

ZnMn₃O₇: A New Layered Cathode Material for Fast-Charging Zinc-Ion Batteries

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n Part 1: Theoretical Methods

Spin-polarized structure relaxations and static energy calculations were based on the density functional theory (DFT) implemented in the Vienna ab initio simulation package (VASP).^[1] The interaction between core electron and nuclei was treated by the projector-augmented wave (PAW) pseudopotentials,^[2] and a cutoff energy of 520 eV was adopted for the plane wave expansion. The Perdew, Burke, and Ernzerhof (PBE)^[3] functional of general gradient approximation (GGA) was applied to describe the exchange and correlation interactions. A Gamma-centered Monkhorst-Pack 4 x 4 x 2 *k*-point grid was employed to sample the Brillouin zones (BZ) of ZnMn₃O₇. The GGA+U formalism with a Hubbard *U* value of 3.8 eV^[4] was exploited to account for the on-site Coulombic interactions of the localized Mn 3*d* electrons. And DFT-D3 proposed by Grimme^[5] was used to describe the van der Waals interactions. The optimization of cell dimension and atomic position was completed until the energy changes between each electronic iteration were less than 1x10⁻⁶ eV and the Hellmann-Feynman forces acting on each atom were lower than 0.02 eV/Å.

Table S1. lattice parameters of ZnMn_3O_7 .

Structure	Lattice parameters						
	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)	$V(\text{Å}^3)$
ZnMn_3O_7	7.5842	7.5842	14.2955	90	90	120	705.36

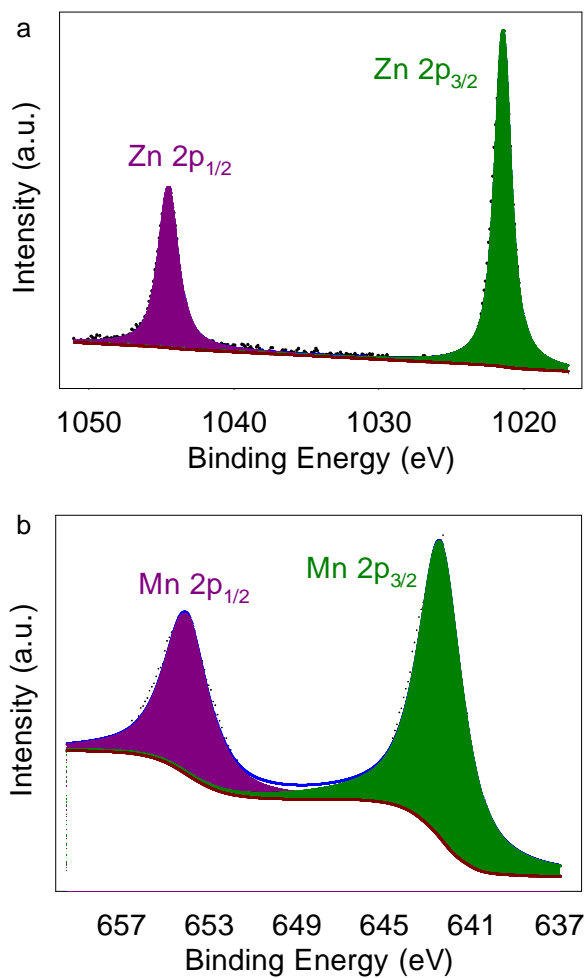


Figure S1. XPS analysis of Zn 2p and Mn 2p.

Table S2. Galvanostatic Cycling Comparisons.

Cathode	Voltage window	Cycle	Capacity retention	Current Density	Reference
ZnMn ₃ O ₇	0.8-1.85 V	100	85.3%	0.85 C	This work
δ-MnO ₂	1.0-1.8 V	100	34.77%	1.3 C	[6]
ZnMn ₂ O ₄	0.8-1.8 V	90	67.87%	0.42 C	[7]
α-MnO ₂	1.0-1.8 V	50	65.67%	0.27 C	[8]
α-MnO ₂	1.0-1.8 V	75	43.90%	0.27 C	[9]
α-MnO ₂	1.0-.85 V	100	53.30%	0.27C	[10]
β-MnO ₂	1.0-1.8 V	50	55%	0.33 C	[11]
β-MnO ₂	0.8-1.9 V	100	81.80%	0.65 C	[12]
α-Mn ₂ O ₃	1.0-1.9 V	30	79.19%	0.32 C	[13]
α-MnO ₂	1.0-1.8 V	100	39.34%	1 C	[14]
δ-MnO ₂	1.0-1.8 V	100	44.40%	0.27 C	[15]
α-MnO ₂	1.0-1.8 V	100	57.40%	0.33 C	[16]
Mn ₃ O ₄	1.0-1.8 V	30	48%	0.44 C	[17]
ZnMn ₂ O ₄	0.8-1.8 V	100	71.30%	1.25 C	[18]
α-MnO ₂	1.0-1.9 V	20	92.30%	0.27 C	[19]
T-MnO ₂	0.7-2.0 V	50	83.30%	0.5 C	[20]
δ-MnO ₂	1.0-1.8 V	100	66.50%	0.33 C	[21]
α-MnO ₂	1.0-.85 V	100	89.70%	0.97 C	[10]
α-MnO ₂	0.7-2.0 V	30	70%	0.2 C	[22]
γ-MnO ₂	1.0-1.8 V	40	63.20%	0.27 C	[23]
O _d -MnO ₂	1.0-1.8 V	100	91.90%	0.65 C	[24]
α-MnO ₂	0.7-2.0 V	50	56.50%	0.5 C	[20]
Na ₃ V ₂ (PO ₄) ₃	0.8-1.7 V	100	83.30%	0.5 C	[25]
ZnHCF	0.8-1.9 V	100	75%	1 C	[26]
LiV ₃ O ₈	0.6-1.2 V	65	74.50%	0.52 C	[27]
H ₃ V ₂ O ₈	0.2-1.6 V	60	84.2%	0.33 C	[28]
CuHCF	0.2-1.2 V	20	77%	0.24 C	[29]

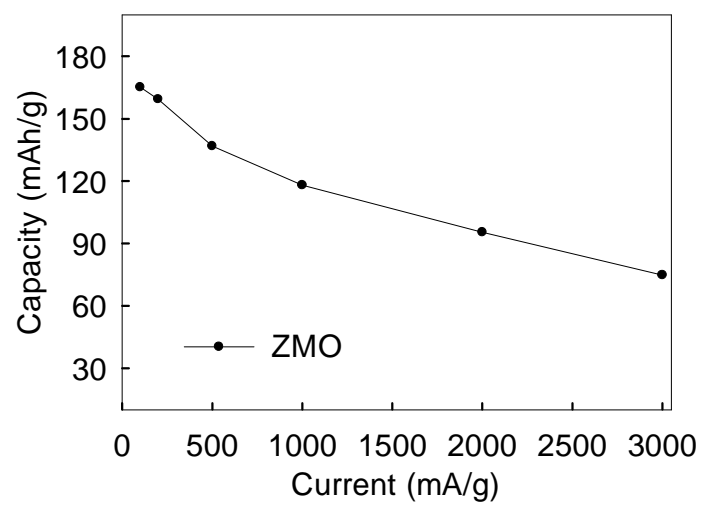


Figure S2. Rate capability of ZMO.

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Table S3. Rate Capability Comparisons.

Cathode	Rate (C) (mA/g)	Capacity (mAh/g) (mA h/g)	Capacity retention (%)	Reference
ZnMn ₃ O ₇	0.1	165	100.00%	This work
	0.2	159	96.51%	
	0.5	137	82.86%	
	1	118	71.53%	
	2	95	57.73%	
	3	75	45.32%	
δ -MnO ₂	0.083	208	100.00%	[15]
	0.166	201	96.63%	
	0.333	150	72.12%	
	0.666	92	44.23%	
	1.333	30	14.42%	
	1.666	7	3.37%	
ZnMn ₂ O ₄	0.05	148	100.00%	[30]
	0.1	121	81.76%	
	0.2	102	68.92%	
	0.5	90	60.81%	
	1	81	54.73%	
	2	78	52.70%	
α -MnO ₂	0.016	353	100.00%	[8]
	0.033	272	77.05%	
	0.066	229	64.87%	
	0.133	200	56.66%	
	0.266	165	46.74%	
	0.533	109	30.88%	
α -MnO ₂	0.308	260	100.00%	[31]
	0.616	207	79.62%	
	1.54	161	61.92%	
	3.08	113	43.46%	
α -MnO ₂	0.016	323	100.00%	[9]
	0.033	273	84.52%	
	0.066	231	71.52%	
	0.133	197	60.99%	
	0.266	163	50.46%	
	0.533	120	37.15%	
Spinel Mn ₃ O ₄	0.2	232	100.00%	[32]
	0.5	195	84.05%	
	1	163	70.26%	
	2	124	53.45%	
β -MnO ₂	0.033	312	100.00%	[11]
	0.066	247	79.17%	
	0.132	193	61.86%	
	0.264	157	50.32%	
	0.528	123	39.42%	
	1.056	86	27.56%	
β -MnO ₂	0.2	258	100.00%	[12]
	0.5	213	82.56%	
	1	188	72.87%	
	2	151	58.53%	
α -Mn ₂ O ₃	0.1	137	100.00%	[13]
	0.2	100	72.99%	
	0.3	86	62.77%	
	0.5	74	54.01%	

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	1	57	41.61%	
	2	38	27.74%	
δ -MnO ₂	0.2	180	100.00%	[6]
	0.4	120	66.67%	
	0.8	75	41.67%	
	1.6	50	27.78%	
α -MnO ₂	0.15	405	100.00%	[14]
	0.3	265	65.43%	
	0.6	166	40.99%	
	1.5	85	20.99%	
	3	40	9.88%	
ZnMn ₂ O ₄	0.05	238	100.00%	[7]
	0.1	232	97.48%	
	0.2	205	86.13%	
	0.5	167	70.17%	
	1	110	46.22%	
	2	75	31.51%	
β -MnO ₂	0.03	285	100.00%	[33]
	0.075	256	89.82%	
	0.15	221	77.54%	
	0.3	193	67.72%	
	0.6	179	62.81%	
	1.2	156	54.74%	
MnO	0.1	267	100.00%	[34]
	0.2	247	92.51%	
	0.4	216	80.90%	
	0.6	172	64.42%	
	0.8	126	47.19%	
	1	95	35.58%	
ZnMn ₂ O ₄	0.1	110	100.00%	[18]
	0.3	105	95.45%	
	0.5	100	90.91%	
	0.8	90	81.82%	
	1	70	63.64%	
	2	50	45.45%	
α -MnO ₂	0.1	350	100.00%	[35]
	0.2	315	90.00%	
	1	230	65.71%	
	2	162	46.29%	
	3	150	42.86%	
T-MnO ₂	0.15	NA	100.00%	[20]
	0.3	NA	92.00%	
	0.9	NA	71.00%	
δ -MnO ₂	0.1	225	100.00%	[21]
	0.2	200	88.89%	
	0.3	180	80.00%	
	0.4	163	72.44%	
	0.5	150	66.67%	
	1	120	53.33%	
δ -MnO ₂	0.012	110	100.00%	[36]
	0.03	98	89.09%	
	0.06	89	80.91%	
	0.15	65	59.09%	
	0.30	30	27.27%	
α -MnO ₂	0.1	242.3	100.00%	[10]

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	0.3	176.4	72.80%	
	0.5	151.5	62.53%	
	1	104.3	43.05%	
	3	58.9	24.31%	
$\text{O}_d\text{-MnO}_2$	0.2	345	100.00%	
	0.5	298	86.38%	[24]
	1	235	68.12%	
	2	170	49.28%	
$\text{Na}_3\text{V}_2(\text{PO}_4)_3$	0.06	65.4	100.00%	
	0.12	60.4	92.35%	
	0.18	56.8	86.85%	[25]
	0.3	52.5	80.28%	
	0.6	45.5	69.57%	
	0.9	39.1	59.79%	
ZnHCF	0.06	65	100.00%	
	0.12	60	92.35%	
	0.18	57	86.85%	[26]
	0.3	53	80.27%	
	0.6	46	69.57%	
	0.9	39	59.79%	
$\text{Na}_{0.33}\text{V}_2\text{O}_5$	0.1	367.1	100.00%	
	0.2	253.7	69.11%	
	0.5	173.4	47.24%	[37]
	0.8	145.3	39.58%	
	1	137.5	37.46%	
	2	96.4	26.26%	
LiV_3O_8	0.016	256	100.00%	
	0.033	230	89.84%	
	0.066	211	82.42%	[27]
	0.133	188	73.44%	
	0.266	148	57.81%	
	0.533	79	30.86%	
$\text{Zn}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$	0.05	200	100.00%	
	0.1	166	83.00%	
	0.3	145	72.50%	[38]
	0.5	122	61.00%	
	0.8	105	52.50%	
	1	84	42.00%	
$\text{K}_2\text{V}_8\text{O}_{21}$	0.3	247	100.00%	
	0.5	226	91.50%	[39]
	1	183	74.09%	
	2	139	56.28%	
NaV_3O_8	0.1	372	100.00%	
	0.2	320	86.02%	
	0.5	277	74.46%	[40]
	1	241.5	64.92%	
	2	205	55.11%	
V_2O_5	0.05	242	100.00%	
	0.1	217	89.67%	
	0.2	192	79.34%	[41]
	0.5	171	70.66%	
	1	156	64.46%	

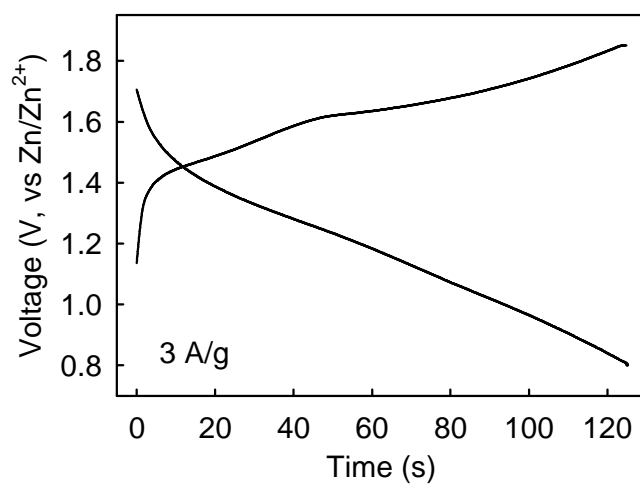


Figure S3. The time-voltage profiles of the fast-charge.

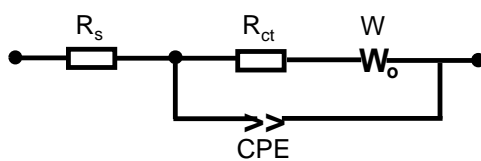


Figure S4. The equivalent circuit model.

Table S4. The Resistance Derived from EIS.

	Resistance/ Ω	Pristine	After 3 cycles
ZnMn ₃ O ₇	R _s	1.355	1.786
	R _{ct}	47.67	45.08

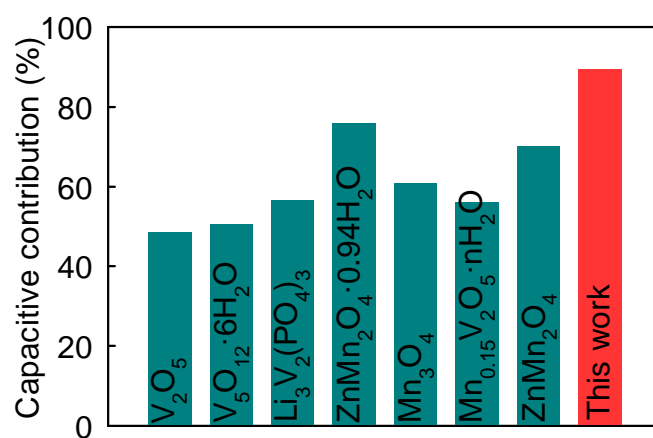


Figure S5. Comparison of capacitive contributions.^[7, 42-47]

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