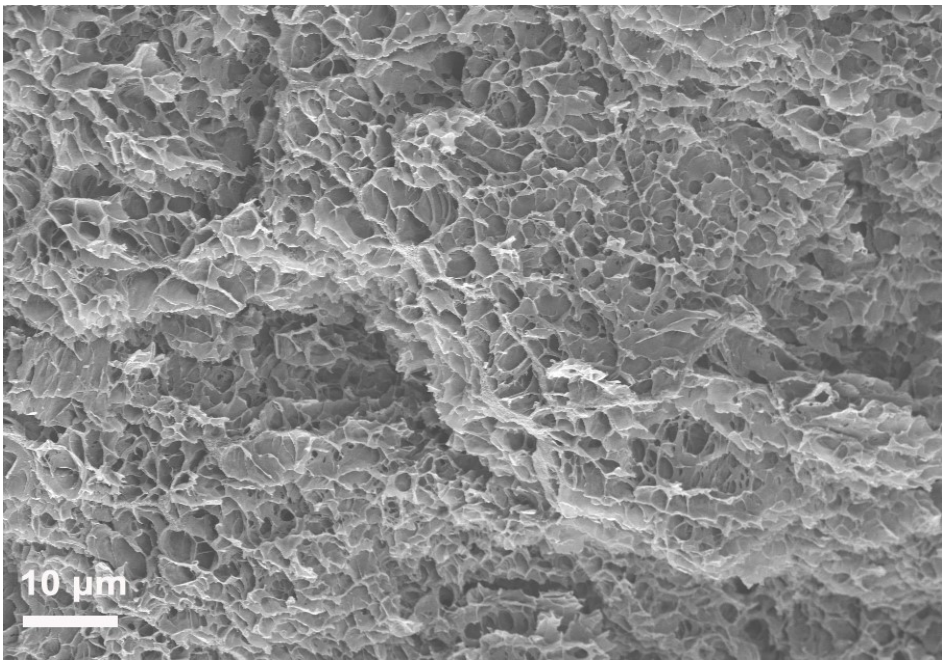

Figure S2 The SEM images of PVA hydrogel.

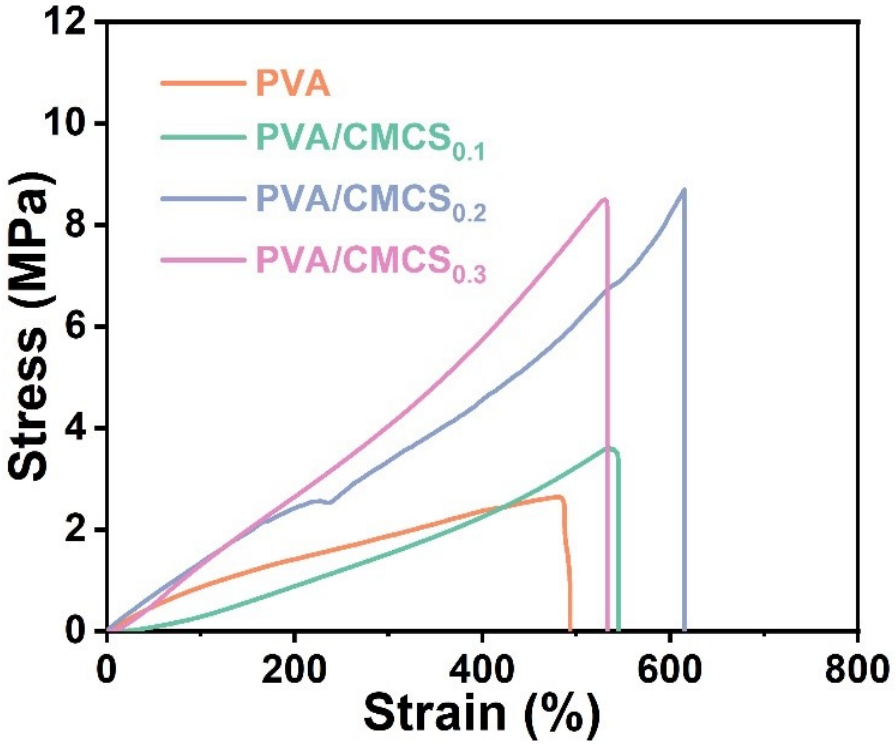

Figure S3 The stress-strain curve of PVA/CMCS gel with different contents of CMCS.

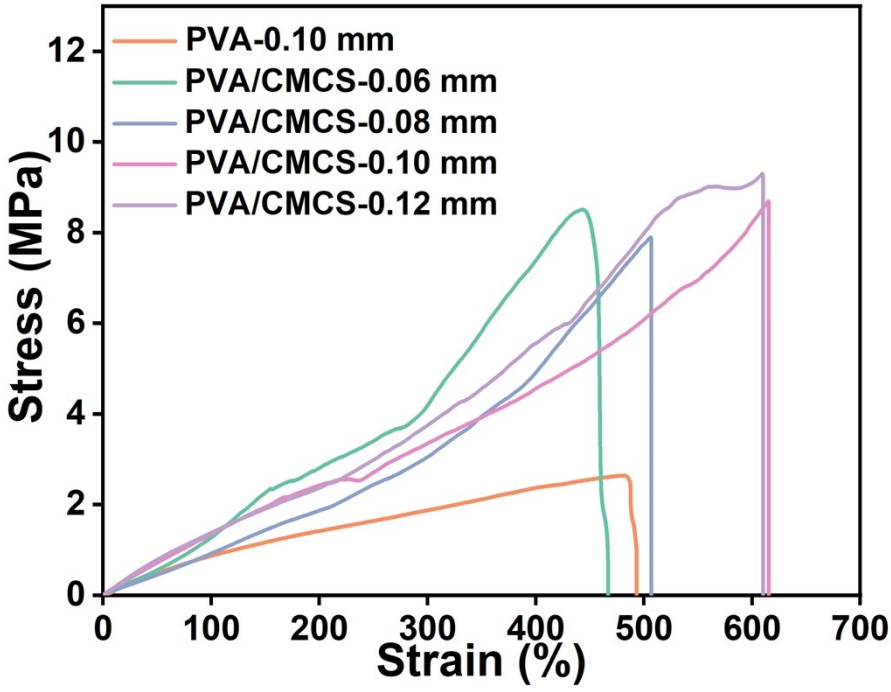

Figure S4 The stress-strain curve of PVA/CMCS gel with different thicknesses.

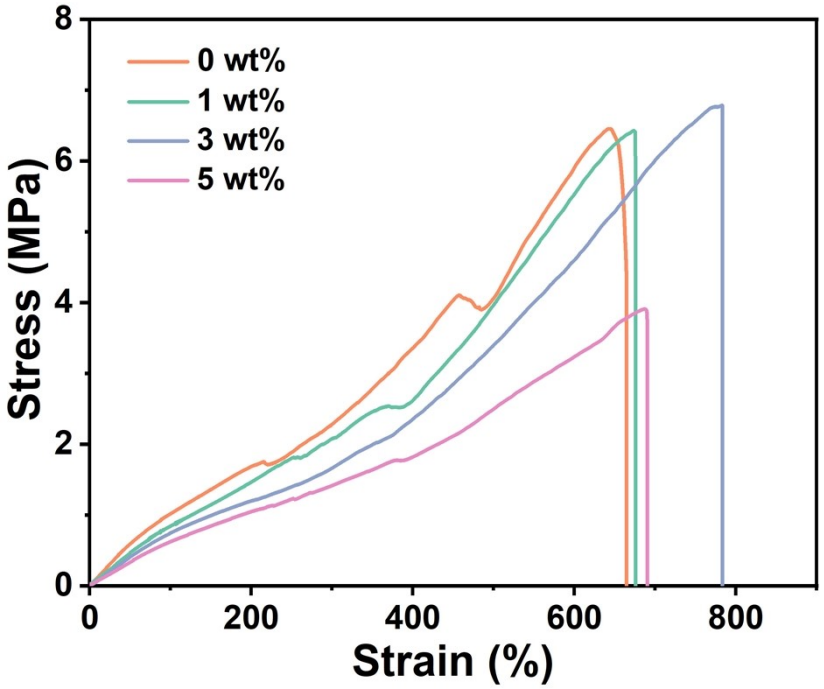

Figure S5 The stress-strain curve of PVA/CMCS-[ViEtIm][Br] gel with different concentrations of [ViEtIm][Br].

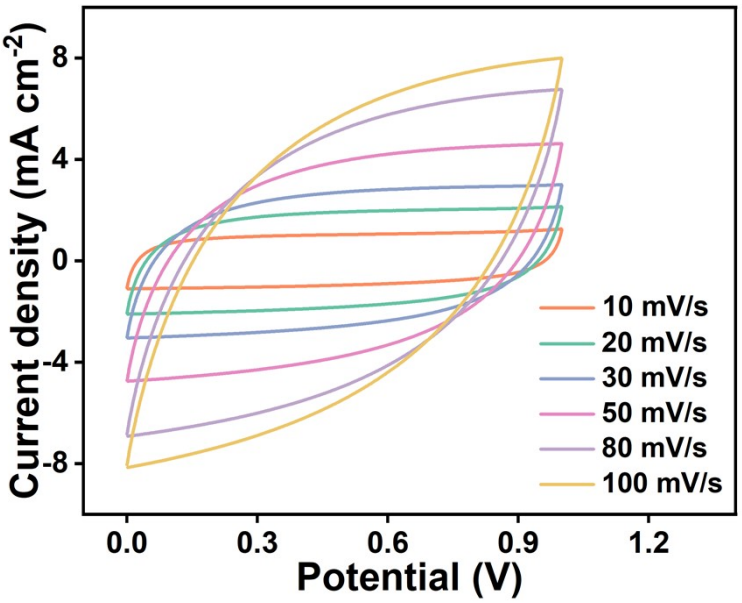

Figure S6 CV curves of PVA/CMCS gel-based supercapacitor with 0.2 g concentrations of CMCS at different scan rates.

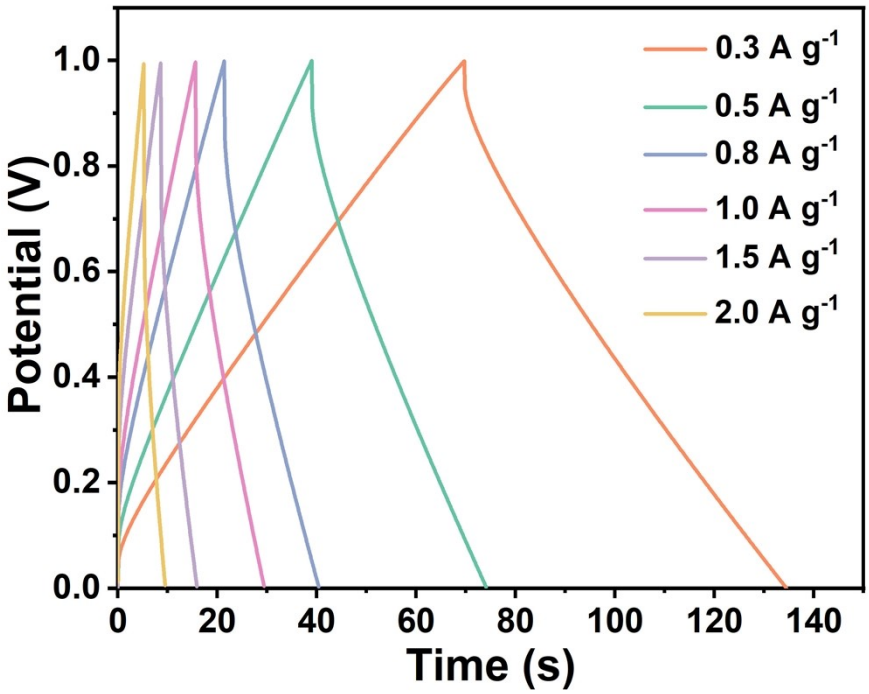

Figure S7 GCD curve of PVA/CMCS gel-based supercapacitor with 0.2 g concentrations of CMCS at different current densities.

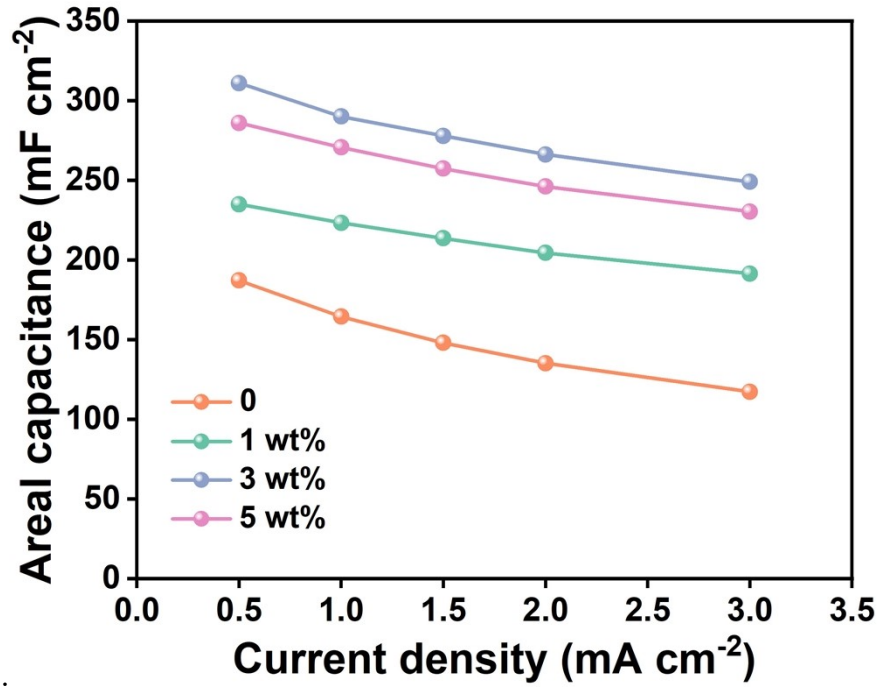

Figure S8 Area capacitance of gel-based capacitors with different [ViEtIm][Br] contents at different current densities.

Table S1. Comparison of electrochemical properties of our device with recently reported gel-based supercapacitors.

Materials	Specific Volume	Cyclic Stability	Specific Capacitance (μ W cm^{-2})	Energy Density (μ Wh cm^{-2})	Refs.
PVA/CMCS-[ViEtIm][Br]	310 mF cm^{-2} (0.5 mA cm^{-2})	87.5% (10000 cycles)	540	78.6	This work
poly(pyrrole-co-aniline) incorporated PVA/PEG	773 mF cm^{-2} (0.2 mA cm^{-2})	-	100	54	[S2]
PANI-PVA/PA hydrogel (electrolyte)-PANI	356.5 mF cm^{-2} (1 mA cm^{-2})	160% (5000 cycles)	800	31.7	[S3]
carbonate-poly(methylmethacrylate)-[EMIM][TFSI]	-	-	352	24.7	[S4]
Graphene paper (PANi-GP)	176 mF cm^{-2} (0.2 mA cm^{-2})	-	250	17.1	[S5]
hydro-thermally reduced graphene oxide nanosheets	-	-	63.7	5.3	[S6]
Sodium Alginate/Zwitterionic	1.96 mF cm^{-2} (50 μ A cm^{-2})	95% (10000 cycles)	290	5.8	[S7]
PAD/H_2SO_4-PANI	228 mF cm^{-2} (5 mA cm^{-2})	-	1500	11.5	[S8]
PVA-CNM-PANI	-	90% (5500 cycles)	~300	~10.3	[S9]

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