Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Construction of Supramolecular-Polymer Hydrogel Electrolyte with Ionic Channels for Flexible Supercapacitors

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Experimental

Materials

All the following chemical reagents were used without further purification. Poly (vinyl Alcohol) (n=approx. 1700 degree of saponification 97.0 to 100.0 mol % Aladdin Co., Japan), 2-Amino-9-beta-D-ribofuranosyl-9H-purine-6-(1H)-one hydrate (guanosine, Energy Chemical Co., China), KOH (Sinopharm Chemical Reagent Co., China), H_3BO_3 (Tianjin Beichen Fangzheng Chemical Reagent Factory Co., China), AC (Activated carbon, Nanjing XFNANO Co., China), PTFE (polytetrafluoroethylene D210C DAIKIN Co., Japan).

Preparation of original PVA gel and PVA-GB gel

Typically, the PVA hydrogel was prepared by dissolving 0.15 g PVA into 3 mL distilled water with stirring at 90 ^oC to form a clear solution. After cool the solution to room temperature, 1 mL KOH (10.7 M) solution was added to the solution drop by drop under stirring. The mixture was placed at room temperature for 72 h to form the original PVA hydrogel.

For the PVA-GB hydrogel, firstly, a certain quality of guanosine (0, 56, 112, 168 mg) and an appropriate amount of KOH (0.6 mM) and H_3BO_3 (1.9 mM) was dissolved in 4 mL deionized water. The mixture was ultrasound 30 s and heated to 90 ^oC to form a transparent solution. Then, mixing the hot solution with the above fresh PVA solution quickly to form a homogeneous mixture. Pour the mixed solution into a PTFE mold and let it polymerize spontaneously at room temperature for 24 hours to get the PVA-GB hydrogel.

Construction of all-solid-state SCs device

For the construction of the symmetric quasi-solid-state SCs device, active carbon (AC) serves as the positive and negative electrodes. The AC electrode material was prepared by mixing AC (80 mg, 80 wt%), carbon black (10 mg, 10 wt%), PTFE (10 mg, 10 wt%) and a few drops of ethanol to form a mixture slurry. Then, the as-prepared slurry was dried at 70 ^oC under vacuum for 12 h to remove the solvent. Finally, the dried slurry was pressed on NF with a mass loading of 2 mg. The device was assembled by employing the PVA-GB gel as electrolyte and separator between two AC electrodes

to form a sandwich structure.

Characterizations

Fourier transform infrared spectroscopy (FTIR) tests were performed on a Thermo Scientific Nicolet IS 50 spectrometer in the wavelength range of 4000-400 cm^{-1}. The crystalline structures were analyzed by X-ray diffraction patterns (XRD, Rigaku Dmaxrc diffractometer with Cu Kα radiation at λ = 1.541 Å). The water content of the samples were examined using Thermogravimetric Analyzer (TG 209 F3, Netzsch). The Morphologies of samples were studied by scanning electron microscopy (SEM, Hitachi S-4800). All the electrochemical measurements were carried out at room temperature on a CHI760E electrochemical working station (Chenhua, Shanghai). LAND cycler (Land Electronic, Wuhan) was employed for electrochemical tests. Then the specific capacitance (C and Cs, F g^{-1}), energy density (E, Wh kg^{-1}) and power density (P, W kg^{-1}), were according to the following equation.

$$C = \frac{I \times \Delta t}{\Delta V}$$

$$Cs = 4 \times C$$

$$E = \frac{C \times \Delta V^2}{7.2}$$

$$P = \frac{3600 \times E}{\Delta t}$$

Table S1. The σ of other previous reports based on hydrogel electrolyte

Electrolytes	**Ionic conductivity/ mS • cm^{-1}**	**References**
PVA/CH_3COONH_4/BmImBr	9.29±0.1	1
PVAG1.0N0.8	62.26	2
PVA-g-PAA/KCl	41	3
PVA-BMIMCl-Li_2SO_4	37	4
B-PVA/KCl/GO	47.5	5
PPDE-LiCl-EV	20	6
AG/PAAm/LiCl	13±0.8	7
PVA–H_2SO_4–ARS	33.3	8
PVDF-HFP/IL/DPA/KI	4.52	9
PKF4	45.56	10
PVA–$LiClO_4$	31.3	11
PVA–H_2SO_4–IC	20.07	12
HA-GPE	74.1	13
PVA-Li_2SO_4-BMIMCl	46	14
PVA-GB	70	This work

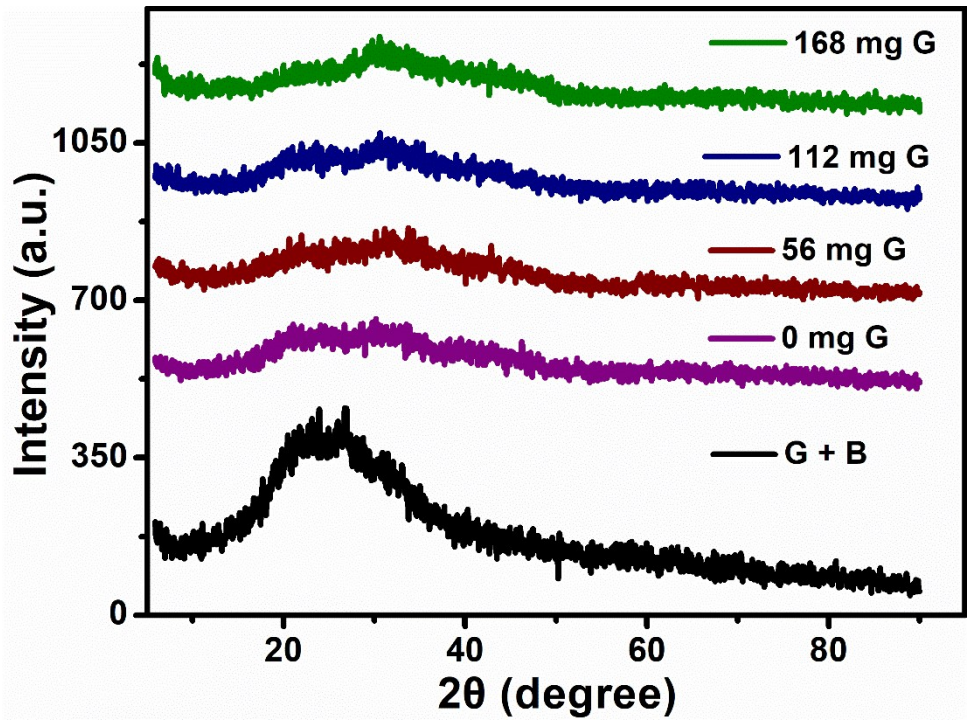

Figure S1. The XRD patterns of PVA-GB hydrogel with different contents G.

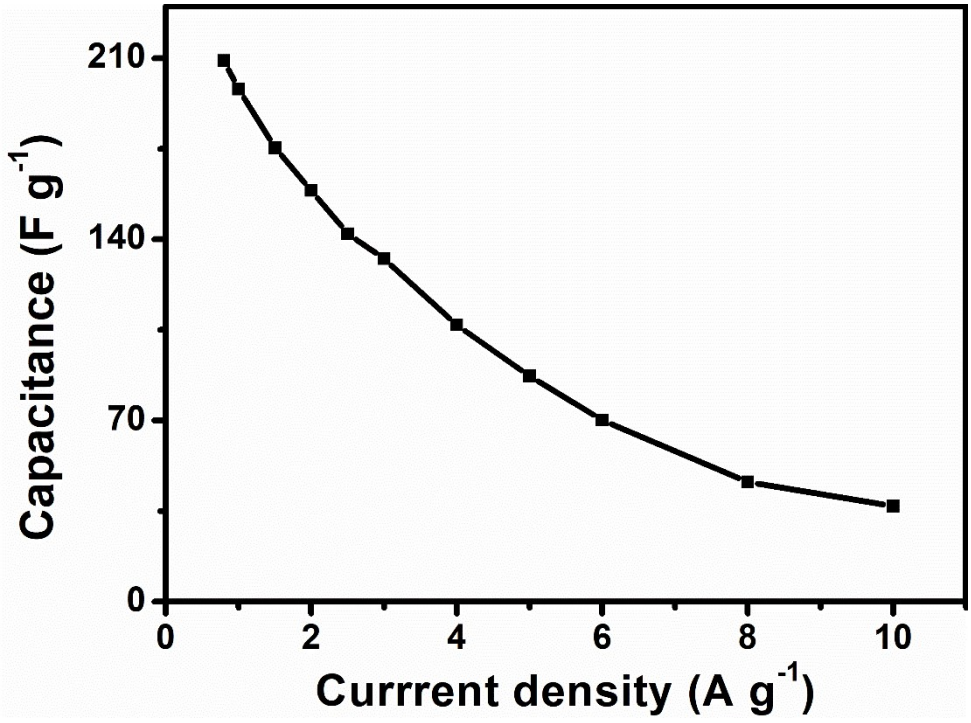

Figure S2. The rate performance of the device with PVA-GB (112 mg G) as the electrolyte at a wide range of current densities.

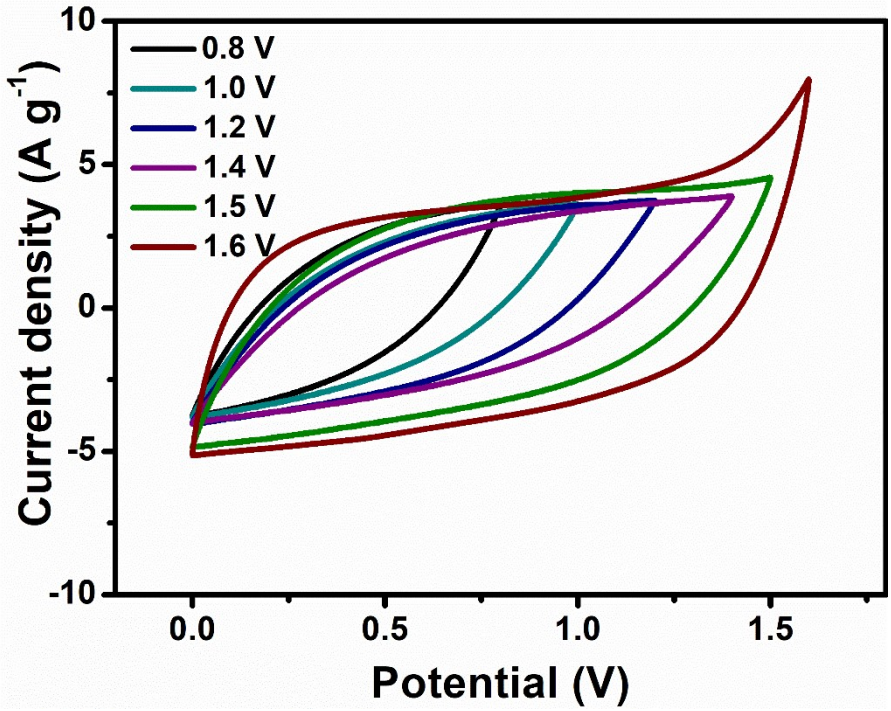

Figure S3. The CV curves at 100 mV s^{-1} with different voltage ranges from 0 ~ 0.8 V to 0 ~ 1.6 V.

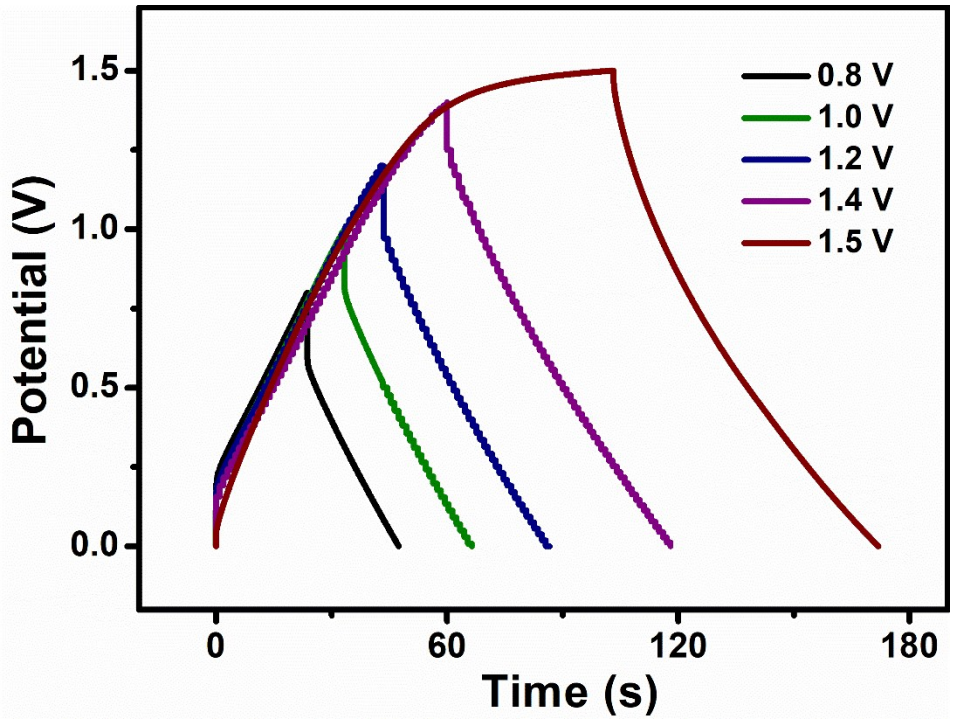

Figure S4. The GCD curves with different voltage ranges at 1 A g^{-1}.

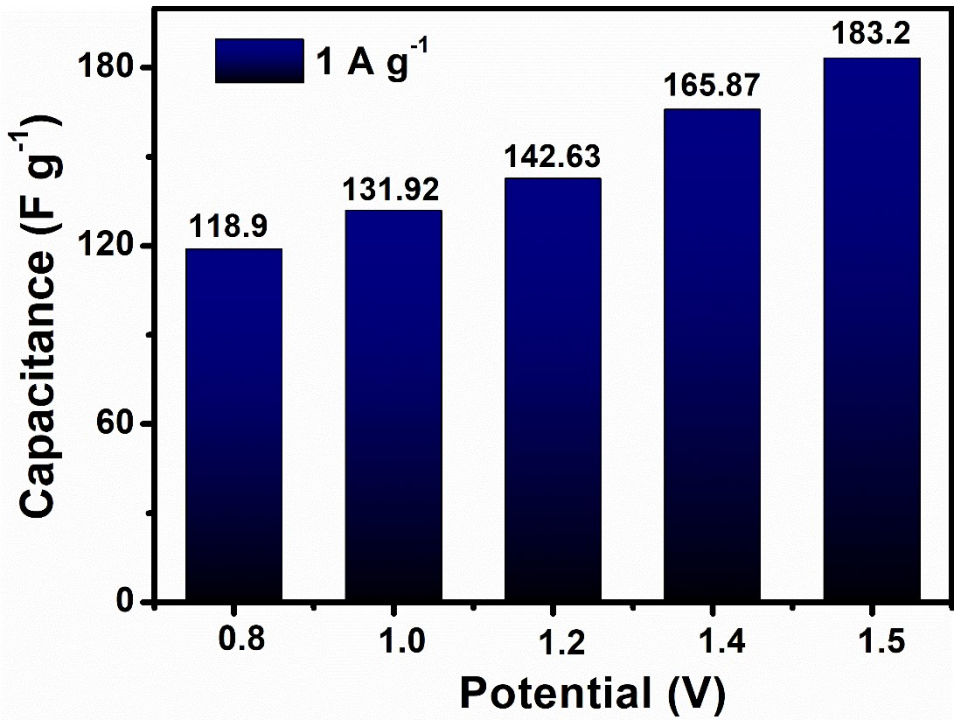

Figure S5. The specific capacitance of the device with different voltage ranges at 1 A g^{-1}: 0 ~ 0.8 V, 0 ~ 1.0 V, 0 ~ 1.2 V, 0 ~ 1.4 V and 0 ~ 1.5 V.

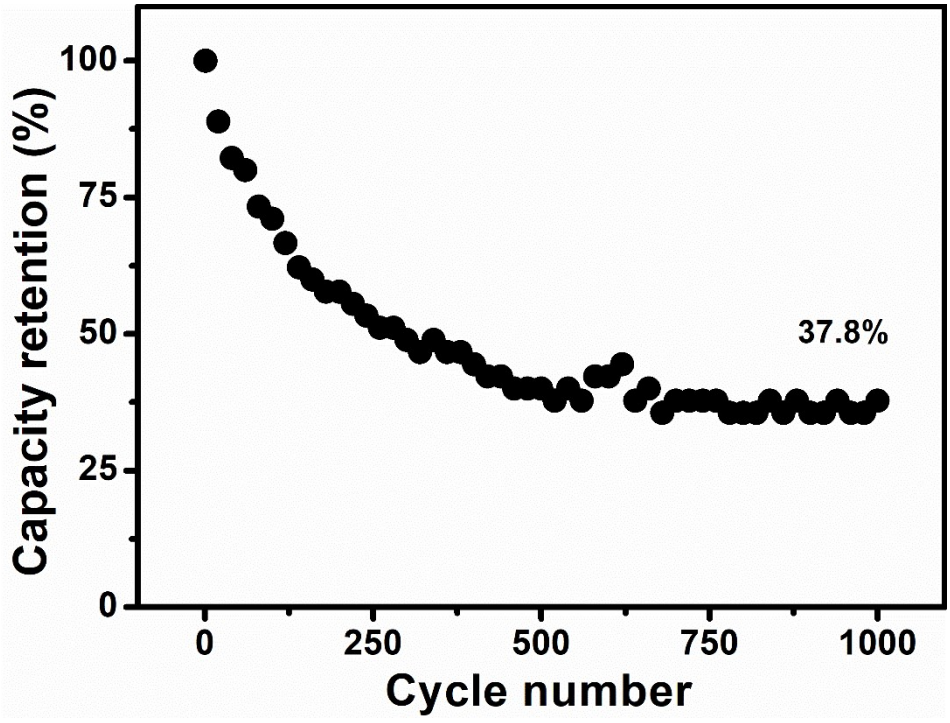

Figure S6. the cycle stability of the PVA hydrogel electrolyte-based device

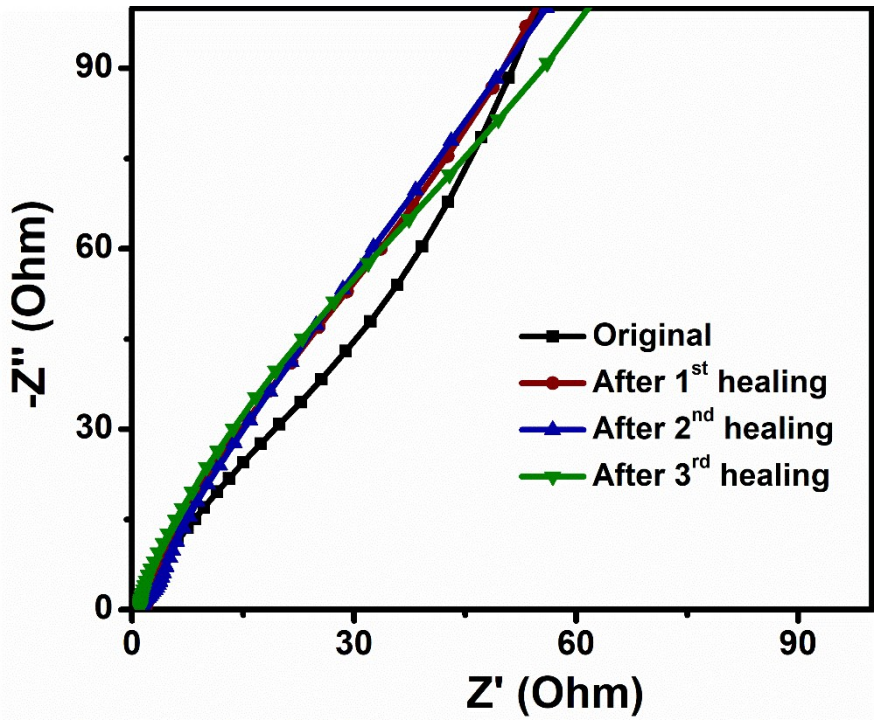

Figure S7. The Nyquist plots after different cut-healing cycles.

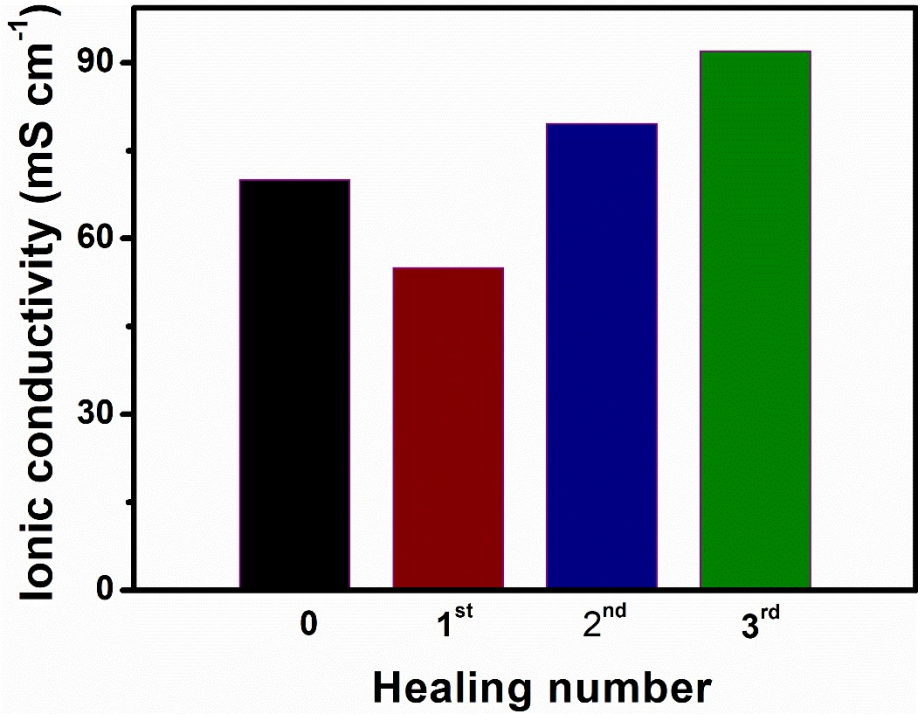

Figure S8. The ionic conductivity after different cut-healing cycles.

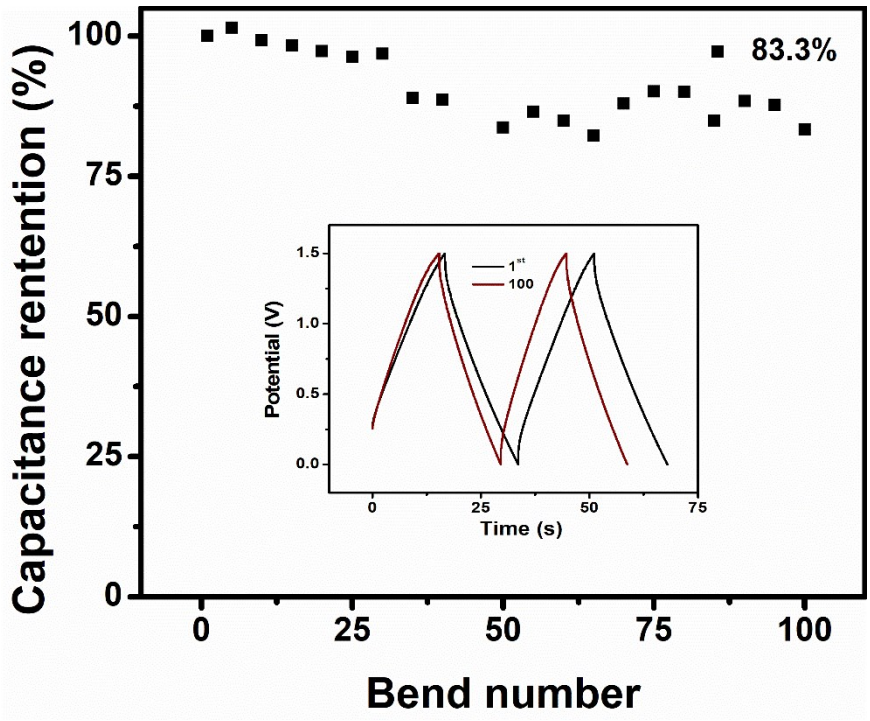

Figure S9. The capacitance retention of the device after different bending times.

Supporting References

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