Supporting Information for

High-Performance Aqueous Zinc-Manganese Battery with Reversible Mn^{2+}/Mn^{4+} Double Redox Achieved by Carbon Coated MnO_x Nanoparticles

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Supplementary Figures

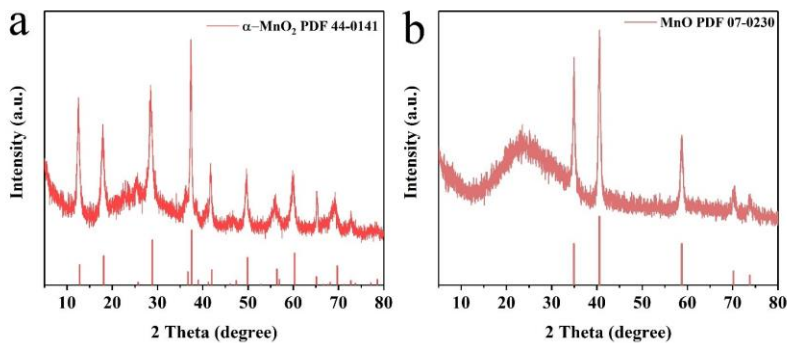

Fig. S1 XRD patterns of **a** α-MnO_2 and **b** MnO

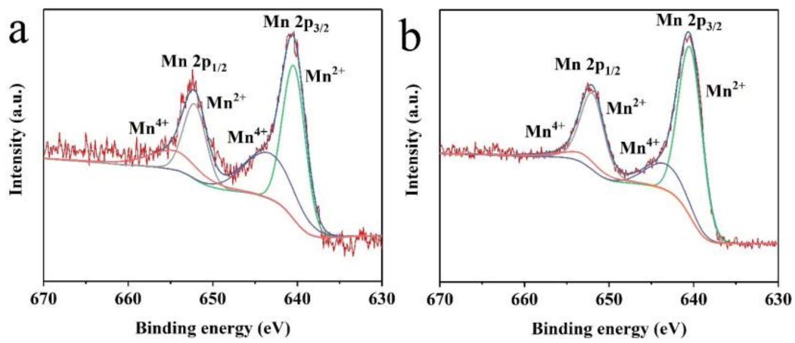

Fig. S2 XPS spectra showing the 2*p* core-level spectra of Mn in **a** MnO_x-1 and **b** MnO_x-3

Table S1 Comparison for electrochemical performances of representative Mn-O cathode materials in Zinc ion battery and our present work

Cathode material	Electrolytes	Capacity	Max Energy density	Max Power density	Capacity retention
α-MnO_2 [S1]	1 M $ZnSO_4$	353 mAh g^{-1} at 16 mA g^{-1}	—	—	63% after 50 cycles
α- MnO_2 [S2]	0.1 M $Zn(NO_3)_2$	210 mAh g^{-1} at 0.5C	—	—	100% after 100 cycles
α- MnO_2 [S3]	1 M $ZnSO_4$	323 mAh g^{-1} at 16 mA g^{-1}	—	—	46% after 75 cycles
α- MnO_2 [S4]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	285 mAh g^{-1} at C/3	~170 Wh kg^{-1} (cathode, anode and electrolyte)	—	92% after 5000 cycles.
β- MnO_2 [S5]	3 M $Zn(CF_3SO_3)_2$ + 0.1 M $Mn(CF_3S_3)_2$	258 mAh g^{-1} at 0.65 C	254 Wh kg^{-1} (based on cathode)	5.9 kW kg^{-1} (based on cathode)	94% after 2000 cycles.
γ- MnO_2 [S6]	1 M $ZnSO_4$	285 mAh g^{-1} at 0.05 mA cm^{-2}	—	—	63% after 45 cycles
ε- MnO_2 [S7]	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	290 mAh g^{-1} at 90 mA g^{-1}	—	—	99.3% after 10000 cycles.
δ- MnO_2 [S8]	1 M $ZnSO_4$	252 mAh g^{-1} at 83 mA g^{-1}	—	—	～44% after 100 cycles
Graphene/α-MnO_2 [S9]	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	382 mAh g^{-1} at 300 mA g^{-1}	406.6 Wh kg^{-1}(based on cathode)	9.5 kW kg^{-1} (based on cathode)	94% after 3000 cycles
MnO_2/PEDOT [S10]	PVA+3 M LiCl+2 M $ZnCl_2$ + 0.4 M $MnSO_4$	367 mA h g^{-1} at 0.74 A g^{-1}	505 Wh kg^{-1} (based on cathode)	8.6 kW kg^{-1} (based on cathode)	83.7% after 300 cycles
Polyaniline-intercalated MnO_2 [S11]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	280 mA h g^{-1} at 200 mA g^{-1}	—	—	100% after 200 cycles
O_d- MnO_2 [S12]	1 M $ZnSO_4$ + 0.2 M $MnSO_4$	345 mAh g^{-1} at 200 mA g^{-1}	470 Wh kg^{-1} (based on cathode)	10 kW kg^{-1} (based on cathode)	84% after 2000 cycles
Mn_2O_3 [S13]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	148 mAh g^{-1} at 100 mA g^{-1}	—	—	～68% after 2000 cycles
Mn_3O_4 [S14]	2 M $ZnSO_4$	239 mAh g^{-1} at 100 mA g^{-1}	—	—	—
MnO_x@N-C [S15]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	385 mAh g^{-1} at 100 mA g^{-1}	—	—	100% after 1600 cycles
D-β-MnO_2 [S16]	3 M $ZnSO_4$ + 0.1 M $MnSO_4$	276 mAh g^{-1} at 100 mA g^{-1}	—	—	94 % after 300 cycles
MnO_2 [S17]	6 M KOH + 0.2 M ZnO + 5 mM vanillin and 3 M H_2SO_4 + 0.1	616 mAh g^{-1} at 100 mA g^{-1}	1,621.7 Wh $kg^{-1}_{MnO_2}$	—	96 % after 200 cycles

	M $MnSO_4$)				
G-MnO_2 [S18]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	321 mAh g^{-1} at 240 mA g^{-1}	—	—	91 % after 300 cycles
P-MnO_{2-x}@VMG [S19]	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	302.8 mAh g^{-1} at 500 mA g^{-1}	—	—	90 % after 1000 cycles
Mn_2O_3 [S20]	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	233 mAh g^{-1} at 300 mA g^{-1}	—	—	89 % after 3000 cycles
Birnessite MnO_2 [S21]	2 M $ZnSO_4$ + 0.5 M $MnSO_4$	279.7 mAh g^{-1} at 300 mA g^{-1}	—	—	61 % after 1500 cycles
Ca_2MnO_4 [S22]	2 M $ZnSO_4$ + 0.1 M $MnSO_4$	250 mAh g^{-1} at 100 mA g^{-1}	—	—	80 % after 1000 cycles
N-MnO_{2-x} [S23]	2 M $ZnSO_4$ + 0.2 M $MnSO_4$	285 mAh g^{-1} at 200 mA g^{-1}	—	—	85.7 % after 1000 cycles
$MnO_2H_{0.16}(H_2O)_{0.27}$ [S24]	1 M $ZnSO_4$ + 0.2 M $MnSO_4$	275.6 mAh g^{-1} at 30.8 mA g^{-1}	228.5 Wh kg^{-1}	—	96 % after 500 cycles
MnO_x (our work)	1 M $ZnSO_4$ + 0.3 M $MnSO_4$	842.5 mAh g^{-1} at 200 mA g^{-1}	1158 Wh kg^{-1} (based on initial cathode)	1.2 kW kg^{-1} (based on initial cathode)	80% after 1500 cycles

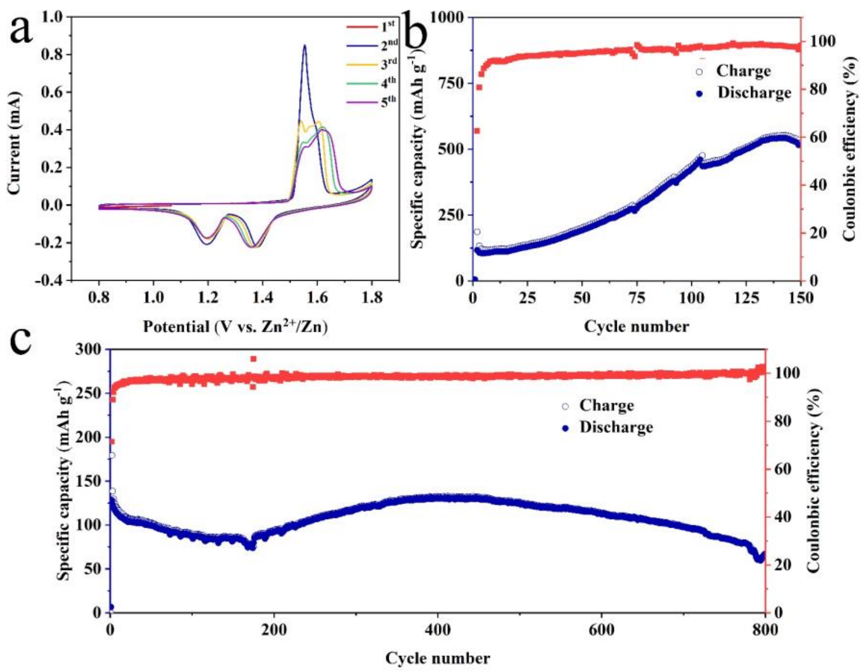

Fig. S3 a CV curses at 0.1 mV s^{-1} in the voltage range of 0.8-1.8 V vs. Zn^{2+}/Zn, **b** cycling performance at 0.2 A g^{-1}; **c** Cycling performance at 1 A g^{-1} of MnO_x-1

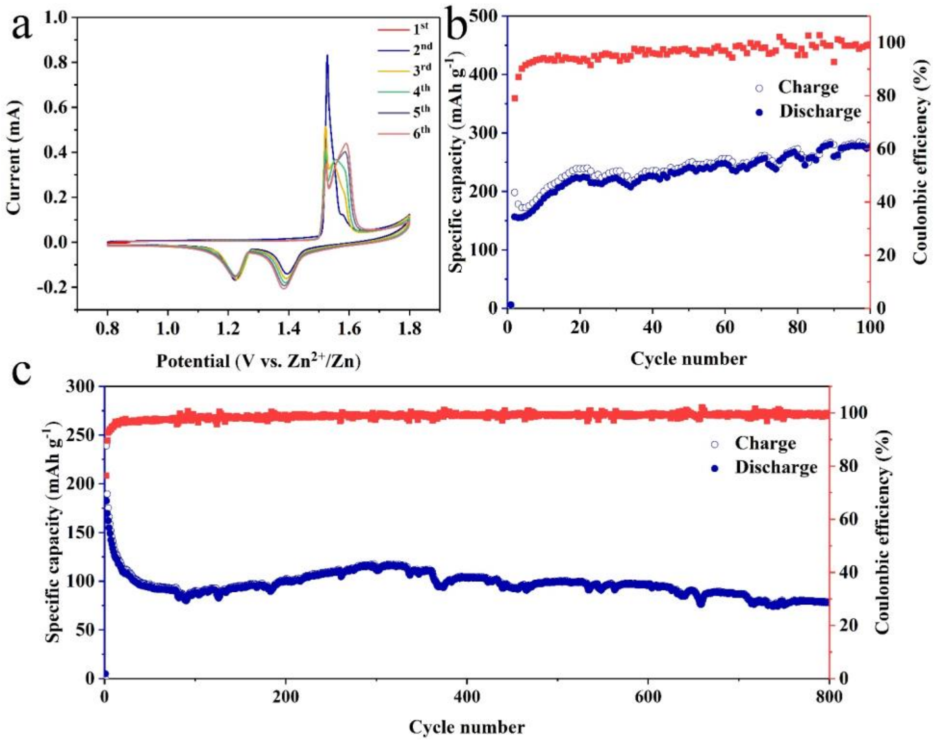

Fig. S4 a CV curses at 0.1 mV s^{-1} in the voltage range of 0.8-1.8 V vs. Zn^{2+}/Zn, **b** cycling performance at 0.2 A g^{-1}; **c** Cycling performance at 0.5 A g^{-1} of MnO_x-3

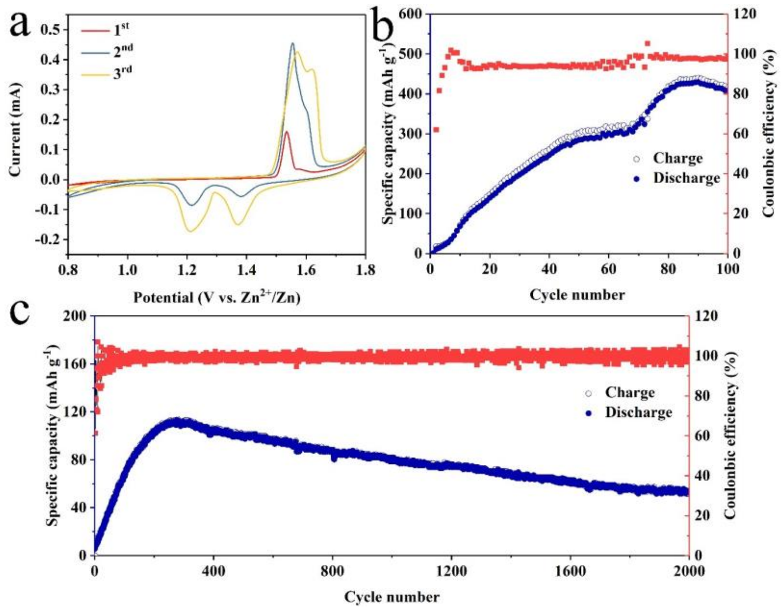

Fig. S5 a CV curses at 0.1 mV s^{-1} in the voltage range of 0.8-1.8 V vs. Zn^{2+}/Zn, **b** cycling performance at 0.2 A g^{-1}; **c** Cycling performance at 1 A g^{-1} of MnO

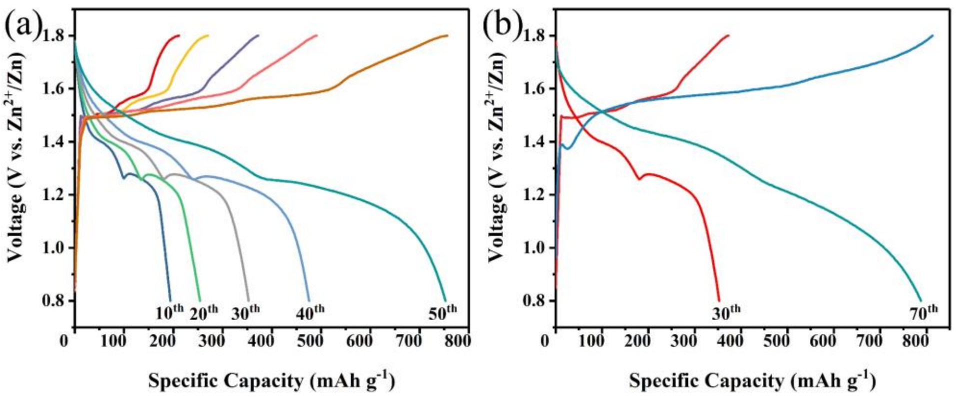

Fig. S6 Voltage profile of MnO_x-2

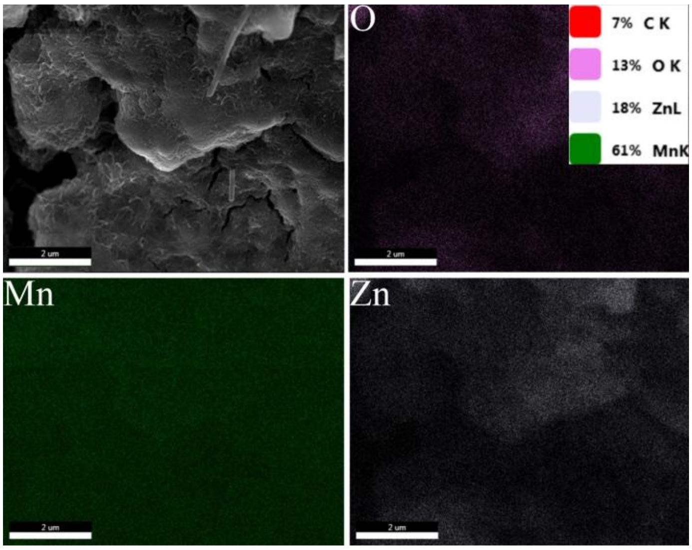

Fig. S7 *ex*-situ SEM image and EDX elemental mapping images of MnO_x-2 when discharging to 1.28 V

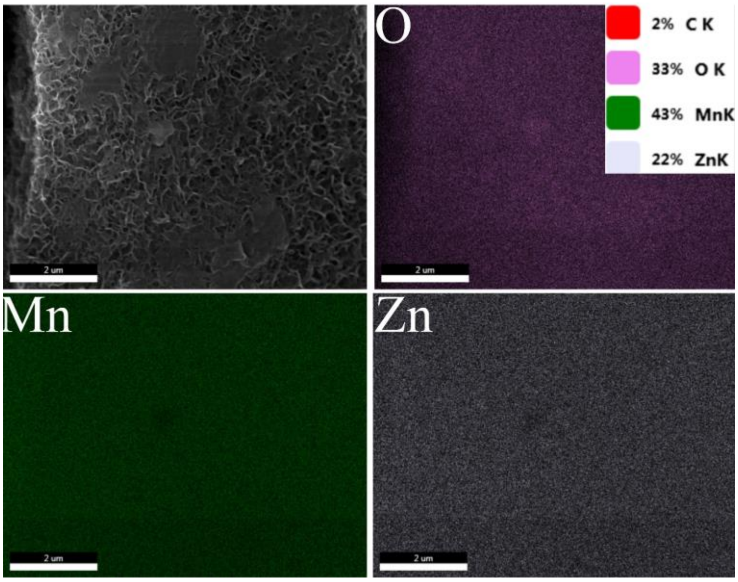

Fig. S8 *ex*-situ SEM image and EDX elemental mapping images of MnO_x-2 when charging to 1.55 V

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