# **Supporting Information**

## Stretchable Solid-State Zinc Ion Battery Based on Cellulose Nanofiber-

## Polyacrylamide Hydrogel Electrolyte and Mg0.23V2O5·1.0H2O

#### Cathode

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**Figure S1.** Photos of CNF and CNF-PAM solid-state electrolytes. (a) CNF dispersed in the solution of 1 M  $Zn(CF_3SO_3)_2$ ; (b), (c) CNF-PAM prepared by the radical polymerization.



Figure S2. Cross-section SEM image of CNF-PAM film.



Figure S3. SEM (a, b) and TEM (c, d) images of CNFs



Figure S4. SEM images of (a, b) freeze-dried PAM film and (c, d) freeze-dried CNF-PAM films



Figure S5. Stress-strain curves of CFC/PAM hydrogel film



**Figure S6.** XRD pattern of  $V_2O_5 \cdot nH_2O$ .



Figure S7. Crystal structure of  $Mg_{0.23}V_2O_5$ ·1.0H<sub>2</sub>O microspheres viewed from c axis.



**Figure S8.** Crystal structure of  $Mg_{0.23}V_2O_5 \cdot 1.0H_2O$  microspheres in (a) xy, and (b) vanadium environments. The V atoms are shown by red ball, and O atoms are shown by yellow balls.



Figure S9. FTIR spectrum of  $Mg_{0.23}V_2O_5$ ·1.0H<sub>2</sub>O microspheres.



Figure S10. SEM images of  $V_2O_5$ ·nH<sub>2</sub>O nanowires.



Figure S11. XPS spectrum of Mg<sub>0.23</sub>V<sub>2</sub>O<sub>5</sub>·1.0H<sub>2</sub>O microspheres. (a) XPS survey, high-resolution XPS spectrum of (b) Mg 2s, (c) V2p, (d) O 1s.



Figure S12. TGA curve of the prepared  $Mg_{0.23}V_2O_5$ ·1.0H<sub>2</sub>O microspheres.



Figure S13. CV curves of MVO/Zn solid-state batteries during the first three cycles at 0.1 mV/s.



Figure S14. Cycling performance of  $V_2O_5 \cdot nH_2O$  at current density of 500 mA/g.



Figure S15. Cycling performance at current density of10 A/g.



Figure S16. Optical photos of (a, b) CNF and (c, d) CNF-PAM films



Figure S17. Optical photos of MVO/Zn solid-state ZIB with burning.



Figure S18. Nyquist plots of solid-state MVO/Zn batteries using CNF-PAM film and CNF film as electrolyte.



Figure S19. The CV curve at 0.1 mV/s. (The shaded area shows the capacitive contributions)



**Figure S20.** GITT profiles of V<sub>2</sub>O<sub>5</sub>·1.0H<sub>2</sub>O based solid-state ZIBs, (e) diffusion versus different Zn<sup>2+</sup> insertion/extraction states,



Figure S21. XRD of MVO at initial and fully discharge states.



**Figure S22.** XPS survey of Mg<sub>0.23</sub>V<sub>2</sub>O<sub>5</sub>·1.0H<sub>2</sub>O microspheres at different charging/discharging states.



**Figure S23.** EDS mapping of  $Mg_{0.23}V_2O_5$ ·1.0H<sub>2</sub>O microspheres after fully discharge.



**Figure S24.** Schematic illustrations of Zn<sup>2+</sup> storage during charge discharge process.

Cathode	Operating voltage	Current rate	Capacity[m Ah/g]	Cycle performance	Ref.
Our MVO/Zn	0.2-1.6 V	5 A/g	216 mAh/g	98.6% retention	
solid-state				after 2000 cycles	
batteries		20 A/g	82.3 mAh/g	70 % retention after 12000 cycles	
VS <sub>2</sub>	0.4-1.0 V	0.2 A/g	120 mAh/g	250 cycles	[22] <sup>1</sup>
$NH_4V_3O_8$	0.2-1.4 V	0.5 A/g	133 mAh/g	200 cycles	[23] <sup>2</sup>
MnO <sub>2</sub> nanocrystallites	0.8-1.9 V	0.131 A/g	135.2 mAh/g	1000 cycles	[24] <sup>3</sup>
zinc orthovanadate	0.4-1.5 V	4 A/g	125 mAh/g	2000 cycles	[25]4
MnO <sub>2</sub>	1.0-2.0 V	1.3 A/g	127 mAh/g	1000 cycles	[26] <sup>5</sup>
α-MnO <sub>2</sub>	0.9-2.0 V	2.772 A/g	100 mAh/g	1000 cycles	[27] <sup>6</sup>
MnO <sub>2</sub> /PEDOT	0.9-1.8 V	1.11 A/g	280 mAh/g	300 cycles	[28] <sup>7</sup>
$NaV_3O_8 \cdot 1.5H_2$	0.3-1.25 V	0.5 A/g	~125 mAh/g	120 cycles	[29]8
MnO <sub>2</sub>	0.9-1.9 V	1.232 A/g	190 mAh/g	1000 cycles	[30]9
VS <sub>2</sub>	0.4-1.0 V	0.5 A/g	128 mAh/g	200 cycles, 91% of initial capacity	[31] <sup>10</sup>
MoS <sub>2</sub>	0.3-1.5 V	1 A/g	~150 mAh/g	500 cycles, 97.7% of initial capacity	[32]11
MnO <sub>2</sub>	0.9-1.9 V	0.924 A/g	$\sim \! 150 \text{ mAh/g}$	100 cycles	[33] <sup>12</sup>
MnO <sub>2</sub>	0.9-1.8 V	2.4 A/g	146 mAh/g	600 cycles, 87% of initial capacity	[34] <sup>13</sup>
FeHCF	0-2.3 V	3 A/g	57 mAh/g	100 cycles	[35] <sup>14</sup>
$Co_{0.247}V_2O_5$ .	0.6-2.2 V	4 A/g	200 mAh/g	5500 cycles, 94.5% of initial cycle	[36] <sup>15</sup>

**Table S1.** Summary of electrochemical performance of cathode materials for solid-stateZIBs.

NiCo	1.2-2.0 V	96 C	70 mAh/g	16000 cycles, 65% of	[37] <sup>16</sup>
				initial cycle	
CoFe(CN) <sub>6</sub>	0.75-2.0 V	2 A/g	110 mAh/g	2000 cycles	[38]17
$\delta$ -MnO <sub>2</sub>	0.9-1.9 V	10 C	$\sim 100 \text{ mAh/g}$	55 cycles	[ <b>39</b> ] <sup>18</sup>

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