

## Supplementary Information

### All-Solid-State Asymmetric Supercapacitors Based on Cobalt Hexacyanoferrate Derived CoS and Activated Carbon†

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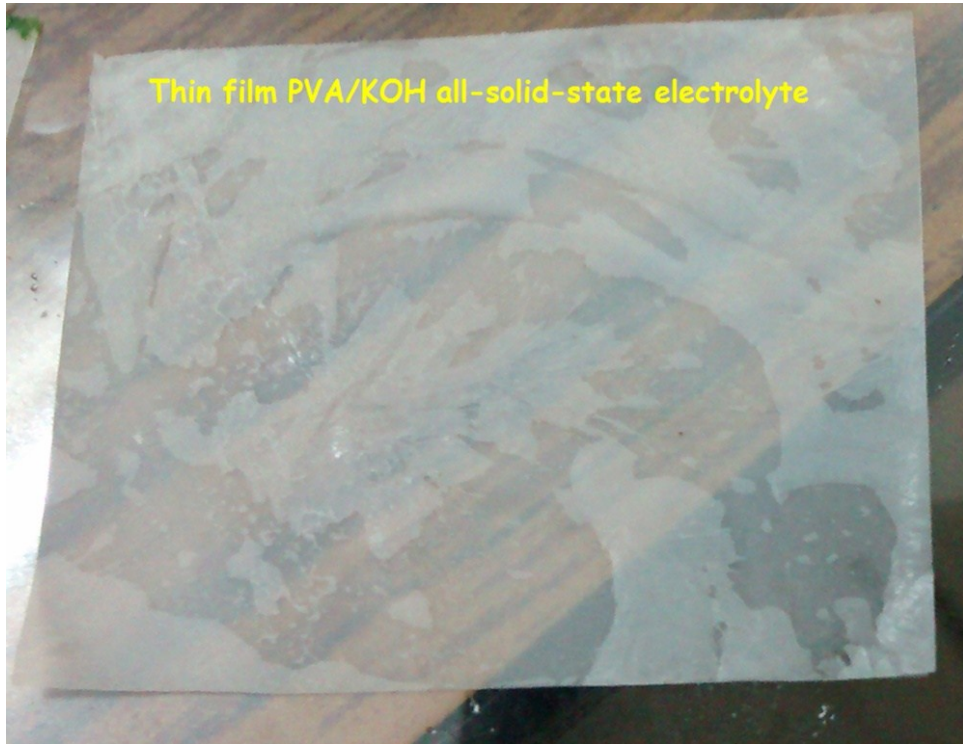
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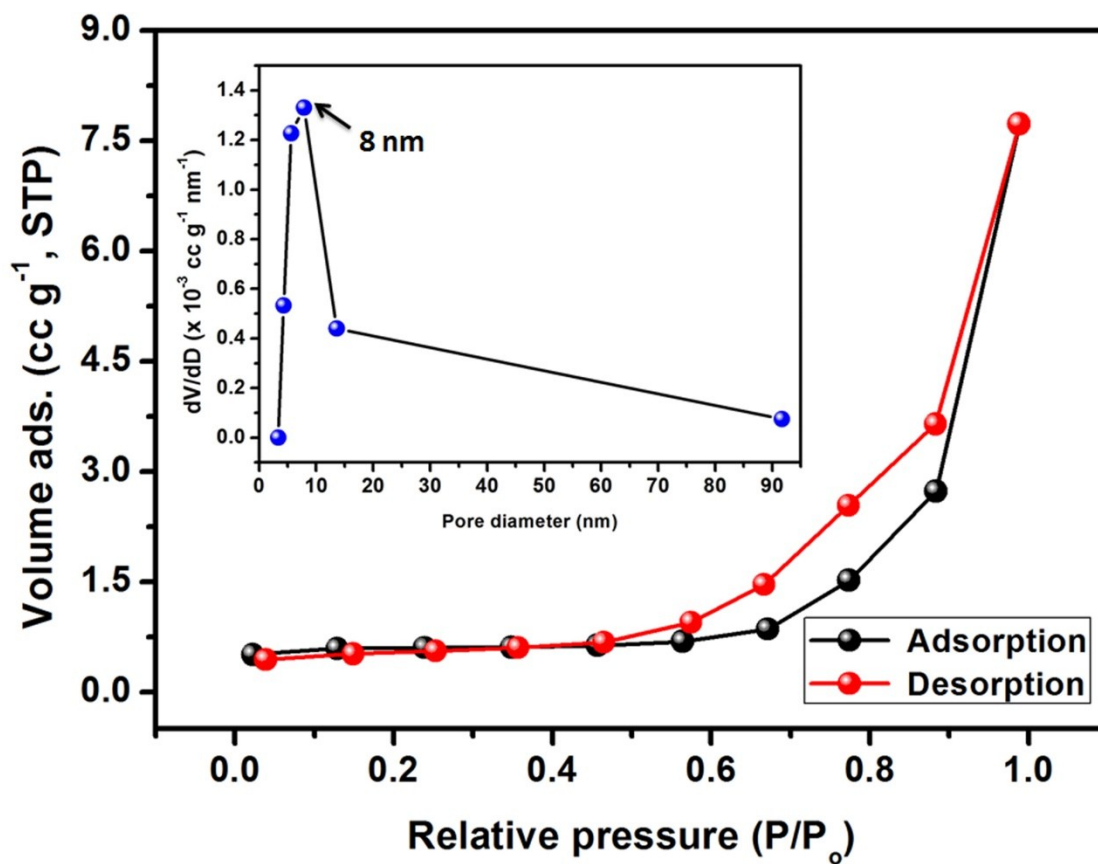
## Supplementary Figures:

**Table S1.** Recent development of cobalt sulfide based electrode materials with different morphology

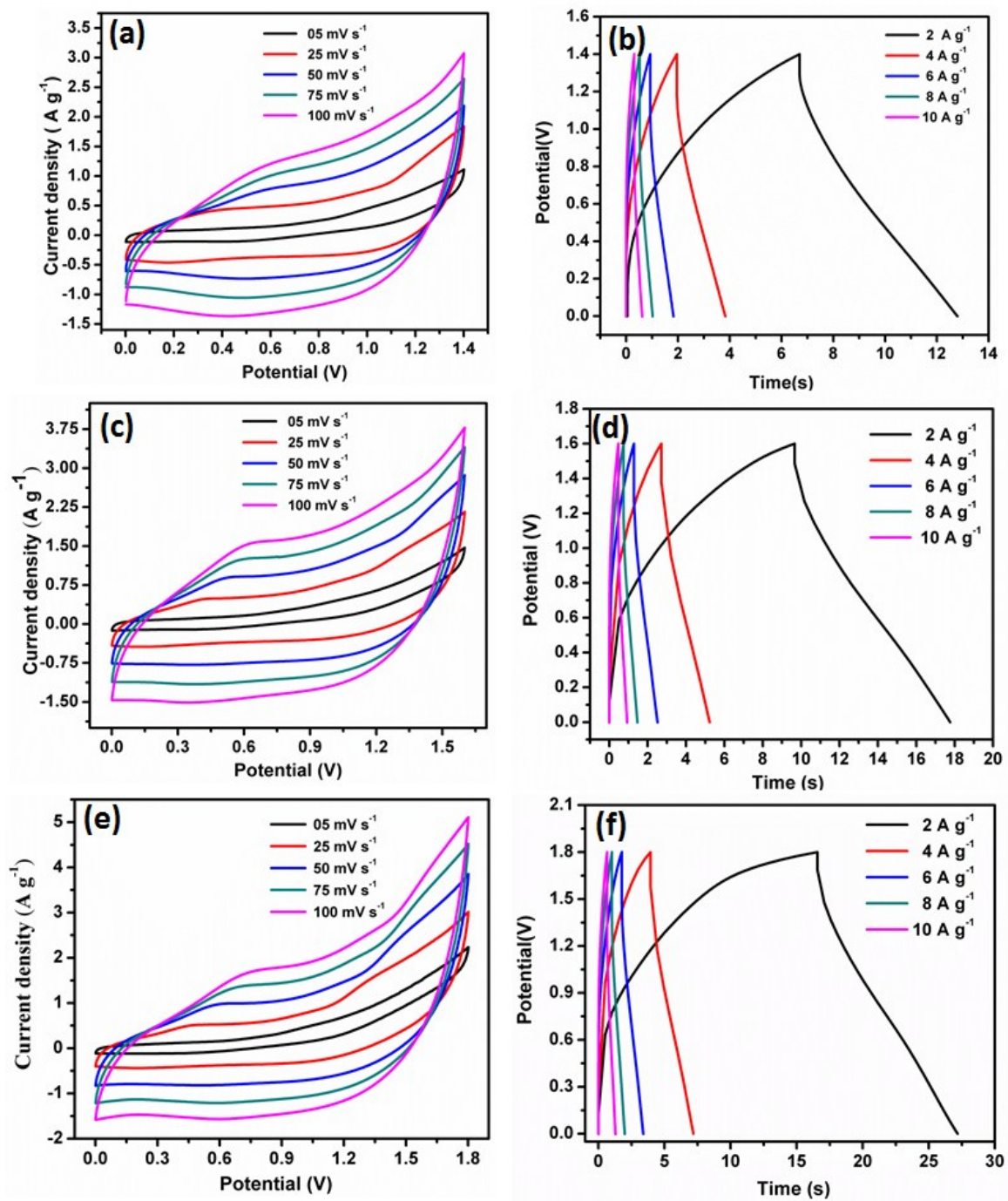
S. No	Materials	Method	Morphology	Reference
1	Co <sub>9</sub> S <sub>8</sub>	Atomic layer deposition	Nanoparticles	1
2	CoS/Graphene	Hydrothermal	Nanoparticles	2
3	CoS <sub>1.097</sub>	Ostwald ripening	Hierarchitectures	3
4	CoS <sub>x</sub> /CNT	Hydrothermal	Core/shell	4
5	CoS	Hydrothermal	Nanowires	5
6	Graphene/CoS	Hydrothermal	Nanoparticles	6
7	CoS <sub>1.097</sub>	Solvothermal	Nanotube	7
8	CoS	Hydrothermal	Nanotube	8
9	Co <sub>9</sub> S <sub>8</sub>	Hydrothermal	Nanoflake	9
10	Co <sub>9</sub> S <sub>8</sub>	Hydrothermal	Octahedra	9
11	CoS	Solvothermal	Dumb-bells	This work



**Figure S1.** Fabricated PVA/KOH all-solid-state electrolyte thin film.



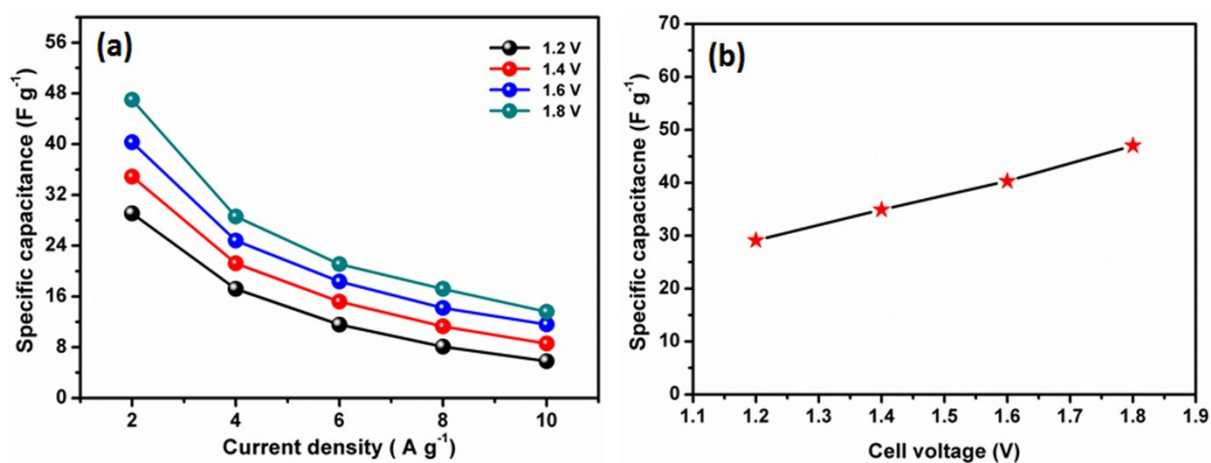
**Figure S2.** BET nitrogen adsorption/desorption isotherm and its corresponding pore-size distribution curve (inset) of CoS nanoparticles.



**Figure S3.** CV and CD profile of a flexible ASC cell in (a & b) 1.4, (c & d) 1.6 and (e & f) 1.8 V cell voltage at different scan rate and current density, respectively.

**Table S2.** Comparison of metal oxide/sulfide based all-solid-state asymmetric supercapacitors

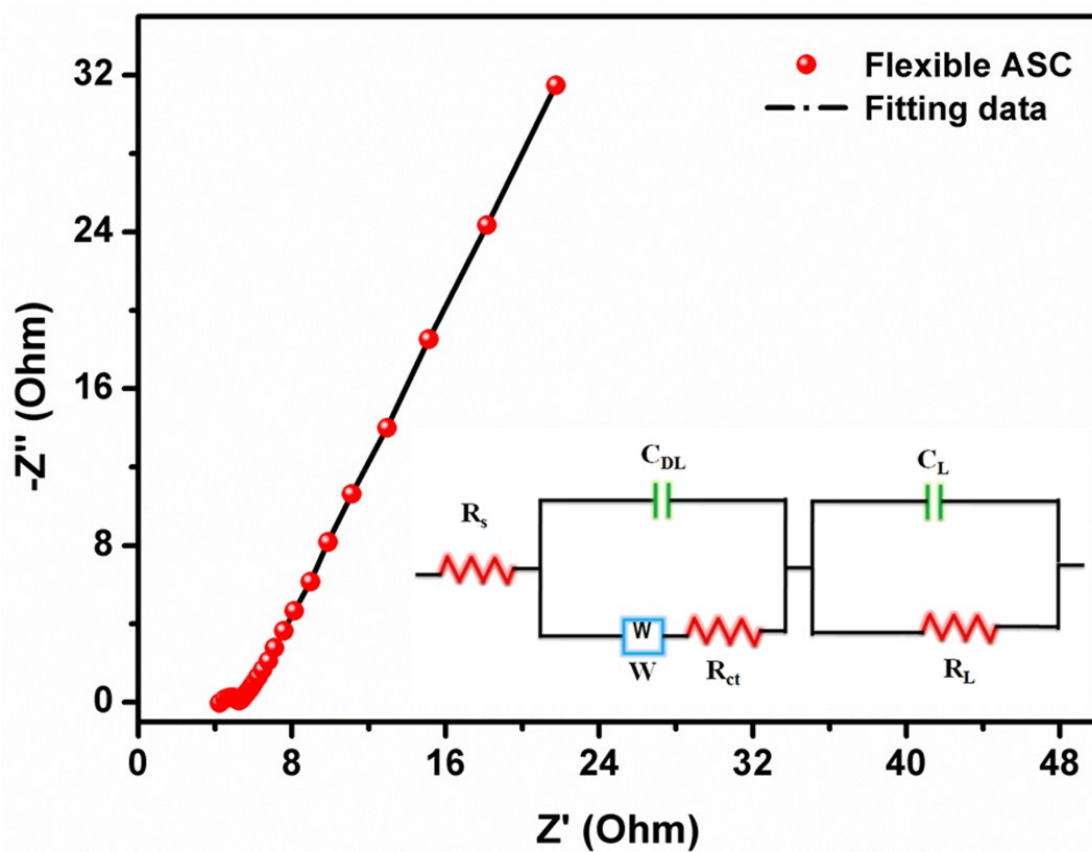
S. No.	Cathode	Anode	Cell Voltage (V)	Current density (A/g)	C <sub>sp</sub> (F/g)	Cycle life (No. of cycle)	Energy density (Wh/kg)	Power density (W/kg)	Journal name	Reference
1.	Co <sub>9</sub> S <sub>8</sub> nanoflake	AC	1.6	1.25	83	89.5% (5000)	31.4	200	<i>J. Mater. Chem. A</i>	9
2.	Co <sub>9</sub> S <sub>8</sub> octahedra	AC	1.6	1.25	18.6	65% (5000)	7	230	<i>J. Mater. Chem. A</i>	9
3.	CoS <sub>x</sub> nanostrip	graphene	1.5	0.001	46.2	84% (3000)	14.68	369	<i>Energy Technol.</i>	10
4.	Co(OH) <sub>2</sub> nanowires	NTAC	1.6	0.0012	38.9	-	13.6	153	<i>J. Power Sources</i>	11
5.	Co <sub>3</sub> O <sub>4</sub> nanowires	Carbon aerogel	1.5	1	57.4	85% (1000)	17.9	750	<i>J. Power Sources</i>	12
6.	MnS	AC	1.6	1	110.4	89.87% (5000)	37.6	181.2	<i>Sci. Rep.</i>	13
7.	Cu(OH) <sub>2</sub>	AC	1.6	4	26.4	90% (5000)	3.68	1253	<i>J. Mater. Chem. A</i>	14
8.	Fe <sub>3</sub> O <sub>4</sub> /Carbon	porous carbon	1.4	1	58.5	70.8% (5000)	18.3	351	<i>ACS Appl. Mater. Interfaces</i>	15
9.	Ni-Co-S	graphene	1.8	2	133	82.2% (20000)	60	1800	<i>J. Mater. Chem. A</i>	16
10.	<b>CoS</b>	<b>AC</b>	<b>1.8</b>	<b>2</b>	<b>47</b>	<b>92% (5000)</b>	<b>5.3</b>	<b>1800</b>	<b>-</b>	<b>This work</b>



**Figure S4.** Specific capacitance of fabricated flexible all-solid-state ASC as a function of (a) different current density and (b) cell voltage.

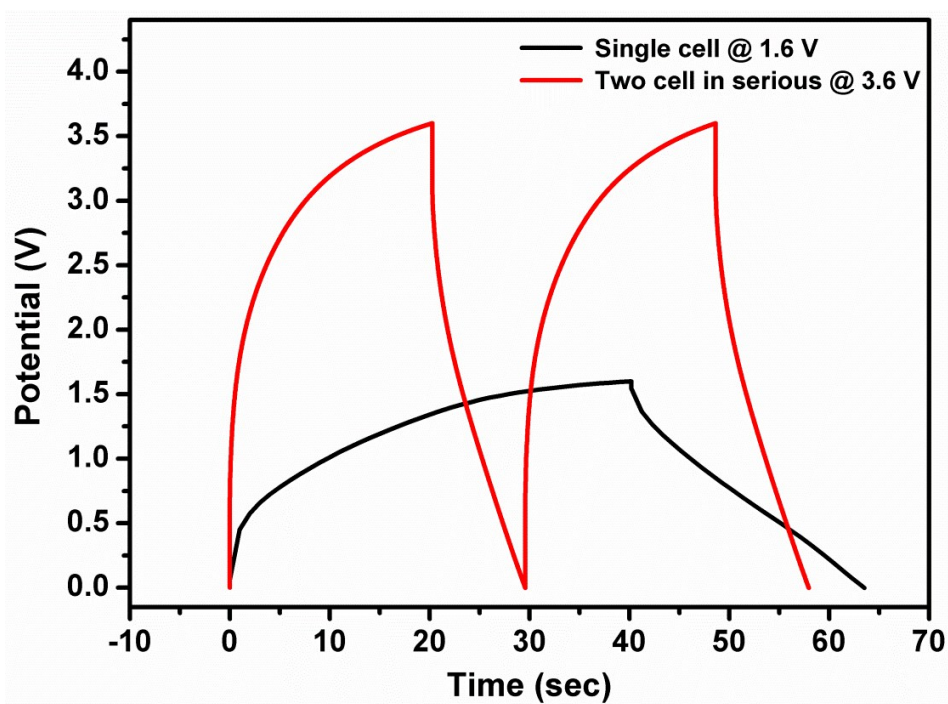
**Table S3.** Specific capacitance of fabricated flexible all-solid-state ASC with different cell voltage at various current densities-

Cell voltage (V)	Specific capacitance $C_{sp}$ (F g <sup>-1</sup> )				
	2 A g <sup>-1</sup>	4 A g <sup>-1</sup>	6 A g <sup>-1</sup>	8 A g <sup>-1</sup>	10A g <sup>-1</sup>
1.2	29.1	17.2	11.6	8.1	5.8
1.4	34.9	21.3	15.2	11.3	8.6
1.6	40.3	24.8	18.4	14.2	11.6
1.8	47	28.6	21.1	17.2	13.6

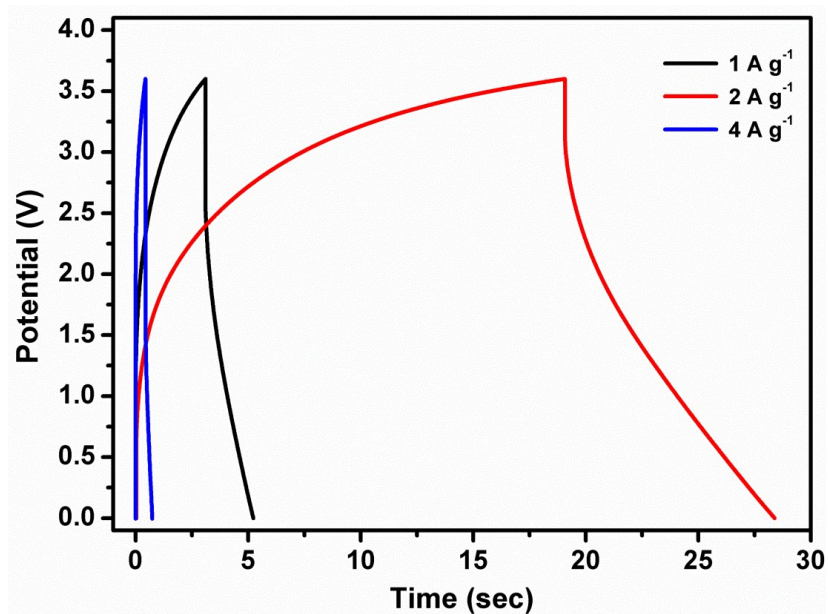


**Figure S5.** EIS Nyquist plots of fabricated flexible all-solid-state ASC and its corresponding equivalent circuits. (inset)





**Figure S6.** CD profile of single asymmetric cell (1.6 V) and two asymmetric cells were connected in series (3.6 V) at  $1 \text{ A g}^{-1}$  current density.



**Figure S7.** CD profile of two asymmetric cells connected in series for 3.6 V at different current densities.

## Reference

- 1 H. Li, Y. Gao, Y. Shao, Y. Su and X. Wang, *Nano Lett.*, 2015, **15**, 6689–6695.
- 2 X. Meng, J. Deng, J. Zhu, H. Bi, E. Kan and X. Wang, *Sci. Rep.*, 2016, **6**, 21717.
- 3 Q. Wang, L. Jiao, H. Du, J. Yang, Q. Huan, W. Peng, Y. Si, Y. Wang and H. Yuan, *CrystEngComm*, 2011, **13**, 6960.
- 4 M. L. Mao, L. Mei, L. C. Wu, Q. H. Li and M. Zhang, *Rsc Adv.*, 2014, **4**, 12050–12056.
- 5 S. Bao, C. M. Li, C. Guo and Y. Qiao, *J. Power Sources*, 2008, **180**, 676–681.
- 6 X. Meng, H. Sun, J. Zhu, H. Bi, Q. Han, X. Liu and X. Wang, *New J. Chem.*, 2016, **40**, 2843–2849.
- 7 S. Liu, C. Mao, Y. Niu, F. Yi, J. Hou, S. Lu, J. Jiang, M. Xu and C. Li, *ACS Appl. Mater. Interfaces*, 2015, **7**, 25568–25573.
- 8 H. Wan, X. Ji, J. Jiang, J. Yu, L. Miao, L. Zhang, S. Bie, H. Chen and Y. Ruan, *J. Power Sources*, 2013, **243**, 396–402.
- 9 R. B. Rakhi, N. a. Alhebshi, D. H. Anjum and H. N. Alshareef, *J. Mater. Chem. A*, 2014, **2**, 16190–16198.
- 10 D. P. Dubal, G. S. Gund, C. D. Lokhande and R. Holze, *Energy Technol.*, 2014, **2**, 401–408.
- 11 Y. Tang, Y. Liu, S. Yu, S. Mu, S. Xiao, Y. Zhao and F. Gao, *J. Power Sources*, 2014, **256**, 160–169.
- 12 W. Liu, X. Li, M. Zhu and X. He, *J. Power Sources*, 2015, **282**, 179–186.
- 13 T. Chen, Y. Tang, Y. Qiao, Z. Liu, W. Guo, J. Song, S. Mu, S. Yu, Y. Zhao and F. Gao, *Sci. Rep.*, 2016, **6**, 23289.
- 14 J. Chen, J. Xu, S. Zhou, N. Zhao and C. Wong, *J. Mater. Chem. A*, 2015, **3**, 17385–17391.
- 15 H. Fan, R. Niu, J. Duan, W. Liu and W. Shen, *ACS Appl. Mater. Interfaces*, 2016, acsami.6b05415.
- 16 J. Shi, X. Li, G. He, L. Zhang and M. Li, *J. Mater. Chem. A*, 2015, **3**, 20619–20626.