Supporting Information

Metallic State Two-dimensional Holey-Structured Co_3FeN Nanosheets as Stable and Bifunctional Electrocatalysts for Zinc-Air Batteries

Hai-Peng Guo, Xuan-Wen Gao, Neng-Fei Yu, Zhi Zheng, Wen-Bin Luo*, Chang Wu, Hua-Kun Liu, Jia-Zhao Wang*

H. P. Guo, Dr. N. F. Yu, Z. Zheng, C. Wu, Prof. H. K. Liu, Prof. J. Z. Wang
Institute for Superconducting and Electronic Materials, University of Wollongong,
Squires Way, Fairy Meadow, NSW 2500, Australia
E-mail: wl368@uowmail.edu.au; jiazhao@uow.edu.au

A/Prof. X. W. Gao, Prof. W. B. Luo
School of Metallurgy, Northeastern University, Shenyang, Liaoning, 110819, China.
E-mail:luowenbin@smm.neu.edu.cn

Dr. N. F. Yu
College of Energy Science and Engineering, Nanjing Tech University, Nanjing, 211800, China

Keywords: two dimensional, nitride electrocatalyst, oxygen evolution reaction, oxygen reduction reaction, zinc-air battery

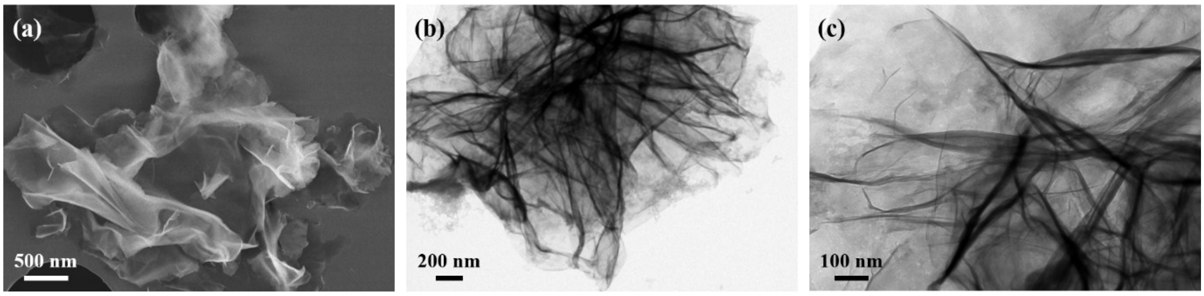

Figure S1. (a) SEM image of Co_3Fe LDH nanosheets. (b) low- and (c) high-magnification TEM images of Co_3Fe LDH nanosheets.

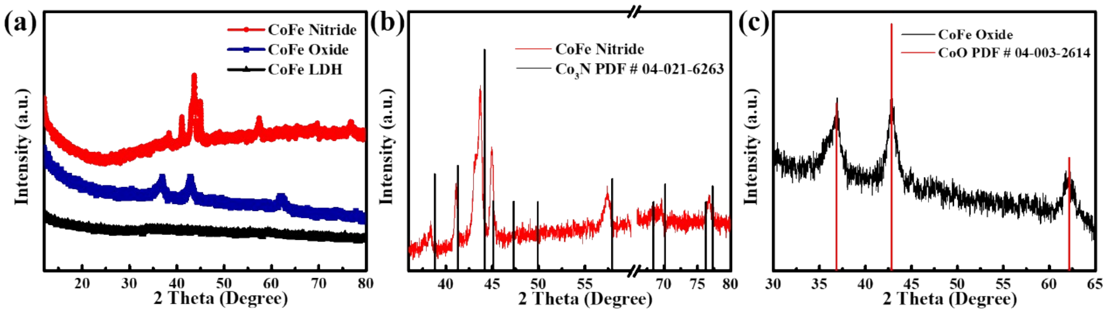

Figure S2. (a) XRD patterns of Co_3FeN, Co_3Fe oxide, and Co_3Fe LDH, (b) comparison of the XRD patterns of Co_3FeN and Co_3N, (c) comparison of the XRD patterns of Co_3Fe oxide and CoO.

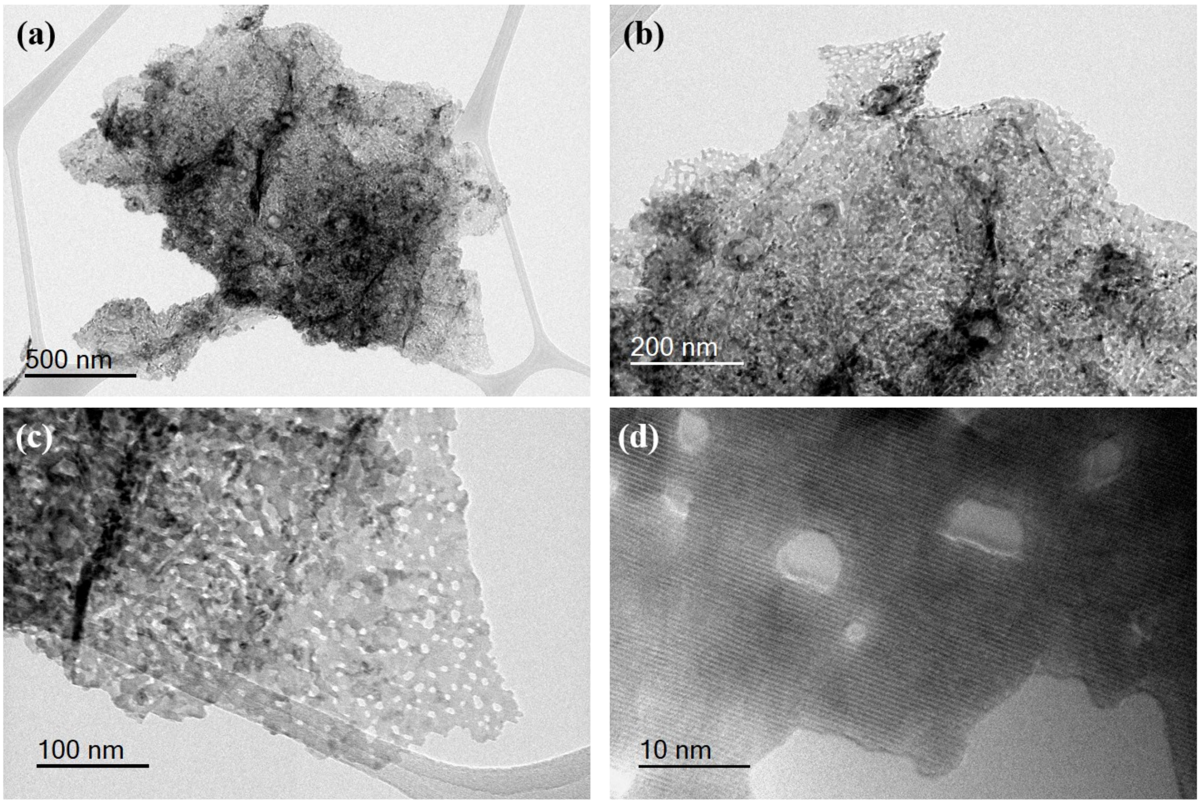

Figure S3. HRTEM images of the 2D holey wrinkled (a, b) Co_3Fe oxide nanosheets; (c, d) Co_3FeN nanosheets.

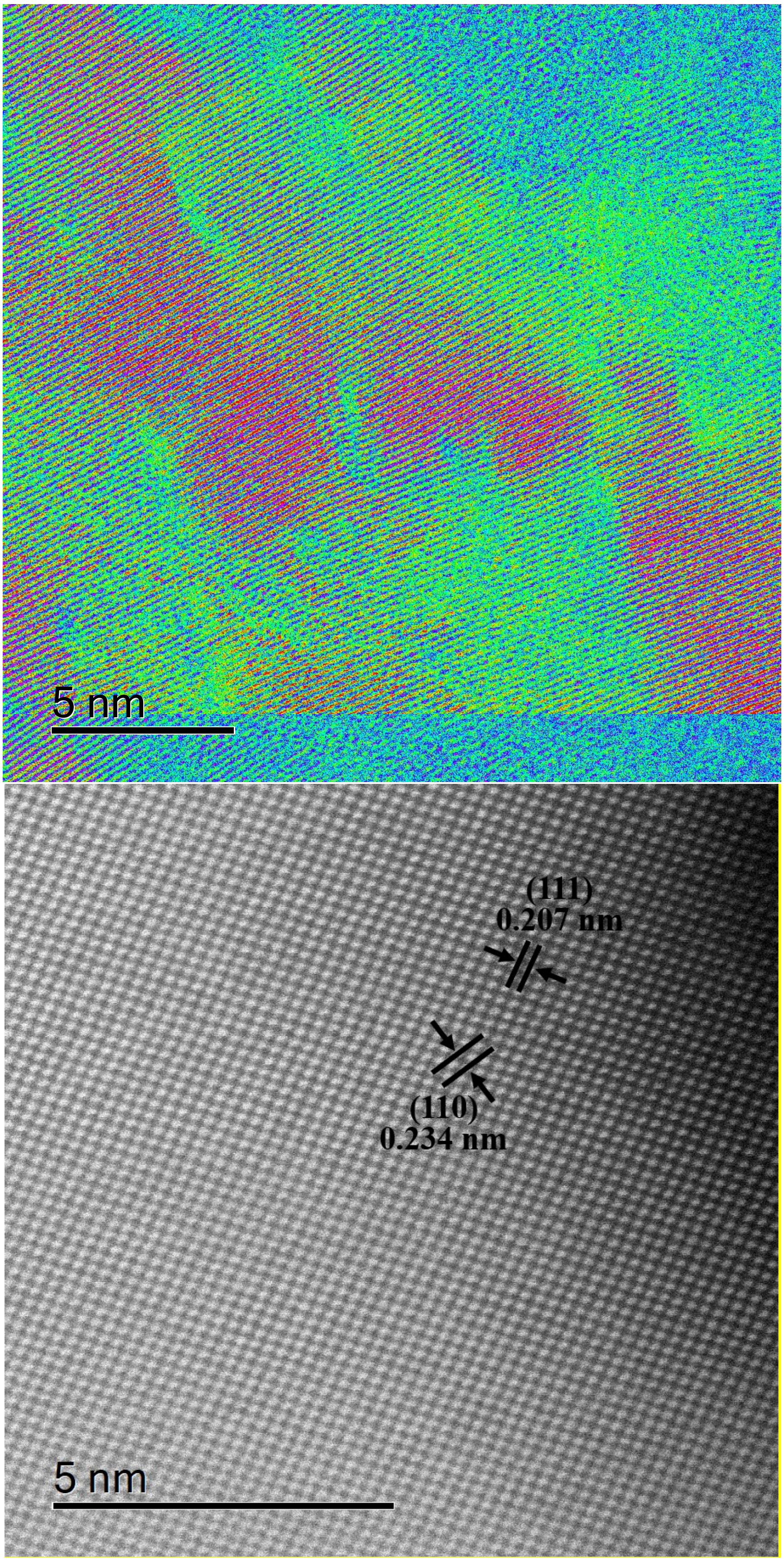

Figure S4. STEM images of 2D holey wrinkled Co_3FeN nanosheets.

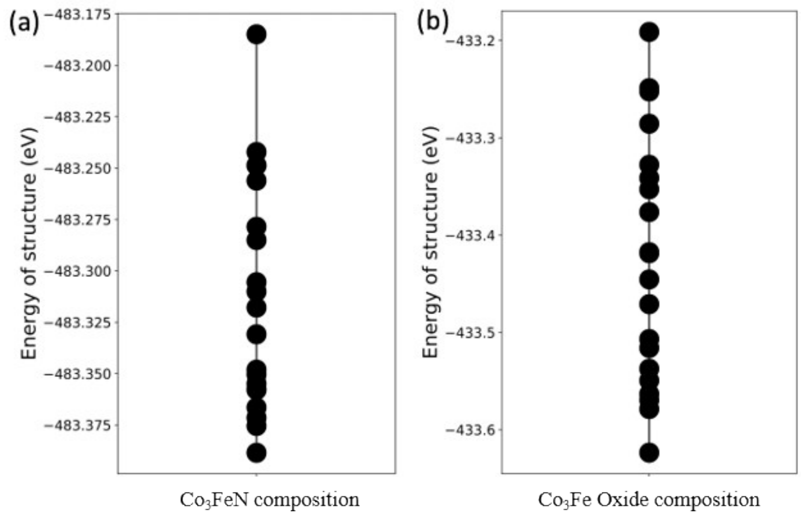

Figure S5. Energy distribution for different structures of (a) Co_3FeN and (b) Co_3Fe oxide.

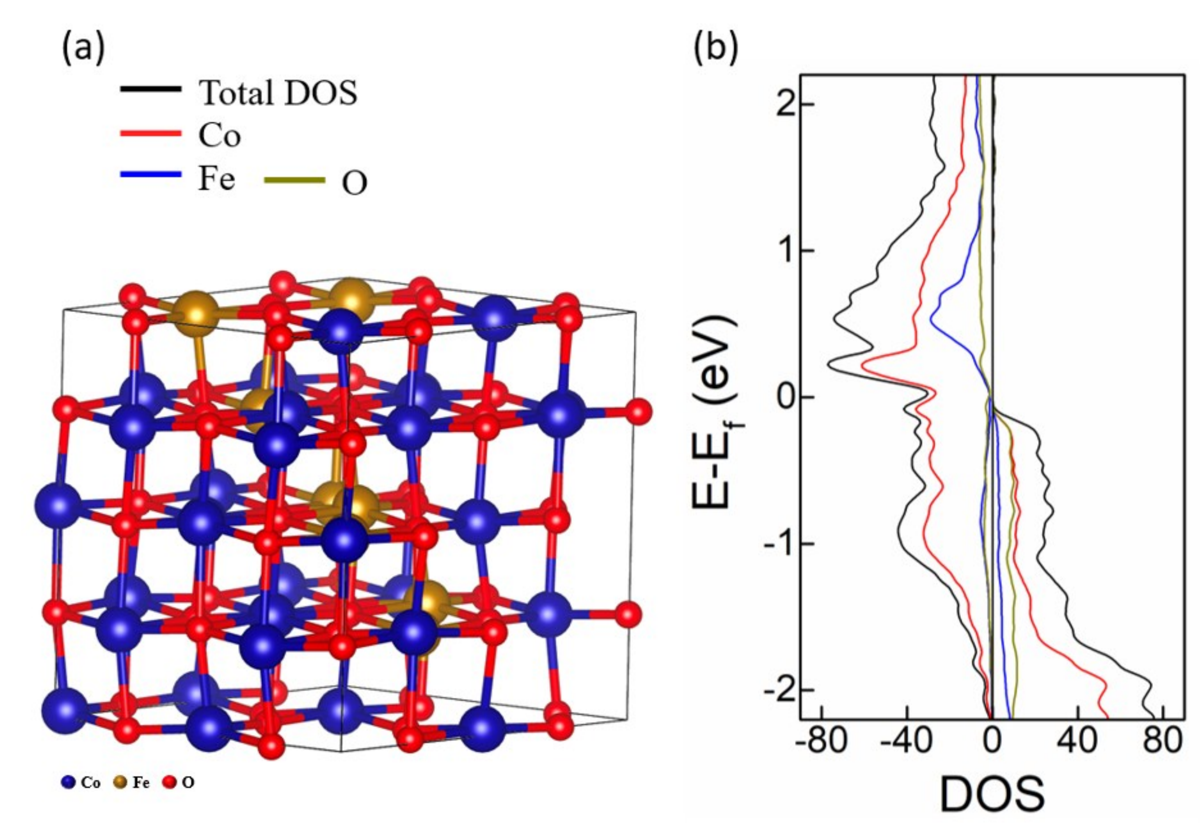

Figure S6. Structure (a) and calculated density of states (b) for Co_3Fe oxide nanosheets.

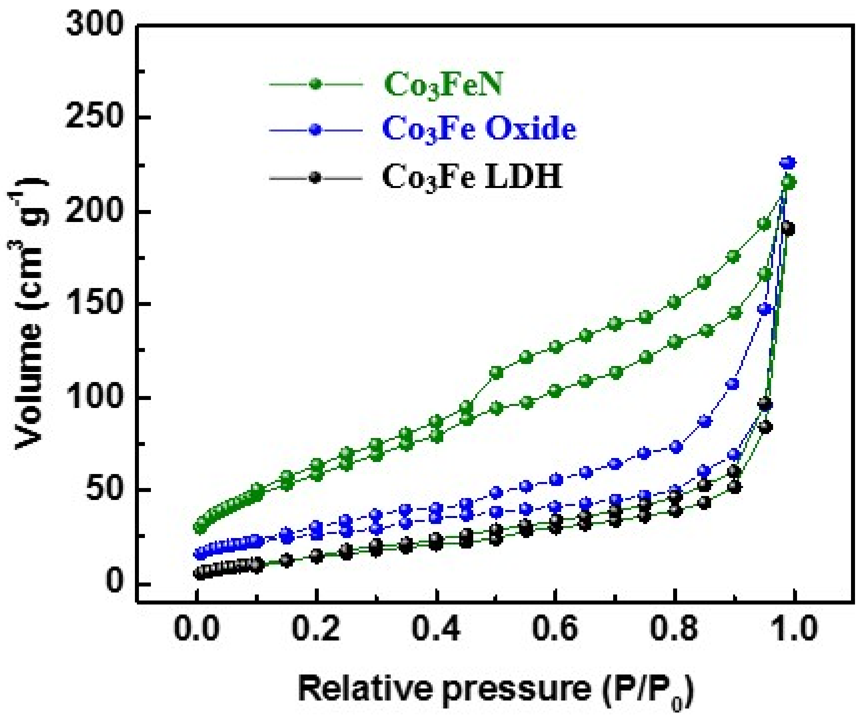

Figure S7. Brunauer-Emmett-Teller (BET) of Co_3FeN, Co_3Fe oxide and Co_3Fe LDH.

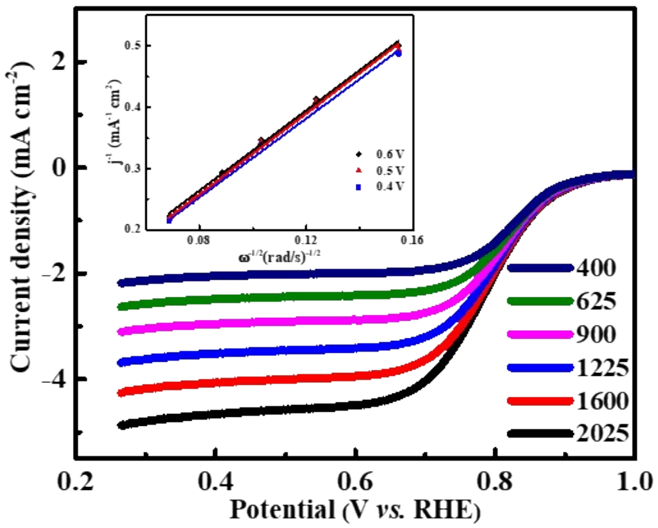

Figure S8. ORR polarization plots for 2D holey wrinkled Co_3FeN nanosheets at various speeds of rotation, the inset shows the K-L plots at different potentials.

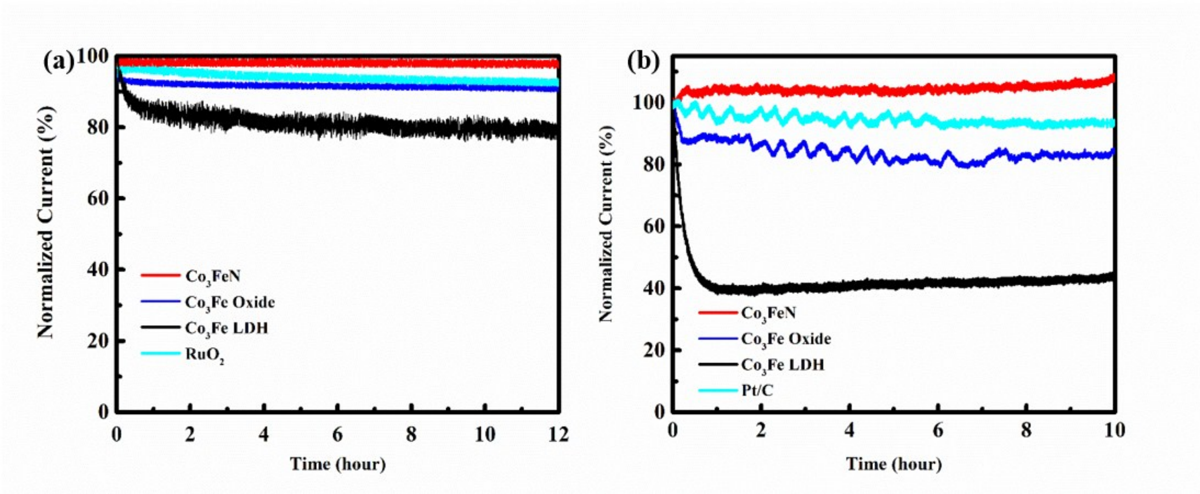

Figure S9. (a) Current-time chronoamperometric responses of RuO_2 and different samples for OER at 1.7 V versus RHE; (b) Current-time chronoamperometric responses of Pt/C and different samples for ORR at 0.75 V versus RHE, in 0.1 M KOH, at a rotation speed of 1600 rpm.

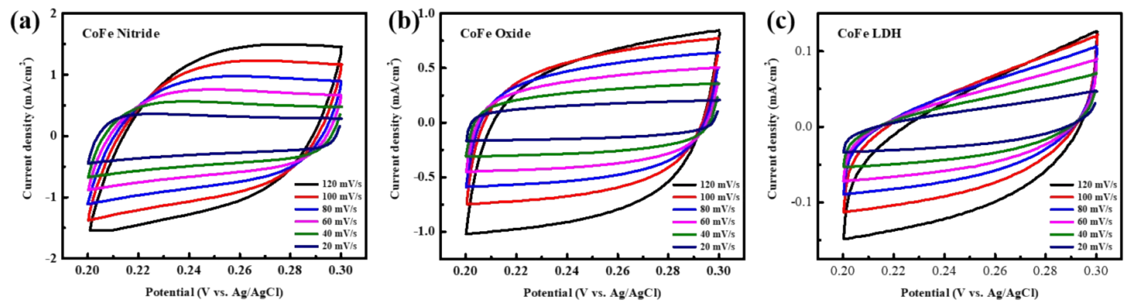

Figure S10. Typical cyclic voltammetry curves of (a) Co_3FeN, (b) Co_3Fe oxide, and (c) Co_3Fe LDH in 0.1M KOH with different scan rates.

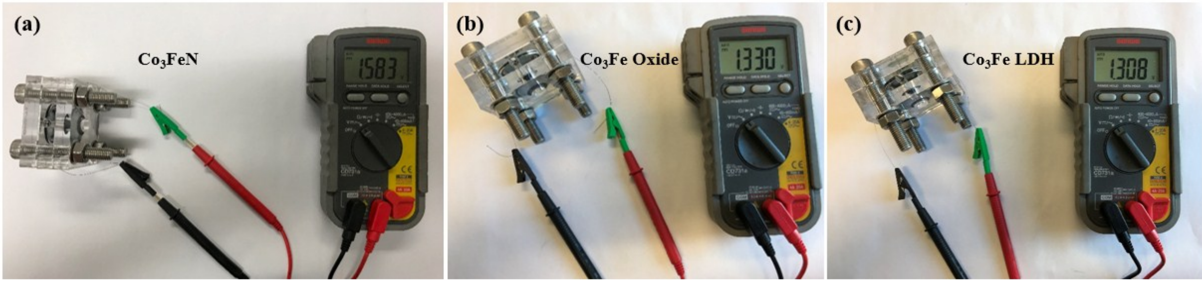

Figure S11. Photograph of the zinc-air battery with 2D holey wrinkled Co_3FeN, Co_3Fe oxide and Co_3Fe LDH as the air electrode catalyst, showing an open-circuit potential.

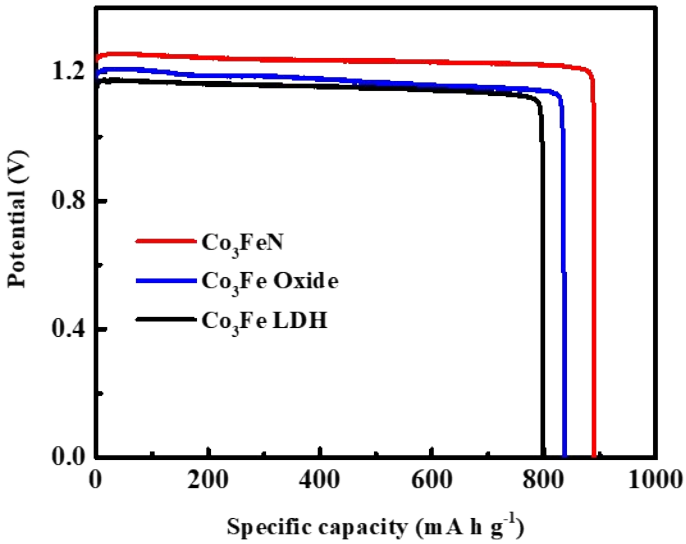

Figure S12. Specific capacities for zinc-air battery with different samples normalized to the consumed mass of Zn at current density of 5 mA cm^{-2}.

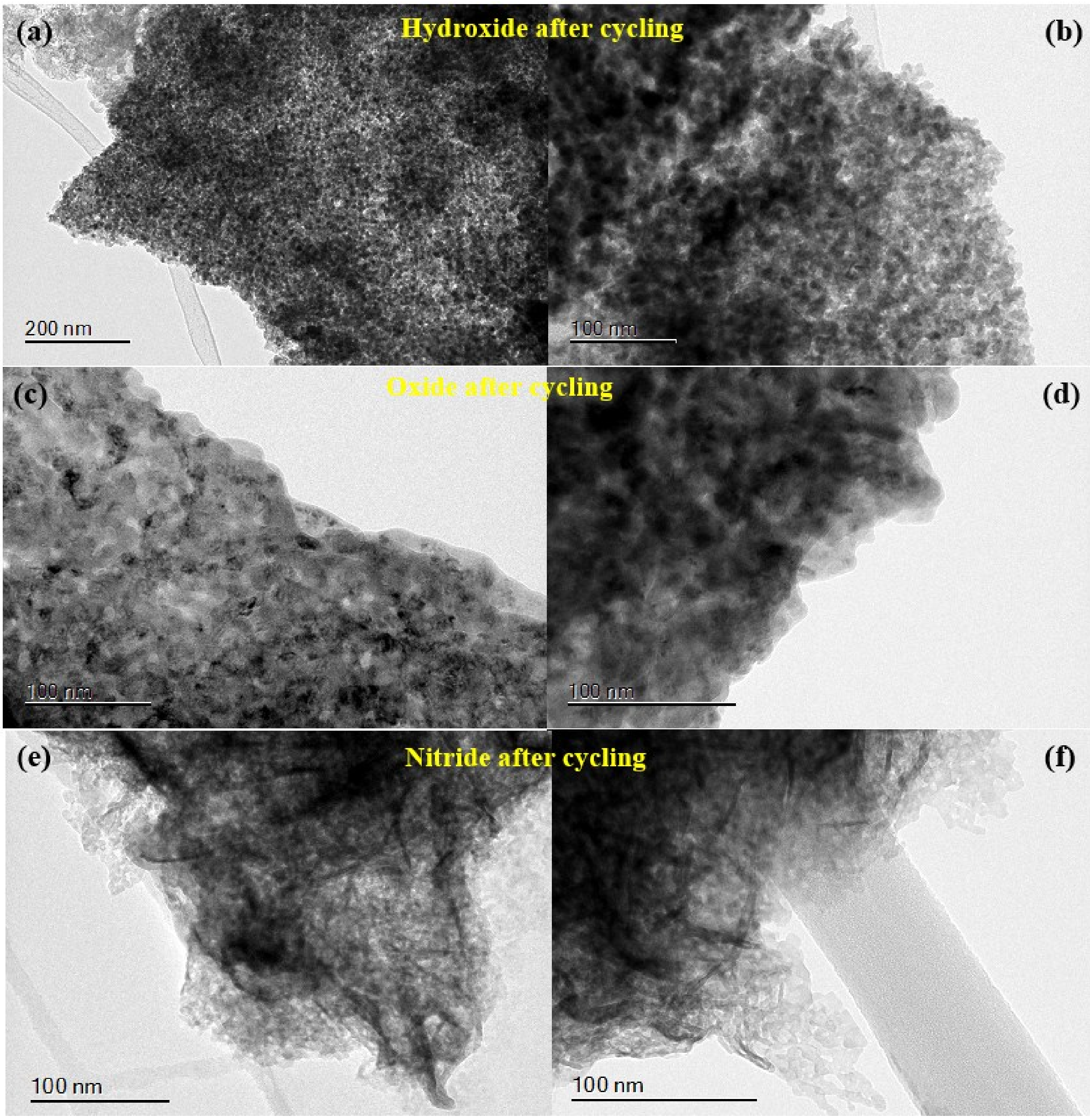

Figure S13. HR-TEM images of the 2D holey wrinkled (a, b) Co_3Fe LDH; (c-d) Co_3Fe oxide; (e, f) Co_3FeN nanosheets after cycling.

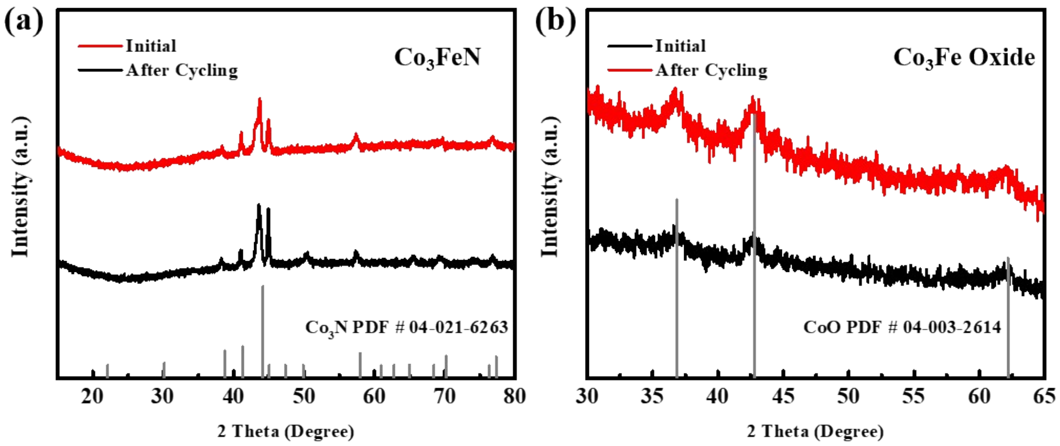

Figure S14. XRD of (a) 2D holey wrinkled Co_3FeN nanosheets and (b) Co_3Fe oxide before and after cycling.

Table S1. *Comparison of the OER, ORR, and Zn-air batteries performance with other electrocatalysts.*

Materials	*OER activity*	*ORR activity*	*Zn-air batteries performance*	*Cycling performance*	*References*
2D holey wrinkled Co_3FeN nanosheets	*Overpotential: 0.42 V @ 0.1 M KOH*	*Onset potential : 0.89 V @ 0.1 M KOH*	*0.90 V and decreased to 0.83 V after 83 hours @ j=5 mA cm^{-2}*	*150 hours (900 cycles) Specific capacity: 890 mA h g^{-1}*	
2D holey wrinkled Co_3Fe oxide nanosheets	*Overpotential:0.52 V @ 0.1 M KOH*	*Onset potential :0.81 V @ 0.1 M KOH*	*0.96 V @ j=5 mA cm^{-2}*	*100 hours (600 cycles) Specific capacity: 836 mA h g^{-1}*	*This work*
2D wrinkled Co_3Fe LDH nanosheets	*Overpotential:0.58 V @ 0.1 M KOH*	*Onset potential :0.82 V @ 0.1 M KOH*	*1.17 V @ j=5 mA cm^{-2}*	*47 hours (280 cycles) Specific capacity: 798 mA h g^{-1}*	
Co_3O_4 nanosheets@N-rGO	*Overpotential: 0.49 V @ 0.1 M KOH*	*Onset potential : 0.90 V @ 0.1 M KOH*	*0.80 V @ j=3 mA cm^{-2}*	*25 hours (75 cycles) Specific capacity: 550 mA h g^{-1}*	*[1]*
N-doped Co_3O_4	*~*	*Onset potential : 0.94 V @ 1 M KOH*	*0.30 V @ j=12.5 mA cm^{-2}*	*28 hours (21 cycles) Specific capacity: 603.7 mA h g^{-1}*	*[2]*
NCNT/$Co_xMn_{1-x}O$	*Overpotential: 0.34 V @ 1M KOH*	*Onset potential : 0.96 V @ 1 M KOH*	*1.18 V @ j=7 mA cm^{-2}*	*12 hours Specific capacity: 581 mA h g^{-1}*	*[3]*
Ni3FeN microspheres	*Overpotential: 0.355 V @ 0.1 M KOH*	*Half-wave potential: 0.78 V @ 0.1 M KOH*	*0.7 V @ j=10 mA cm^{-2}*	*100 hours (310 cycles)*	*[4]*
FeNi3N/N-doped Graphene	*Overpotential: 0.41 V @ 0.1 M KOH*	*Onset potential : 0.88 V @ 0.1 M KOH*	*0.78 V @ j=10 mA cm^{-2}*	*140 hours (840 cycles) Specific capacity: 785.2 mA h g^{-1}*	*[5]*
MnO/Co/PGC	*Overpotential: 0.307 V @ 1 M KOH*	*Onset potential : 0.95 V @ 0.1 M KOH*	*~0.75 V @ j=10 mA cm^{-2}*	*350 cycles Specific capacity: 873 mA h g^{-1}*	*[6]*

Meso/micro-FeCo-N$_x$-CN	*Overpotential: 0.37 V @ 1 M KOH*	*Onset potential : 0.954 V @ 0.1 M KOH*	*0.75 V @ j=5 mA cm^{-2}*	*44 hours (22 cycles)*	*[7]*
Co_9S_8@NSCM	*Overpotential: 0.37 V @ 0.1 M KOH*	*Onset potential : 0.97 V @ 0.1 M KOH*	*0.81 V @ j=10 mA cm^{-2}*	*140 hours (840 cycles) Specific capacity: 810 mA h g^{-1}*	*[8]*
$CaMnO_3$ Nanotubes	*Overpotential: 0.47 V @ 0.1 M KOH*	*Onset potential : 0.915 V @ 0.1 M KOH*	*0.68 V @ j=10 mA cm^{-2}*	*~ 13 hours (120 cycles)*	*[9]*

Supplementary References:

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