

Supporting Information

Inverse-spinel Mg_2MnO_4 cathode for high-performance and flexible aqueous zinc-ion battery

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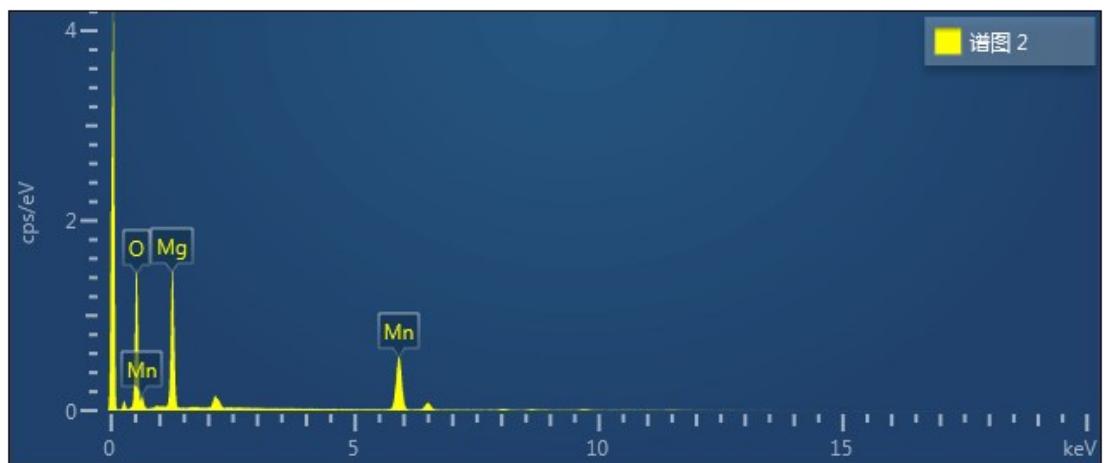


Figure S1. The SEM-EDS of as-synthesized Mg_2MnO_4 sample.

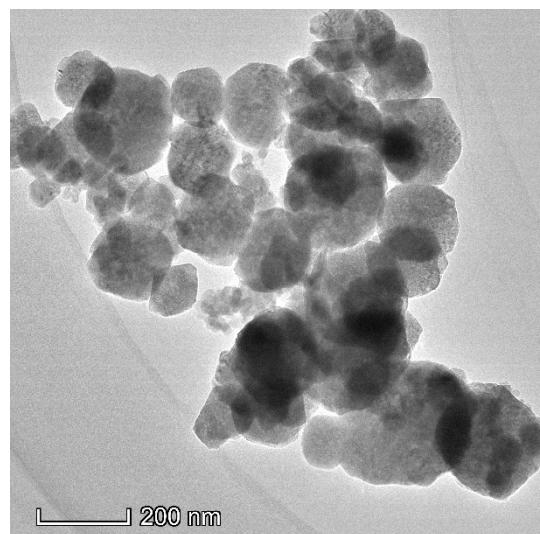


Figure S2. HRTEM image of the as-obtained Mg_2MnO_4 sample.

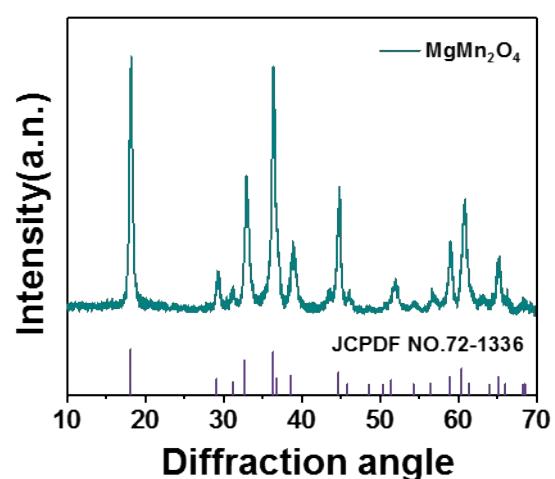


Figure S3. XRD pattern of the as-obtained MgMn_2O_4 sample.

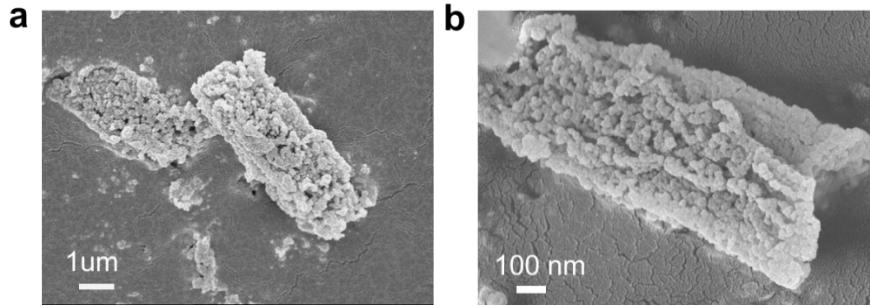


Figure S4. SEM images of the as-obtained MgMn_2O_4 .

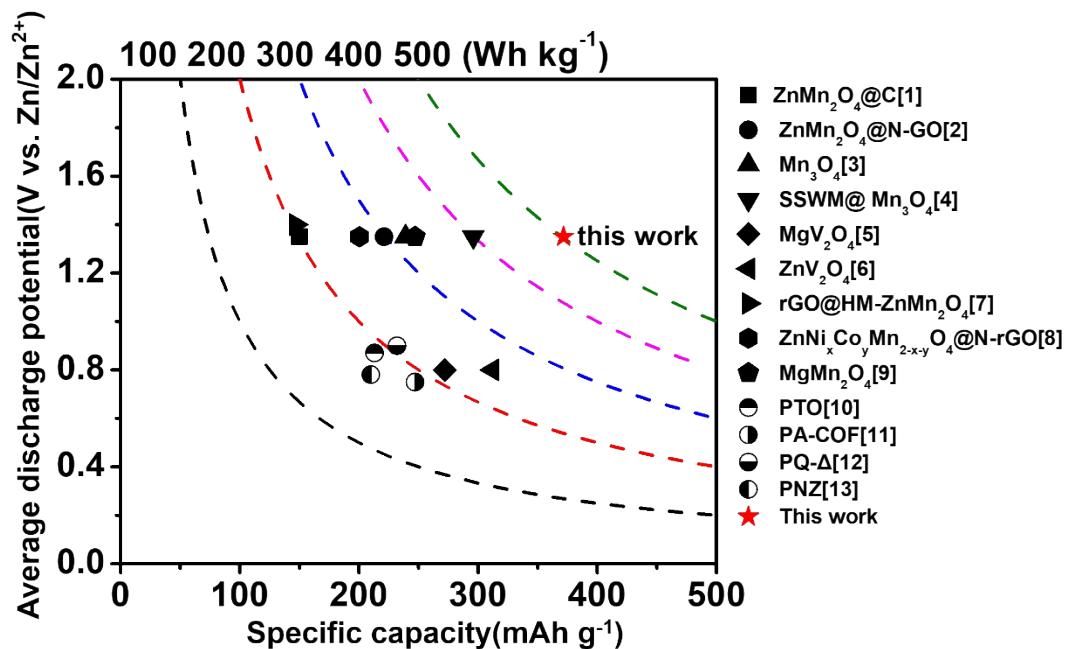


Figure S5. A comparison of average discharge potential, specific capacity and energy density between this work and reported materials.

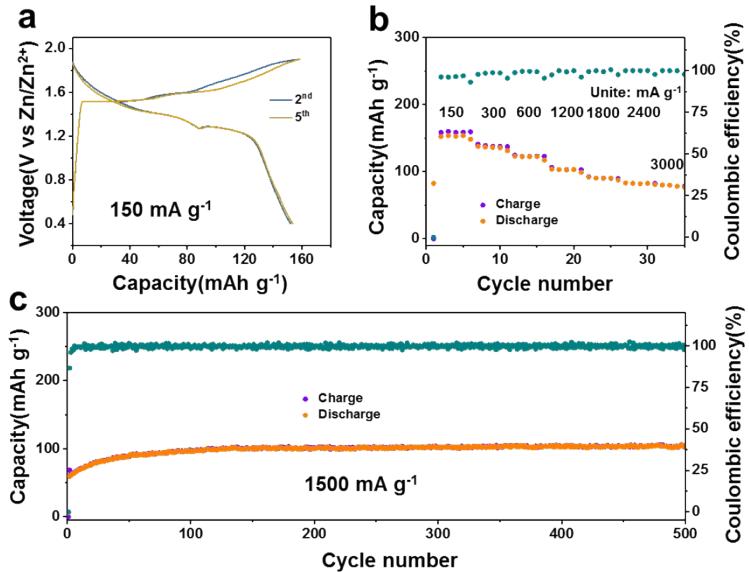


Figure S6. a) The charge-discharge curves of Zn//MgMn₂O₄ battery. b) Rate capacity of Zn//MgMn₂O₄ battery. c) Cycling stability of Zn//MgMn₂O₄ battery.

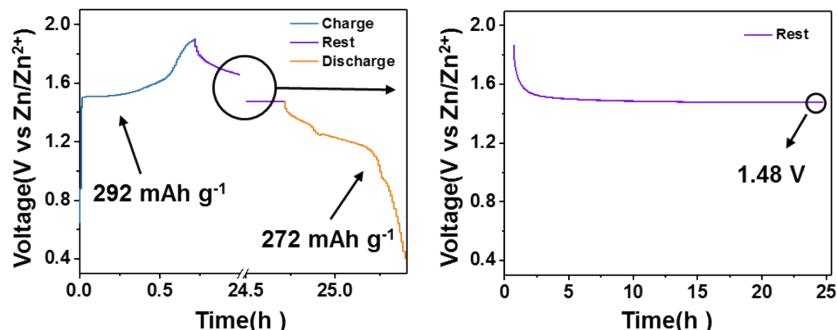


Figure S7. Electrochemical stability of the full battery surveyed by self-discharge experiments. Specifically, the full battery was fully charged to 1.9 V, then fully discharged to 0.4 V after rest for 24 h.

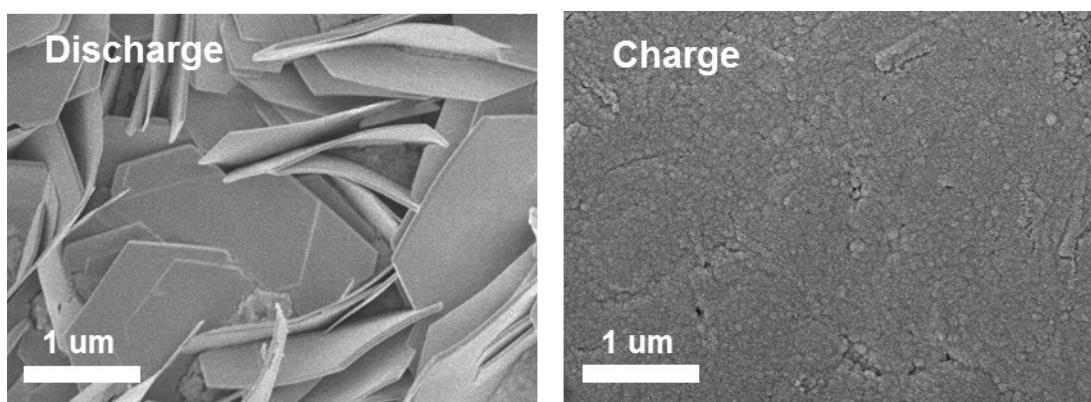


Figure S8. a~b) The ex-situ SEM patterns of Mg₂MnO₄ electrodes. a) Discharge to 0.4 V; b) Charge to 1.9 V.

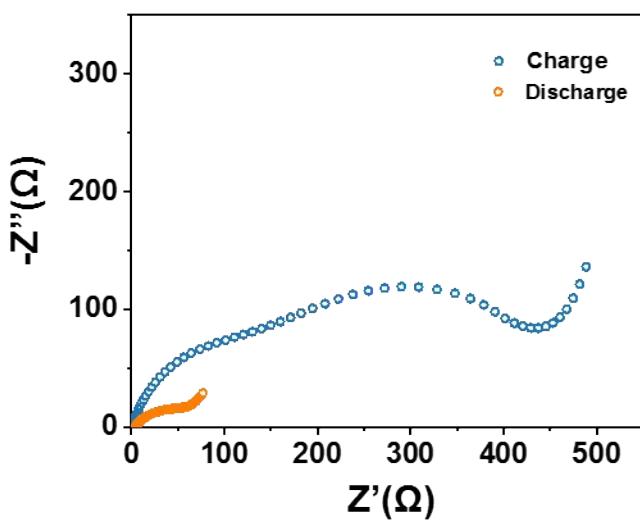


Figure S9. Typical Nyquist plots for Zn//Mg₂MnO₄ battery at different state.

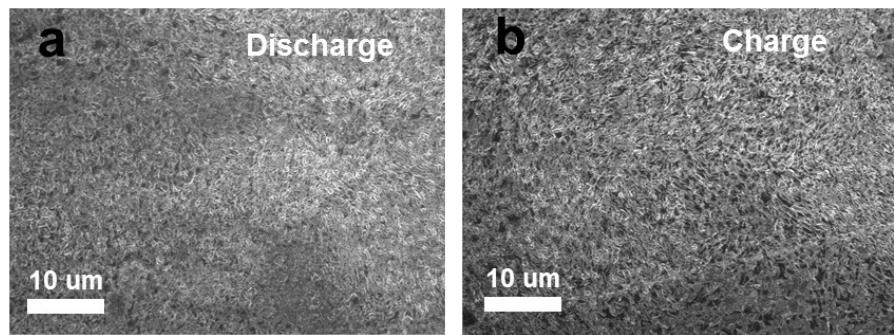


Figure S10. a~b) The ex-situ SEM patterns of Zn anode. a) Discharge to 0.4 V; b) Charge to 1.9 V.

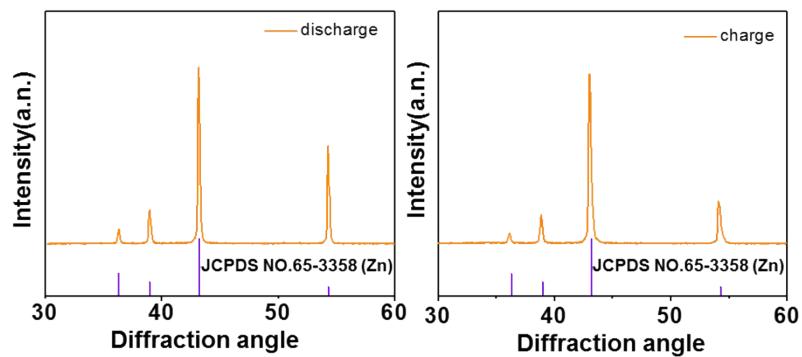


Figure S11. a~b) The ex-situ XRD patterns of Zn anode. a) Discharge to 0.4 V; b) Charge to 1.9 V.

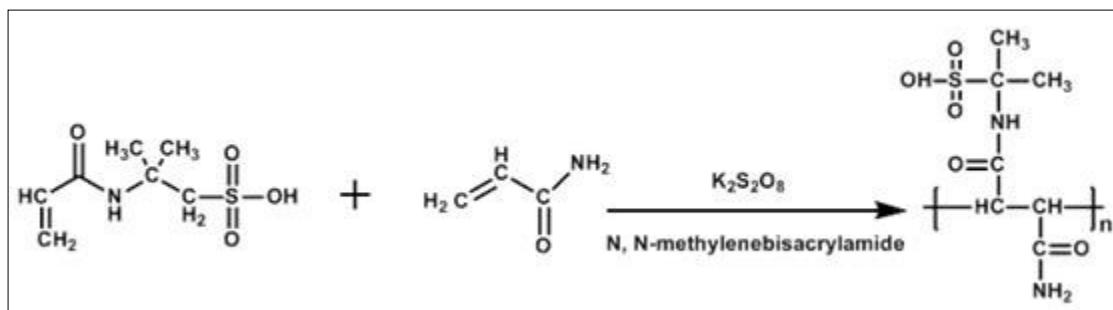


Figure S12. The structural formula for the polymerization.

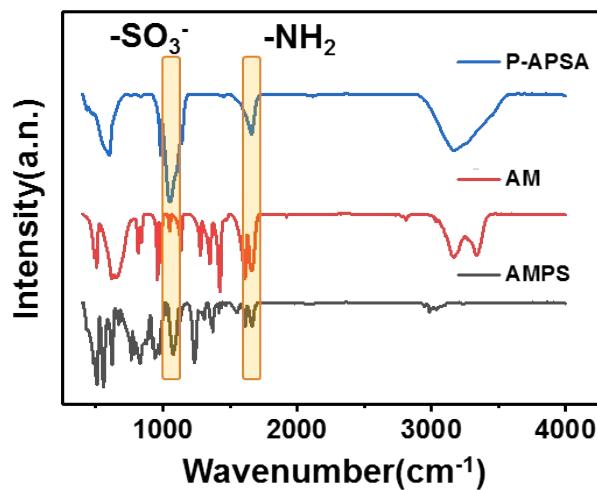


Figure S13. The ATR-FTIR spectra of AM, AMPS, and P-APSA sample.



Figure S14. The stretching test of P-APSA sample.

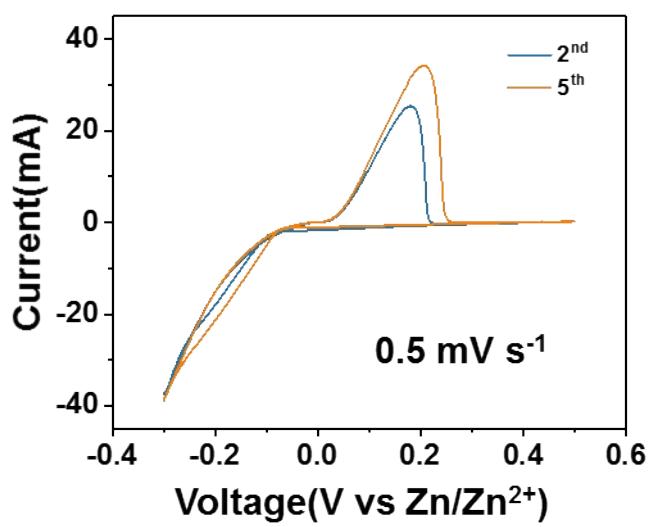


Figure S15. Reversible Zn^{2+} plating/stripping behaviour on SS

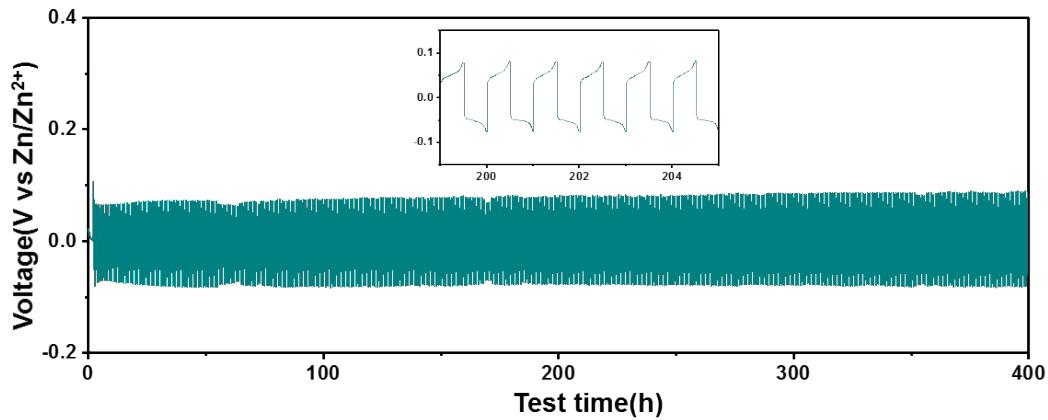


Figure S16. Cycling stability of $\text{Zn}/\text{P-APSA}/\text{Zn}$ symmetric cell.

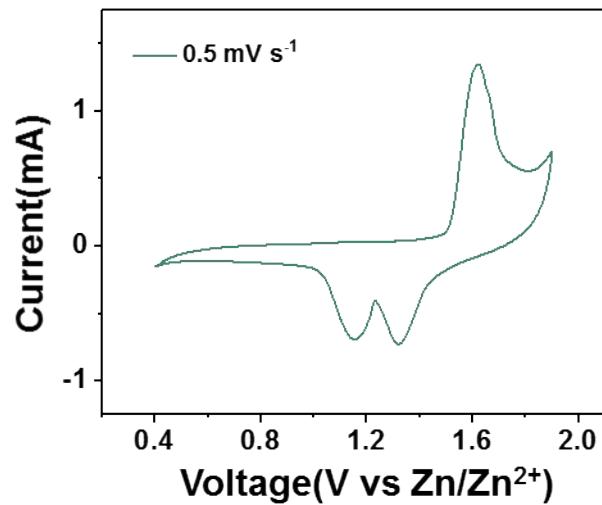


Figure S17. The CV curves of Zn//P-APSA//Mg₂MnO₄ battery.

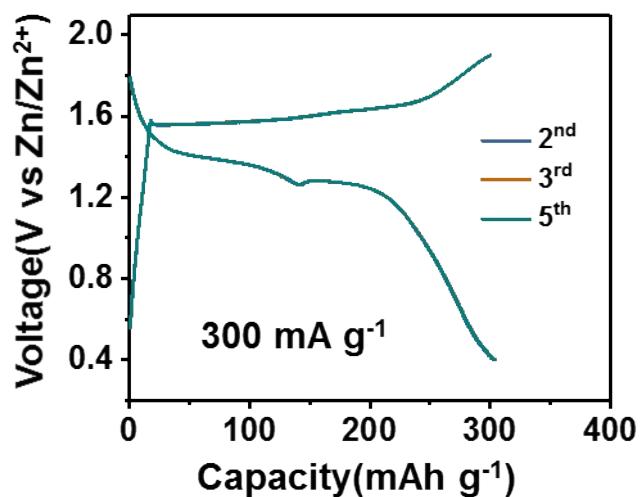


Figure S18. The charge-discharge curves of Zn//P-APSA//Mg₂MnO₄ battery.

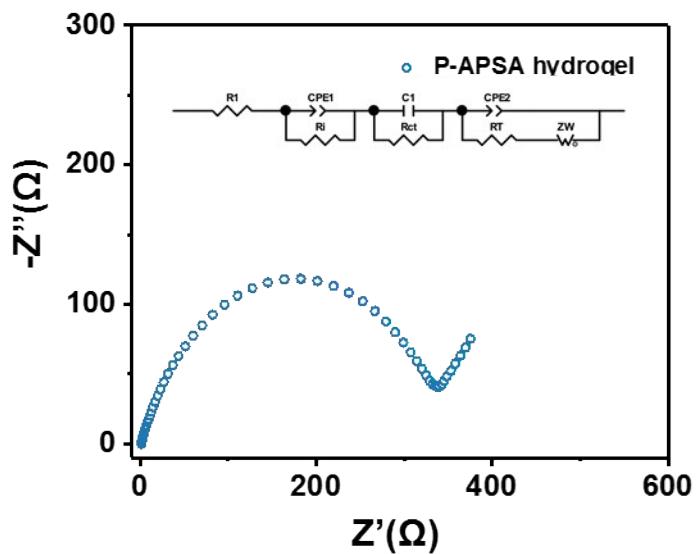


Figure S19. Typical Nyquist plots for Zn//P-APSA// Mg_2MnO_4 battery.

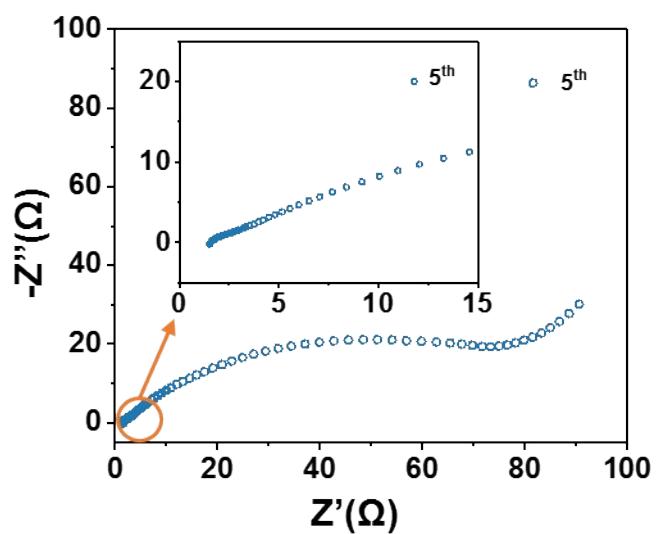


Figure S20. Typical Nyquist plots for Zn//P-APSA// Mg_2MnO_4 battery after five cycles.

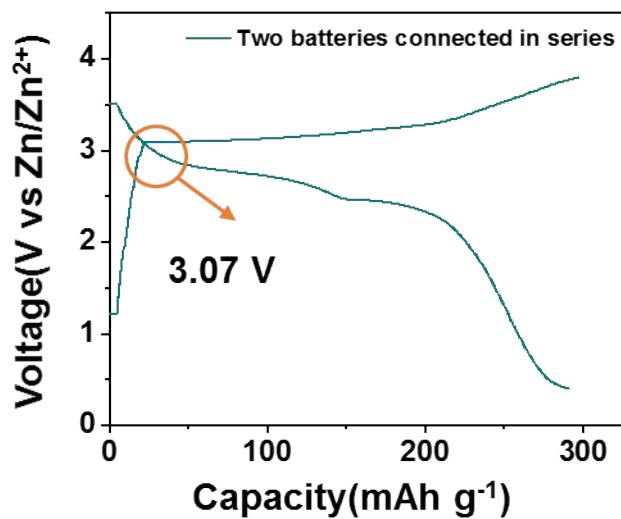


Figure S21. The charge-discharge curve of two batteries connected in series.

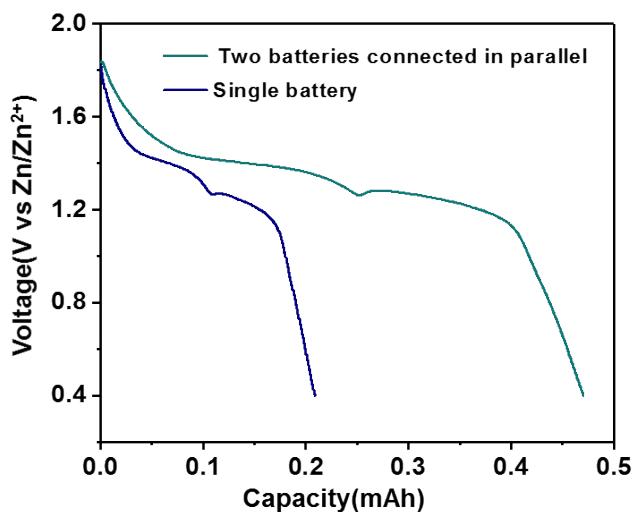


Figure S22. The discharge curves of single battery and two batteries connected in parallel.

Table S1 The SEM-EDS of as-synthesis Mg_2MnO_4 sample.

Technology		SEM-EDS		ICP-OES
Element	wt%	wt% sigma	Atom%	wt%
O	36.77	0.29	55.06	
Mg	31.62	0.25	31.16	35.907%
Mn	31.61	0.27	13.78	36.105%
Mole ration of Mn:Mg	0.442:1			0.445:1

Table S2 A comparison of electrochemical performance between this work and reported materials.

Electrode	Electrolyte	Specific capacity	Cycling stability	Ref
$ZnMn_2O_4@C$	3 M $Zn(CF_3SO_3)_2$	150 mAh g ⁻¹ at 50 mA g ⁻¹	94% after 500 cycles at 500 mA g ⁻¹	¹
$ZnMn_2O_4@N-GO$	1M $ZnSO_4$ + 0.05M $MnSO_4$	221 mAh g ⁻¹ at 100 mA g ⁻¹	97.4% after 2500 cycles at 1000 mA g ⁻¹	²
Mn_3O_4	2 M $ZnSO_4$	239.2 mAh g ⁻¹ at 100 mA g ⁻¹	72.2% after 300 cycles at 500 mA g ⁻¹	³
$SSWM@Mn_3O_4$	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	296 mAh g ⁻¹ at 100 mA g ⁻¹	60% after 500 cycles at 500 mA g ⁻¹	⁴
MgV_2O_4	2 M $Zn(TFSI)_2$	272 mAh g ⁻¹ at 200 mA g ⁻¹	74% after 500 cycles at 4000 mA g ⁻¹	⁵
ZnV_2O_4	2 M $Zn(ClO_4)_2$	312 mAh g ⁻¹ at 0.5 C	82% after 1000 cycles at 10C	⁶
$rGO@HM-ZnMn_2O_4$	1M $ZnSO_4$ +0.05M $MnSO_4$	146.9 mAh g ⁻¹ at 300 mA g ⁻¹	88% after 650 cycles at 1000 mA g ⁻¹	⁷
$ZnNi_xCo_yMn_{2-x-y}O_4@N-rGO$	2M $ZnSO_4$ + 0.2M $MnSO_4$	200.5 mAh g ⁻¹ at 10 mA g ⁻¹	79% after 900 cycles at 1000 mA g ⁻¹	⁸
$MgMn_2O_4$	1M $ZnSO_4$ + 1M $MgSO_4$ + 0.1 M $MnSO_4$	247 mAh g ⁻¹ at 50 mA g ⁻¹	80% after 500 cycles at 500 mA g ⁻¹	⁹
Mg_2MnO_4	2M $ZnSO_4$ + 0.1M $MnSO_4$	371.7 mAh g ⁻¹ at 150 mA g ⁻¹	85% after 2000 cycles at 3000 mA g ⁻¹ (compared to the discharge capacity after activation)	This work

Reference:

- N. Zhang, F. Cheng, Y. Liu, Q. Zhao, K. Lei, C. Chen, X. Liu and J. Chen, *J. Am. Chem. Soc.*, 2016, **138**, 12894-12901.
- L. Chen, Z. Yang, H. Qin, X. Zeng and J. Meng, *J. Power Sources*, 2019, **425**, 162-169.
- J. Hao, J. Mou, J. Zhang, L. Dong, W. Liu, C. Xu and F. Kang, *Electrochim. Acta*, 2018, **259**, 170-178.
- C. Zhu, G. Fang, J. Zhou, J. Guo, Z. Wang, C. Wang, J. Li, Y. Tang and S. Liang, *J. Mater. Chem. A*, 2018, **6**,

- 9677-9683.
- 5. W. Tang, B. Lan, C. Tang, Q. An, L. Chen, W. Zhang, C. Zuo, S. Dong and P. Luo, *ACS Sustainable Chem. Eng.*, 2020, **8**, 3681-3688.
 - 6. Y. Liu, C. Li, J. Xu, M. Ou, C. Fang, S. Sun, Y. Qiu, J. Peng, G. Lu, Q. Li, J. Han and Y. Huang, *Nano Energy*, 2020, **67**, 104211.
 - 7. L. Chen, Z. Yang, H. Qin, X. Zeng, J. Meng and H. Chen, *Electrochim. Acta*, 2019, **317**, 155-163.
 - 8. Y. Tao, Z. Li, L. Tang, X. Pu, T. Cao, D. Cheng, Q. Xu, H. Liu, Y. Wang and Y. Xia, *Electrochim. Acta*, 2020, **331**, 135296.
 - 9. V. Soundharajan, B. Sambandam, S. Kim, V. Mathew, J. Jo, S. Kim, J. Lee, S. Islam, K. Kim, Y.-K. Sun and J. Kim, *ACS Energy Lett.*, 2018, **3**, 1998-2004.
 - 10. Z. Guo, Y. Ma, X. Dong, J. Huang, Y. Wang, Y. Xia, *Angew. Chem. Int. Ed.*, 2018, **57**, 11737-11741.
 - 11. W. Wang, V. S. Kale, Z. Cao, S. Kandambeth, W. Zhang, J. Ming, P. T. Parvatkar, E. Abou-Hamad, O. Shekhah, L. Cavallo, M. Eddaoudi, H. N. Alshareef, *ACS Energy Lett.* 2020, **5**, 2256-2264.
 - 12. Q. Wang, Y. Liu, P. Chen, *J. Power Sources*, 2020, **468**, 228401.
 - 13. K. W. Nam, H. Kim, Y. Beldjoudi, T. W. Kwon, D. J. Kim, J. F. Stoddart, *J. Am. Chem. Soc.*, 2020, **142**, 2541-2548.