Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Fully integrated design of a stretchable kirigami-inspired micro-sized zinc-sulfur battery

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Mechanical and electrochemical characterization

An Instron 5543 single column table top tensile tester with 1 kN load cell was utilized to measure the mechanical properties of the stretchable kirigami-inspired Zn-S battery. All the electrochemical tests were conducted by a CH Instrument 660E Bipotentiostat. For non-linear discharge curves, specific capacitance was calculated by equation S1. The capacitance (C_{sp}), capacity (Q), and efficiency (η) were obtained by applying Eqs. (S1), (S2), and (S3).

$$C_{sp} = \frac{2\ i.A\ t_d}{m_{act} \cdot \Delta V^2} \qquad (S1)$$

$$Q = t_d(s)\frac{i}{3.6 m_{act}} \qquad (S2)$$

$$\text{Efficiency} = \frac{t_d}{t_c} \times 100 \qquad (S3)$$

The energy density (*E*) and power density (*P*) were respectively calculated by questions S4 and S5 [1].

$$E = \left(\frac{C_{sp}}{7.2}\right)(\Delta V)^2\ (mWh/g) \qquad (S4)$$

$$P = 3600.E/\Delta t\ (mW/g) \qquad (S5)$$

In the equations, i is the current, m_{act} is the cumulative masses of the anode and cathode electrodes, t_d is the discharge time, t_c is the charge time, ΔV is the scanning potential window and

A is the electrode surface area. Energy and power densities were calculated based on the total mass of the cathode material.

Tables

Table S1. Discharge and charge reactions in the stretchable Zn-S battery

Discharge Reactions	$\boldsymbol{S + Zn^{2+} + 2e^- \rightarrow ZnS}$ $\boldsymbol{S + Zn^{2+} + I_2 + 4e^- \rightarrow ZnS + 2I^-}$ $\boldsymbol{I^- + I_2 \rightarrow I_3^-}$
Charge Reactions	$ZnS \rightarrow S + Zn^{2+} + 2e^-$ $2ZnS + 4H_2O \rightarrow S + 2Zn^{2+} + SO_4^{2-} + 8H^+ + 10e^-$ $ZnS + I_3^- \rightarrow S + Zn^{2+} + 1.5\, I_2 + 3e^-$ $2ZnS + 4H_2O + I_3^- \rightarrow S + 2Zn^{2+} + SO_4^{2-} + +1.5\, I_2 + 8H^+ + 11e^-$

Table S2. Nitrogen adsorption (BET) results of porous CNFs

	BET surface area m^2/g	Average pore diameter (nm)	Total pore volume cm^3/g
KOH-treated AC (AC)	1241.634	2.38	0.7391
AC-S	2.948	13.40	0.009876
As-received AC	467.55	2.31	0.27

Table S3. Nitrogen adsorption (BET) results of porous CNFs

Energy storage type	Configuration	**Stretchability [%]**	Gravimetric, areal and volumetric **e**nergy density	Reference
Crumpled Graphene Supercapacitor	2D planar	140	–	[2]
Graphene Supercapacitor	3D-Textile	140	–	[3]
Carbon nanotubes – fabric Supercapacitor	3D -Textile	220	20 Wh/kg	[4]
Manganese dioxide-Carbon nanotubes Supercapacitor	3D -Textile	220	2.6×10^{-3} mWh/cm^2	[5]
Polypyrrole-fabric Supercapacitor	3D -Textile	200	11.1 Wh/kg	[6]
Polypyrrole /reduced graphene oxide fabric Supercapacitor	3D -Textile	150	2.53 Wh/kg	[7]
Manganese dioxide nanowires Supercapacitor	3D - Kirigami	600	21.07×10^{-3} mWh/cm^2	[8]

Carbon nanotubes Supercapacitor	3D - Textile	130	20 Wh/kg	[9]
Polypyrrole-Carbon nanotubes Supercapacitor	3D -Textile	180	61.3 µWh/cm^2	[10]
Carbon nanotubes Supercapacitor	3D -Textile	150	–	[11]
Graphite-cellular paper Supercapacitor	3D -Origami	130	–	[12]
Graphene-decorated Metallic textile Supercapacitor	3D -Textile	290	6.1×10^{-3} Wh/cm^3	[13]
Graphene Supercapacitor	3D -Kirigami	382.5	–	[14]
Polypyrrole-Manganese dioxide-Carbon nanotubes Supercapacitor	3D -Textile	121	31.1 Wh/kg	[15]
$Zn-MnO_2$	Fiber-like	300	447 mWh g^{-1} (98.5 % retention after 500 cycles)	[16]
Zn−Ag	2D planar	100	3.93 mWh cm^{-2} (140 % after 30 cycles at 100 % strain)	[17]
Li-ion Batteries	Origami	1300	0.53 mWh cm^{-2}	[18]
Li-ion Batteries	2D planar	50	8.14 mWh cm^{-2} (85 % after 60 cycles)	[19]
Li-ion Batteries	Fiber-like	600	201 mWh g^{-1} (90 %, after 50 cycles)	[20]
Li-ion Batteries	Fiber-like	100	345 mWh g^{-1} (84 % after 200 cycles)	[21]
Li-ion Batteries	Fiber-like	100	231 mWh g^{-1} (92.1 % after 100 cycles)	[22]
Li-air	Serpentine	100	2540 Wh kg^{-1}	[23]
Al-air	Fiber-like	30	935 mA h g^{-1}	[24]
Na-ion battery	3D -Textile	50	278 mWh g^{-1} (85 % after 60 cycles0	[25]
Zinc-sulfur battery	Kirigami	200	184.8 mWh/g (19.6 % increase after 10,000 stretch/release cycles under 200% under 200% tensile strain)	This study
Zinc-sulfur battery	Kirigami	200	184.8 mWh/g (6.3 % drop after 10,000 stretch/bending cycles of 200% strain and 180° bending angle)	This study

Table S4. AC impedance equivalent circuit fitting parameters

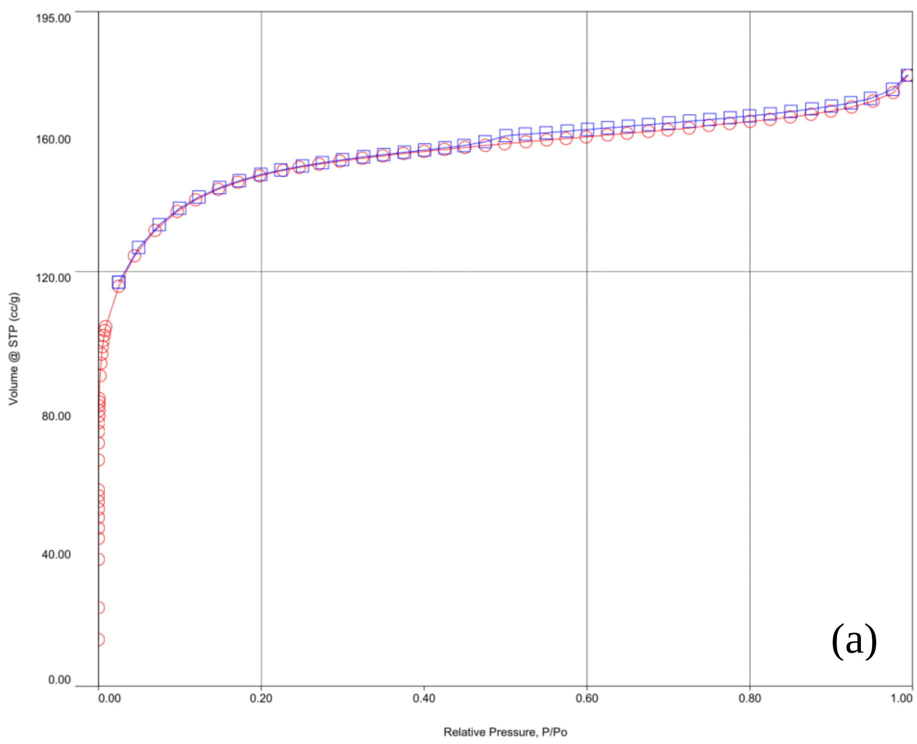
	R_s (Ω)	R_i	R_{ct}	W
0% strain and 1st cycle	7.28	6.27	12.81	0.03
50% strain and 1st cycle	7.12	5.80	11.55	0.03
100% strain and 1st cycle	7.86	5.39	9.7	0.03

150% strain and 1st cycle	6.71	5.15	9.80	0.03
200% strain and 1st cycle	6.09	2.64	6.02	0.03
0 % strain and 800th cycle	14.16	10.15	23.32	0.01
200 % strain and 800th cycle	13.88	9.52	18.63	0.01

Table S5. AC impedance equivalent circuit fitting parameters

	R_s (Ω)	R_i	R_{ct}	W
0% strain and 1st cycle at -25 °C	22.2	24.39	13.46	0.03
0% strain and 1st cycle at -25 °C	23.24	23.27	13.84	0.03

Figures



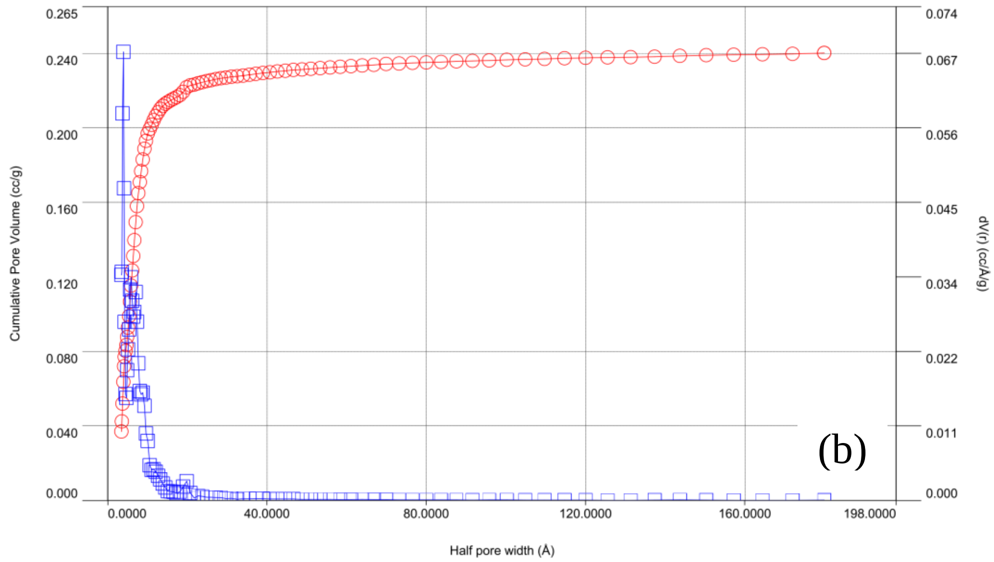


Fig. S1. (a) Adsorption-desorption isotherms and (b) distribution of pore size of as-received AC.

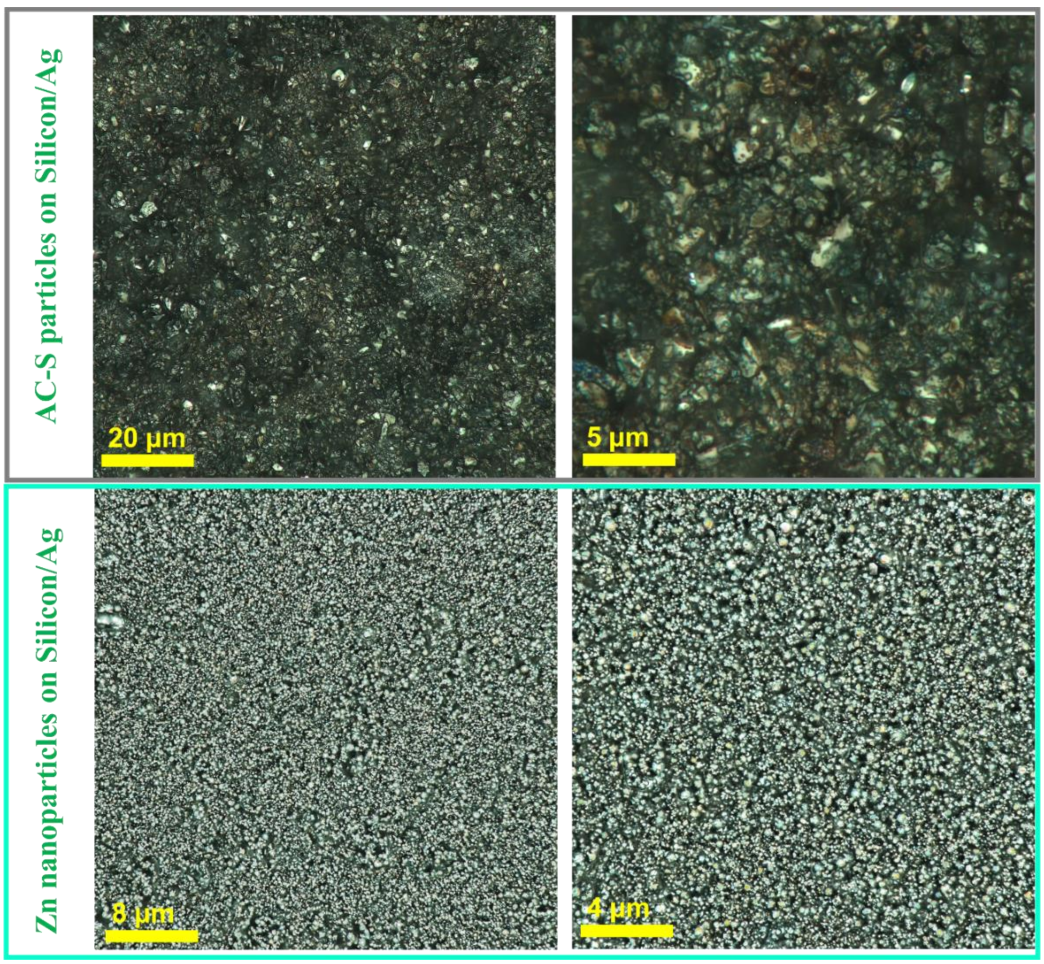


Fig. S2. The optical images of AC-S and zinc nanoparticles distributed on the current collector.

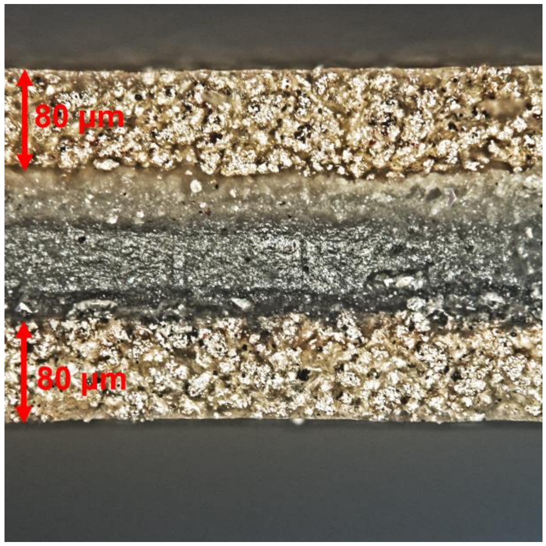
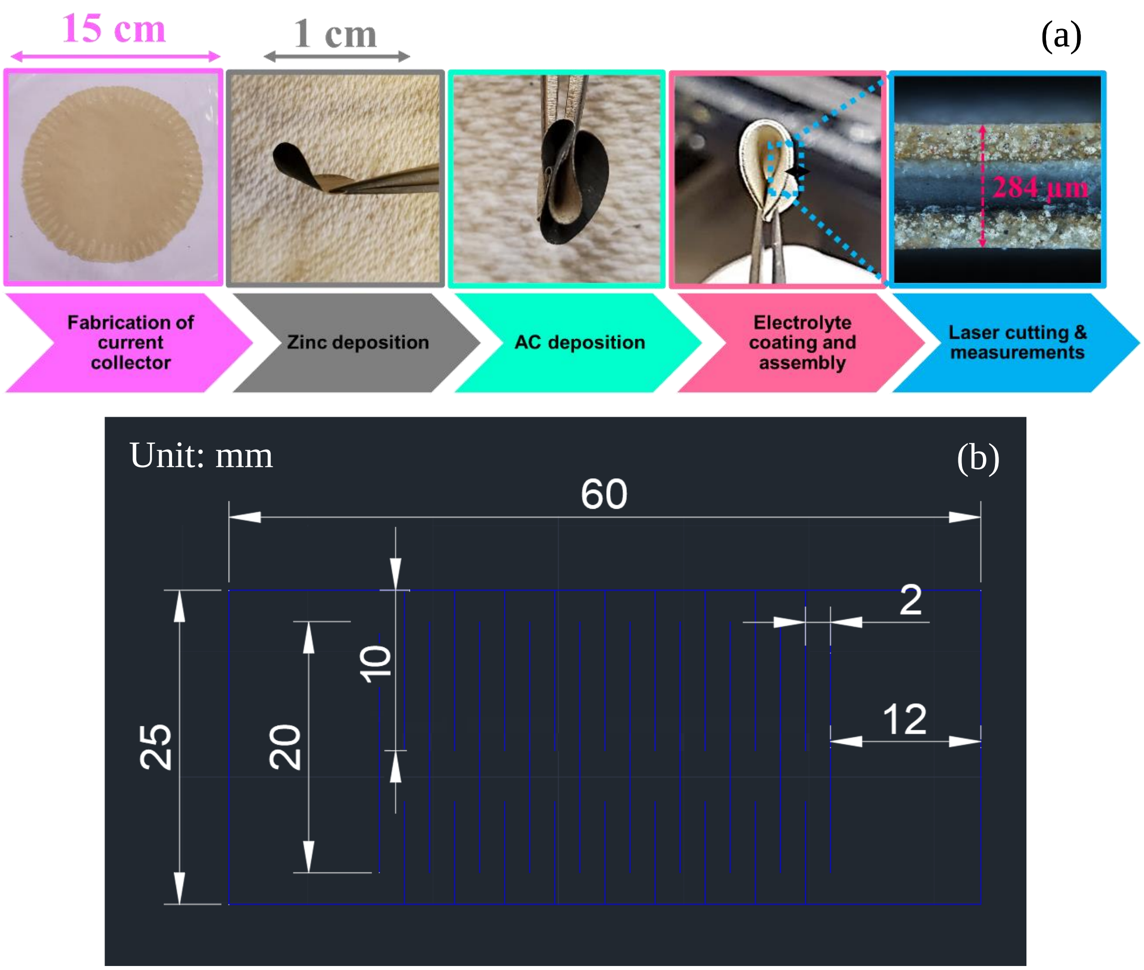


Fig. S3. The optical images of kirigami- inspired Zn-S cell. The thin film of cured silver-filled silicone adhesive possessed an average thickness of 80 ± 20 µm.



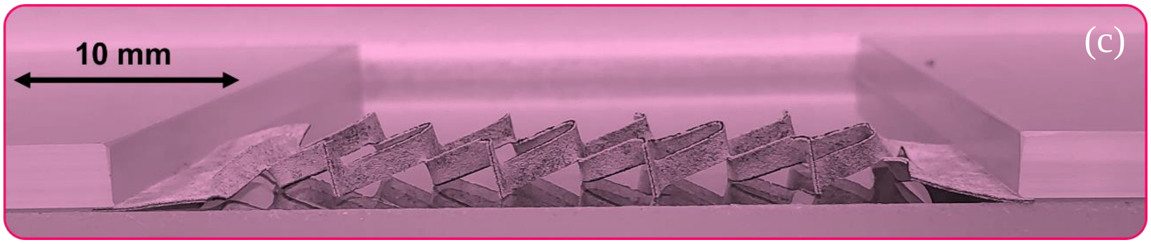


Fig. S4. (a) The photographs of the stretchable kirigami-inspired Zn-S battery in different stages of fabrication. (b) Dimensions and patterns for laser cutting. (c) 3D structure of the kirigami-inspired Zn-S battery.

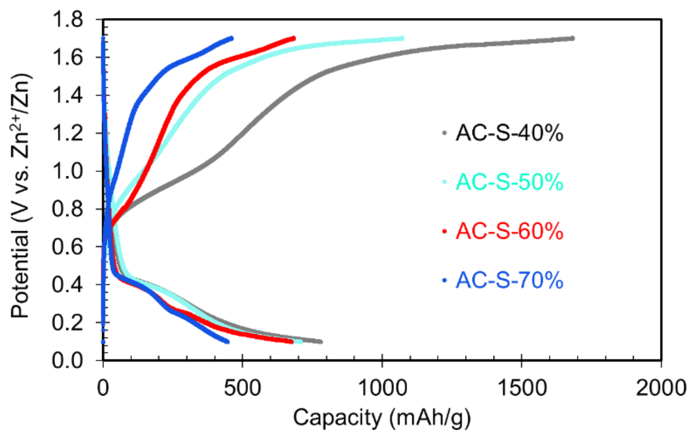


Fig. S5. Charge and discharge profiles of Zn-S battery for samples with different sulfur contents at 0.25 A/g.

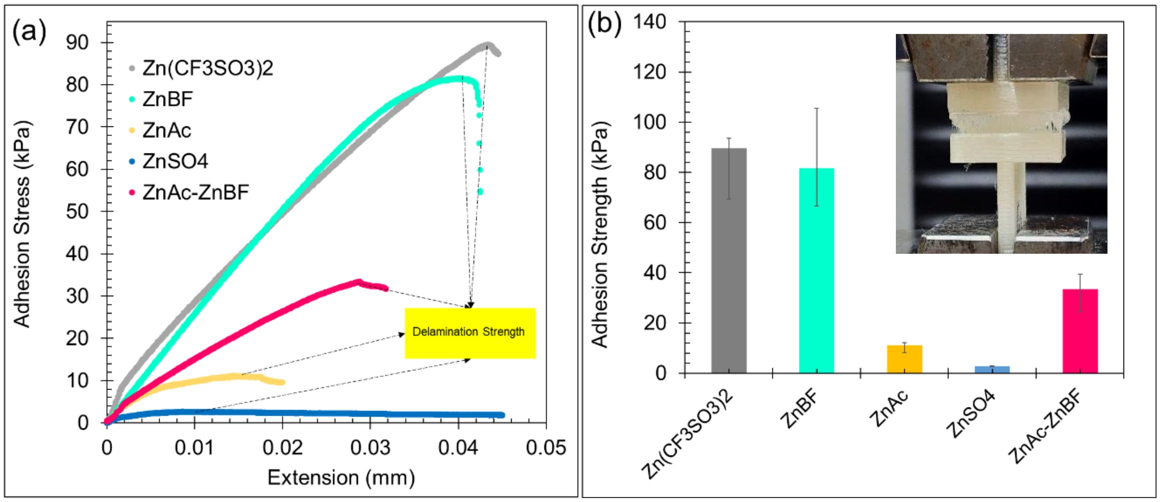


Fig. S6. (a) Typical stress–extension plots and (b) adhesion strength of different zinc salt-containing hydrogels.

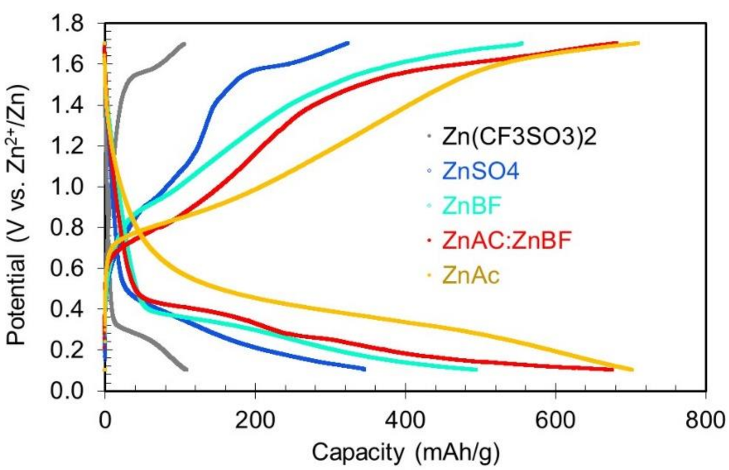


Fig. S7. Charge and discharge profiles of Zn-S battery in the presence of different electrolytes at 0.25 A/g.

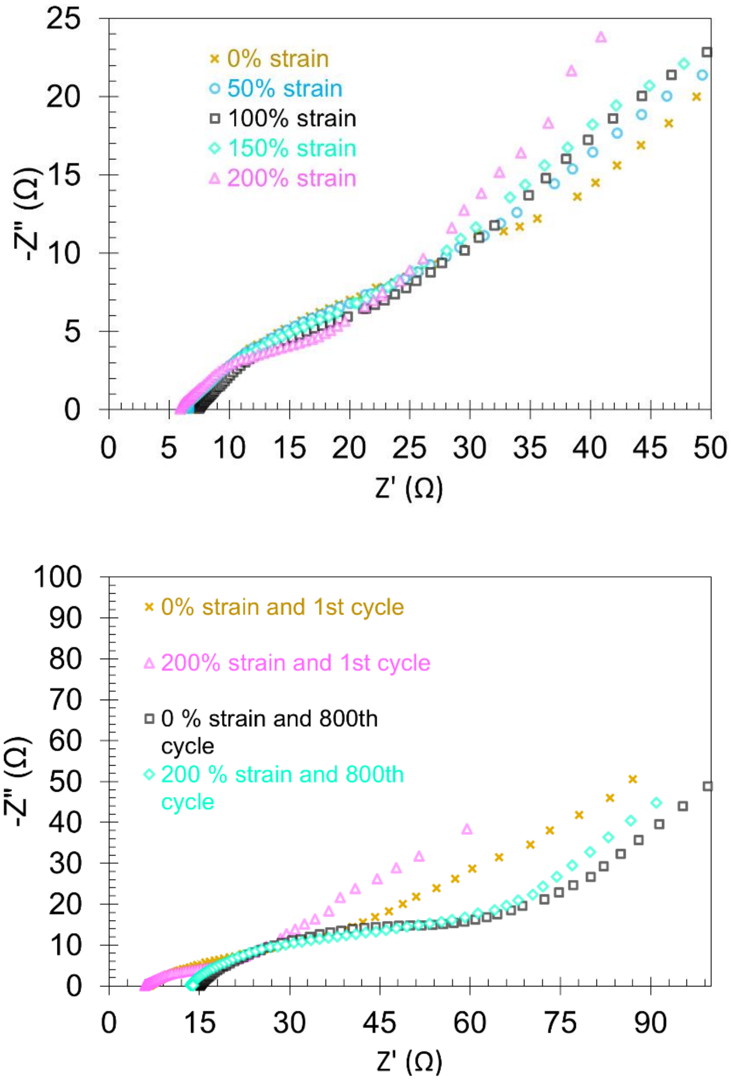


Fig. S8. Nyquist plots of kirigami-inspired Zn-S battery as a function of (a) strain and (b) cycle.

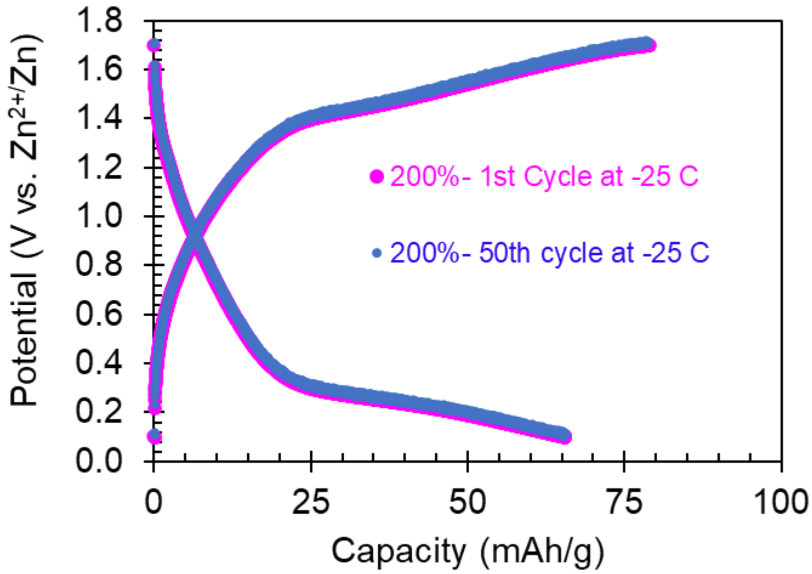


Fig. S9. Charge and discharge profiles of the kirigami-inspired Zn-S battery subjected to 1 and 50 stretching cycles under 200% tensile strain at -25 °C.

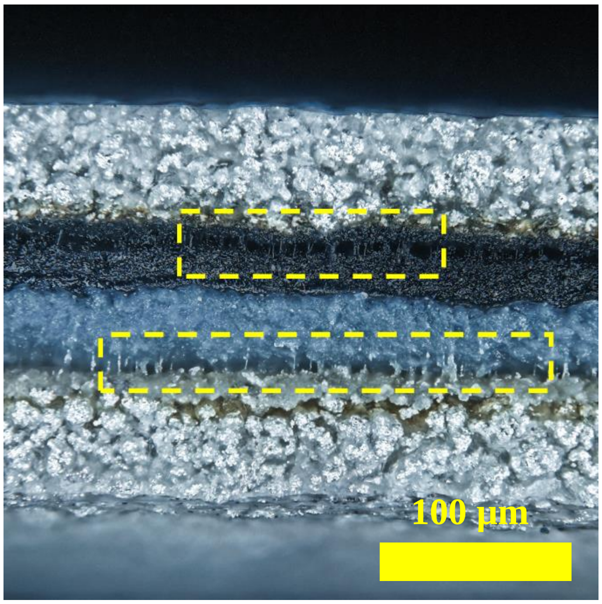


Fig. S10. The optical image of the partial delamination between electrodes and current collector at -25 °C. We observed a handful of delaminated sites only at -25 °C.

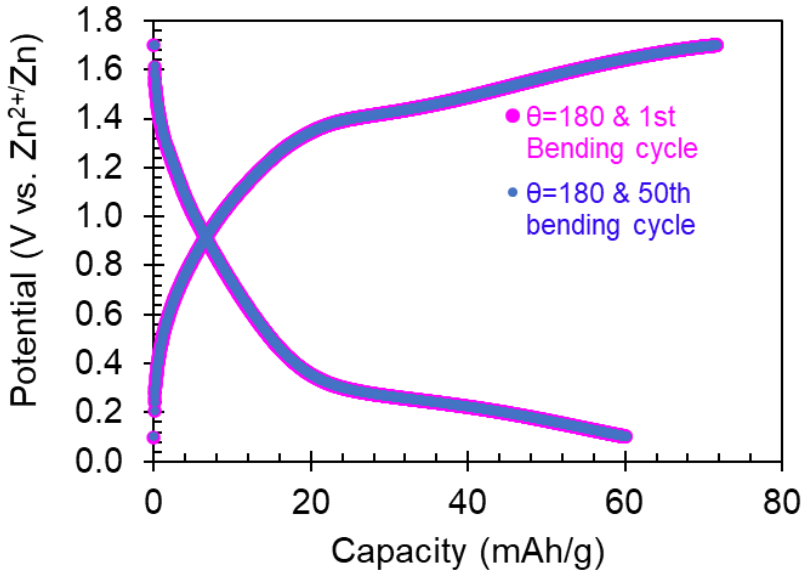


Fig. S11. Charge and discharge profiles of the kirigami-inspired Zn-S battery subjected to 1 and 50 bending cycles under 180° at -25 °C.

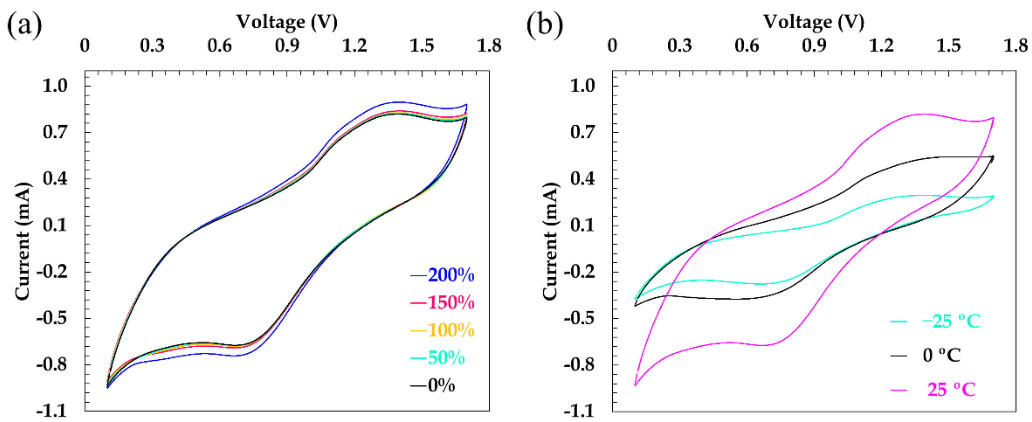


Fig. S12. CV charts of the kirigami-inspired Zn-S battery as a function of (a) tensile strain at 25 °C and 1 mV/s and (b) temperature at 1 mV/s.

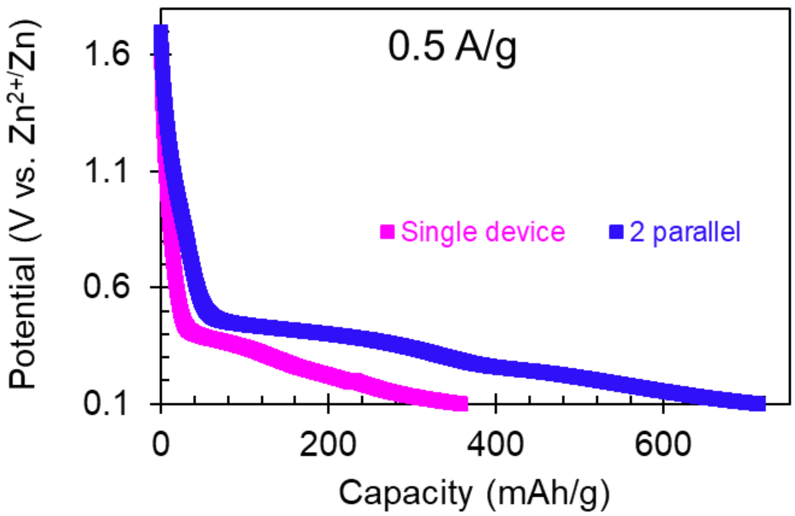


Fig. 13. Discharge curves of two Zn-S assembled in a parallel design vs. a single device at 0.5 A/g.

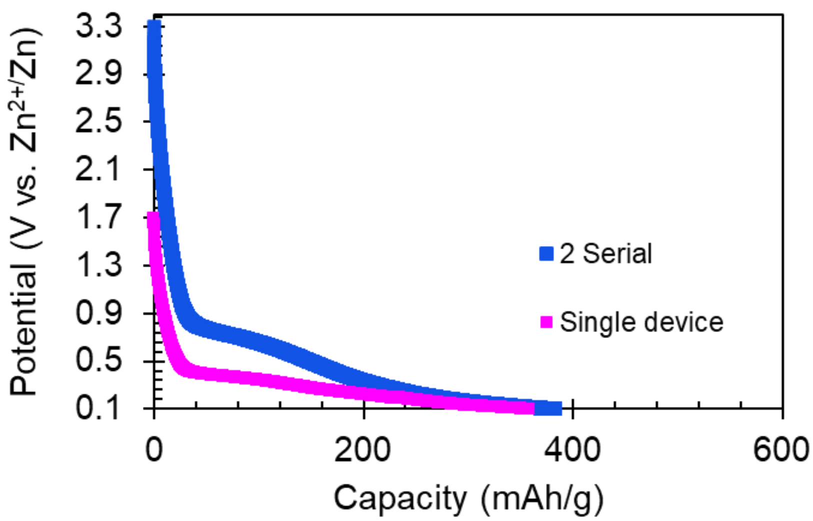


Fig. 14. Discharge curves of two Zn-S assembled in a serial design vs. a single device at 0.5 A/g.

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