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# SiOC Nanolayers Wrapped 3D Interconnected Graphene Sponge as High-Performance Anodes for Lithium Ion Batteries

Zhiyuan Sang[a], Zhihao Zhao[a], Dong Su[a*],Peishuang Miao[a], Fengrui Zhang[a], Huiming Ji[a*], Xiao Yan[b]

[a] Key Laboratory for Advanced Ceramics and Machining Technology of Ministry of Education, School of Materials Science and Engineering, Tianjin University, Tianjin 300350, P.R. China

[b] Guangdong Key Laboratory of Membrane Materials and Membrane Separation, Guangzhou Institute of Advanced Technology, Chinese Academy of Sciences, Guangzhou 511452, China

[*]Corresponding author: Dong Su (E-mail: sudong@tju.edu.cn, Tel: 0086-13920782557)
[*]Corresponding author: Huiming Ji (E-mail: jihuiming@tju.edu.cn, Tel: 13642050311)

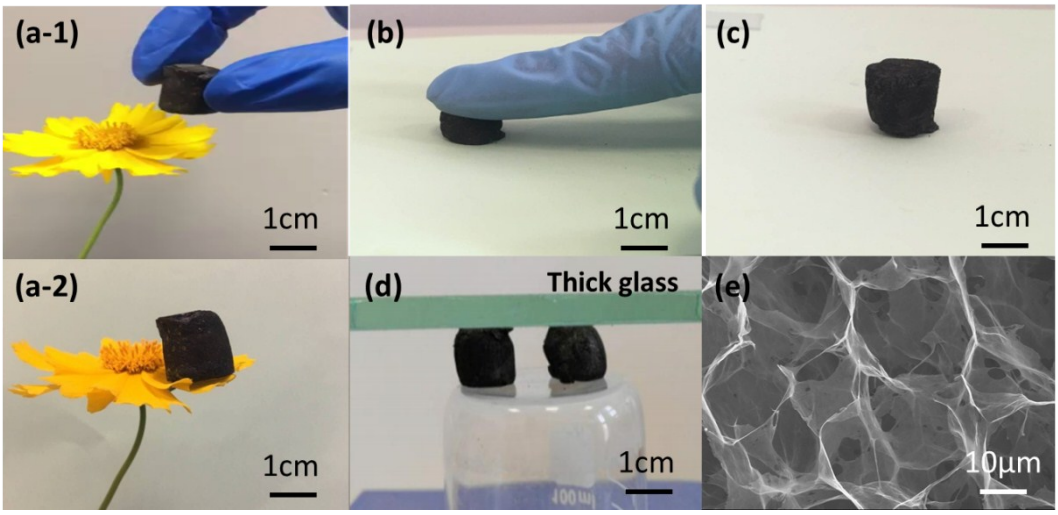



Fig. S1 Ultra-light , high-strength and compressibility of the 3D-GNS

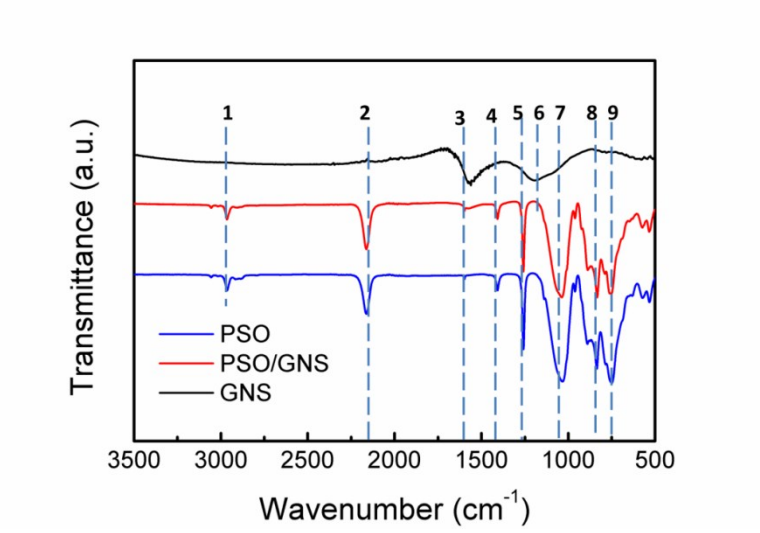



Fig. S2 Fourier transform infrared (FTIR) spectrum of the 3D-GNS, PSO and 3D-PSO/GNS.

1. C-H 2. Si-H 3.C=C 4. Si-$CH_3$/O-H 5. Si−$CH_3$ /C-OH 6. C-O/C-O-C 7. Si-O 8-9. Si-C

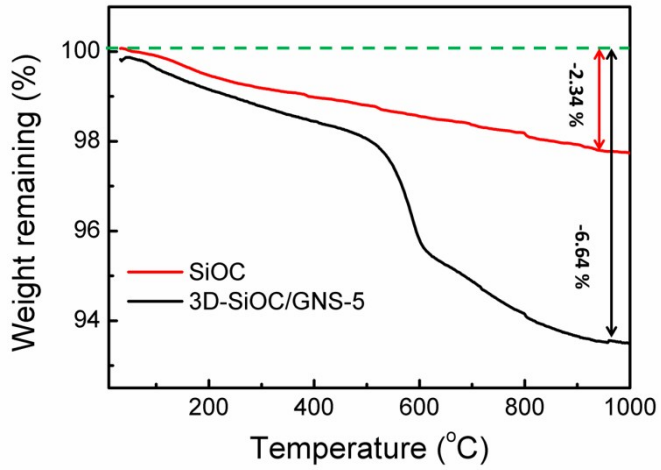



Fig. S3 TG analysis of the SiOC and 3D-GNS/SiOC-5.

Thermogravity analysis (TGA, Netzsch STA 449F3, Germany) was carried with a heating rate of 10℃/min under air to determined the ratio of GNS in the nanocomposites.

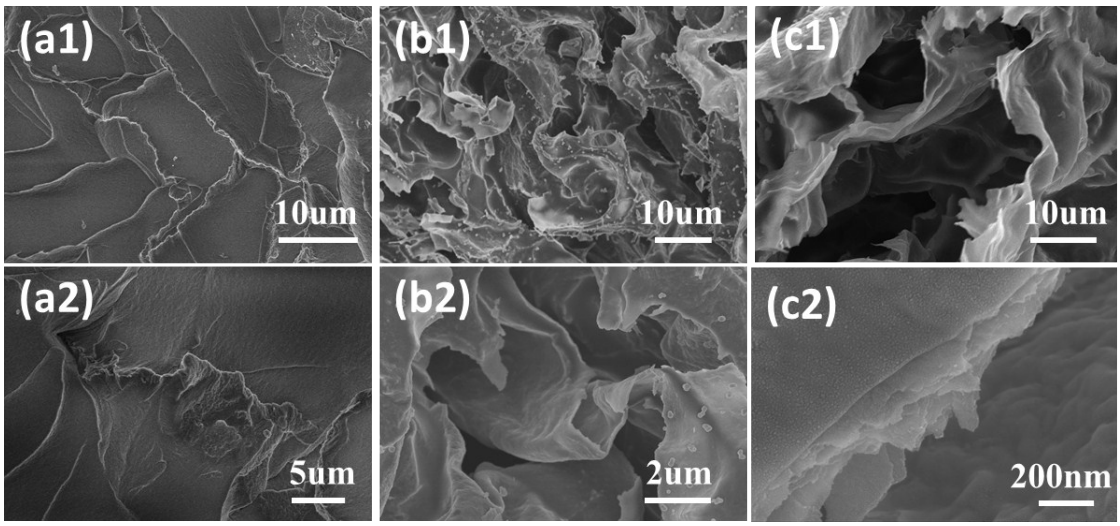



Fig. S4 SEM morphologies of 3D-GNS/SiOC with different content of GNS: (a) 3D-GNS/SiOC-L (1wt.%-GNS) (b) 3D-GNS/SiOC (5wt.%-GNS) (c) 3D-GNS/SiOC-H (10wt.%-GNS)

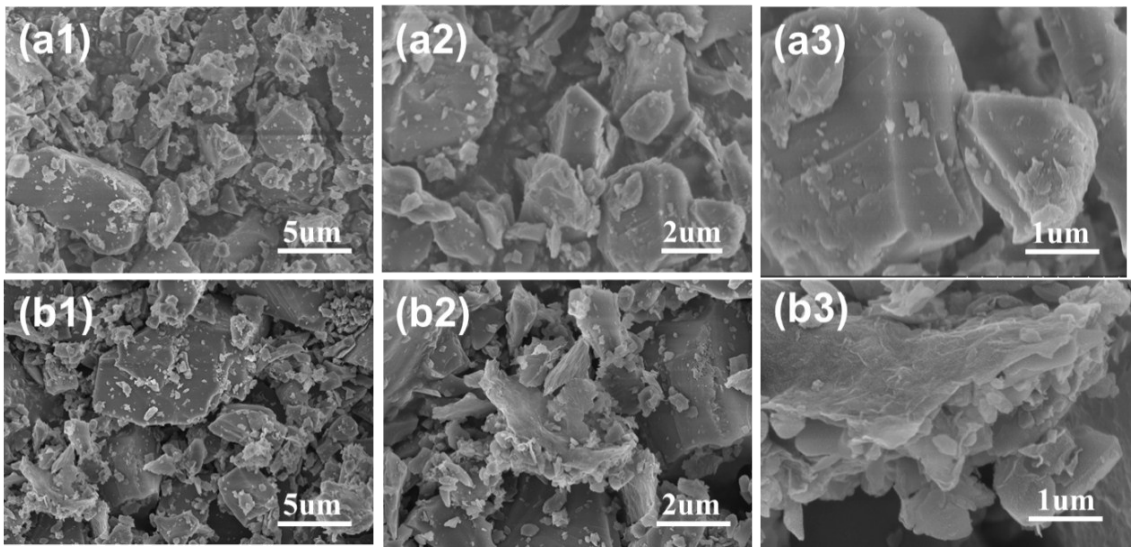
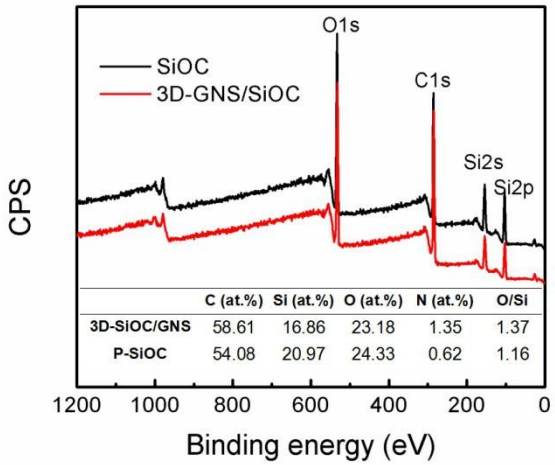



Fig. S5 SEM morphologies of (a) SiOC and (b) m-GNS/SiOC




| | C (at.%) | Si (at.%) | O (at.%) | N (at.%) | O/Si |
|---|---|---|---|---|---|
| 3D-SiOC/GNS | 58.61 | 16.86 | 23.18 | 1.35 | 1.37 |
| P-SiOC | 54.08 | 20.97 | 24.33 | 0.62 | 1.16 |

Fig. S6 XPS survey spectrums of SiOC and 3D-GNS/SiOC.

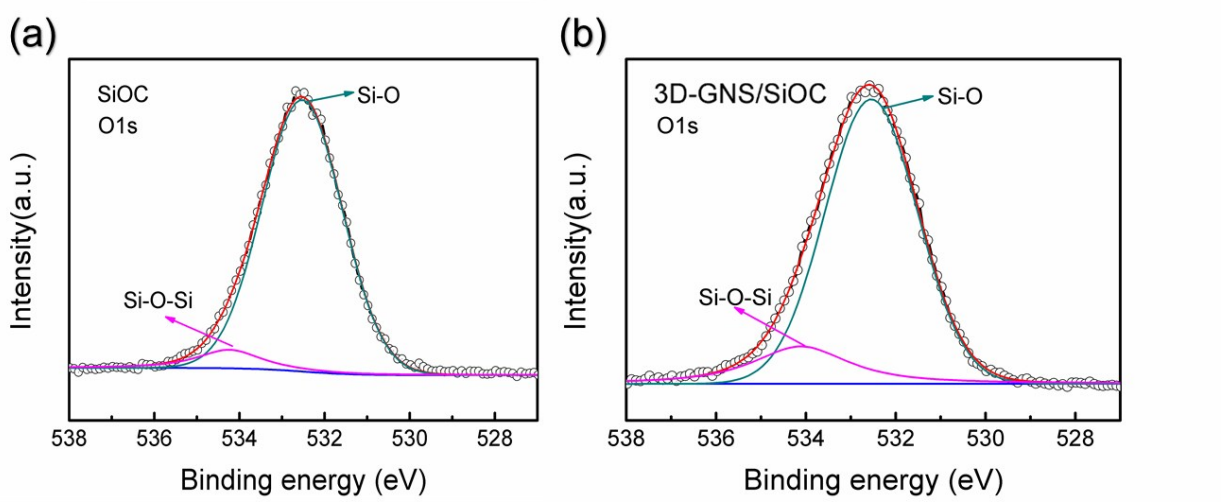



Fig. S7 High-resolution XPS spectra of O1s of SiOC and 3D-GNS/SiOC.

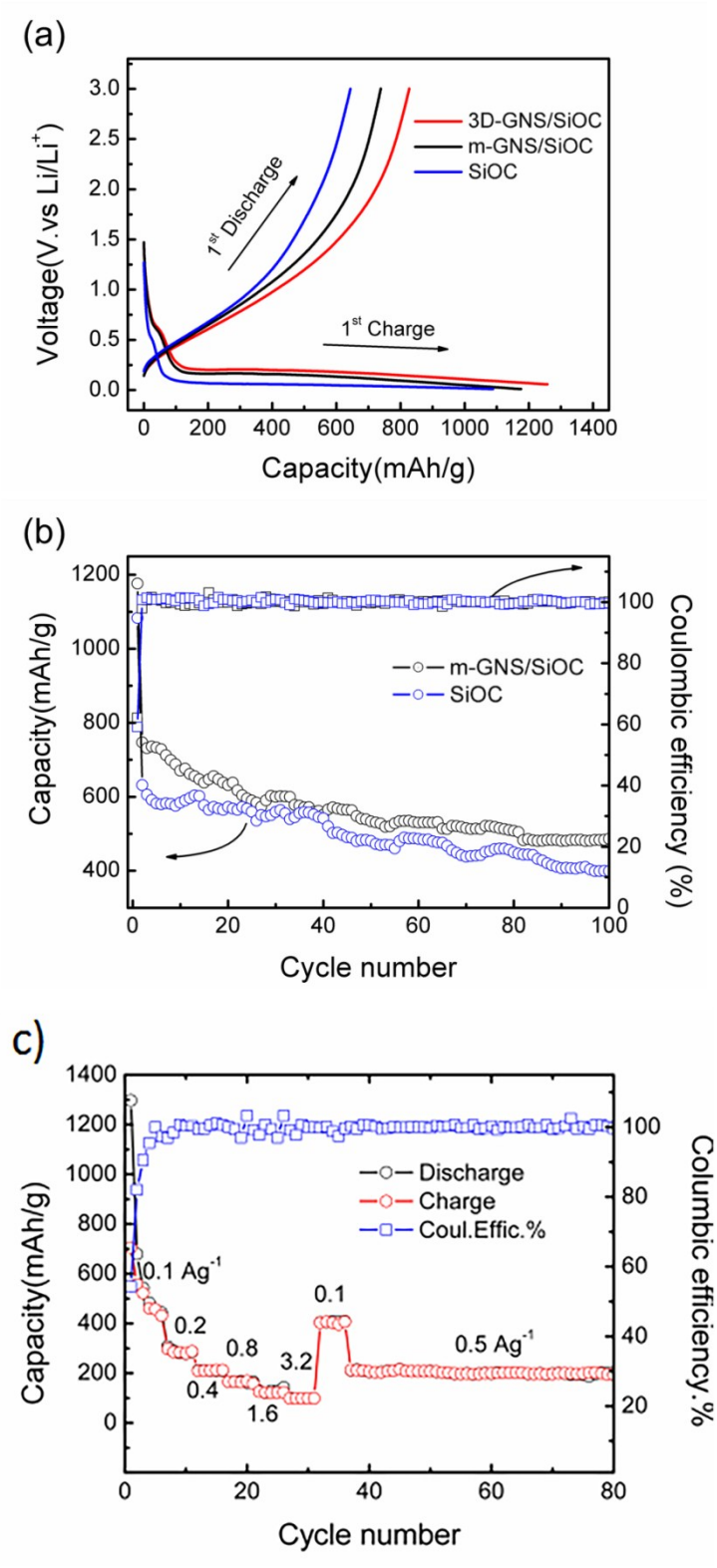



Fig. S8 Electrochemical performances:(a) discharge/charge profiles of p-SiOC, m-GNS/SiOC and 3D-GNS/SiOC at 0.1 A/g (b) the cycling performance of SiOC, m-GNS/SiOC at 0.1 A/g. (c) the rate and cycling performance of 3D-GNS.

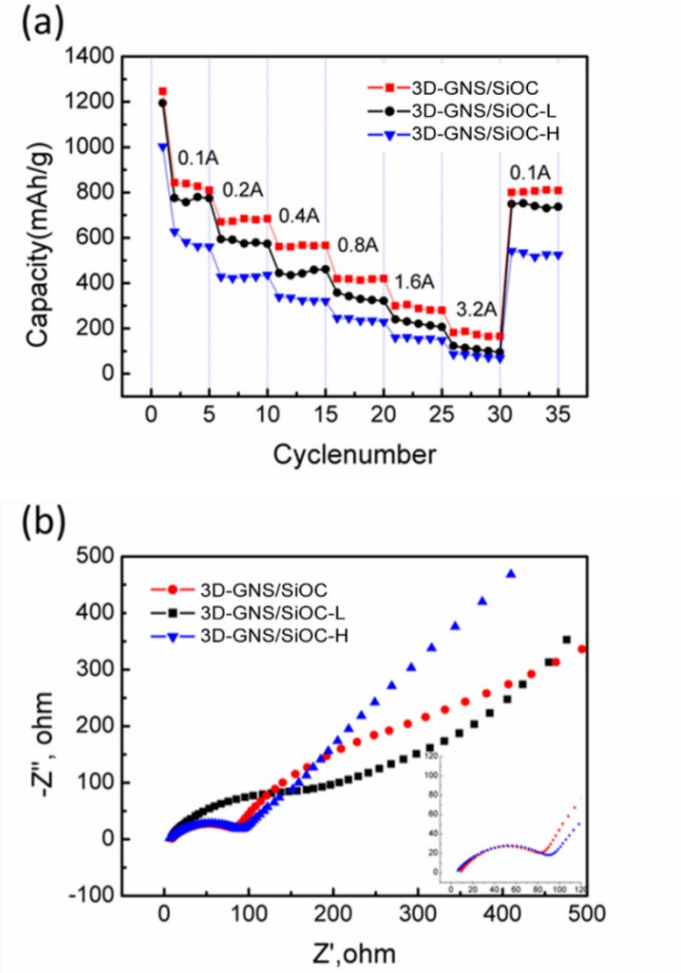



Fig. S9 The electrochemical performance of 3D-GNS/SiOC, 3D-GNS/SiOC-L and 3D-GNS/SiOC-H: (a) the rate performance (b) the EIS plots.

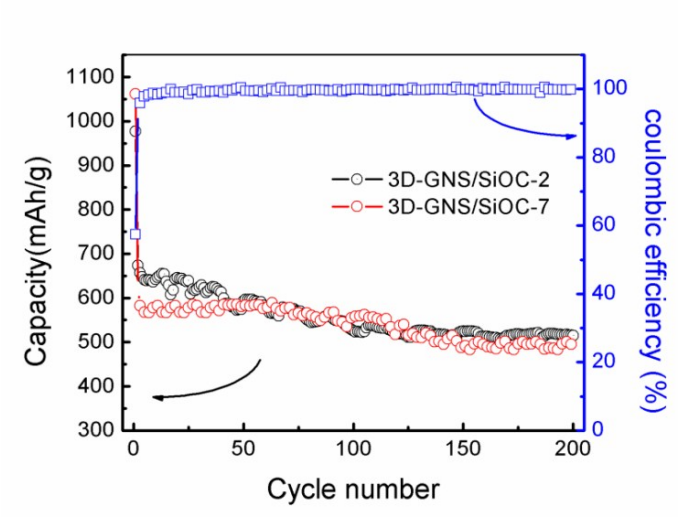



Fig. S10 The cycling performance of 3D-GNS/SiOC-2 and 3D-GNS/SiOC-7 at 0.5A $g^{-1}$ (3D-GNS/SiOC-X, where X shows the percent of GNS loading about 2wt.% and 7wt.%, respectively).

Table S1. Peak assignment of Si2p of SiOC and 3D-GNS/SiOC

| Sample | Binding Energy (eV) | Assignment | Atom% |
|---|---|---|---|
| P-SiOC | 101.2 | $SiOC_3$ | 16.88 |
| | 102.1 | $SiO_2C_2$ | 35.12 |
| | 103.0 | $SiO_3C$ | 33.44 |
| | 104.2 | $SiO_4$ | 14.56 |
| 3D-GNS/SiOC | 101.4 | $SiOC_3$ | 13.53 |
| | 102.0 | $SiO_2C_2$ | 28.25 |
| | 103.1 | $SiO_3C$ | 37.82 |
| | 104.4 | $SiO_4$ | 20.40 |

Table S2. Peak assignment of C1s and O1s of SiOC and 3D-GNS/SiOC, respectively

| Sample | Element | Binding Energy (eV) | Assignment | Atom% |
|---|---|---|---|---|
| P-SiOC | C 1s | 284.6 | $sp^2/sp^3$ C-C | 77.05 |
| | | 286.5 | C-O | 6.13 |
| | | 283.7 | C-Si | 16.82 |
| | O1s | 532.5 | Si-O | 93.08 |
| | | 534.2 | Si-O-Si | 6.92 |
| 3D-GNS/SiOC | C 1s | 284.6 | $sp^2/sp^3$ C-C | 80.88 |
| | | 286.4 | C-O | 14.38 |
| | | 283.8 | C-Si | 4.74 |
| | O1s | 532.5 | Si-O | 84.10 |
| | | 534.1 | Si-O-Si | 15.90 |

Table S3. EIS fitting results of SiOC, m-GNS/SiOC and 3D-GNS/SiOC anode

| Parameter | SiOC | m-GNS/SiOC | 3D-GNS/SiOC |
|---|---|---|---|
| $R_o$/ Ω | 9.397 | 4.864 | 3.069 |
| $R_{ct}$/ Ω | 161.4 | 81.76 | 44.7 |
| CPE-T/μF | 14.329 | 29.1 | 31.45 |
| Ws-R/ Ω | 25554 | 22609 | 8991 |

Table S4. Comparison of electrochemical properties of the 3D-GNS/SiOC composites with other Si-O-C anode materials for LIBs

| Samples | Current density | Cycles | Capacity (mAh $g^{-1}$) | Ref./Year |
|---|---|---|---|---|
| C-rich SiOC | 74 mA $g^{-1}$ | 30 | 460 | 2016[1] |
| HF-etched SiOC | 200 mA $g^{-1}$ | 100 | 572 | 2016[2] |
| SiOC aerogel | 360 mA $g^{-1}$ | 50 | 600 | 2015[3] |
| SiOC/C fiber | 50 mA $g^{-1}$ | 60 | 669 | 2014[4] |
| SiOC/BN nanotubes | 100 mA $g^{-1}$ | 25 | 450 | 2017[5] |
| SiOC-CNT Shell/Core | 100 mA $g^{-1}$ | 40 | 686.3 | 2013[6] |
| Intercalated SiOC/GNS | 50 mA $g^{-1}$ | 90 | 582 | 2015[7] |
| Layered SiOC/GNS | 40 mA $g^{-1}$ | 20 | 357 | 2009[8] |
| SiOC/GNS Paper | 100 mA $g^{-1}$ | 1020 | 588 | 2016[9] |
| SiOC/GNS Paper | 100 mA $g^{-1}$ | 30 | 400 | 2016[10] |
| SiOx-C/GNS | 100 mA $g^{-1}$ | 250 | 630 | 2015[11] |
| 3D $SiO_2$@Graphene Aerogel | 500 mA $g^{-1}$ | 300 | 300 | 2015[12] |
| **3D- GNS/SiOC** | **500 mA $g^{-1}$** | **200** | **586.6** | **This work** |
| | **100 mA $g^{-1}$** | **100** | **701.5** | **This work** |

## Reference


1. J. Kaspar, M. Graczyk-Zajac, S. Choudhury and R. Riedel, *Electrochim. Acta*, 2016, **216**, 196-202
2. M. Ma, H. Wang, M. Niu, L. Su, X. Fan, J. Deng, Y. Zhang and X. Du, *RSC Adv.*, 2016, **6**, 43316-43321.
3. V. S. Pradeep, D. G. Ayana, M. Graczyk-Zajac, G. D. Soraru and R. Riedel, *Electrochim. Acta,*, 2015, **157**, 41-45
4. Li, Y.; Hu, Y.; Lu, Y.; Zhang, S.; Xu, G.; Fu, K.; Li, S.; Chen, C.; Zhou, L.; Xia, X.; Zhang, X. *J. Power Sources,* **2014,** 254, 33-38.
5. Abass, M. A., Syed, A. A., Gervais, C., & Singh, G, *Rsc Adv.,* 2017, **7**, 21576-21584.
6. R. Bhandavat and G. Singh, *J. Phys. Chem. C*, 2013, **117**, 11899-11905.
7. Y. Ren, B. Yang, X. Huang, F. Chu, J. Qiu and J. Ding, *Solid State Ionics*, 2015, **278**, 198-202
8. F. Ji, Y. L. Li, J. M. Feng, D. Su, Y. Y. Wen, Y. Feng and F. Hou, *J. Mater. Chem.*, 2009, **19**, 9063-9067.
9. L. David, R. Bhandavat, U. Barrera and G. Singh, *Nat. Commun.,* 2016, **7**, 10998.
10. L. David, K. M. Shareef, M. A. Abass and G. Singh, *Rsc Adv.*, 2016, **6**, 53894-53902.
11. M. Q. Li, Y. Yu, J. Li, B. L. Chen, A. Konarov and P.Chen, J*. Power Sources,* 2015, **293**, 976-982.
12. J. Meng, Y. Cao, Y. Suo, Y. Liu, J. Zhang and X. Zheng, *Electrochim. Acta*, 2015, **176,** 1001-1009.