

## Supporting Information

# Atomic layer deposited zinc oxysulfide anodes in Li-ion batteries: An efficient solution for electrochemical instability and low conductivity

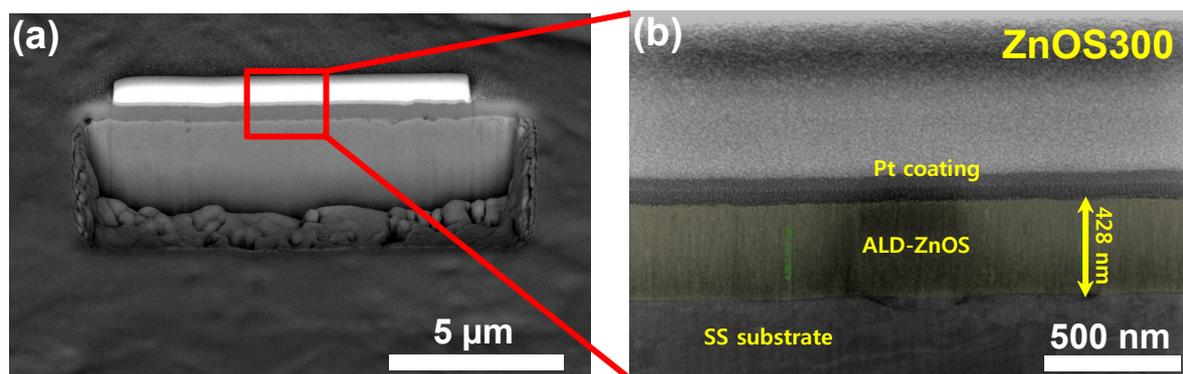
Soumyadeep Sinha,<sup>a</sup> Hari V. Ramasamy,<sup>b</sup> Dip K. Nandi,<sup>c</sup> Pravin N. Didwal,<sup>a</sup> Jae Yu Cho,<sup>a</sup> Chan-Jin Park,<sup>a</sup> Yun-Sung Lee,<sup>b</sup> Soo-Hyun Kim,<sup>c</sup> and Jaeyeong Heo<sup>a\*</sup>

<sup>a</sup>Department of Materials Science and Engineering, and Optoelectronics Convergence Research Center, Chonnam National University, Gwangju 61186, Republic of Korea

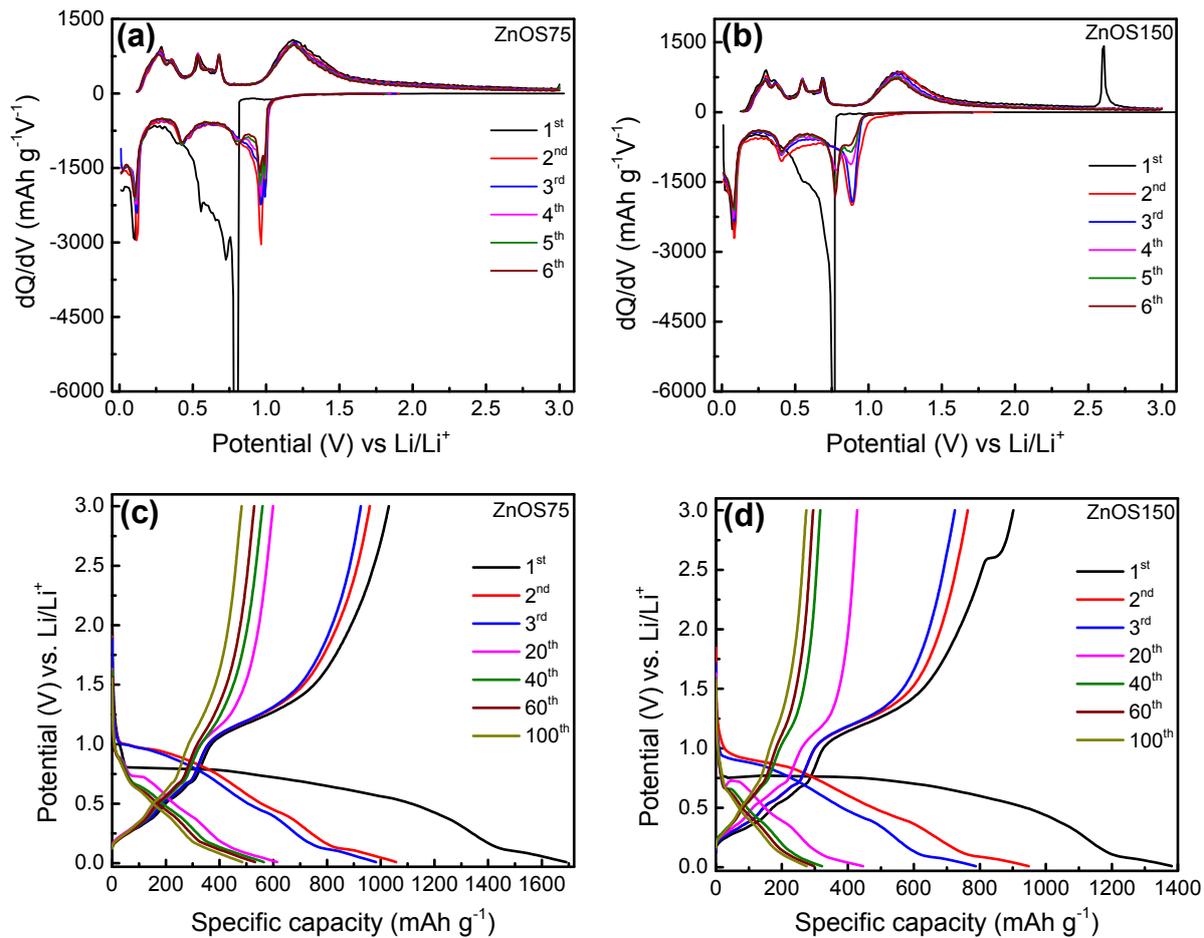
<sup>b</sup>School of Chemical Engineering, Chonnam National University, Gwangju 61186, Republic of Korea

<sup>c</sup>School of Materials Science and Engineering, Yeungnam University, 214-1, Dae-dong, Gyeongsan-si 38541, Republic of Korea

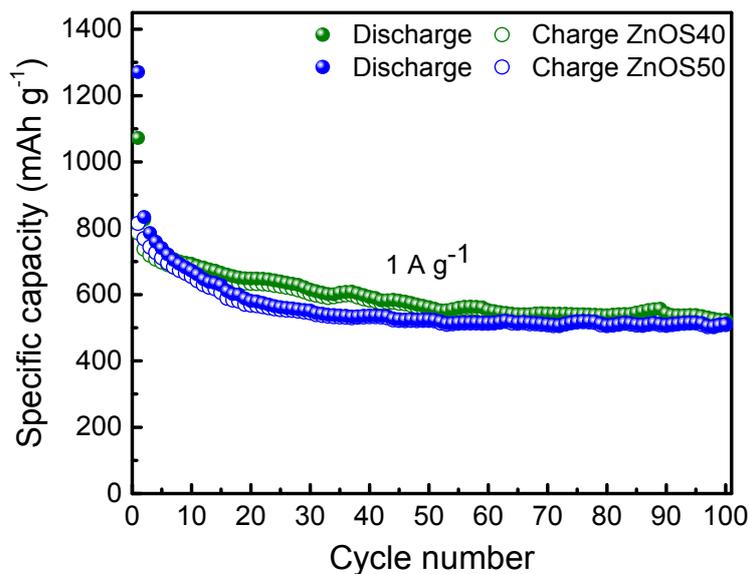
\*Corresponding author E-mail: jheo@jnu.ac.kr



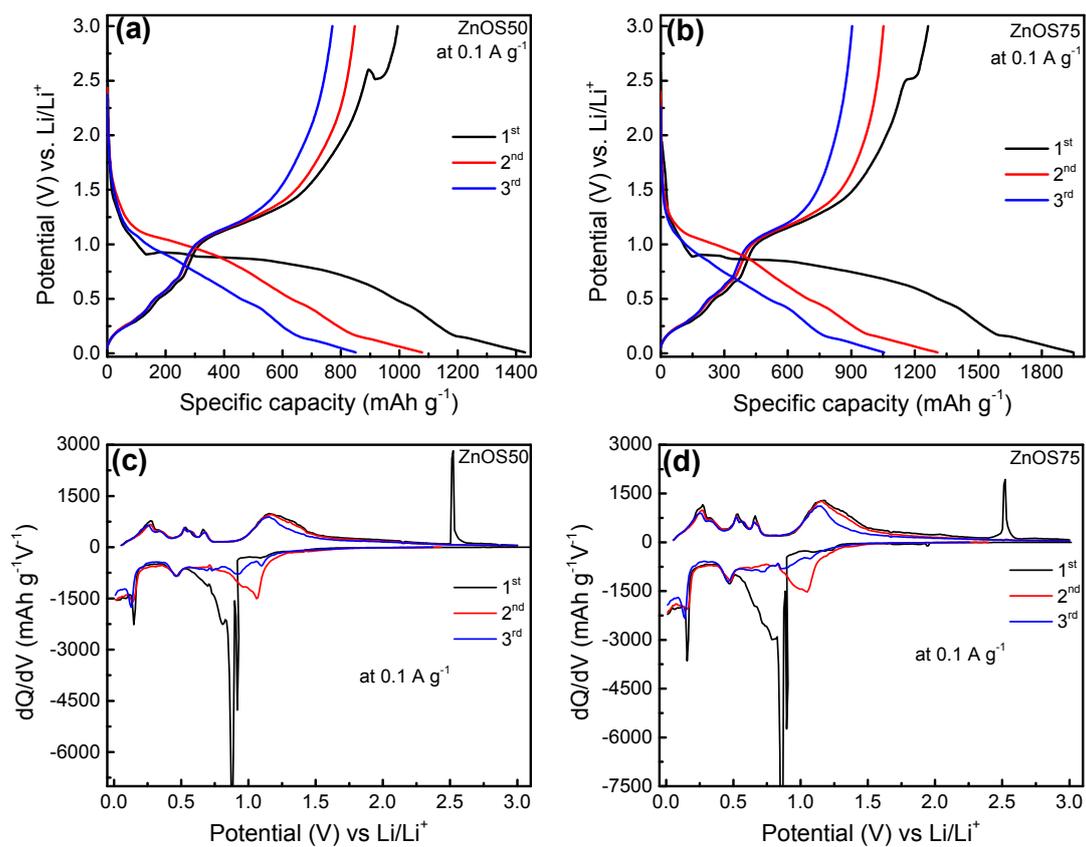
**Fig. S1** (a-b) Cross-sectional SEM images of ZnOS300 on a SS substrate obtained by using FIB-SEM



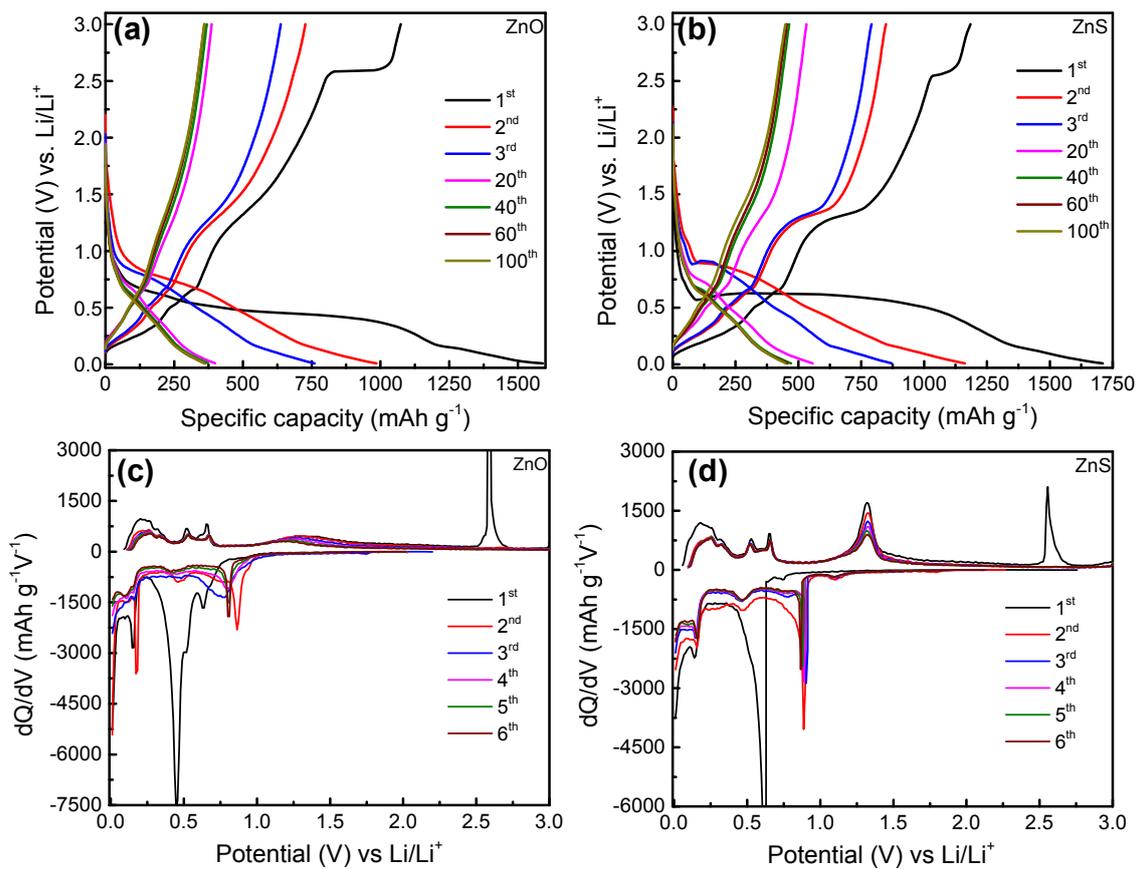
**Fig. S2** dQ/dV plots for the first six consecutive cycles, as derived from the charge-discharge profiles of the (a, c) ZnOS75 and (b, d) ZnOS150 anodes in LIBs at 1 A g<sup>-1</sup>.



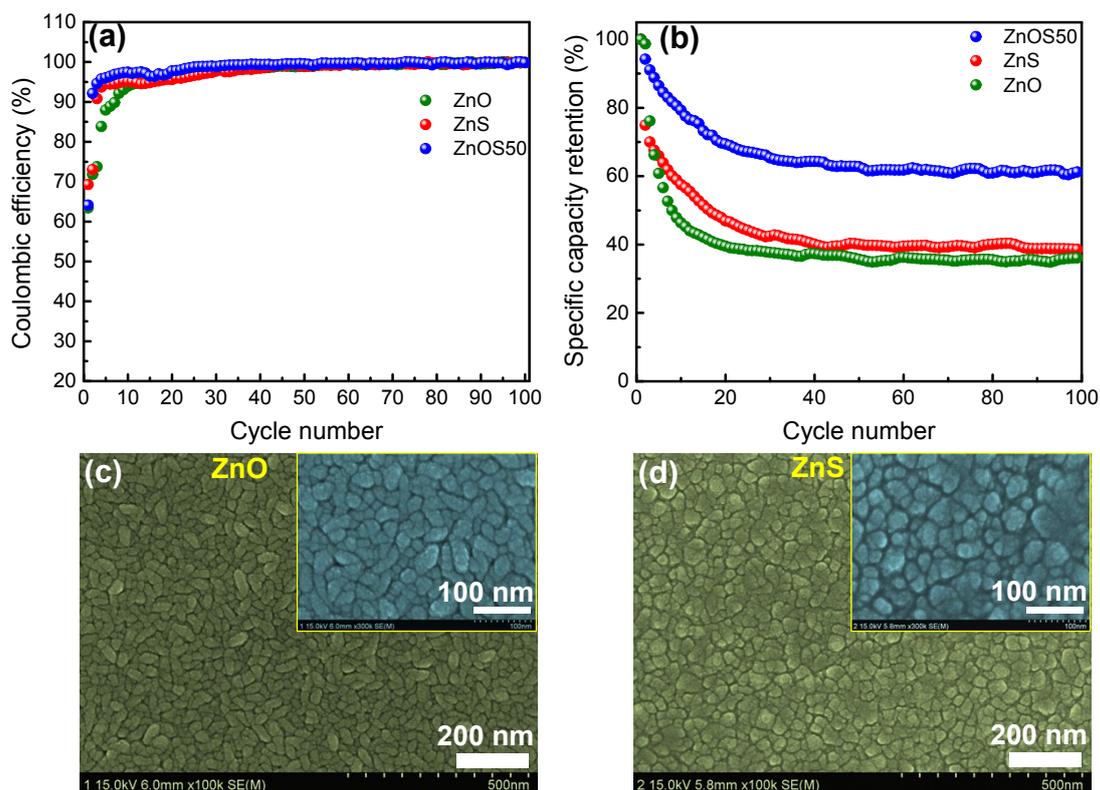
**Fig. S3** Cycling performance comparison of the ZnOS40 and ZnOS50 electrodes in LIBs at  $1 \text{ A g}^{-1}$ .



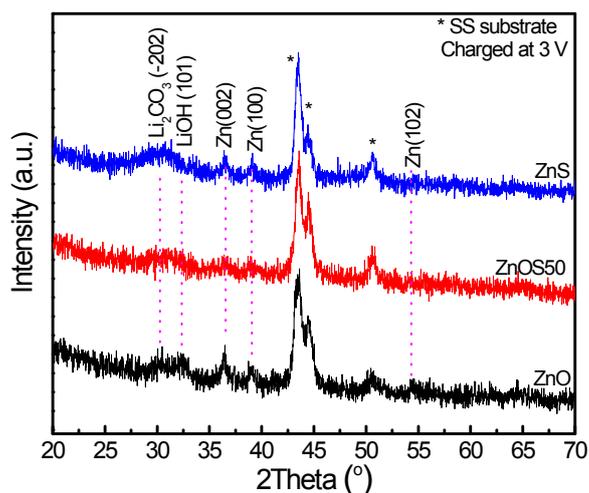
**Fig. S4** Charge-discharge profiles at  $0.1 \text{ A g}^{-1}$  current density and the corresponding  $dQ/dV$  plots for the first three consecutive cycles of the (a, c) ZnOS50 and (b, d) ZnOS75 anodes in LIBs.



**Fig. S5** Charge-discharge profiles for 100 cycles at 1 A g<sup>-1</sup> current density and the corresponding dQ/dV plots for the first six consecutive cycles of the pristine (a, c) ZnO and (b, d) ZnS anodes in LIBs.



**Fig. S6** (a) Coulombic efficiency and (b) specific capacity retention corresponding to 100 charge-discharge cycles for the ZnO, ZnS, and ZnOS50 anodes at  $1 \text{ A g}^{-1}$  current density. Plan-view FESEM images of (c) ZnO and (d) ZnS on SS substrate before the charge-discharge process. The insets represent high-magnification images.



**Fig. S7** *Ex situ* XRD patterns of pristine ZnO, ZnS, and ZnOS50 electrodes in LIBs after complete charge at 3.0 V.

**Table S1.** A comparison of the electrochemical performances of a few ZnO, ZnS, and ZnOS-based anode materials for LIBs, including the results of this study.

\*Obtained from rate studies

Anode materials	Material growth process	Electrochemical performance		Ref	
		Stable specific capacity (mAh g <sup>-1</sup> )	Specific capacity at current density* (mAh g <sup>-1</sup> )		
ZnO thin film/SS	PLD	220, 40 cycles at 20 μA cm <sup>-2</sup>	–	1	
yolk-shell ZnO-C microspheres	Chemical solution reaction	520, 150 cycles at 100 mA g <sup>-1</sup>	212 at 1 A g <sup>-1</sup>	2	
Graphite-coated ZnO nanosheets/Pt/SS	Hydrothermal	600, 100 cycles at 1 A g <sup>-1</sup>	–	3	
ZnO-C-rGO nanofiber	Co-axial electrospinning	618.9, 100 cycles at 50 mA g <sup>-1</sup>	472 at 500 mA g <sup>-1</sup>	4	
ZnO@ZnO QDs/C core-shell NRAs/CC	Chemical route	699, 100 cycles at 500 mA g <sup>-1</sup>	530 at 1 A g <sup>-1</sup>	5	
<b>ZnO</b>	Al <sub>2</sub> O <sub>3</sub> /ZnO-Gr/Cu foil	ALD	487, 100 cycles at 100 mA g <sup>-1</sup>	415 at 1 A g <sup>-1</sup>	6
	ZnO QD/graphene/Cu foil	ALD	540, 100 cycles at 100 mA g <sup>-1</sup>	400 at 1 A g <sup>-1</sup>	7
	ZnO-CB	ALD	1026, 500 cycles at 100 mA g <sup>-1</sup>	1213 at 1 A g <sup>-1</sup>	8
	ZnO/expanded graphite composite	ALD	436.4, 500 cycles at 200 mA g <sup>-1</sup>	417 at 800 mA g <sup>-1</sup>	9
	3D-C/ZnO NMs foam composite	ALD	260,700 cycles at 2 A g <sup>-1</sup> and 180, 500 cycles at 5 A g <sup>-1</sup>	288 at 1 A g <sup>-1</sup>	10
	ZnS/graphene composites/Cu foil	Solvothermal	570, 200 cycles at 200 mA g <sup>-1</sup>	418 at 1 A g <sup>-1</sup>	11
	ZnS/porous carbon matrix/Cu foil	Chemical route	438, 300 cycles at 100 mA g <sup>-1</sup>	180 at 1 A g <sup>-1</sup>	12
<b>ZnS</b>	ZnS nanocrystal/RGO composite/ Cu sheet	Hydrothermal	776, 100 cycles at 100 mA g <sup>-1</sup>	–	13
	Core-shell-ZnS/C nanocomposite/Cu foil	Solvothermal	750, 300 cycles at 500 mA g <sup>-1</sup>	–	14
	ZnS decorated on N-doped porous carbon polyhedra/Cu foil	Co-precipitation	856.8, 1000 cycles at 1 A g <sup>-1</sup>	–	15
	ZnS/C/ Cu foil	Ball milling	570, 150 cycles at 100 mA g <sup>-1</sup>	–	16
<b>ZnOS</b>	ZnO/SS		285,40 cycles at 2 μA cm <sup>-2</sup>		
	ZnS/SS		312,40 cycles at 2 μA cm <sup>-2</sup>		
	O-rich ZnOS/SS	PLD	373,40 cycles at 2 μA cm <sup>-2</sup>	–	17
	S-rich ZnOS/SS		600,40 cycles at 2 μA cm <sup>-2</sup>		

ZnO/SS		360, 100 cycles at 1 A g <sup>-1</sup> (~ 40 μA cm <sup>-2</sup> )		
ZnS/SS	ALD	449.2, 100 cycles at 1 A g <sup>-1</sup> (~ 40 μA cm <sup>-2</sup> )	–	This work
O-rich ZnOS/SS		510.3, 100 cycles at 1 A g <sup>-1</sup> (~ 40 μA cm <sup>-2</sup> )		

## References

1. Z.-W. Fu, F. Huang, Y. Zhang, Y. Chu and Q.-Z. Qin *J. Electrochem. Soc.*, 2003, **150**, A714-A720.
2. Q. Xie, X. Zhang, X. Wu, H. Wu, X. Liu, G. Yue, Y. Yang and D.-L. Peng, *Electrochim. Acta*, 2014, **125**, 659-665.
3. E. Quartarone, V. Dall'Asta, A. Resmini, C. Tealdi, I. G. Tredici, U. A. Tamburini and P. Mustarelli, *J. Power Sources*, 2016, **320**, 314-321.
4. Shilpa, B. M. Basavaraja, S. B. Majumder and A. Sharma, *J. Mater. Chem. A*, 2015, **3**, 5344-5351.
5. G. Zhang, S. Hou, H. Zhang, W. Zeng, F. Yan, C. C. Li and H. Duan, *Adv. Mater.*, 2015, **27**, 2400-2405.
6. M. Yu, A. Wang, Y. Wang, C. Li and G. Shi, *Nanoscale*, 2014, **6**, 11419-11424.
7. X. Sun, C. Zhou, M. Xie, H. Sun, T. Hu, F. Lu, S. M. Scott, S. M. George and J. Lian, *J. Mater. Chem. A*, 2014, **2**, 7319-7326.
8. S. Lu, H. Wang, J. Zhou, X. Wu and W. Qin, *Nanoscale*, 2017, **9**, 1184-1192.
9. Y. Li, Y. Zhao, G. Huang, B. Xu, B. Wang, R. Pan, C. Men and Y. Mei, *ACS Appl. Mater. Interfaces*, 2017, **9**, 38522-38529.
10. Y. Zhao, G. Huang, Y. Li, R. Edy, P. Gao, H. Tang, Z. Bao and Y. Mei, *J. Mater. Chem. A*, 2018, DOI: 10.1039/C8TA00940F.
11. M. Mao, L. Jiang, L. Wu, M. Zhang and T. Wang, *J. Mater. Chem. A*, 2015, **3**, 13384-13389.
12. Y. Fu, Z. Zhang, X. Yang, Y. Gan and W. Chen, *RSC Adv.*, 2015, **5**, 86941-86944.
13. Y. Feng, Y. Zhang, Y. Wei, X. Song, Y. Fu and V. S. Battaglia, *Phys. Chem. Chem. Phys.*, 2016, **18**, 30630-30642.
14. X. Du, H. Zhao, Y. Lu, Z. Zhang, A. Kulka and K. Świerczek, *Electrochim. Acta*, 2017, **228**, 100-106.
15. J. Li, D. Yan, X. Zhang, S. Hou, T. Lu, Y. Yao and L. Pan, *J. Mater. Chem. A*, 2017, **5**, 20428-20438.
16. A.-R. Park, K.-J. Jeon and C.-M. Park, *Electrochim. Acta*, 2018, **265**, 107-114.
17. H. Xu, Y.-N. Zhou, F. Lu and Z.-W. Fu, *J. Electrochem. Soc.*, 2011, **158**, A285-A290.