

Supporting Information

Configuration of Gradient-Porous Ultrathin FeCo₂S₄ Nanosheets Vertical Aligned on Ni Foam as Noncarbonaceous Freestanding Oxygen Electrode for Lithium-Oxygen Batteries

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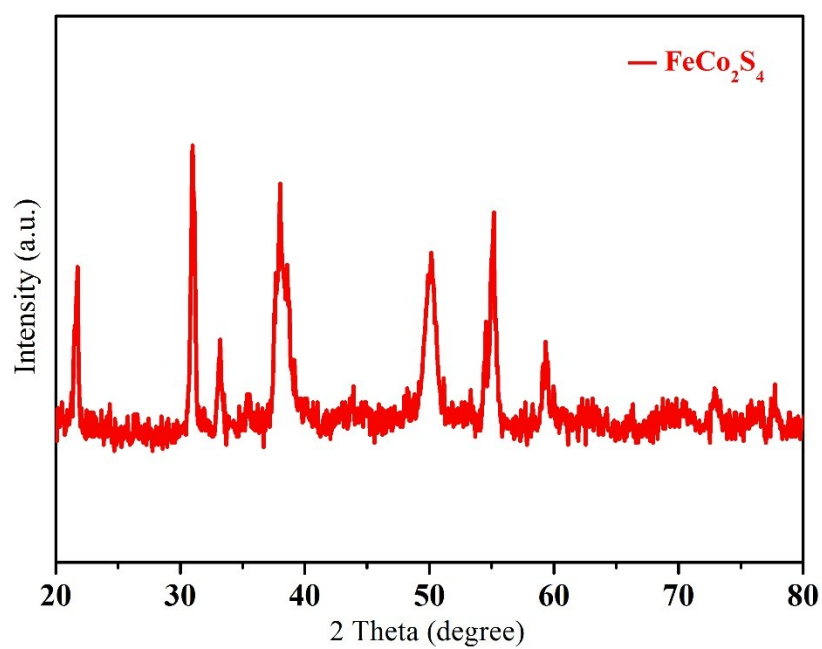


Fig. S1 XRD pattern of FeCo₂S₄ powder.

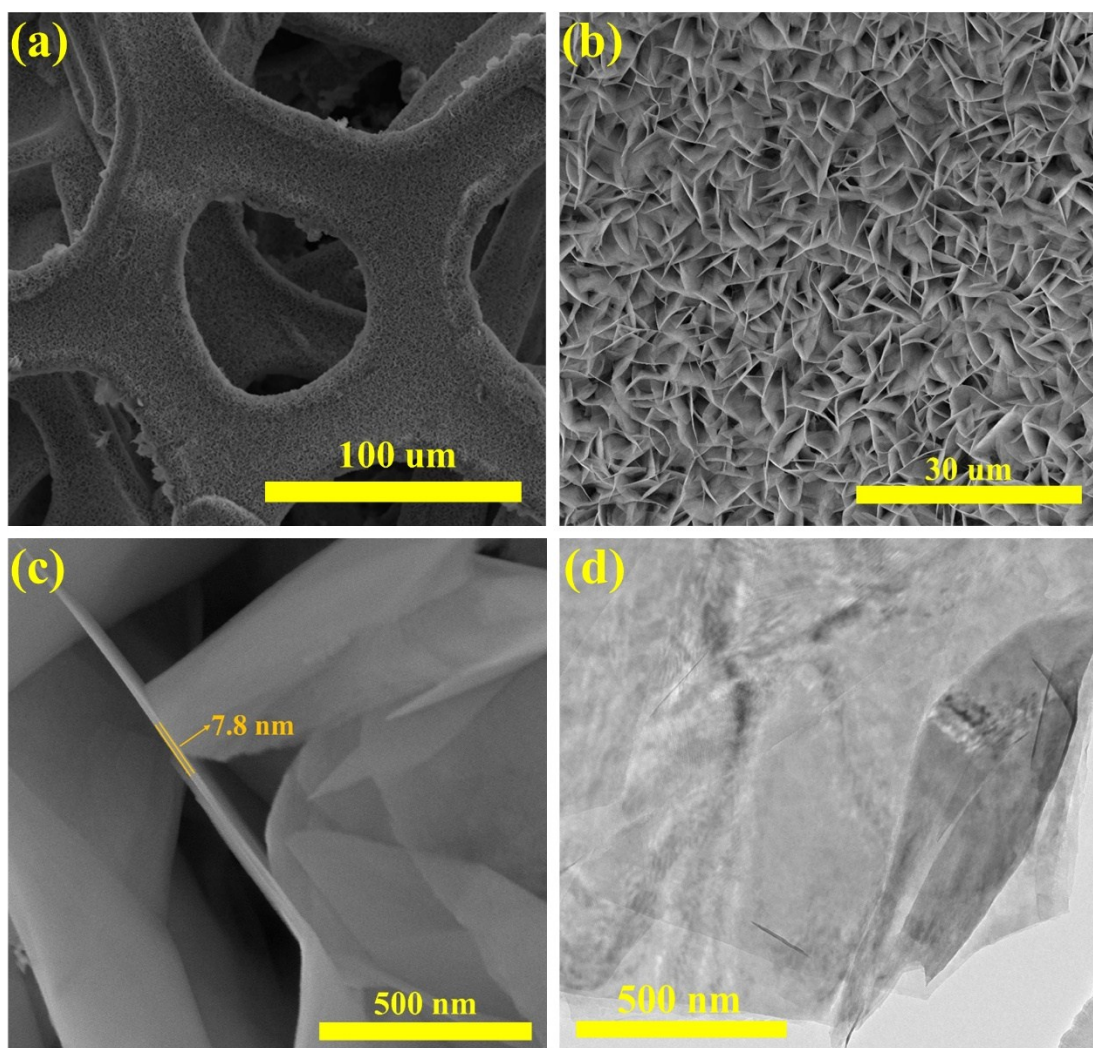


Fig. S2 (a-c) FESEM of $\text{FeCo}_2\text{S}_4@\text{Ni}$ with different resolution, (d) TEM image of FeCo_2S_4 nanosheet.

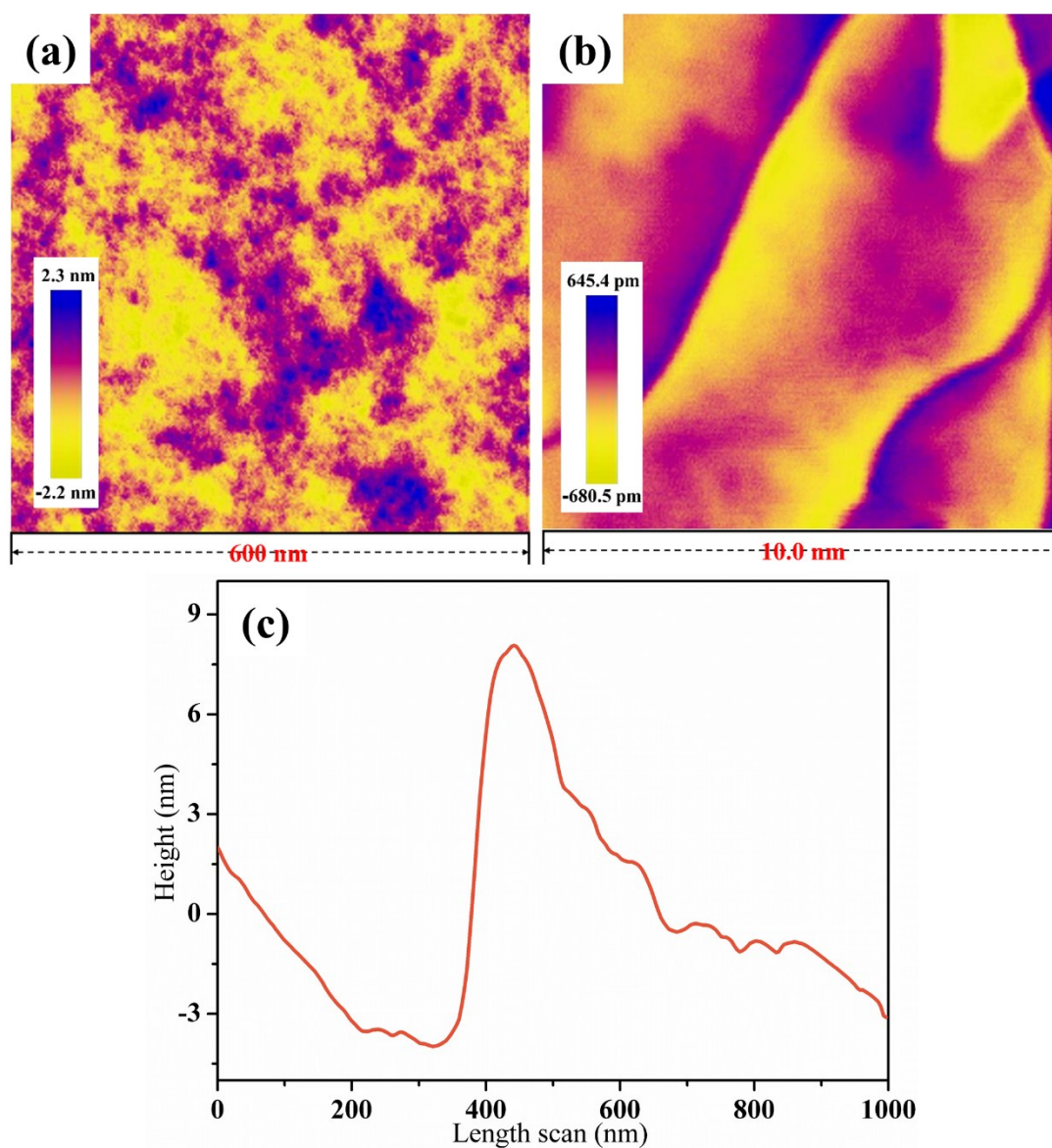


Fig. S3 (a-b) Typical 2D AFM images of FeCo_2S_4 nanosheets; (c) Line scanning of a diluted sample to measure the thickness of the sheets.

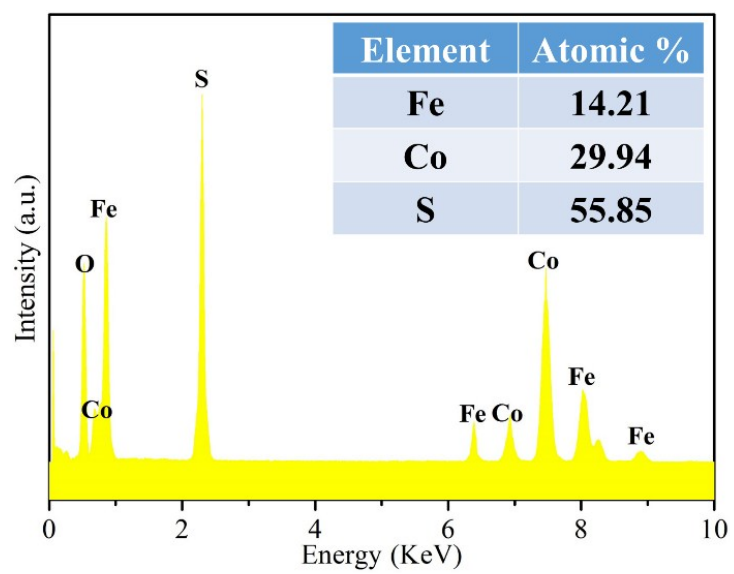


Fig. S4 EDS spectrum of FeCo₂S₄@Ni.

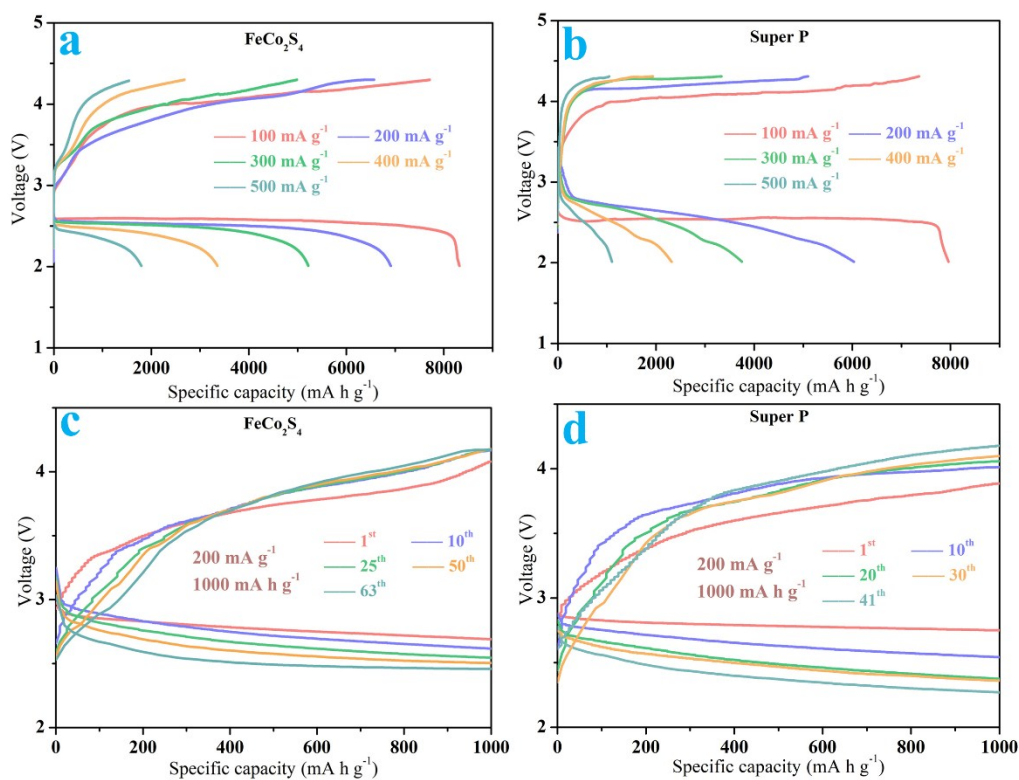


Fig. S5 (a) FeCo₂S₄ powder and (b) Super P based Li-O₂ cells at various current densities (100, 200, 300, 400, 500 mA g⁻¹); (c) and (d) FeCo₂S₄ powder, and Super P electrodes at a current density of 200 mA g⁻¹ and with a restricted capacity of 1000 mA h g⁻¹.

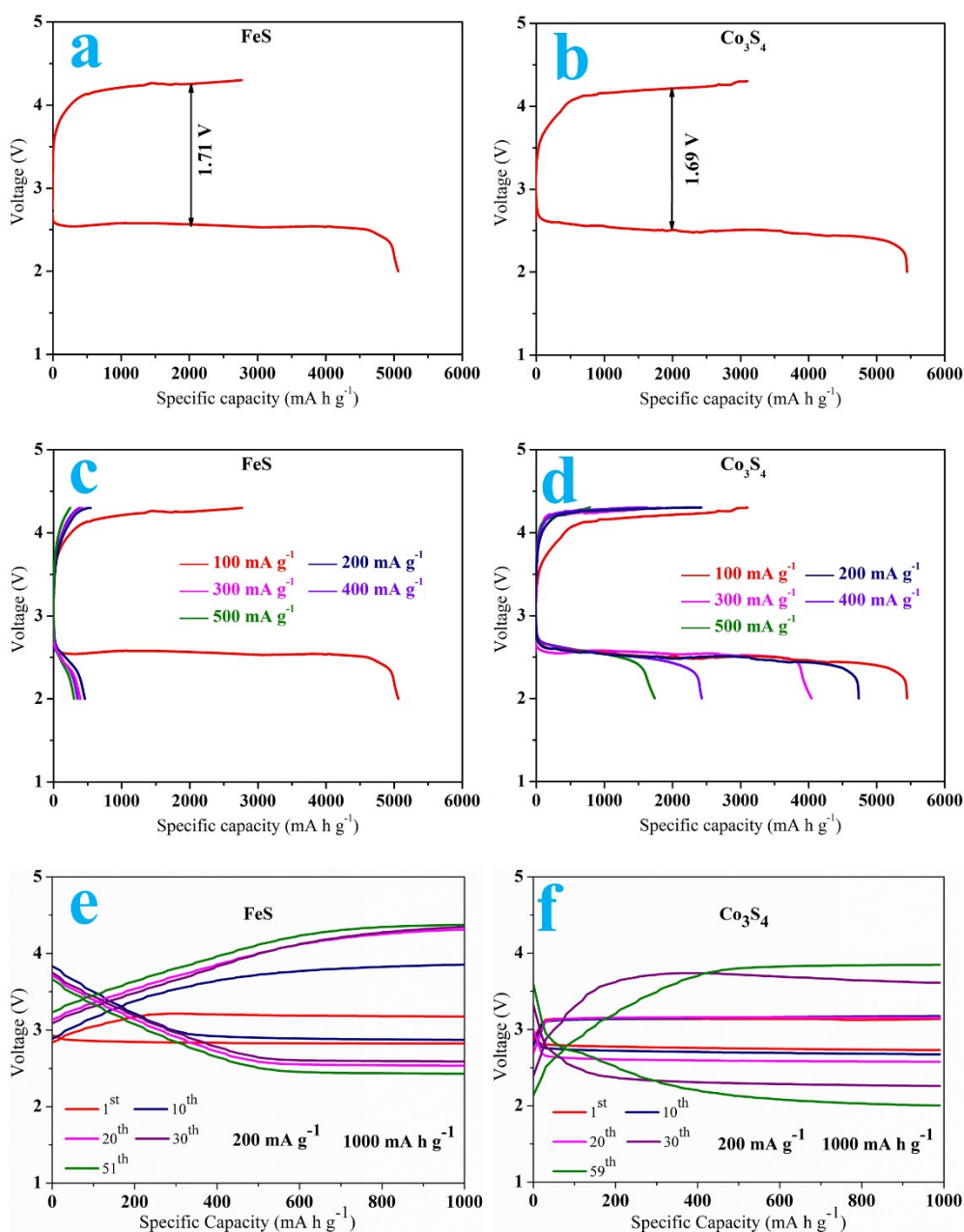


Fig. S6 (a) and (b) the fully discharge/charge profiles of Li-O₂ cells based on FeS powder, Co₃S₄ powder electrodes at a current density of 100 mA g⁻¹; (c) and (d) FeS powder, and Co₃S₄ powder based Li-O₂ cells at various current densities (100, 200, 300, 400, 500 mA g⁻¹); (e) and (f) FeS powder, and Co₃S₄ powder electrodes at a current density of 200 mA g⁻¹ and with a restricted capacity of 1000 mA h g⁻¹.

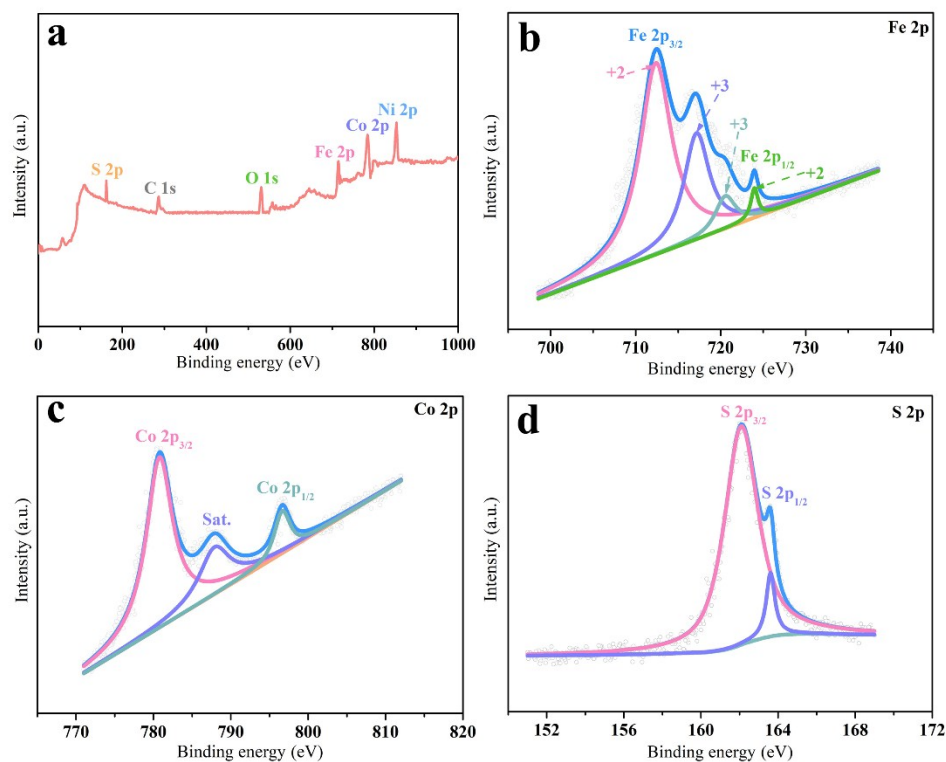


Fig. S7 XPS of $\text{FeCo}_2\text{S}_4@\text{Ni}$ after 10th recharge: (a) survey spectrum, (b) Fe 2p, (c) Co 2p, (d) S 2p spectrum.

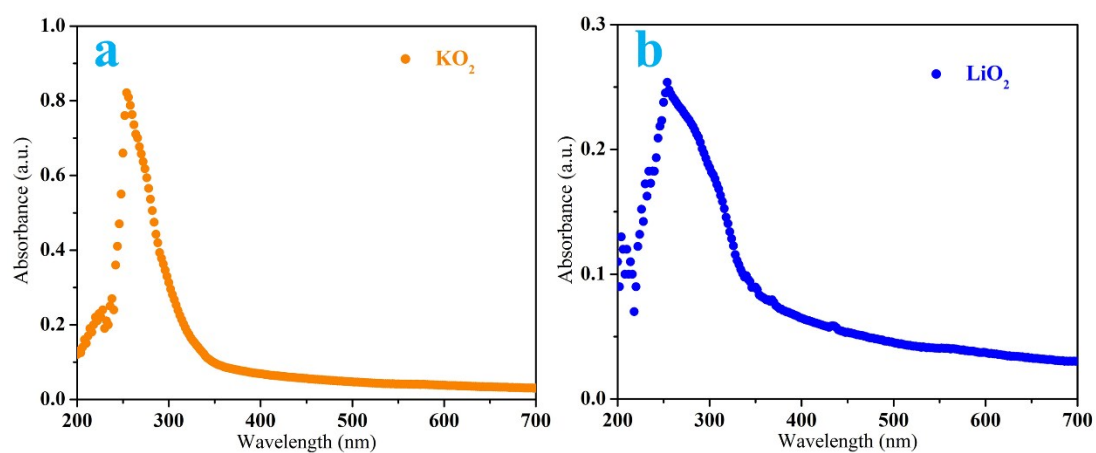


Fig. S8 Ultraviolet-visible spectrum of (a) KO_2 dissolved in DMSO solution and DMSO-extracted superoxide products from the discharged $\text{FeCo}_2\text{S}_4@\text{Ni}$ -cathode based Li-O_2 battery after 10 hours (b).

Table S1. Recent reported results of different types of oxygen electrode electrochemical performance.

Material	Current density	Overpotential (V)	Cycling Performance (Cycles/Limited capacity)	First Discharge Capacity (mA h g ⁻¹)	Ref
MnCo ₂ S ₄ nanosheets	200 mA g ⁻¹	0.82	100/500	10760	S1
CoFe ₂ O ₄ @MWCNTs	0.2 mA cm ⁻²	0.85	15/No limited capacity	1245	S2
ZnCo ₂ O ₄ /S WCNT	0.1 mA cm ⁻²	0.54	50/800	2750	S3
ZnCo ₂ O ₄ microspheres	100 mA g ⁻¹	1.15	35/1000	6024	S4
NiFe ₂ O ₄	100 mA	—	100/1000	~980	S5
NiCo ₂ S ₄	100 mA g ⁻¹	—	144/500	3977	S6
FeCo ₂ S ₄ @Ni	100 mA g ⁻¹	0.81	236/1000	8801	This work

Reference

- S1 Z. Sadighi, J. P. Liu, F. Ciucci, J. K. Kim, *Nanoscale*. 2018, **10**, 15588-15599.
S2 H. Zheng, C. Bo, J. Li, Y. Sun, Y. Liu, *J. Alloy. Compd.* 2017, **695**, 3435-3444.
S3 B. Liu, W. Xu, P. Yan, P. Bhattacharya, R. Cao. M. E. Bowden, M. H. Engelhard, C. M. Wang, J. G. Zhang, *ChemsusChem*. 2015, **8**, 3697-3703.
S4 P.F. Li, W. Sun, Q.L. Yu, M.J. Guan, J.S. Qiao, Z.H. Wang, D. Rooney, K.N. Sun, *Mater. Lett.* 2018, **158**, 84-87.
S5 W. M. Liu, T. T. Gao, Y. Yang, Q. Sun, Z. W. Fu, *Phys. Chem. Chem. Phys.* 2013, **15**, 15806–15810.
S6 M.T. Xu, X.Y. Hou, X.B. Yu, Z.F. Ma, J. Yang, X.X. Yuan, *J. Electrochem. Soc.* 2019, **166**, F406-F413.