

## Supplementary

# Chemically engineered alloy anode enabling fully reversible conversion reaction: Design of C-Sn bonded aerofilm anode

*Sun-Sik Kim,<sup>‡a</sup> Chenrayan Senthil,<sup>‡a</sup> Sung Mi Jung<sup>b</sup> and Hyun Young Jung<sup>\*ac</sup>*

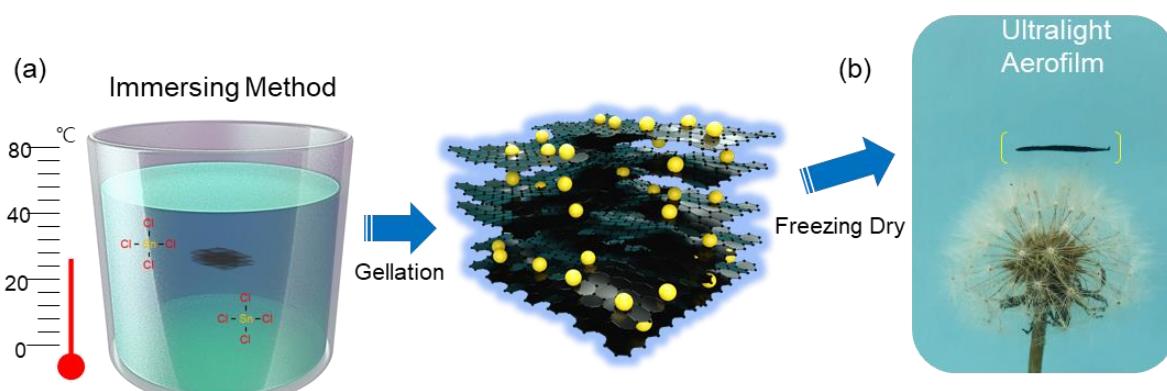
<sup>a</sup> Department of Energy Engineering, Gyeongsang National University, Jinju-si, Gyeongnam 52725, South Korea

<sup>b</sup> Environmental Fate & Exposure Research Group, Korea Institute of Toxicology, Jinju-si, Gyeongnam 52834, South Korea

<sup>c</sup> Future Convergence Technology Research Institute, Gyeongsang National University, Jinju-si, Gyeongnam 52725, South Korea

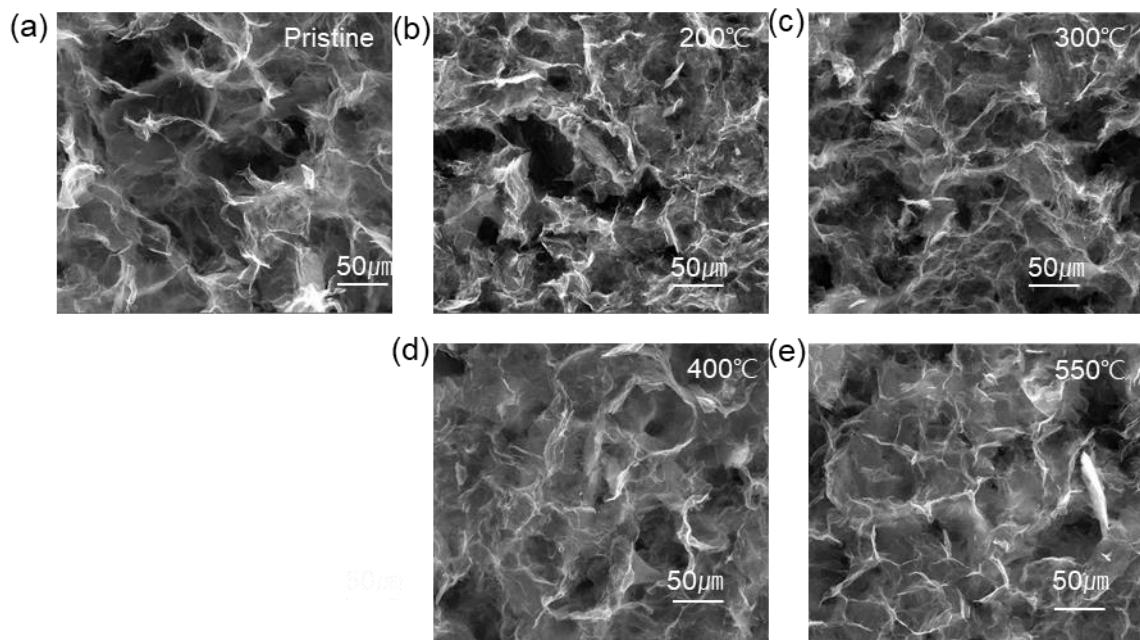
\* To whom correspondence should be addressed. E-mail: [hyjung@gnu.ac.kr](mailto:hyjung@gnu.ac.kr) (Prof. H.Y. Jung)

‡ Authors equally contributed

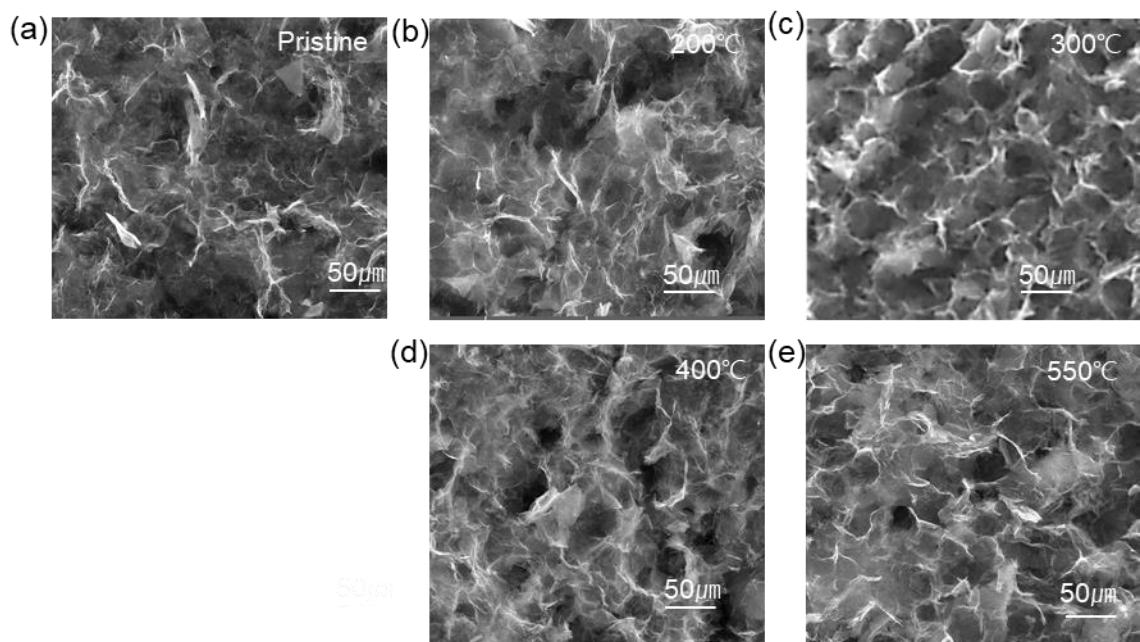


**Fig. S1.** (a) Formation of C-Sn bonded aerofilm anode by a simple immersion technique. (b) Ultralight nature of aerofilm anode placed over a dandelion flower.

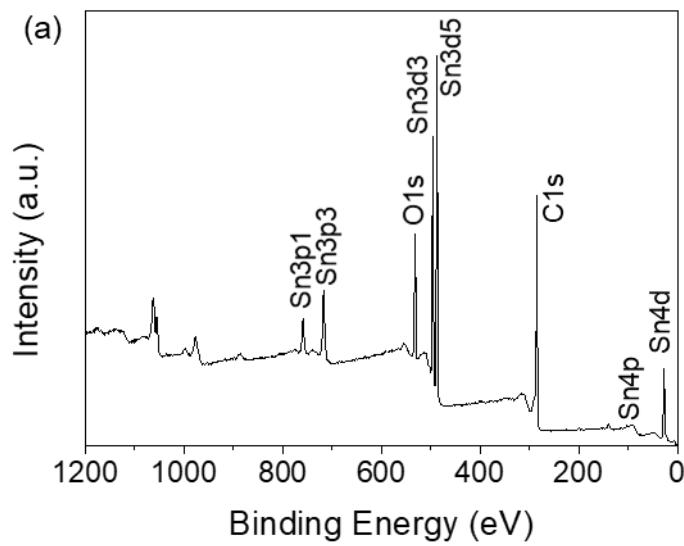
Spot 1



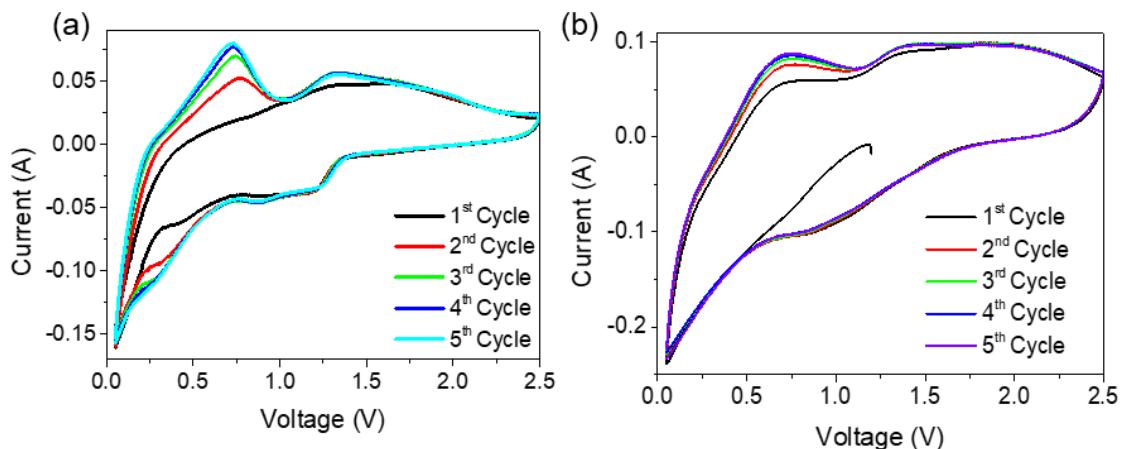
Spot 2



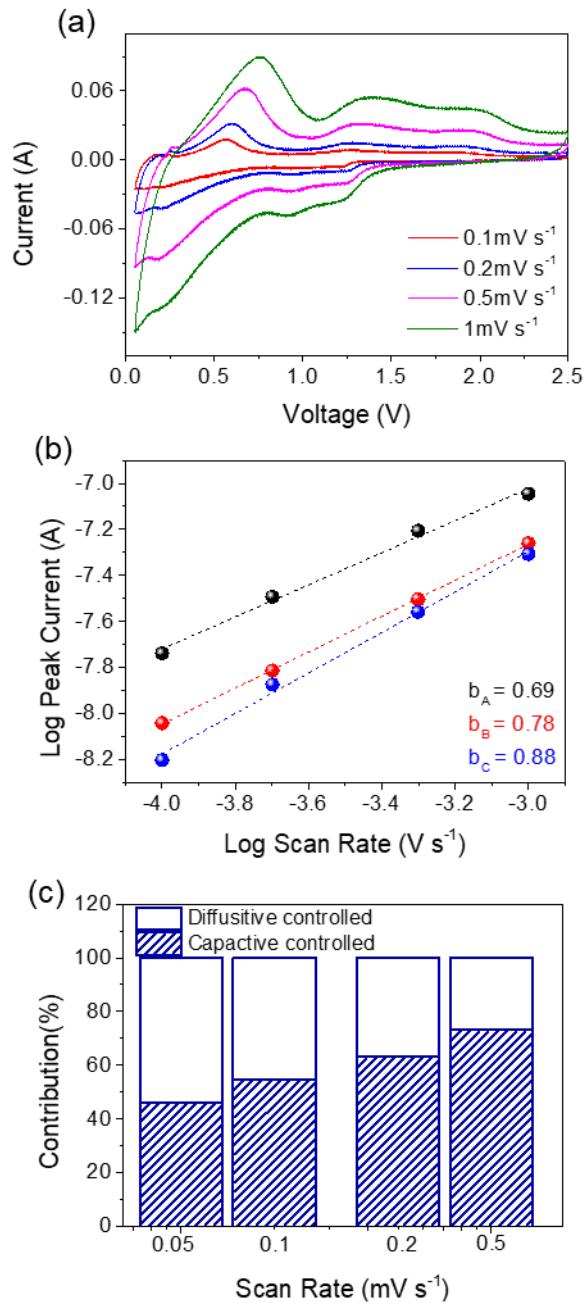
**Fig. S2.** SEM micrographs at different spots (spot 1 and spot 2) depicting the morphological understanding in SnO<sub>2</sub> aerofilm anode annealed at 200, 300, 400, and 550°C compared with no annealed anode.



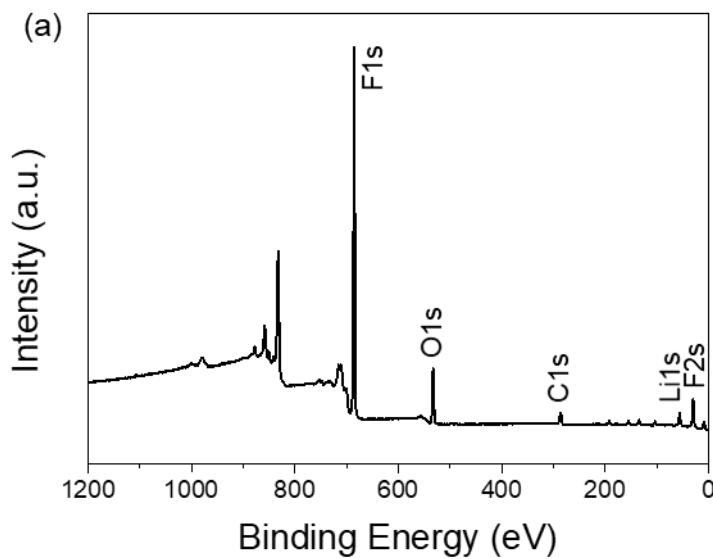
**Fig. S3.** (a) XPS survey for C-Sn bonded aerofilm anode.



**Fig. S4.** Cyclic voltammetry curves during the initial five cycles at  $1\text{mV s}^{-1}$  (a) C-O-Sn and (b) C-Sn bonded anode.



**Fig. S5.** (a) Cyclic voltammetry curves of C-O-Sn bonded anode at various scan rates of 0.1, 0.2, 0.5, 1.0 mV<sup>-1</sup>. (b) b-value from (a). (c) Diffusion and capacitive controlled Li storage contribution.



**Fig. S6.** XPS survey of cycled and lithiated C-Sn bonded aerofilm anode.

## Cell parameters

The specific capacity was calculated based on the mass of  $\text{SnO}_2$ , which is considered as the active material. The details of cell parameters are as follows,

Mass of G- $\text{SnO}_2$  : 1.56 – 2.1 mg/cm<sup>2</sup>,

Mass of  $\text{SnO}_2$  : 1.11 g – 1.49 mg/cm<sup>2</sup>,

Current rate : 1 C (0.79 A/g based on theoretical capacity of  $\text{SnO}_2$ ).

**Table S1.** Comparison of resistance and diffusion coefficient for C-Sn and C-O-Sn aerofilm anode on EIS.

	Rs (kΩ)	Rsei (kΩ)	Rct (kΩ)	D <sub>Li</sub> (cm <sup>2</sup> s <sup>-1</sup> )
C-O-Sn	12.1	0.75	0.36	1.67E-11
C-Sn	6.73	0.49	0.153	1.54E-10

**Table S2.** Electrochemical performance comparison of previously reported graphene-SnO<sub>2</sub> composite electrode for Li-ion battery.

Electrode (Content)	Synthetic method	Voltage Window (V)	1 <sup>st</sup> Cycle Capacity (mAh g <sup>-1</sup> )	2 <sup>nd</sup> Cycle Capacity (mAh g <sup>-1</sup> )	Condition (A g <sup>-1</sup> )	Retention rate (1 <sup>st</sup> and 2 <sup>nd</sup> Cycle)	Ref.
<b>Graphene-SnO<sub>2</sub> aerofilm</b>	Immersing	0.005~2.5	1183	1158	0.158	98%	Our Work
			958	951	0.790	99%	
Graphene/Sn @ carbonaceous foam	Hydrothermal	0.001~3	~1300	~800	0.1	62%	S1
Sn@3D graphene networks	Freeze drying and CVD	0.005~3	~1800	~1200	0.2	67%	S2
SnO <sub>2</sub> quantum dots@GO	Hydrothermal	0.01~3	~1500	~1000	0.1	67%	S3
F-doped SnO <sub>2</sub> @rGO	Hydrothermal	0.01~3	~2000	~1250	0.1	63%	S4
SnO <sub>2</sub> /graphene	hydrolysis	0.01~2	~980	~920	0.067	94%	S5
SnO <sub>2</sub> /CNT-GN composite	Hydrothermal	0.01~3	~1800	~1100	0.2	61%	S6
3D annealed SnO <sub>2</sub> /graphene foams	Hydrothermal	0.01~3	~1650	~1000	0.2	61%	S7
Tin graphene tube	Hydrothermal	0.01~2.5	~1300	~900	0.2	69%	S8
N-Doped Gr_SnO <sub>2</sub>	Solution	0.05~3	~1450	~900	0.05	62%	S9
Graphene Nanoribbon and Nanostructured SnO <sub>2</sub>	Solution refluxing	0.01~2.5	~1500	~1110	0.1	74%	S10
SnO <sub>2</sub> /RGO	Hydrothermal	0.01~3	~1542	~837	0.5	54%	S11

**Table S3.** Cycle stability comparison of previously reported graphene-SnO<sub>2</sub> composite electrode for Li-ion battery.

Electrode (Content)	Voltage Window (V)	Cyclability (mAh g <sup>-1</sup> )	Rate performance (mAh g <sup>-1</sup> )	Ref.
Graphene-SnO <sub>2</sub> aerofilm	0.005~2.5	1255 (200cycle at 158mA g <sup>-1</sup> )	553 (1500cycle at 0.79A g <sup>-1</sup> )	Our Work
Graphene/Sn@carbonaceous foam	0.001~3	777 (100 cycles at 100 mA g <sup>-1</sup> )	506 (500cycle at 400 mA g <sup>-1</sup> ) 270 (500cycle at 3200 mA g <sup>-1</sup> )	S1
Graphene/Sn @ carbonaceous foam	0.005~3	1,089 (100cycle at 200mA g <sup>-1</sup> )	459 (at 5 A/g) 270 (at 10 A g <sup>-1</sup> )	S2
Sn@3D graphene networks	0.01~3	112 (100 cycles at 100 mA g <sup>-1</sup> )	417 (2,000 cycle at 2 A g <sup>-1</sup> )	S3
SnO <sub>2</sub> quantum dots@GO	0.01~3	1,037 (150 cycles at 100 mA g <sup>-1</sup> )	860 (at 1 A/g) 770 (at 2 A g <sup>-1</sup> )	S4
F-doped SnO <sub>2</sub> @rGO	0.01~2	840 (30 cycle at 67mA g <sup>-1</sup> )	590 (50cycle at 400mA g <sup>-1</sup> ) 270 (50cycle at 1000mA g <sup>-1</sup> )	S5
SnO <sub>2</sub> /graphene	0.01~3	809 (100cycle at 200mA g <sup>-1</sup> )	787 (at 500mA g <sup>-1</sup> )	S6
SnO <sub>2</sub> /CNT-GN composite	0.01~3	984.2 (at 200mA g <sup>-1</sup> )	533.7 (150cycle at 1A g <sup>-1</sup> )	S7
3D annealed SnO <sub>2</sub> /graphene sheet foams	0.01~2.5	916 (500cycle at 200mA g <sup>-1</sup> )	810 (500cycle at 0.5A g <sup>-1</sup> )	S8
tin graphene tube	0.05~3	894 (at 50mA g <sup>-1</sup> )	-	S9
N-Doped Gr_SnO <sub>2</sub>	0.01~2.5	825 (50cycle at 100mA g <sup>-1</sup> )	580 (at 2A g <sup>-1</sup> )	S10
Graphene Nanoribbon and Nanostructured SnO <sub>2</sub>	0.01~3	708 (150cycle at 500mA g <sup>-1</sup> )	520 (at 1A g <sup>-1</sup> )	S11

**Table S4.** Comparison of SnO<sub>2</sub> aerofilm anode with commercial graphite

Electrode (Content)	Voltage Window (V)	Cyclability (mAh g <sup>-1</sup> )	Rate performance (mAh g <sup>-1</sup> )	Ref.
Commercial graphite	0~2	260 (100cycle at 200mA g <sup>-1</sup> )	153 (300cycle at 1A g <sup>-1</sup> )	S12
Graphene	0.01~3.5	540 (at 50mA g <sup>-1</sup> )	-	S13
CNT	0.01~3	460 (at 20mA g <sup>-1</sup> )	-	S14
N doping carbon	0.01~3	2053 (80cycle at 100mA g <sup>-1</sup> )	879 (1000cycle at 5A g <sup>-1</sup> )	S15
<b>C-Sn bonded aerofilm</b>	<b>0.005~2.5</b>	<b>1255 (200cycle at 158mA g<sup>-1</sup>)</b>	<b>553 (1500cycle at 0.79A g<sup>-1</sup>)</b>	<b>Our Work</b>

## References

- S1. B. Luo, T. Qiu, D. Ye, L. Wang, L. Zhi, Tin nanoparticles encapsulated in graphene backboned carbonaceous foams as high-performance anodes for lithium-ion and sodium-ion storage, *Nano Energy* 22 (2016) 232-240, <https://doi.org/10.1016/j.nanoen.2016.02.024>.
- S2. J. Qin, C. He, N. Zhao, Z. Wang, C. Shi, E.-Z. Liu, J. Li, Graphene networks anchored with Sn@ graphene as lithium ion battery anode, *ACS Nano* 8 (2014) 1728-1738, <https://doi.org/10.1021/nn406105n>.
- S3. K. Zhao, L. Zhang, R. Xia, Y. Dong, W. Xu, C. Niu, L. He, M. Yan, L. Qu, L. Mai, SnO<sub>2</sub> Quantum dots@graphene oxide as a high-rate and long-life anode material for lithium-ion batteries, *small* 12 (2016) 588-594, <https://doi.org/10.1002/smll.201502183>.
- S4. D. Cui, Z. Zheng, X. Peng, T. Li, T. Sun, L. Yuan, Fluorine-doped SnO<sub>2</sub> nanoparticles anchored on reduced graphene oxide as a high-performance lithium ion battery anode, *J. Power Sources* 362 (2017) 20-26, <https://doi.org/10.1016/j.jpowsour.2017.07.024>.
- S5. X. Wang, X. Zhou, K. Yao, J. Zhang, Z. Liu, A SnO<sub>2</sub>/graphene composite as a high stability electrode for lithium ion batteries, *Carbon* 49 (2011) 133-139, <https://doi.org/10.1016/j.carbon.2010.08.052>.
- S6. M.-S. Wang, Z.-Q. Wang, Z.-L. Yang, Y. Huang, J. Zheng, X. Li, Carbon nanotube-graphene nanosheet conductive framework supported SnO<sub>2</sub> aerogel as a high performance anode for lithium ion battery, *Electrochim. Acta* 240 (2017) 7-15, <https://doi.org/10.1016/j.electacta.2017.04.031>.
- S7. M. Zhang, Z. Sun, T. Zhang, D. Sui, Y. Ma, Y. Chen, Excellent cycling stability with high SnO<sub>2</sub> loading on a three-dimensional graphene network for lithium ion batteries, *Carbon* 102 (2016) 32-38, <https://doi.org/10.1016/j.carbon.2016.02.032>.
- S8. A. Dhanabalan, X. Li, R. Agrawal, C. Chen, C. Wang, Fabrication and characterization of SnO<sub>2</sub>/graphene composites as high capacity anodes for Li-ion batteries, *Nanomaterials* 3 (2013) 606-614, <https://doi.org/10.3390/nano3040606>.
- S9. R. Tian, Y. Zhang, Z. Chen, H. Duan, B. Xu, Y. Guo, H. Kang, H. Li, H. Liu, The effect of annealing on a 3D SnO<sub>2</sub>/graphene foam as an advanced lithium-ion battery anode, *Sci. Rep.* 6 (2016) 1-9, <https://doi.org/10.1038/srep19195> (2016).

- S10. Z.-F. Li, Q. Liu, Y. Liu, F. Yang, L. Xin, Y. Zhou, H. Zhang, L. Stanciu, J. Xie, Facile preparation of graphene/SnO<sub>2</sub> xerogel hybrids as the anode material in Li-ion batteries, ACS Appl. Mater. Interfaces 7 (2015) 27087-27095, <https://doi.org/10.1021/acsami.5b05819>.
- S11. Q. Tan, Z. Kong, X. Chen, L. Zhang, X. Hu, M. Mu, H. Sun, X. Shao, X. Guan, M. Gao, Synthesis of SnO<sub>2</sub>/graphene composite anode materials for lithium-ion batteries, App. Surf. Sci. 485 (2019) 314-322. <https://doi.org/10.1016/j.apsusc.2019.04.225>.
- S12. J.Kim, S.M.N.Jeghan, G.Lee, Superior fast-charging capability of graphite anode via facile surface treatment for lithium-ion batteries, Microporous Mesoporous Mater. 305 (2020) 110325. <https://doi.org/10.1016/j.micromeso.2020.110325>.
- S13. P.Lian, X.Zhu, S.Liang, Z.Li, W.Yang, H.Wang, Large reversible capacity of high quality graphene sheets as an anode material for lithium-ion batteries, Electrochim. Acta 55 (2010) 3909-3914. <https://doi:10.1016/j.electacta.2010.02.025>.
- S14. A.S.Claye, J.E.Fischer, C.B.Huffman, A.G.Rinzler, R.E.Smalley, Solid-State Electrochemistry of the Li Single Wall Carbon Nanotube System, J. Electrochem. Soc. 147 (2000) 2845-2852.
- S15. Y.Yang, S.Jin, Z.Zhang, Z.Du, H.Liu, J.Yang, H.Xu, H.Ji, Nitrogen-Doped Hollow Carbon Nanospheres for High-Performance Li-Ion Batteries, ACS Appl. Mater. Interfaces 9 (2017) 14180-14186. <https://doi.org/10.1021/acsami.6b14840>.