## Inorganic Salt Reinforce Zn<sup>2+</sup>-conducting Solid-state

## **Electrolyte for Ultra-stable Zn Metal Battery**

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Sample	Gelatin content [w/w%]	Salt content [w/w%]	Water content [w/w%]
GSE-0.0	9.09	12.75	78.16
GSE-1.0	5.96	14.85	79.19
GSE-1.5	9.85	20.98	69.17
GSE-2.0	15.71	30.25	54.04
GSE-2.5	18.90	35.75	45.35
GSE-3.0	20.30	34.71	44.98
GSE-3.5	20.83	34.98	44.19

**Table S1**. Component (gelatin, salt, and water) content of GSE-x (x=0, 1.0, 1.5, 2.0, 2.5, 3.0, and 3.5)

Data Point	Electrolyte	Zinc anode	Current density (mA cm <sup>-2</sup> )	Cumulative capacity plated (mAh cm <sup>-2</sup> )	Ref.
1	PVA	Zn foil	0.1	80	12
2	Alginate/PAAM	Electroplated z	zinc 0.2	16	15
3	Silica	Zn foil	0.2	300	17
4	PEO	Zinc powder	0.1	17	19
5	PEO-PPO-PEO	Zinc powder	0.1	22	19
6	Gelatin	Zn foil	0.2	160	20
7	Gelatin	Zn/SS	2	600	28
8	Liquid	C@Zn	2.5	600	29
9	Liquid	TiO <sub>2</sub> @Zn	1	150	30
10	Liquid	PA@Zn	0.5	4000	31
This work	GSE-2.5	Zn foil	5	2000	This work

**Table S2.** Cycling performance reported in Fig. 2e.



Figure S1. The LSV curves of Zn/GSE-x/SS (SS means stainless steel; x=0.0, 1.5, 2.0, 2.5, 3.0, 3.5).



Figure S2. Relaxed (left) and elongated (right) state of the GSE-3.5



**Figure S3**. Comparison of properties (tensile strength, melting point, and ionic conductivity) of GSE-x (x=0, 1.0, 1.5, 2.0, 2.5, 3.0, and 3.5).



**Figure S4**. Galvanostatic cycling of Zn/GSE-2.5-0.235 mm/Zn, Zn/GSE-2.5-0.513 mm/Zn solid-state symmetric cells at current density of 5 mA cm<sup>-2</sup>.



Figure S5. XRD patterns of Zn electrodes in the insets of Figure 2e and 2f.



**Figure S6**. (a) XRD pattern; (b) SEM image of  $\alpha$ -MnO<sub>2</sub>/CNT composite. Scale bar: 200 nm



Figure S7. SEM image of acid-treated CNT. Scale bar: 200 nm



**Figure S8**. (a) TEM and (b) HRTEM images of  $\alpha$ -MnO<sub>2</sub>/CNT composite.



**Figure S9**. HAADF-STEM image and corresponding EDX mapping image of MnO<sub>2</sub>/CNT composite. Scale bar: 200 nm



Figure S10. TG curves of (a)  $MnO_2$ ,  $MnO_2/CNT$  and (b) acid-treated CNT



Figure 11. Cycle performance of (a) Zn/GSE-0/MnO<sub>2</sub>, (b) Zn/GSE-3.5/MnO<sub>2</sub> at 5C (1C=308 mA  $g^{-1}$ )



**Figure S12**. Rate performance of the Zn/GSE-0/MnO<sub>2</sub>, Zn/GSE-2.5/MnO<sub>2</sub>, and Zn/GSE-3.5/MnO<sub>2</sub> batteries at various rates (1C=308 mA g<sup>-1</sup>)



**Figure S13**. Charge-discharge voltage profiles of Zn/GSE-2.5-0.235 mm/MnO<sub>2</sub> and Zn/GSE-2.5-0.513 mm/MnO<sub>2</sub> batteries at the second cycle.



Figure S14. SEM at high magnification of the flask-like products in the fully discharged sample at state 3 in Figure 4a and the corresponding EDS mapping results. Scale bar, 5  $\mu$ m



**Figure S15**. HAADF-STEM image and corresponding EDX mapping of the particleshape products in the fully discharged sample at state 3 in Fig. 4a. Scale bar, 100 nm



**Figure S16**. Electrochemical performance of the solid-state Zn/GSE-2.5/MnO<sub>2</sub> ZMBs under (a) being bent; (b) being hammered.