

**Jahn–Teller Distortion Boosts Holey NiMn-Hydroxide Nanosheets Ultrahigh Areal
Capacity and Cycling Robustness for Flexible Energy Storage Devices**

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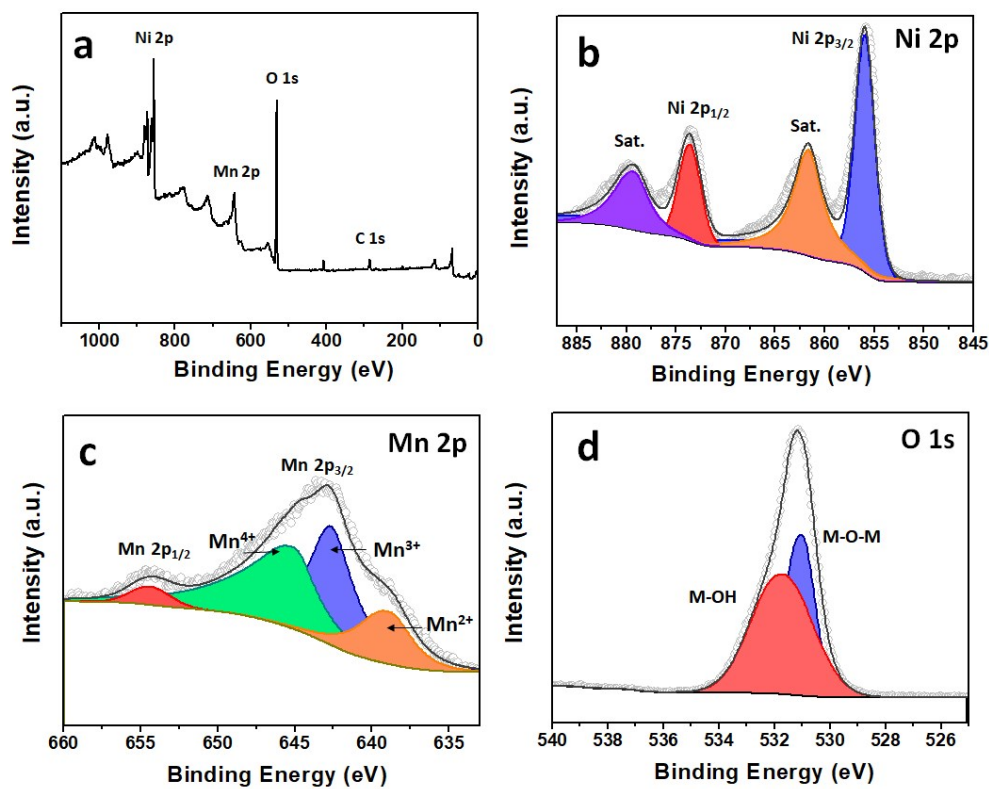


Fig. S1 (a) XPS surface survey. (b) Ni 2p, (c) Mn 2p, and (d) O 1s core-level XPS spectra of NiMn-OH-50%.

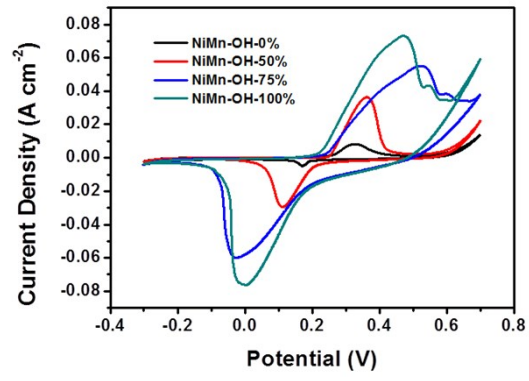


Fig. S2 CV curves of NiMn-OH-0%, NiMn-OH-50%, NiMn-OH-75%, and NiMn-OH-100%.

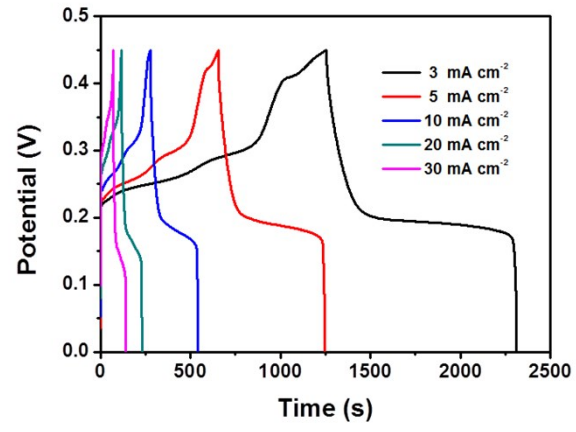


Fig. S3 GCD curves for NiMn-OH-100% at different currents.

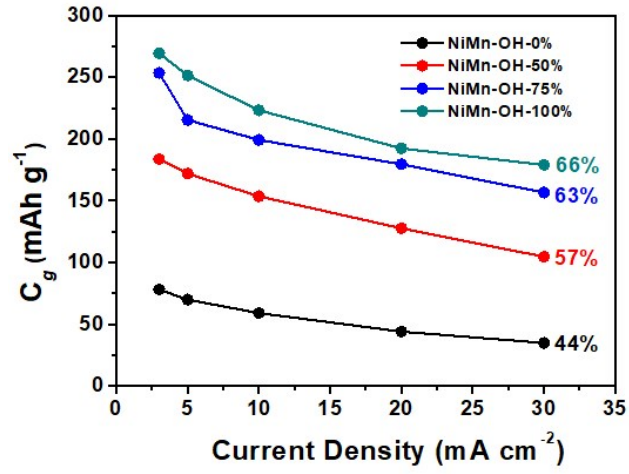


Fig. S4 The capacities of NiMn-OH-0%, NiMn-OH-50%, NiMn-OH-75%, and NiMn-OH-100% at different current densities.

Table S1 Performance comparison of our electrode and others reported in literatures.

Electroactive materials	Areal capacity ($\mu\text{Ah cm}^{-2}$)	Mass capacity (mAh g^{-1})	Current density	Rate capability	Reference
NiMn-OH	881	269	3 mA cm^{-2} (0.92 A g^{-1}) ¹⁾	66% (30 mA cm^{-2})	This work
CoNiO ₂ @Ni(OH) ₂	674	539	1 mA cm^{-2}	70% (10 mA cm^{-2})	1
H-CoO _x @Ni(OH) ₂	820.4	335.5	5.2 mA cm^{-2} (2 A g^{-1})	68% (50 A g^{-1})	2
Ni ₃ S ₂ @Ni(OH) ₂	588.2	158.5	5.1 A g^{-1}	38% (19.8 A g^{-1})	3
Cu-Doped NiCo-LDH/CuO	310	193.3	1.5 mA cm^{-2}	79% (40 mA cm^{-2})	4
Cu ₃ Mo ₂ O ₉ NCAs	449.5	112.3	2 mA cm^{-2}	44% (30 mA cm^{-2})	5
CuCo ₂ O ₄ @CoMoO ₄	500	-	2 mA cm^{-2}	66.7% (80 mA cm^{-2})	6
CuCo ₂ O ₄ @Ni(OH) ₂	439	-	2 mA cm^{-2}	82.7% (80 mA cm^{-2})	7
Ni _{0.54} Co _{0.46} O ₂ nanosheet	438	238	1 mA cm^{-2}	90.3% (80 mA cm^{-2})	8
Ni _{0.54} Co _{0.46} O ₂ nanowire	394.1	218.9	1 mA cm^{-2}	55% (40 mA cm^{-2})	8
NiCo ₂ S ₄ @Ni(OH) ₂	680	240.3	5 mA cm^{-2}	94.9% (100 mA cm^{-2})	9
NiS	410	177.1	2.3 mA cm^{-2}	49% (10 A g^{-1})	10

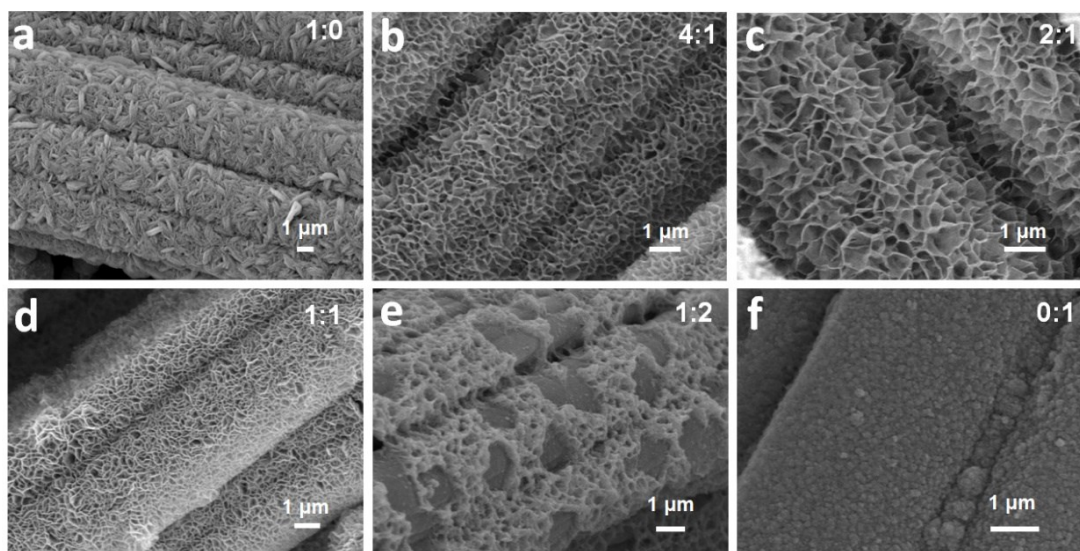


Fig. S5 SEM images of the products from different Ni and Mn feeding mole ratios: (a) 1:0, (b) 4:1, (c) 2:1, (d) 1:1, (e) 1:2, (f) 0:1.

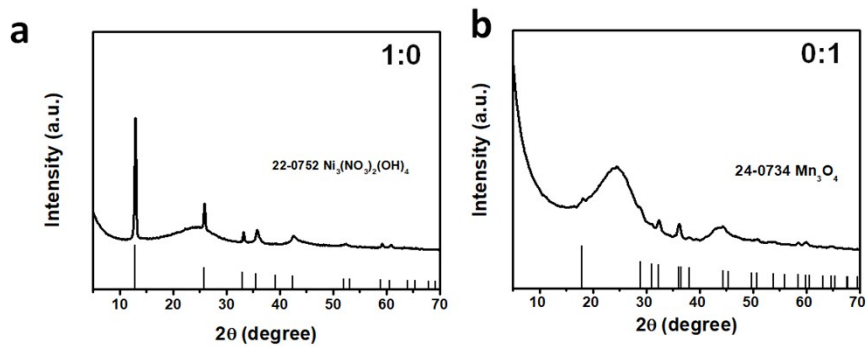


Fig. S6 XRD pattern of the products from Ni and Mn feeding ratio of (a) 1:0 and (b) 0:1.

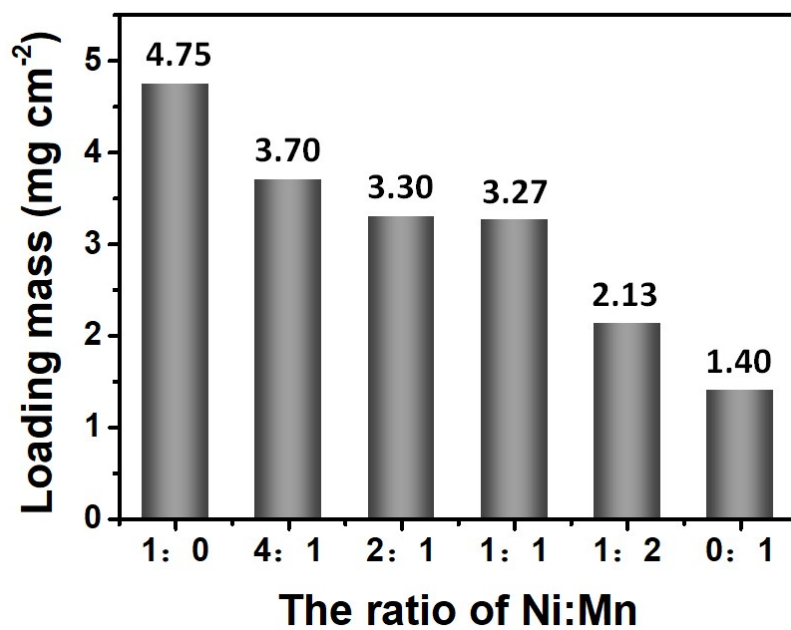


Fig. S7 The mass loading of solvothermal products as a function of different Ni:Mn ratio.

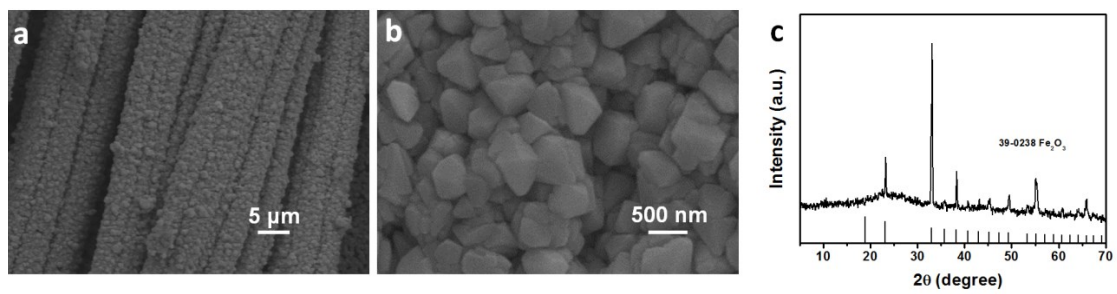


Fig. S8 (a, b) SEM images and (c) XRD pattern of $\text{Fe}_2\text{O}_3/\text{CC}$.

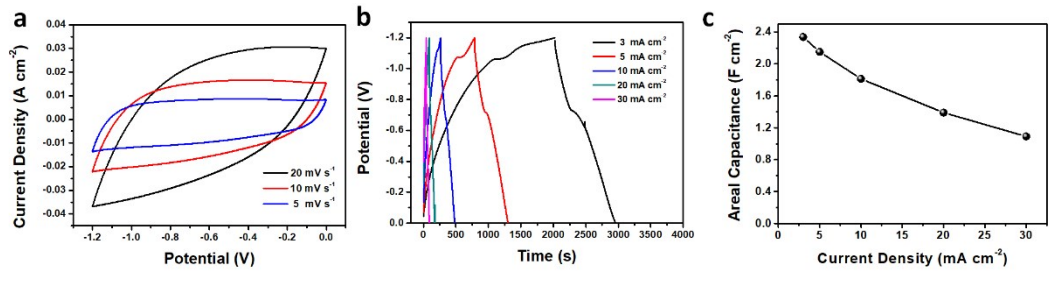


Fig. S9 (a) CV and (b) GCD curves of $\text{Fe}_2\text{O}_3/\text{CC}$ electrode. (c) The areal capacitance of $\text{Fe}_2\text{O}_3/\text{CC}$ electrode at different current densities.

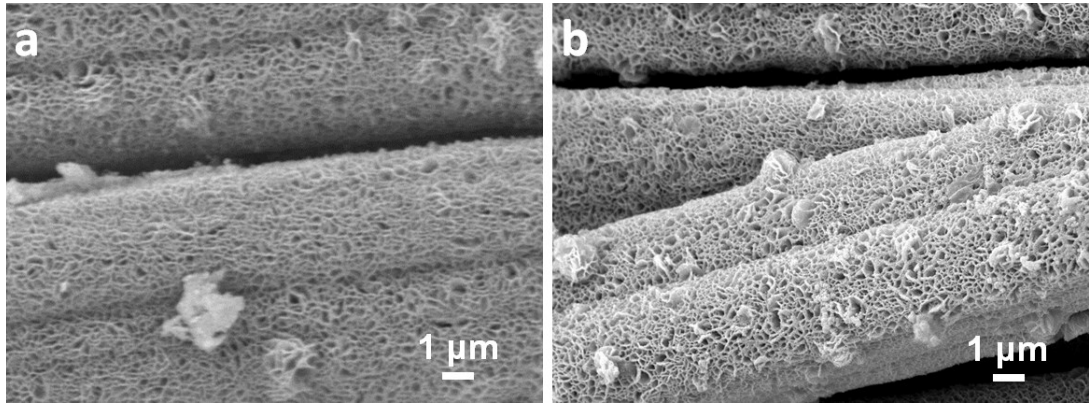


Fig. S10 SEM images of NiMn-OH-100% electrode (a) before and (b) after 10000 GCD cycles.

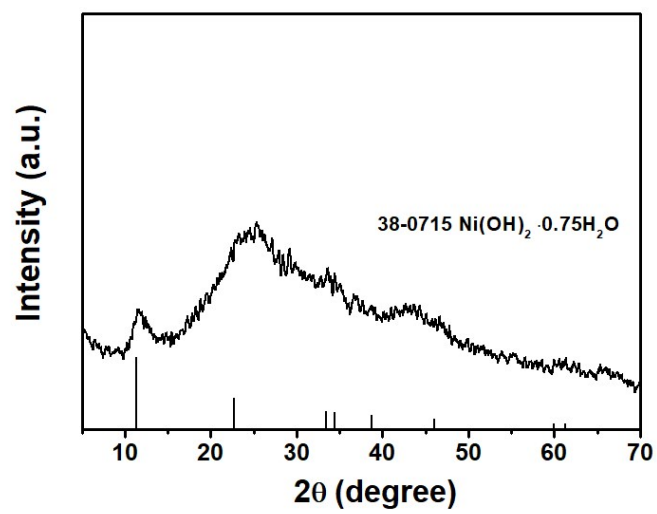


Fig. S11 XRD pattern of NiMn-OH-100% electrode after 10000 GCD cycles.

Reference

- 1 Q. Li, Q. Zhang, J. Sun, C. Liu, J. Guo, B. He, Z. Zhou, P. Man, C. Li, L. Xie and Y. Yao, *Adv. Sci.*, 2019, **6**, 1801379.
- 2 J. Zhu, L. Huang, Y. Xiao, L. Shen, Q. Chen and W. Shi, *Nanoscale*, 2014, **6**, 6772-6781.
- 3 W. Zhou, X. Cao, Z. Zeng, W. Shi, Y. Zhu, Q. Yan, H. Liu, J. Wang and H. Zhang, *Energy Environ. Sci.*, 2013, **6**, 2216-2221.
- 4 Y. Guo, X. Hong, Y. Wang, Q. Li, J. Meng, R. Dai, X. Liu, L. He and L. Mai, *Adv. Funct. Mater.*, 2019, **29**, 1809004.
- 5 S. M. Cha, S. Chandra Sekhar, R. Bhimanaboina and J. S. Yu, *Inorg. chem.*, 2018, **57**, 8440-8450.
- 6 J. Xie, Z. Zhan, S. Zhang, G. Li, H. Xia, Y. Yang and J. Xiong, *Mate. Lett.*, 2018, **226**, 30-33.
- 7 Z. Zhan, S. Chen, J. Xie, Y. Yang and J. Xiong, *J. Alloy. Compd.*, 2017, **722**, 928-937.
- 8 Y. Jiang, L. Zhang, H. Zhang, C. Zhang and S. Liu, *J. Power Sources*, 2016, **329**, 473-483.
- 9 Y. Yang, D. Cheng, S. Chen, Y. Guan and J. Xiong, *Electrochim. Acta*, 2016, **193**, 116-127.
- 10 J. Xu, L. Zhang, G. Xu, Z. Sun, C. Zhang, X. Ma, C. Qi, L. Zhang and D. Jia, *Appl. Surf. Sci.*, 2018, **434**, 112-119.