Supporting information

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Experimental section

Preparation of epoxy-based polymer electrolyte

Zinc triflate with 0.2g was dissolved into polymer PEGDGE, and then the plasticizer PC was added. The complexes were mixed by the magnetic stir for at least 3 hours to obtain a homogeneous transparent solution. Curing agent TETA is added and stirred for an extra 5 minutes after the zinc salt was entirely dissolved in the polymer. The solid polymer film was fabricated by casting with a PTFE mold which was in the disc shape with 15 mm diameter and 1-2 mm thickness. As to ensure the polymer was fully polymerised, the entire polymer was cured at 80°C for 24 hours with the heat and cooling rate 3°C/min.

Electrode fabrication

For general electrochemical tests, cathode materials were fabricated by casting the commercial active material β -MnO₂ on a hydrophilic carbon paper. Super P carbon and polyvinylidene difluoride (PVDF) were mixed with the active material in the weight ratio of 2:1:7. The casted carbon paper was dried in a vacuum oven for 12 h at 70°C. The average mass loading for the dried cathode is 1.5~2.5 mg cm⁻². Regarding the anode, zinc metal foil was used after fully polishing to remove the surface ZnO.

In terms of the device, both cathode and anode materials were casted on the woven carbon cloth. The mixture of the electrode materials was in the same ratio as mentioned above, whilst zinc powder is applied instead of the zinc metal. The casted carbon paper was dried in a vacuum oven for 12 h at 70°C.

Material characterisation

X-ray diffraction (XRD) patterns were examined by a STOE SEIFERT diffractometer with detected angular range of $2^{\circ} < 2\theta < 45^{\circ}$ under the radiation source of metal Mo. X-ray photoelectron spectra (XPS) were obtained from Thermo scientific K-alpha photoelectron spectrometer. Optical microscopy used to exploit the morphology were tested by ZEISS Axio Lab.A1. Fourier transform infrared spectroscopy was measured by Shimadzu IRTracer-100 from the wavenumber from 400 to 4000 cm⁻¹. Thermogravimetric analyser (TGA) (PerkinElmer TGA 4000 System) was used to determine the crystal water of the cathode material and the decomposition temperature of the polymer electrolytes. The mass of the active materials was weighed accurately by an analytical balance (Ohaus; δ = 0.01 mg). Raman was obtained on a Raman Spectroscopy (Renishaw Raman microscope spectrometer with the laser wavelength of 514.5 nm).

Electrochemical characterisation

Battery galvanic charge-discharge tests were measured by NEWARE battery testing systems. Cyclic Voltammetry (CV) and Electrochemical Impedance Spectroscopy (EIS) were investigated by the VMP3 Biologic electrochemical workstation and Gamry 1010E, respectively. EIS was test under the frequency 100 kHz to 0.01 Hz.

Solid/hydrogel	Ionic	Cathode	Anode	Capacity	Energy	Cycling	characteristics	Ref.
electrolyte	conductivities			(mAh g ⁻¹)	density	stability		
	(S cm ⁻¹)							
Fumed silica	8.1×10 ⁻³	Zn ₂ (OH)VO ₄	Zn	204	115 Wh kg ⁻¹	5.1 kW	Avoid dendrite	1
				(0.5C)		kg^{-1}		
PVA	1.26 ×10 ⁻²	MnO ₂ @PEDOT	Zn	366.6	$504.9 \text{ Wh } \text{kg}^{-1}$	8.6 kW	Flexible	2
				(0.74 A g ⁻¹)		kg^{-1}		
PAM	0.12	Zn/Co ₃ O ₄	Zn	205	360.8 Wh kg ⁻¹	-	Flexible	3
				(0.5 A g ⁻¹)			wearable	
Gelatin-g-PAM	1.76 ×10 ⁻²	α-MnO2@CNT	Zn@CNT	306	6.18	148.2	Flexible	4
				(1C)	mW h cm ⁻²	mW cm ⁻²	Stretchable	
CMC	-	ZnHCF	Zn	100.2	195.39	-	Fibre	5
				(0.1 A cm ⁻	mW h cm ⁻²		flexible	
				1)				
Xanthan gum	1.46 ×10 ⁻²	MnO_2	Zn	260	$364 \text{ Wh } \text{kg}^{-1}$	2.5 kW	flexible	6
				(1C)		kg^{-1}		
PVDF	3.94×10-3	γ -MnO ₂	Zn	105	-	-	flexible	7
				(10µA cm ⁻				
				²)				
PEO	1.1 ×10 ⁻⁵	-	-	-	-	-	-	8
PEO & Al ₂ O ₃	2×10-5	-	-	-	-	-	-	8

Table S1 Summary of polymer electrolytes for zinc ion batteries

Material characterisations



Figure S1. DSC check for the polymer electrolyte



Figure S2. Schematic diagram of polymerisation mechanism



Figure S3. (a) SEM image of the polymer electrolyte, (b) EDX mapping

			Spectrum 8
	Element	Weight %	Atomic %
	0	71.56	80.60
	F	15.66	14.86
	S	3.56	2,00
	Zn	9.22	2.54
9	• •		
<u>í</u> í í í í í í í í í í í í í í í í í í	^s ç¦Q		
0 1 2 Full Scale 1970 cts Cursor: -0.0	3)34 (905 cts)	4	5 6 keV

Figure S4. EDX elemental ratio



Figure S5. CV curve for Zn/EBSPE/Zn



Figure S6. Zinc deposition on the electrodes



Figure S7. Vacuum assisted resin transfer moulding

The polymer electrolyte in the liquid state formed a film on each component by laminating in a certain order. Then the entire structure was vacuum sealed and placed in an autoclave for the polymerisation. PEGDGE is a biodegradable epoxy resin, which has good surface crosslinking ability it strengthens the device from the mold from peeling. Therefore, peel ply, breather and releasing films which are normally used in carbon fibre composite manufacturing process were placed on the top of the laminates. Peel plies are used to easily peel off the device after the polymer polymerisation and leave a smooth surface; breathers are used to absorb extra liquid resin during curing and enable air bubble escape when vacuum is applied and the formed films exhibit no release characteristics and are typically placed directly with the laminate to avoiding the bonding to the mould.



Figure S8. XPS spectrums for the EBSPE

No.	PEGDGE (g)	PC (g)	TETA (g)	ZnOf (g)	R_b (Ω)	σ (S cm ⁻¹)
1	2.00	0.00	0.15	0.20	266	2.13×10 ⁻⁴
2	2.00	1.00	0.15	0.20	193	2.93×10 ⁻⁴
3	2.00	2.00	0.15	0.20	150	3.77×10 ⁻⁴
4	2.00	3.00	0.15	0.2	205	2.88×10-4

Table S2 Summary of ionic conductivities for different amount of PC



Figure S9 (a) Ionic conductivity with concentration of PC; (b) Ionic conductivity with concentration of ZnOTf

As shown in Figure S9a, there is a decrease of ionic conductivity when the content of PC is over an optimum value 45% (wt%) which is consistent with the outcome reported by Nithya. Regarding the concentration of ZnOTf, concentration of ZnOTf decreases as the increasing of PC content, however the ionic conductivities increase as the reduction of ZnOTf concentration. Hence, there is a slight influence of the ZnOTf concentration to the ionic conductivity when PC is 45% (wt%) in the polymer electrolyte. While once the PC concentration is greater than 50% in the polymer, the low content ratio of ZnOTf in the complexes results the decrease in the ionic conductivity.



Figure S10. Raman spectra for solid polymer electrolyte (ABSPE) in comparison of pure PEGDGE resin



Figure S11. TGA for alginate hydrogel electrolyte



Figure S12. Adhesive ability of the polymer (polymer is sticky to the rod)

Electrochemical performances



Figure S13. (a) Charge/discharge rate test under current density of 0.1 A g^{-1} , 0.2 A g^{-1} , 0.5 A g^{-1} , 1 A g^{-1} , 2 A g^{-1} , 5 A g^{-1} , (b) Charge/discharge profiles under different current densities, (c) cycling performance under 0.5 A g^{-1} , (d) electrochemical performance under deformations.



Figure S14. (a) SEM image of pristine Zn anode before cycling, (b) SEM image of Zn anode after the cycling test.

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