Supporting Information

One-pot Synthesis of Double-network Hydrogel Electrolyte with Extraordinarily Excellent Mechanical Properties for Highly Compressible and Bendable Flexible Supercapacitor

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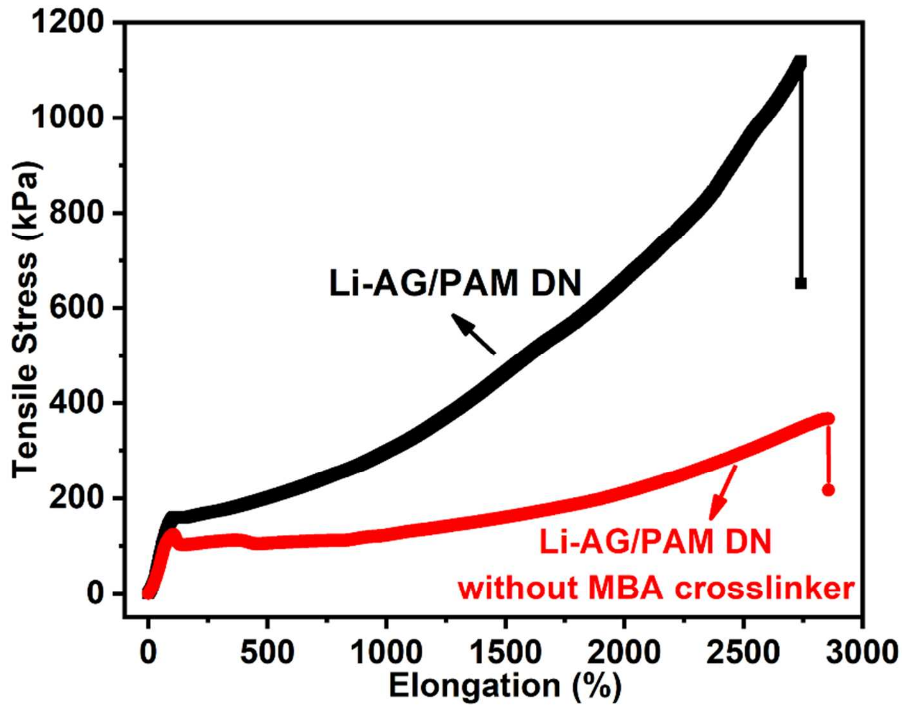

Figure S1. Tensile stress-elongation cures of the Li-AG/PAM DN hydrogel electrolyte and the hydrogel electrolyte synthesized by the same method of the Li-AG/PAM DN hydrogel electrolyte without the addition of MBA crosslinker.

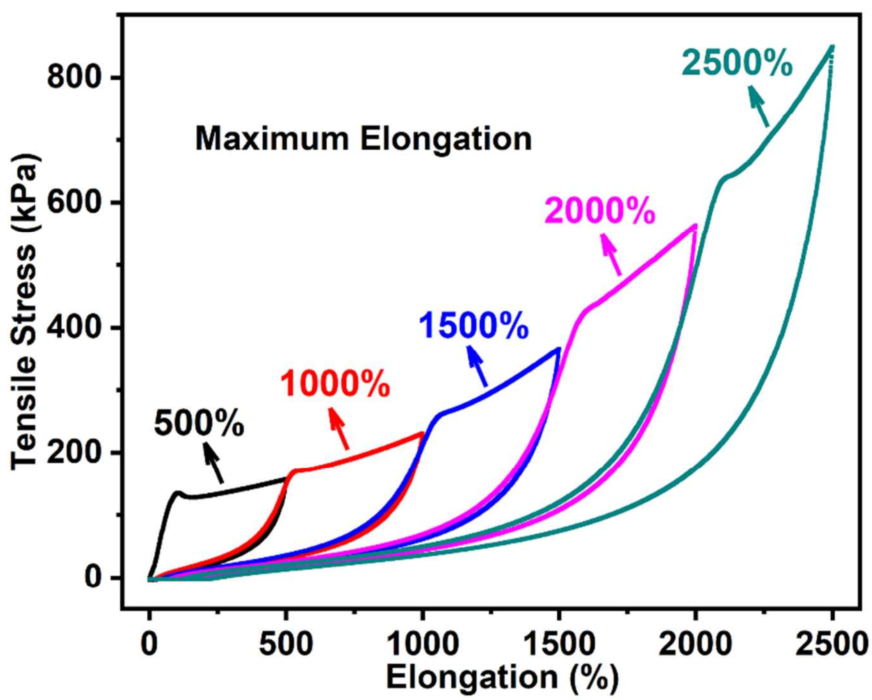

Figure S2. Successive cyclic tensile tests of the Li-AG/PAM DN hydrogel electrolyte at a maximum elongation of 500%, 1000%, 1500%, 2000%, and 2500%, respectively.

Table S1. Energy dissipation values of the Li-AG/PAM DN hydrogel electrolyte during cyclic tensile tests at different maximum elongations.

Maximum Elongation (%)	500	1000	1500	2000	2500
Energy Dissipation (MJ/m^3) [a]	45.96	70.73	106.68	168.56	247.70

[a] During each tensile cycle, hydrogel electrolyte specimens were stretched to a maximum elongation ($\lambda_{\max}$) and then returned to the original height, so the energy dissipation (E_d) was estimated by area between the loading-unloading curves ($E_d = \oint_0^{\lambda_{\max}} \sigma \, d\lambda$), where σ and λ are tensile stress and elongation, respectively.

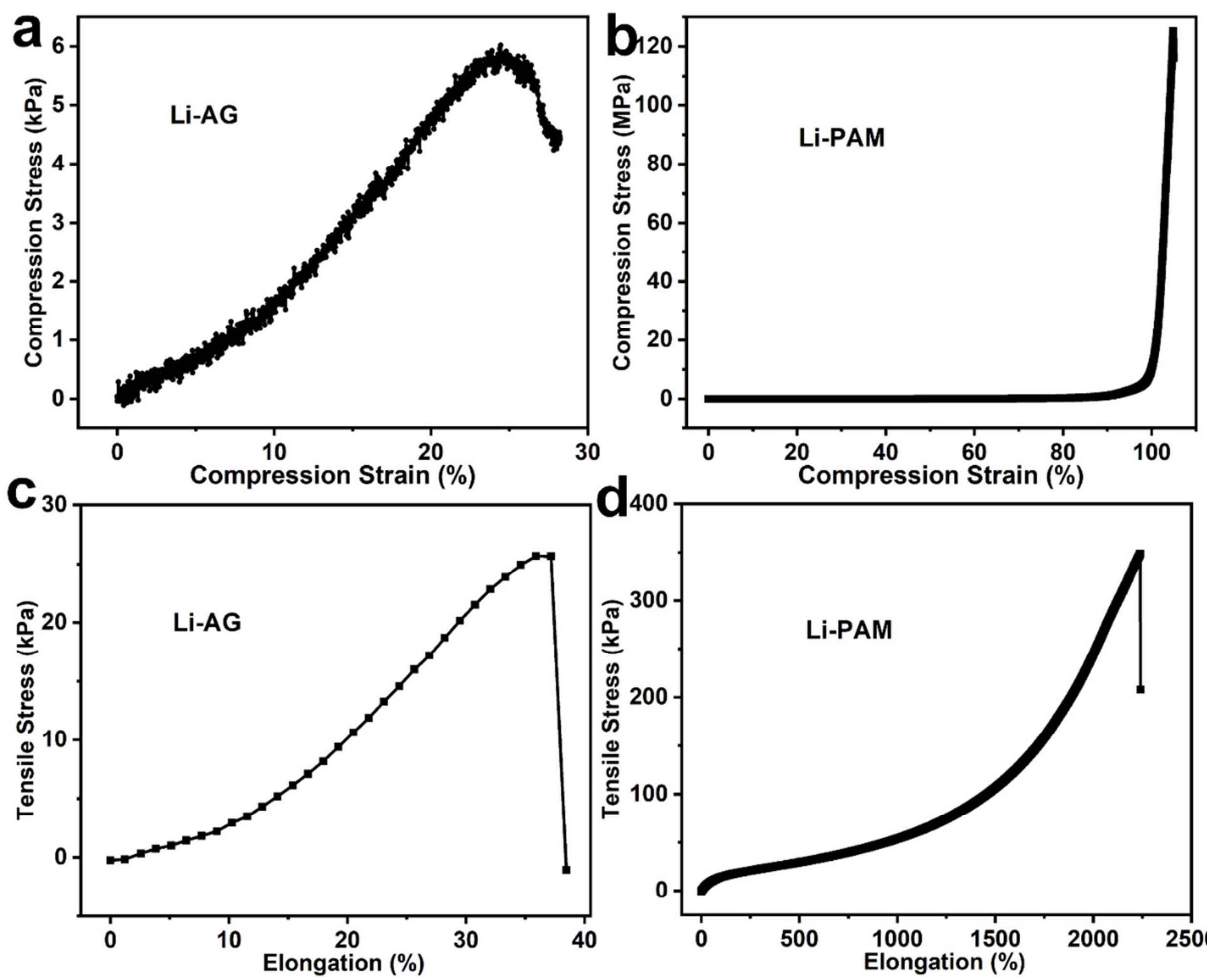

Figure S3. Compression stress-compression strain curves of (a) the Li-AG hydrogel electrolyte and (b) the Li-PAM hydrogel electrolyte and tensile stress-elongation curves of (c) the Li-AG hydrogel electrolyte and (d) the Li-PAM hydrogel electrolyte.

Table S2. Mechanical properties of the Li-AG hydrogel electrolyte and the Li-PAM hydrogel electrolyte.

Hydrogel electrolyte	Tensile properties		Compression properties	
	Tensile strength (kPa)	Elongation at break (%)	Compression strength (MPa)	Fracture compression strain (%)
Li-AG	27±2 [a]	34±3	0.0058±0.0007	27±3
Li-PAM	319±44	2186±91	131±5	above 99.9 [b]
Li-AG/PAM DN	1103±24	2780±55	150±7	above 99.9

[a] All Error bars show standard deviation; sample size n = 3.

[b] The fracture compression strain of hydrogel electrolytes is too high to be precisely measured.

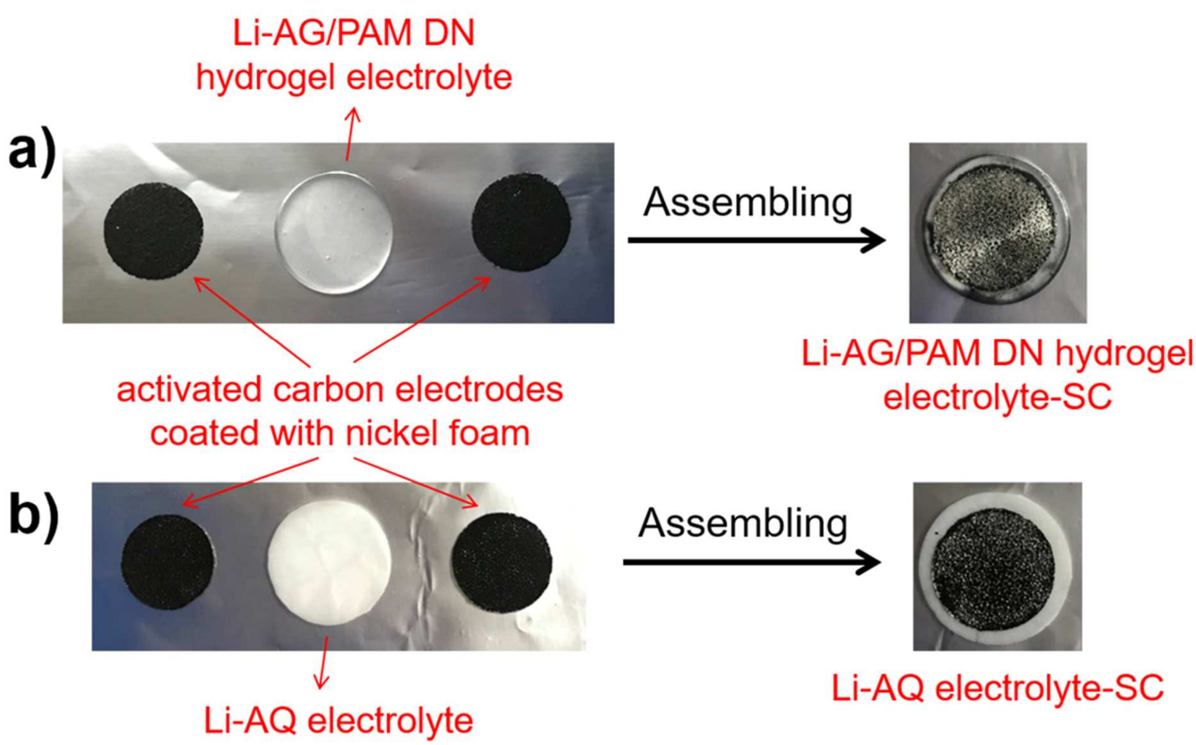

Figure S4. Assembly process diagram of (a) the Li-AG/PAM DN hydrogel electrolyte-SC and (b) the Li-AQ electrolyte-SC.

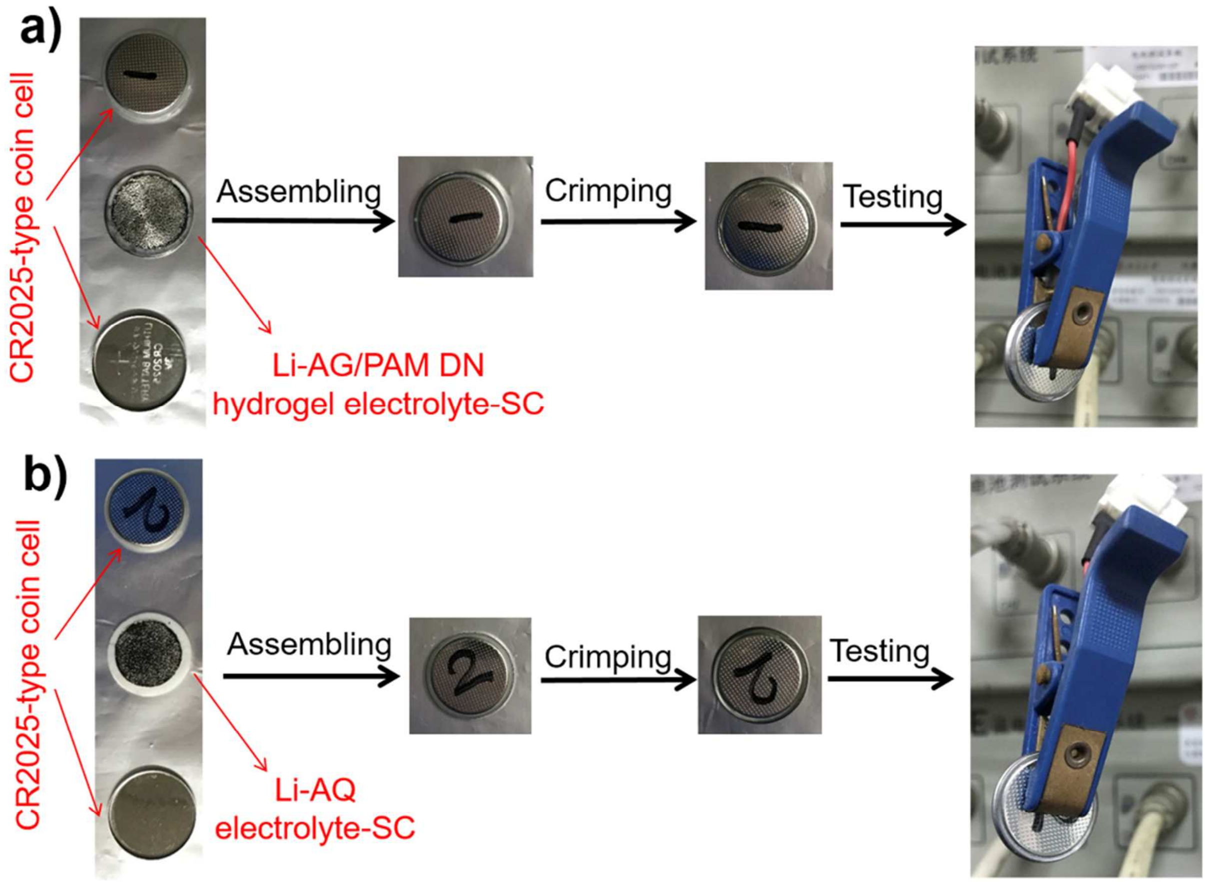

Figure S5. Assembly process diagram of the two-electrode testing configuration for (a) the Li-AG/PAM DN hydrogel electrolyte-SC and (b) the Li-AQ electrolyte-SC.

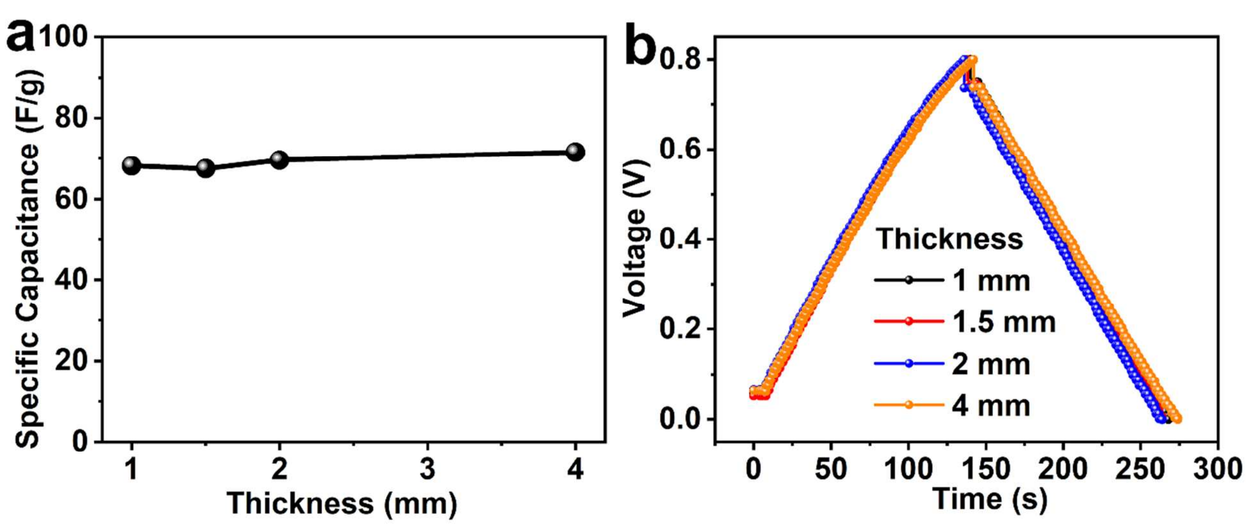

Figure S6. (a) Specific capacitance and (b) GCD curve of the Li-AG/PAM DN hydrogel electrolyte-SC applying various thickness of the Li-AG/PAM DN hydrogel electrolyte at a current density of 0.2 A g^{-1}.

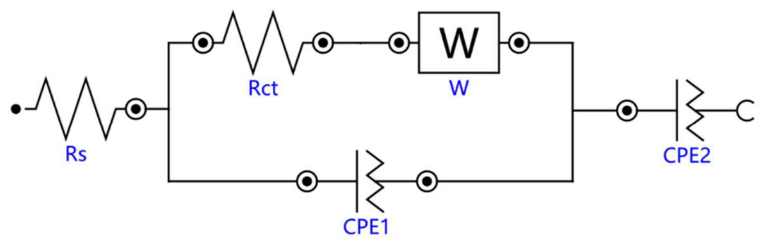

Figure S7. Equivalent circuit model and fitting parameters of the AG/PAM DN hydrogel electrolyte-SC and the Li-AQ electrolyte-SC. Both *R*s and *R*ct represent resistance. Both *CPE*1 and *CPE*2 represent constant phase element (CPE). *W* represents Warburg impedance.

Table S3. The values of *R*s, *R*ct, and *W* of the AG/PAM DN hydrogel electrolyte-SC and the Li-AQ electrolyte-SC fitted by Nova 2.1.2 software.

	***R*s (Ω)**	***R*ct (Ω)**	**Y_0 (Ω^{-1} $s^{0.5}$) of *W* [a]**
Li-AG/PAM DN hydrogel electrolyte-SC	3.33	0.58	0.17
Li-AQ electrolyte-SC	2.07	1.08	0.06

[a] The W (Ω) is defined as $W = \frac{1}{Y_0(jw)^{0.5}}$, where j, w (s^{-1}), Y_0 (Ω^{-1} $s^{0.5}$) are imaginary unit, angular frequency, and coefficient of the *W*, respectively.

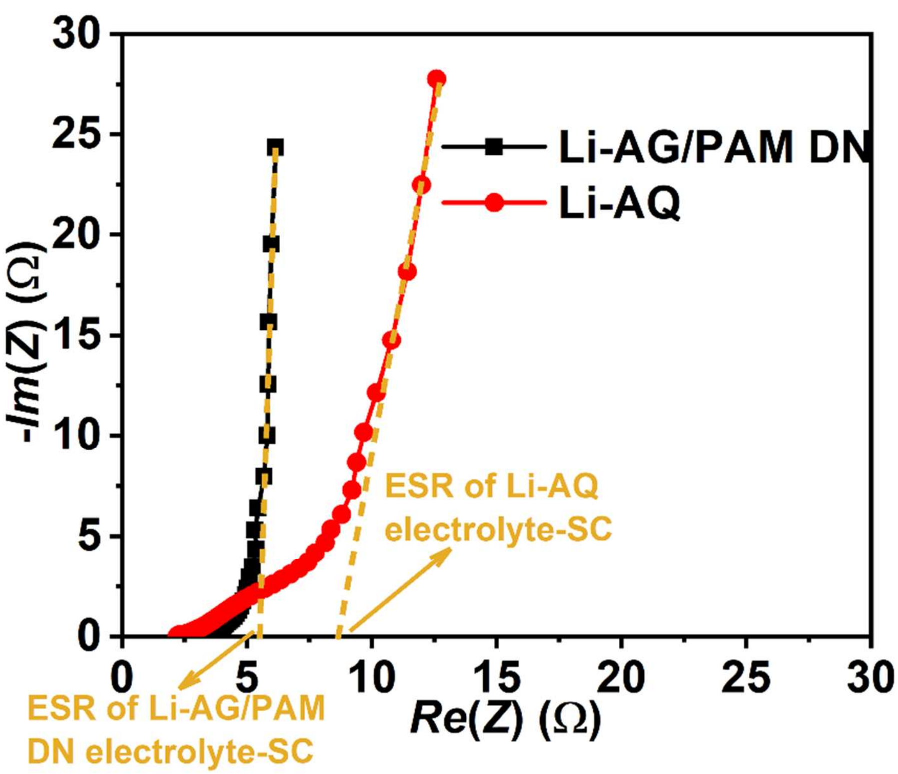

Figure S8. *ESR* of the Li-AG/PAM DN hydrogel electrolyte-SC and the Li-AQ electrolyte-SC obtained from the Nyquist plots in a frequency range from 10^6 to 10^{-2} Hz.

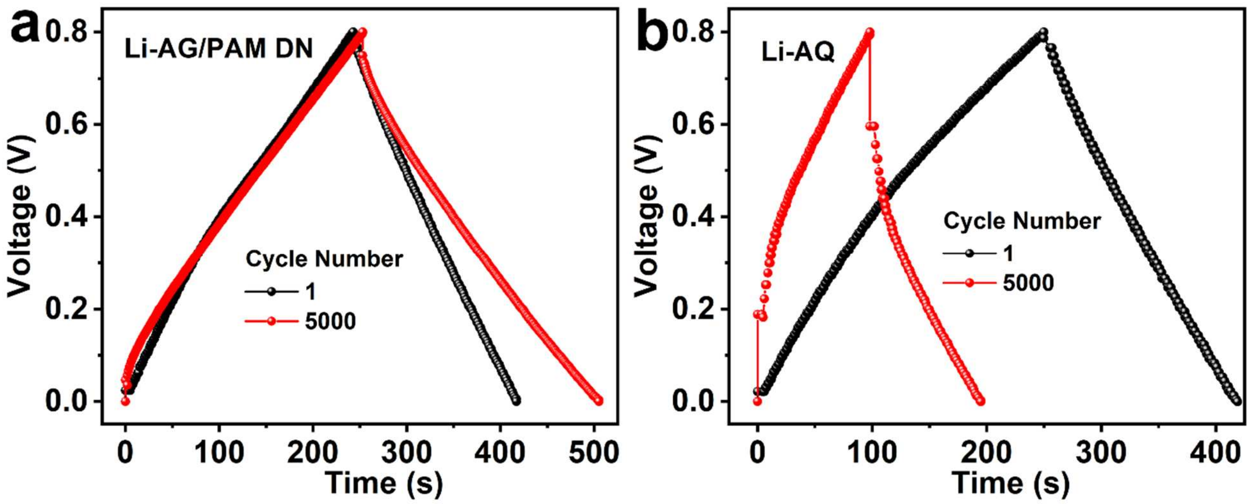

Figure S9. GCD curves of (a) the Li-AG/PAM DN hydrogel electrolyte-SC and (b) the Li-AQ electrolyte-SC before and after 5000 cycles at a current density of 0.2 A g^{-1}.

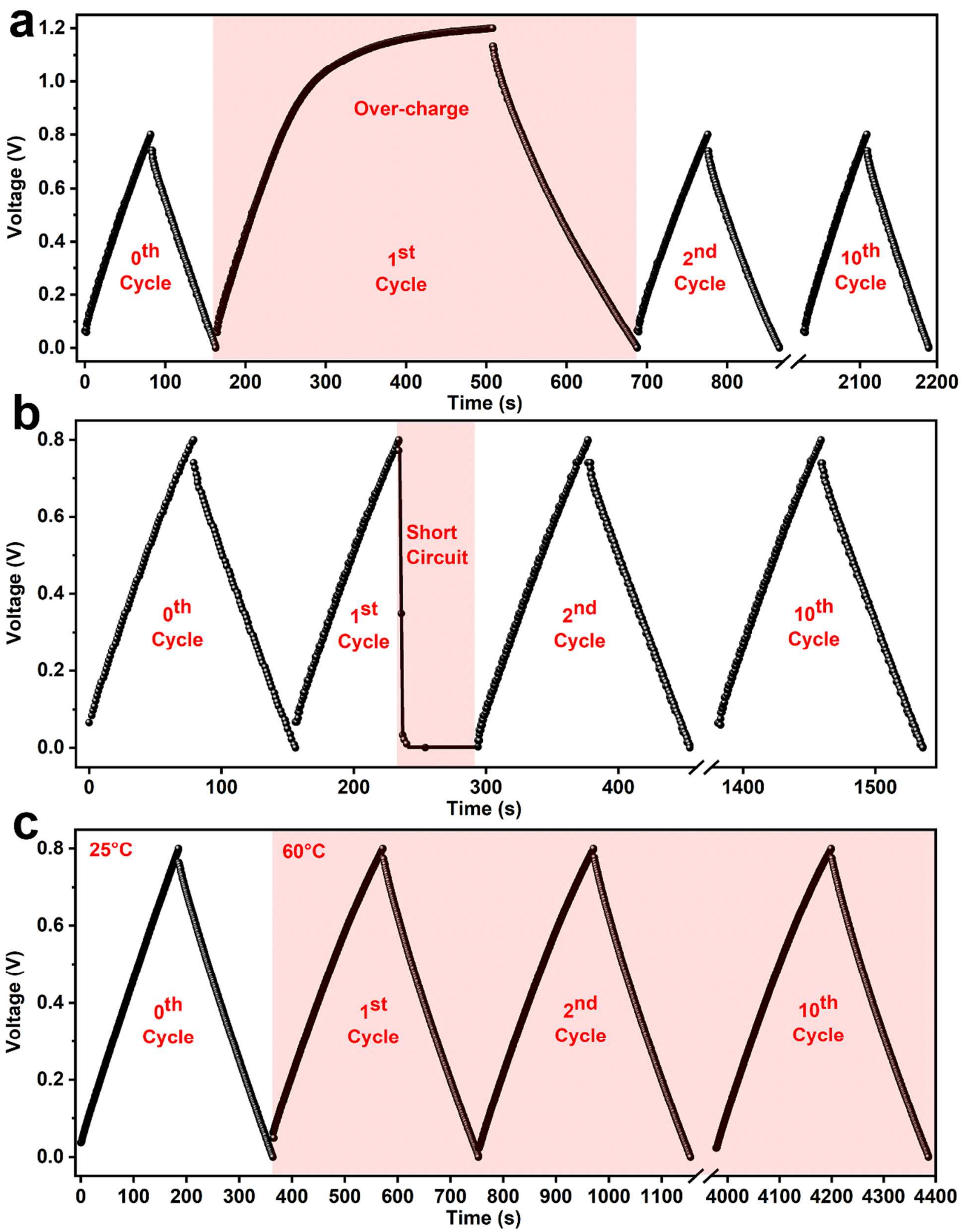

Figure S10. GCD curves of the Li-AG/PAM DN hydrogel electrolyte-SC on cycling after (a) over-charge test at a current density of 0.3 A g^{-1}, (b) short circuit test at a current density of 0.3 A g^{-1}, and (c) high temperature of 60 °C test at a current density of 0.2 A g^{-1}.

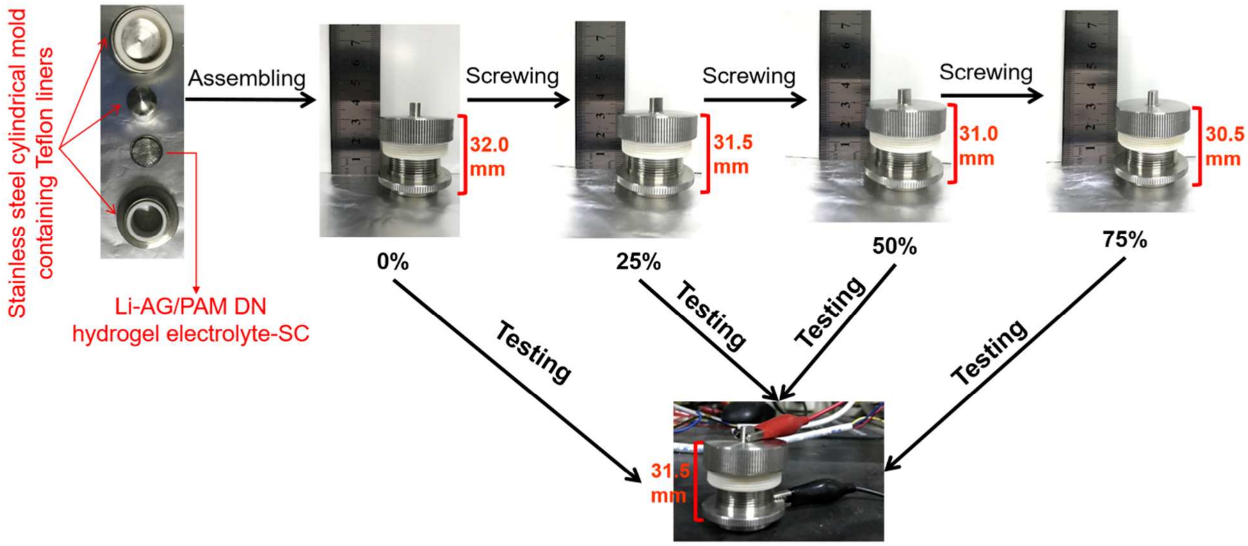

Figure S11. Assembly process diagram of the Li-AG/PAM DN hydrogel electrolyte-SC for compression tests. The bottom of the photo shows the SC during electrochemical tests under a compression strain of 25%.

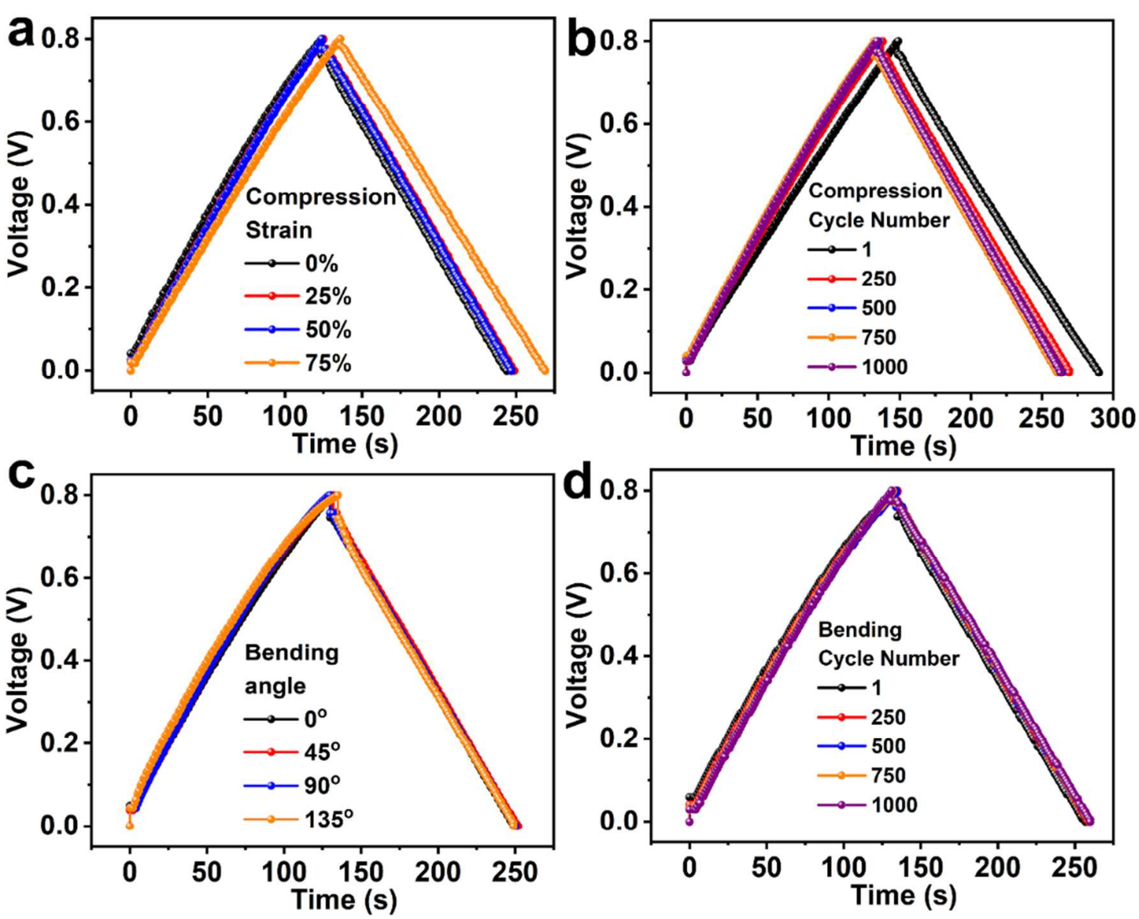

Figure S12. GCD curves of the Li-AG/PAM DN hydrogel electrolyte-SC at various (a) compression strain, (b) compression cycles, (c) bending angle, and (d) bending cycles.

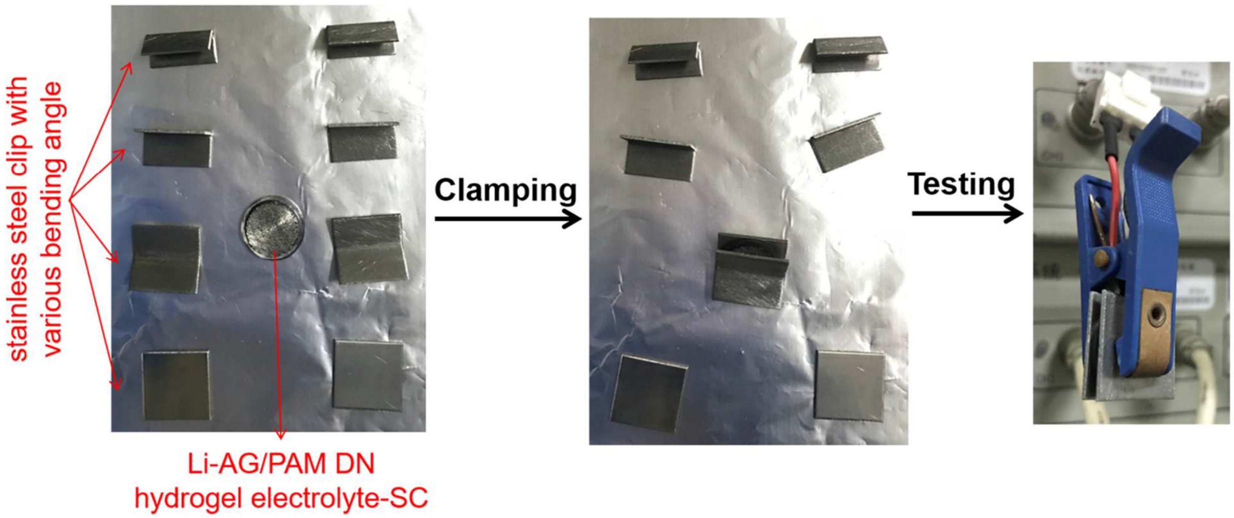

Figure S13. Assembly process diagram of the Li-AG/PAM DN hydrogel electrolyte-SC for bending tests. The most right photo shows how electrochemical tests were carried out in the SC bended to 45^{o}.