

Electronic supplementary information (ESI):

**Dealloying-directed synthesis of efficient mesoporous CoFe-based
catalysts towards oxygen evolution reaction and overall water
splitting**

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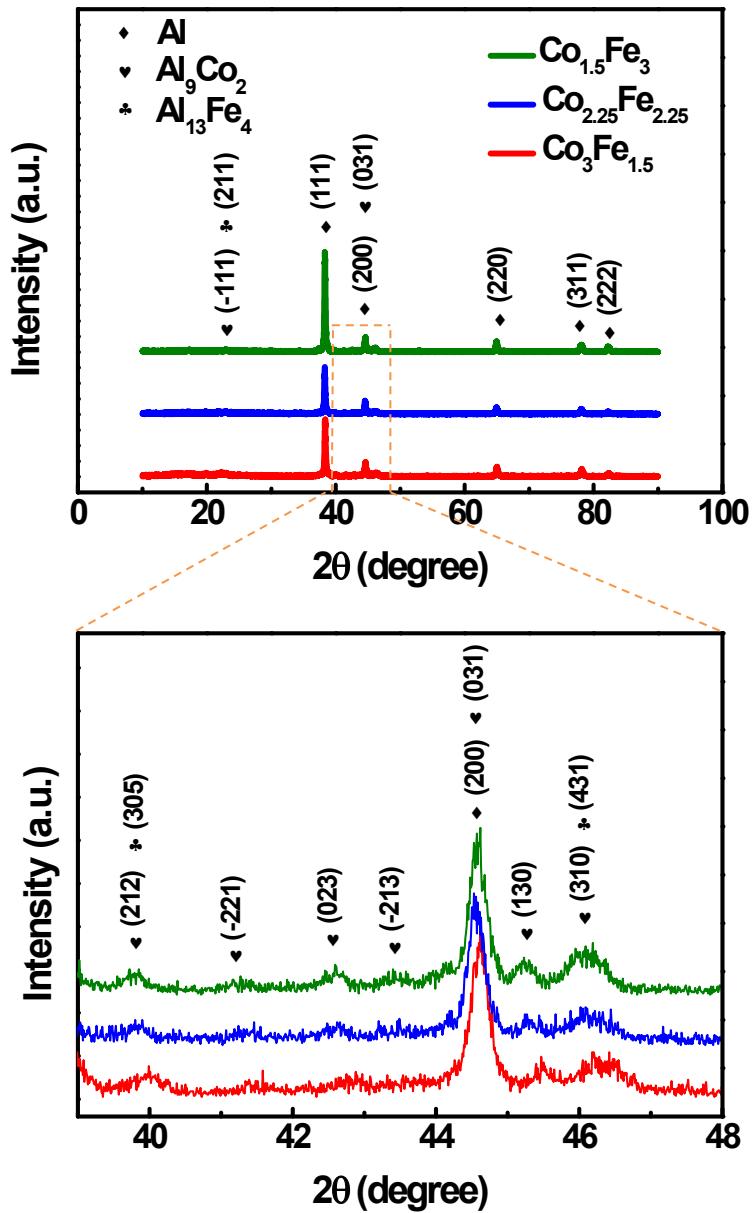


Figure S1. XRD patterns and magnified XRD patterns with the degree ranging from 39° to 48° of the $\text{Al}_{95.5}\text{Co}_x\text{Fe}_{4.5-x}$ ($x=1.5, 2.25, 3$) alloy ribbons.

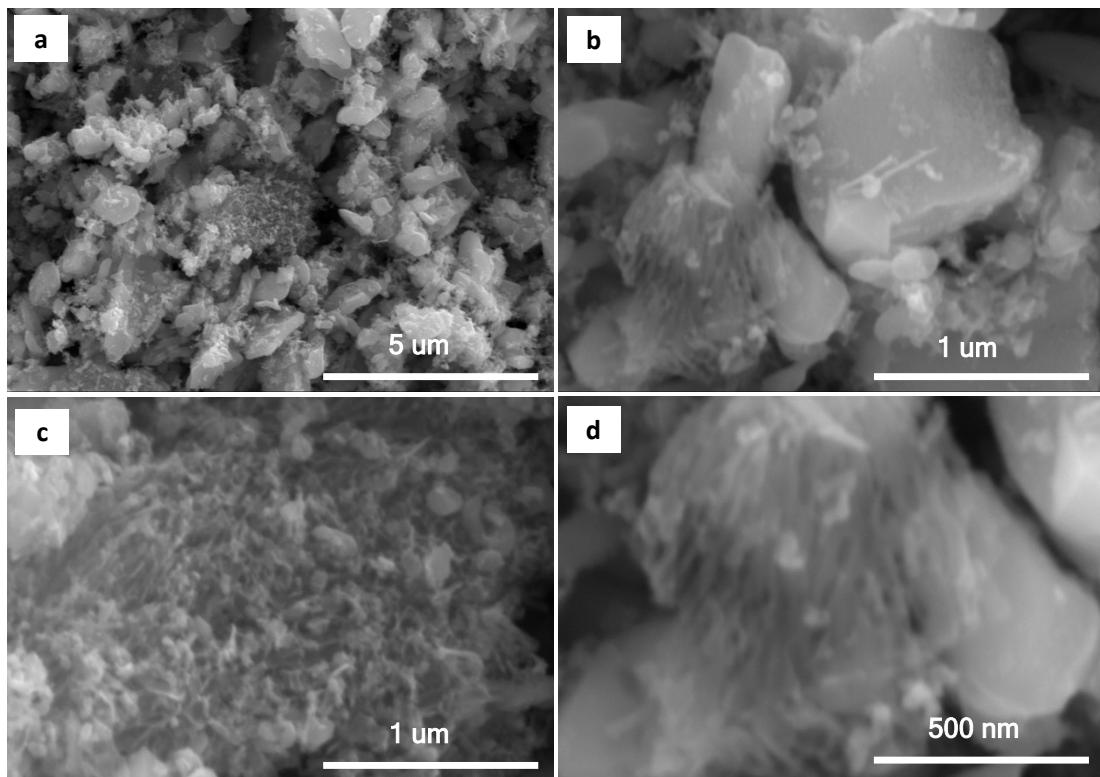


Figure S2. SEM images of the $\text{Co}_{1.5}\text{Fe}_3\text{-O}$ catalyst with different magnifications.

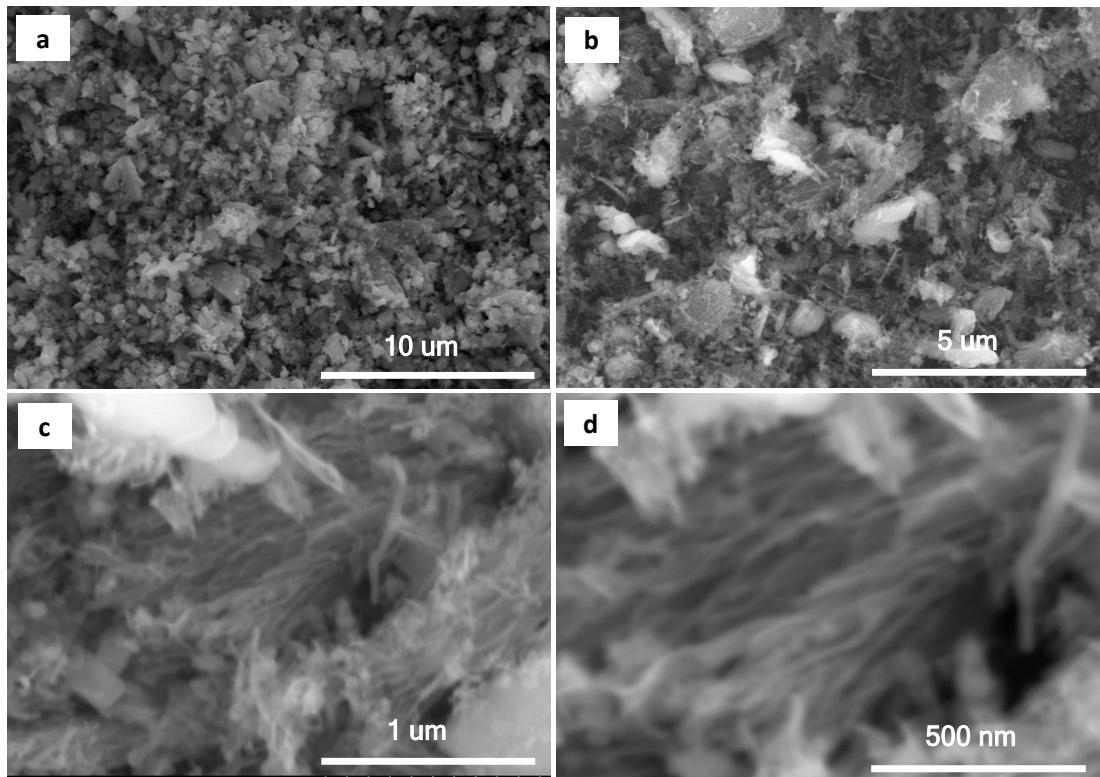


Figure S3. SEM images of the $\text{Co}_{2.25}\text{Fe}_{2.25}\text{-O}$ catalyst with different magnifications.

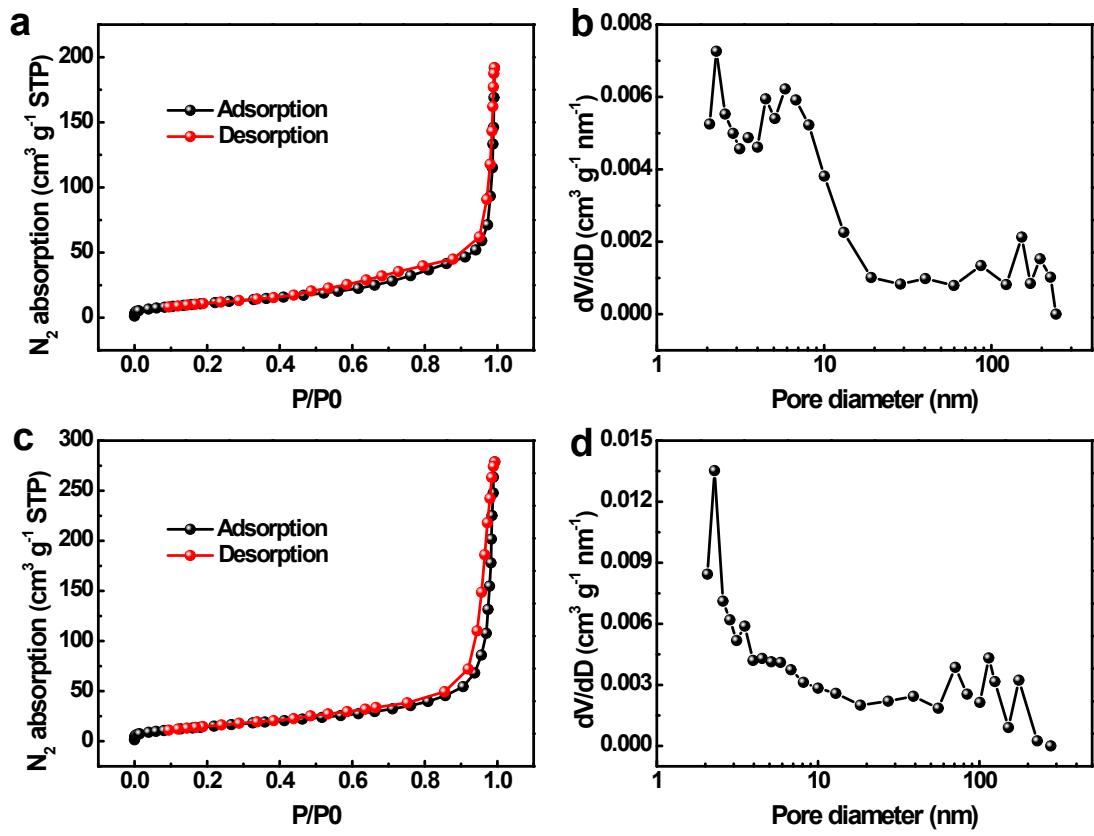


Figure S4. N₂ adsorption-desorption isotherms of the (a) Co_{1.5}Fe₃-O and (c) Co_{2.25}Fe_{2.25}-O catalysts. Pore size distribution of the (b) Co_{1.5}Fe₃-O and (d) Co_{2.25}Fe_{2.25}-O catalysts.

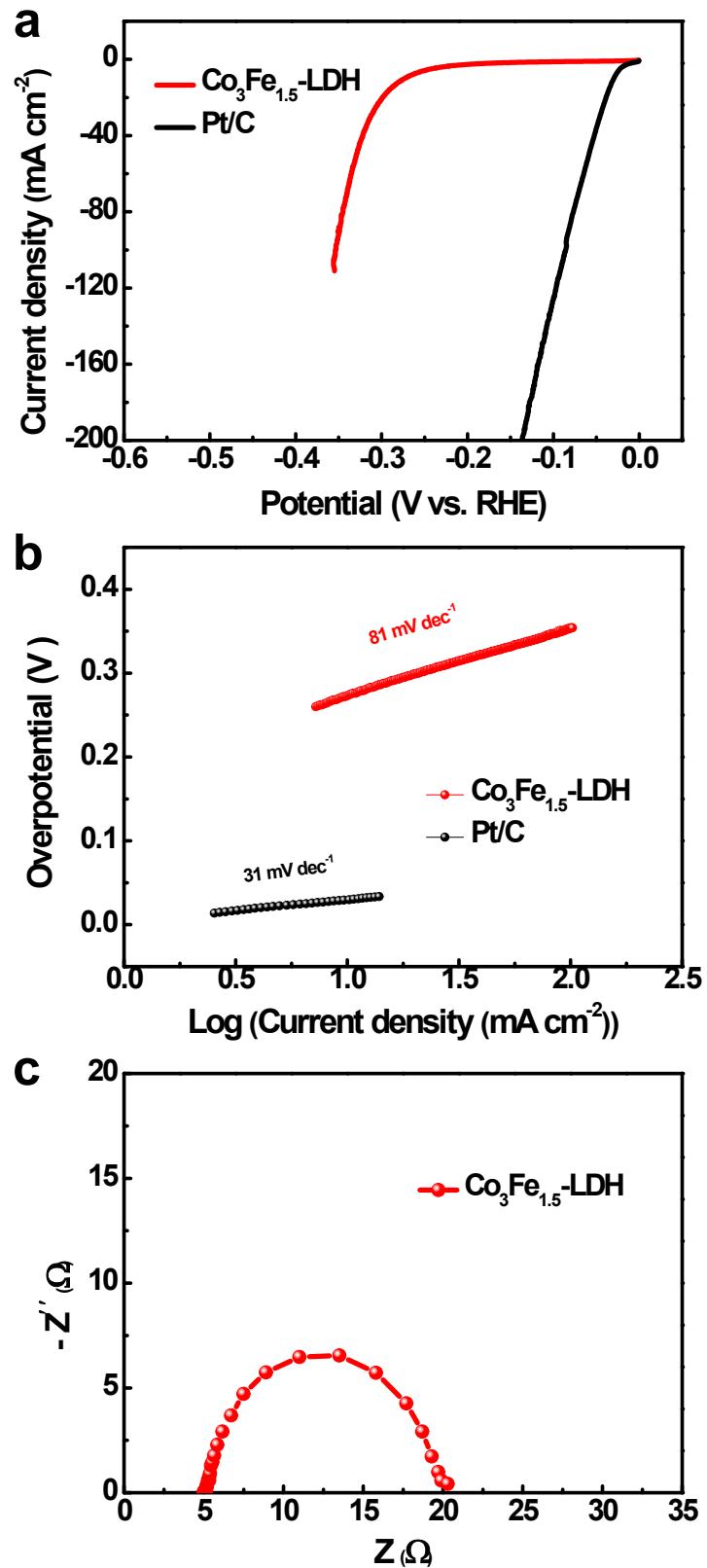


Figure S5. (a) HER polarization curves and (b) the corresponding Tafel curves of the $\text{Co}_3\text{Fe}_{1.5}\text{-LDH}$ and Pt/C catalysts. (c) EIS of the $\text{Co}_3\text{Fe}_{1.5}\text{-LDH}$ catalyst loaded on GC at -0.3 V vs. RHE.

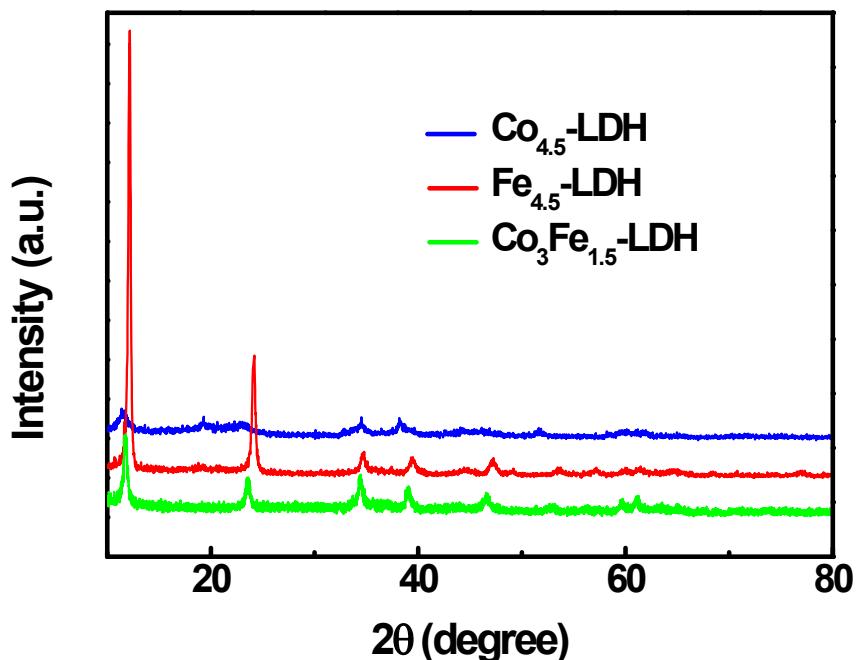


Figure S6. XRD patterns of the $\text{Co}_{4.5}\text{-LDH}$, $\text{Fe}_{4.5}\text{-LDH}$ and $\text{Co}_3\text{Fe}_{1.5}\text{-LDH}$ catalysts prepared from the $\text{Al}_{95.5}\text{Co}_{4.5}$, $\text{Al}_{95.5}\text{Fe}_{4.5}$ and $\text{Al}_{95.5}\text{Co}_3\text{Fe}_{1.5}$ precursor ribbons, respectively.

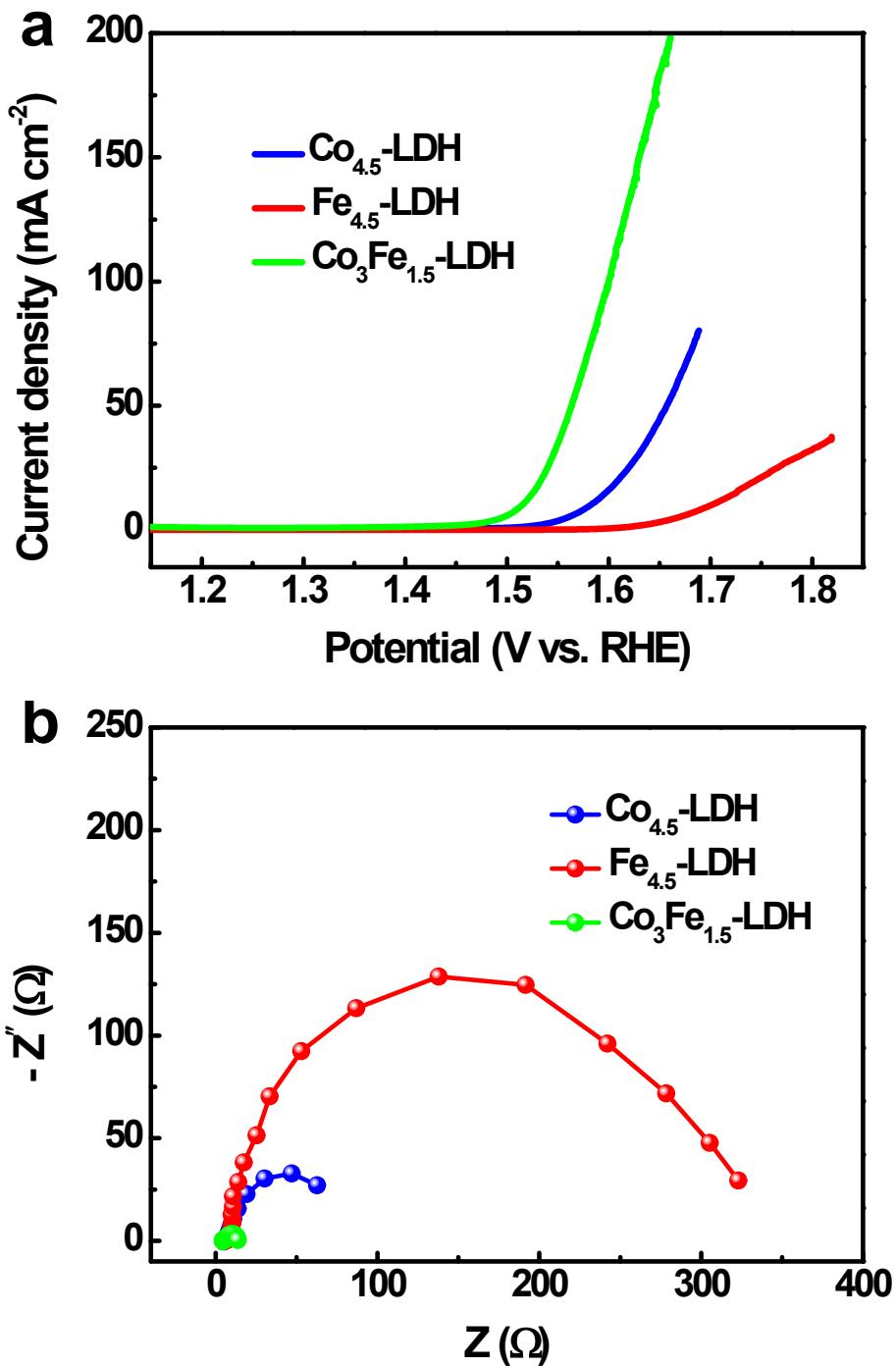


Figure S7. (a) OER polarization curves and (b) EIS of the Co_{4.5}-LDH, Fe_{4.5}-LDH and Co₃Fe_{1.5}-LDH catalysts loaded on GC at 1.54 V vs. RHE.

Table S1 OER activity comparison of our catalysts with other reported CoFe-based active catalysts.

Catalysts/Substrates	Onset potential (V vs. RHE)	η at 10 mA/cm ² (V)	Tafel slope mV/dec	Reference
Co₃Fe_{1.5}/GC	1.356	0.284	60	This work
Co₃Fe_{1.5}-LDH/GC	1.464	0.286	45	This work
CoFe ₂ O ₄ /CP	not available	0.477	51	1
LiCo _{0.8} Fe _{0.2} O ₂ /GC	1.49	0.34	50	2
(Co _{0.54} Fe _{0.46}) ₂ P/GC	1.43	0.37	not available	3
CoFe ₂ O ₄ /GC	1.5	0.314	not available	4
CoFeO _x /Au	1.51	0.32	36	5
CoFePO/NF	not available	0.275	51.7	6
CoFe-LDH/GC	1.49	0.34	43	7
W _{0.5} Co _{0.4} Fe _{0.1} /NF	not available	0.26	32	8
LDH FeCo/GC	not available	0.33	85	9
CoFe LDHs-Ar/GC	1.457	0.266	37.9	10

GC : glassy carbon. CP : carbon paper. NF : Ni foam.

Table S2 The simulated values of solution resistance (R_s) and charge-transfer resistance (R_{ct}) of the CoFe-based catalysts obtained from the electrical equivalent circuit model in the inset of Figure 5c.

Samples	R_s (Ω)	R_{ct} (Ω)
$\text{Co}_{1.5}\text{Fe}_3\text{-O}$	4.62	17.16
$\text{Co}_{2.25}\text{Fe}_{2.25}\text{-O}$	4.55	15.36
$\text{Co}_3\text{Fe}_{1.5}\text{-O}$	4.58	9.61
$\text{Co}_3\text{Fe}_{1.5}\text{-LDH}$	4.72	8.05

Notes and Reference

- 1 V. Maruthapandian, M. Mathankumar, V. Saraswathy, B. Subramanian and S. Muralidharan, *ACS Appl. Mater. Interfaces*, 2017, **9**, 13132-13141.
- 2 Y. Zhu, W. Zhou, Y. Chen, J. Yu, M. Liu and Z. Shao, *Adv. Mater.*, 2015, **27**, 7150-7155.
- 3 A. Mendoza-Garcia, D. Su and S. Sun, *Nanoscale*, 2016, **8**, 3244-3247.
- 4 Y. Liu, J. Li, F. Li, W. Li, H. Yang, X. Zhang, Y. Liu and J. Ma, *J. Mater. Chem. A*, 2016, **4**, 4472-4478.
- 5 C. G. Morales-Guio, L. Liardet and X. Hu, *J. Am. Chem. Soc.*, 2016, **138**, 8946-8957.
- 6 J. Duan, S. Chen, A. Vasileff and S. Z. Qiao, *ACS Nano*, 2016, **10**, 8738-8745.
- 7 X. Han, C. Yu, J. Yang, C. Zhao, H. Huang, Z. Liu, P. M. Ajayan and J. Qiu, *Adv. Mater. Interfaces*, 2016, **3**, 1500782.
- 8 Y. Pi, Q. Shao, P. Wang, F. Lv, S. Guo, J. Guo and X. Huang, *Angew. Chem.*, 2017, **56**, 4502-4506.
- 9 B. Zhang, X. L. Zheng, O. Voznyy, R. Comin, M. Bajdich, M. García-Melchor, L. L. Han, J. X. Xu, M. Liu, L. R. Zheng, F. P. García de Arquer, C. T. Dinh, F. J. Fan, M. j. Yuan, E. Yassitepe, N. Chen, T. Regier, P. f. Liu, Y. h. Li, P. D. Luna, A. Janmohamed, H. L. Xin, H. g. Yang, A. Vojvodic, and E. H. Sargent, *Science*, 2016, **352**, 333-337.
- 10 Y. Wang, Y. Zhang, Z. Liu, C. Xie, S. Feng, D. Liu, M. Shao and S. Wang, *Angew. Chem.*, 2017, **56**, 5867-5871.