Supporting Information for

# Simultaneously Regulating Uniform $Zn^{2+}$ Flux and Electron Conduction by MOF/rGO Interlayers for High-Performance Zn Anodes

Ziqi Wang[1, #], Liubing Dong[1, #] Weiyuan Huang[2], Hao Jia[3], Qinghe Zhao[2], Yidi Wang[3], Bin Fei[3, *], Feng Pan[2, *]

[1]Department of Materials Science and Engineering, College of Chemistry and Materials Science, Jinan University, Guangzhou 510632, P. R. China

[2]School of Advanced Materials, Peking University Shenzhen Graduate School, Shenzhen 518055, P. R. China

[3]Nano Center, Institute of Textiles and Clothing, Hong Kong Polytechnic University, Hunghom, Kowloon, Hong Kong 999077, P. R. China

[#]Ziqi Wang and Liubing Dong contributed equally to this work

*Corresponding authors. E-mail: panfeng@pkusz.edu.cn (Feng Pan), bin.fei@polyu.edu.hk (Bin Fei)

## Supplementary Figures

**Fig. S1** Photograph of the as prepared Janus separators

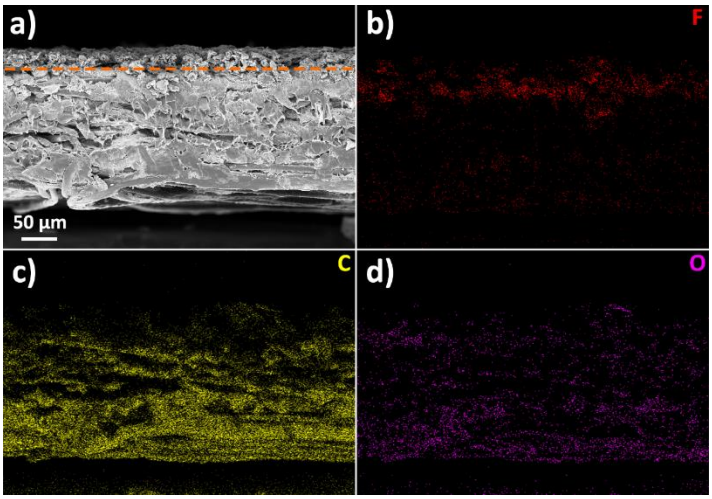


**Fig. S2 a** SEM image and **b-d** corresponding EDS elemental mappings for the cross-section view of the Janus separator

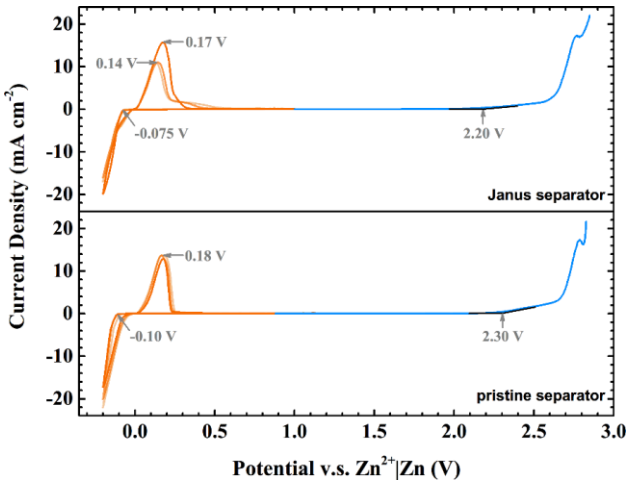


**Fig. S3** LSV (blue line) and CV (deep to light orange lines represent the 1st to 3rd cycles) profiles for the electrochemical window test with pristine and Janus separator in 2 M $ZnSO_4$ aqueous electrolyte. The scan rate is 0.2 mV $s^{-1}$

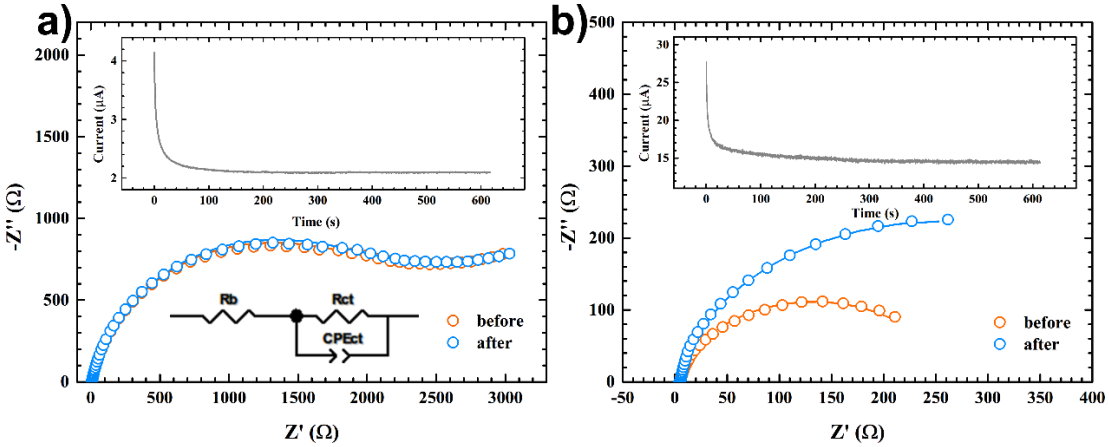


**Fig. S4** $Zn^{2+}$ transference number tests for **a** pristine and **b** Janus separator in 2 M $ZnSO_4$ electrolyte

The $Zn^{2+}$ transference number was calculated based on the equation $t_{Zn^{2+}} = \frac{I_S(\Delta V - I_0 R_0)}{I_0(\Delta V - I_S R_S)}$, where $I_0$/ $I_s$ and $R_0$/ $R_s$ are the initial/ steady current and interfacial resistance, respectively. $\Delta V$ is set as 10 mV. The $I_0$/ $I_s$, and $R_0$/ $R_s$ for pristine and Janus separator are 4.15/2.09 μA, 1477/1570 Ω and 27.7/14.4 μA, 256.2/505 Ω, respectively.

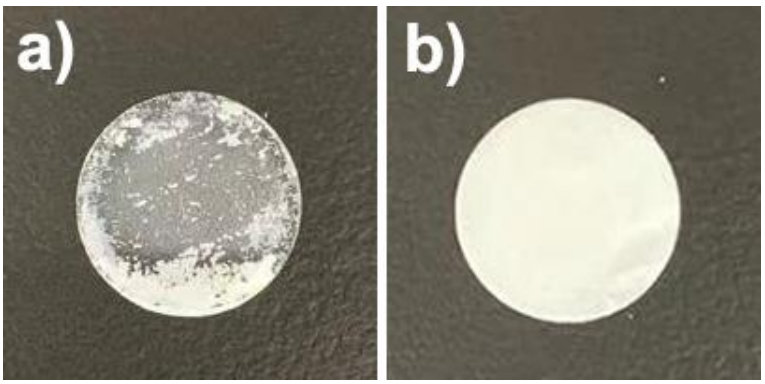


**Fig. S5** Photographs of the Zn anode after cycling: **a** upon disassembly and **b** after ultrasonic cleaning

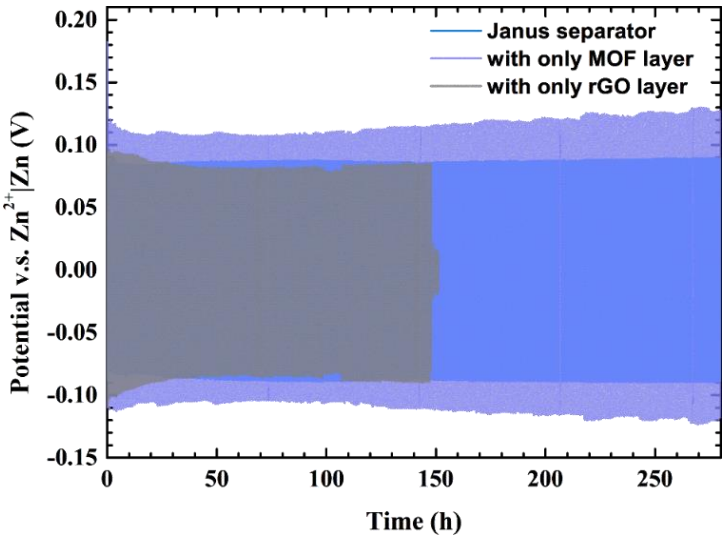


**Fig. S6** Cycling performance of the Zn|Zn symmetric cells using the separator with only MOF or rGO layer at 2.0 mA $cm^{-2}$, 1 mAh $cm^{-2}$

**Fig. S7** Photograph of the pristine separator of Zn|Cu cell after short-circuit

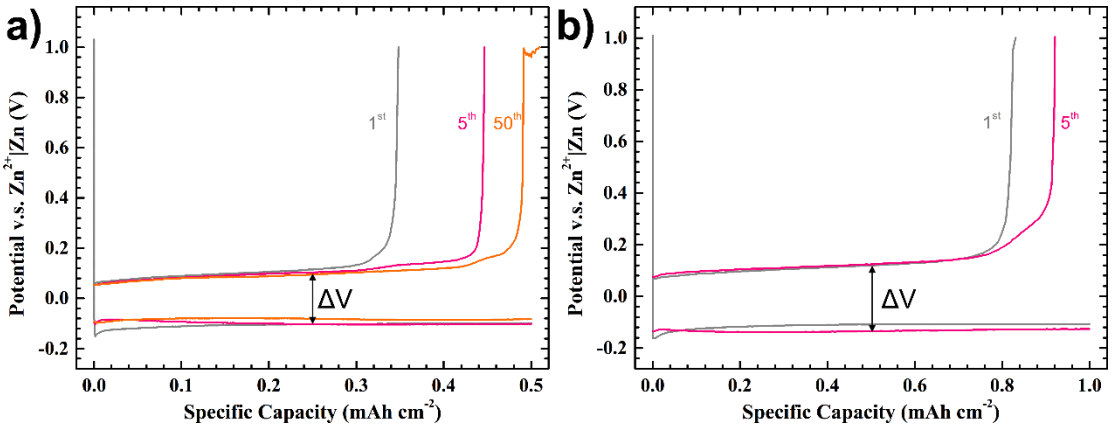


**Fig. S8** Voltage profiles of the Zn|Cu cell with pristine separator for the selected cycles at **a** 0.5 and **b** 2.0 mA $cm^{-2}$

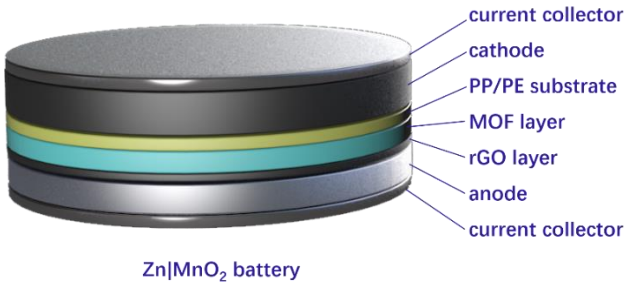


**Fig. S9** Illustration of Zn|$MnO_2$ battery configuration

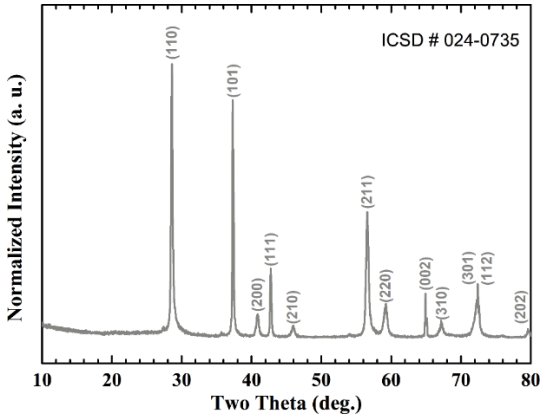


**Fig. S10** XRD pattern of the synthesized β-$MnO_2$@rGO

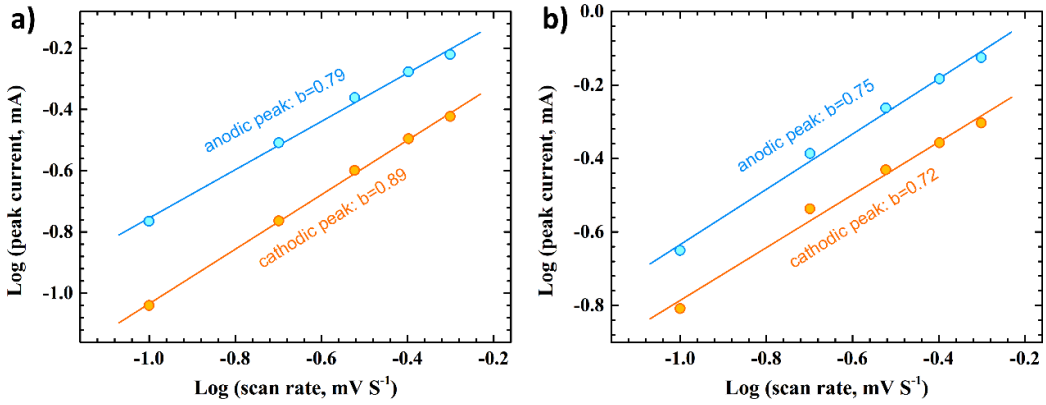


**Fig. S11** Calculation of *b* values for the cells with **a** Janus and **b** pristine separators

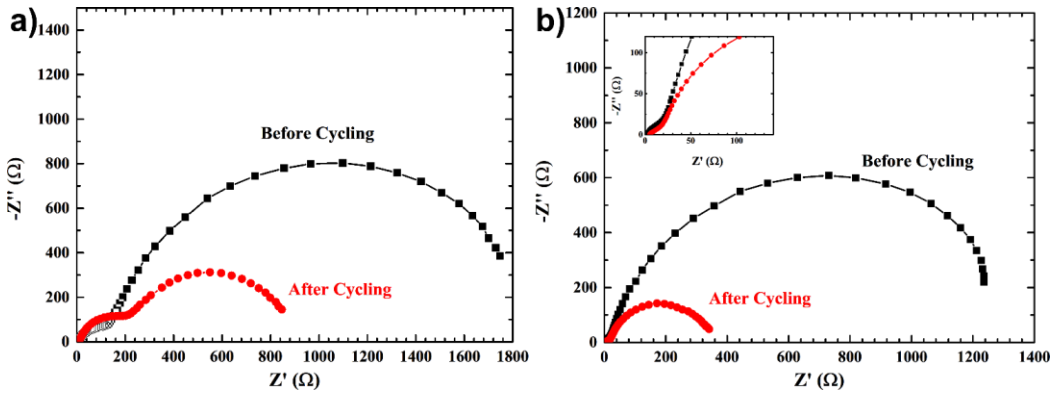


**Fig. S12** EIS profiles of the batteries with **a** pristine and **b** Janus separators before cycling and after 50 cycles at 4 A $g^{-1}$ charge/discharge rate

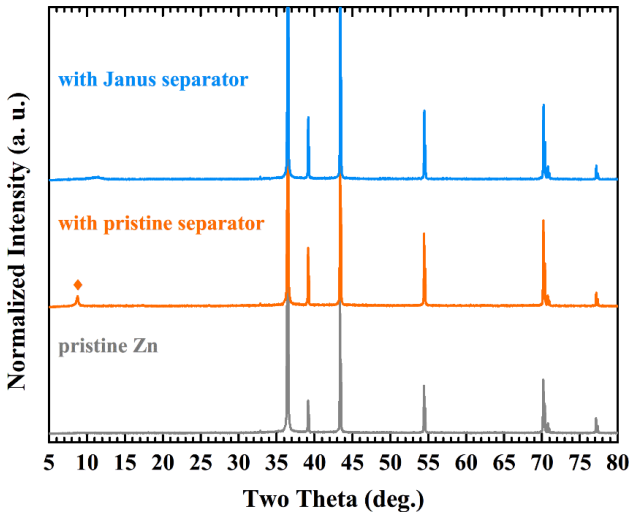


**Fig. S13** XRD for the Zn anodes of the disassembled cells. The refection peak of zinc hydroxide sulphate ($Zn_4SO_4(OH)_6 \cdot 4H_2O$, PDF No. 44-0673) is marked with a diamond

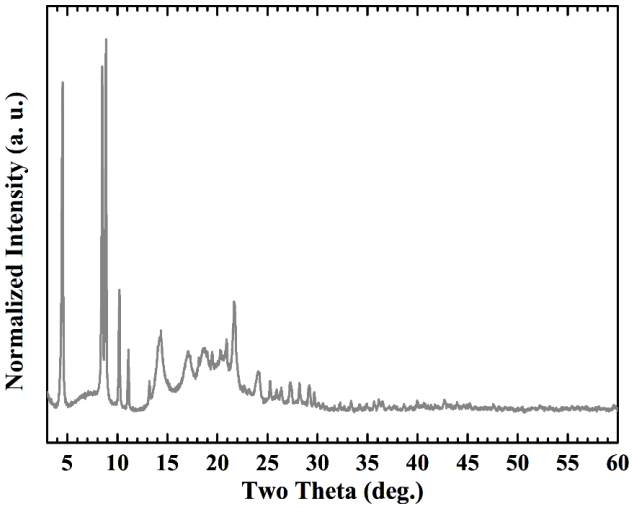


**Fig. S14** XRD of the Janus separator after battery cycling