

Supporting Information

Low-temperature Molten Salt Ion Regeneration Strategy towards Green and Efficient Spent Graphite Recycle

Wenchao Zhu^a, Gonggang Liu^{a}, Jinbo Hu^a, Geng Su^a, Miaohua Liu^a, Xianjun Li^a,
Binghui Xu^{b*}*

- a.* Hunan Province Key Laboratory of Materials Surface & Interface Science and Technology,
College of Materials Science and Engineering, Central South University of Forestry and
Technology, Changsha 410004, China.
- b.* Institute of Materials for Energy and Environment, College of Materials Science and
Engineering, Qingdao University, Qingdao 266071, China.

Email: *liugonggang@csuft.edu.cn* (G. Liu); *xubinghuiqdu@qdu.edu.cn* (B. Xu)

1. Supplementary Fig.s and Tables

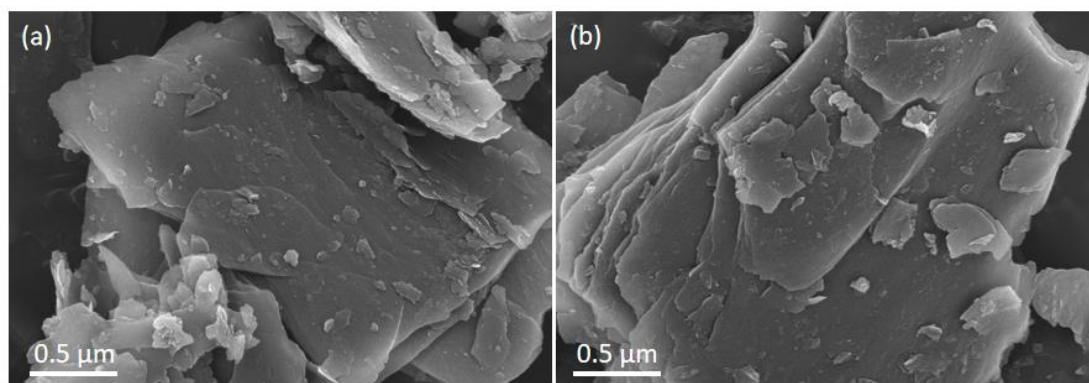


Fig. S1 SEM images of (a) SG and (b) CPG.

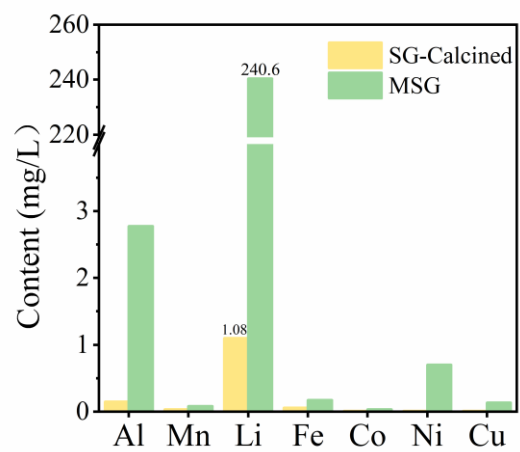


Fig. S2 ICP–OES test results of leaching solutions from SG-Calcined and MSG.

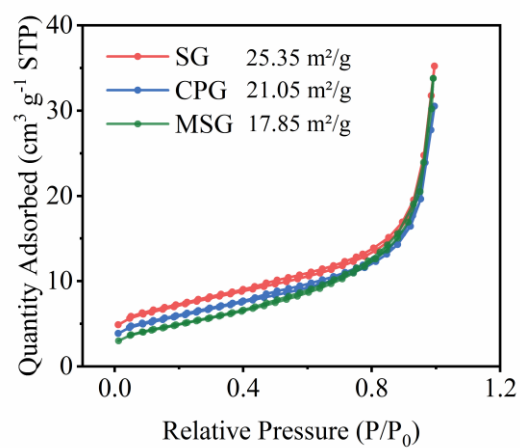


Fig. S3 N₂ adsorption and desorption curves of SG,CPG and MSG.

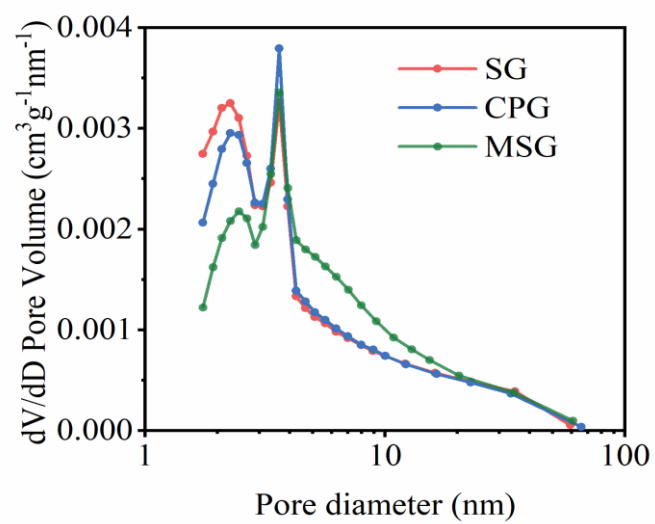


Fig. S4 Pore size distribution curves of SG,CPG and MSG.

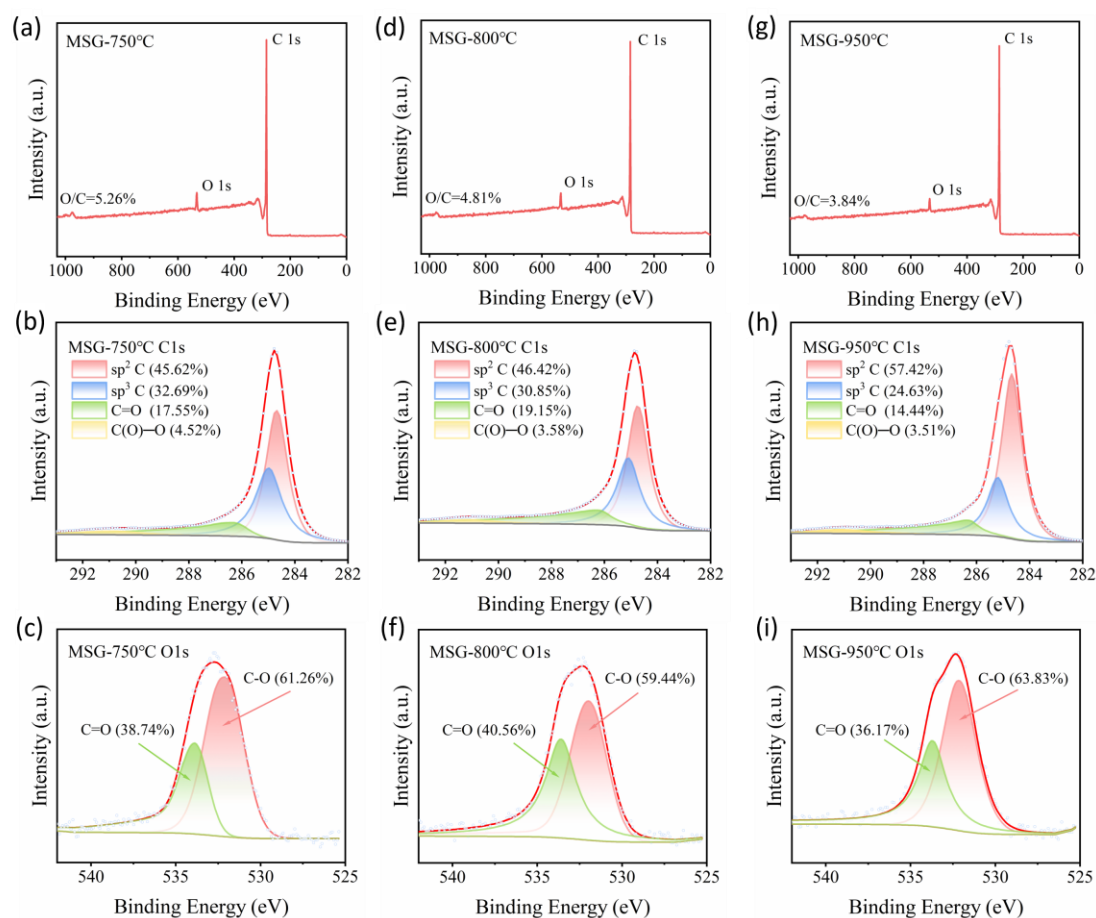


Fig. S5 Survey XPS and high-resolution C 1s and O 1s spectra for MSG-750 °C (a-c), MSG-800 °C (d-f) and MSG-950 °C (g-i).

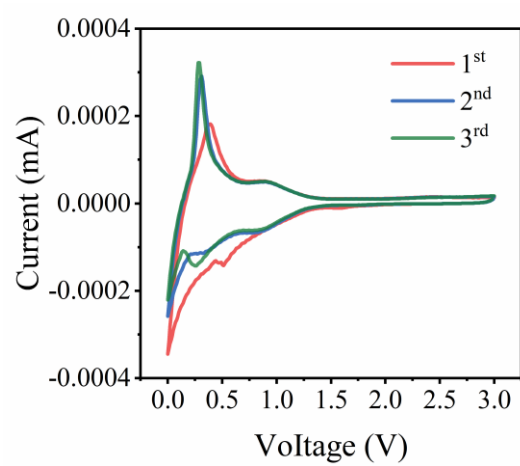


Fig. S6 Cyclic voltammetry curves from 1st to 3rd at 0.1 mV/s of SG.

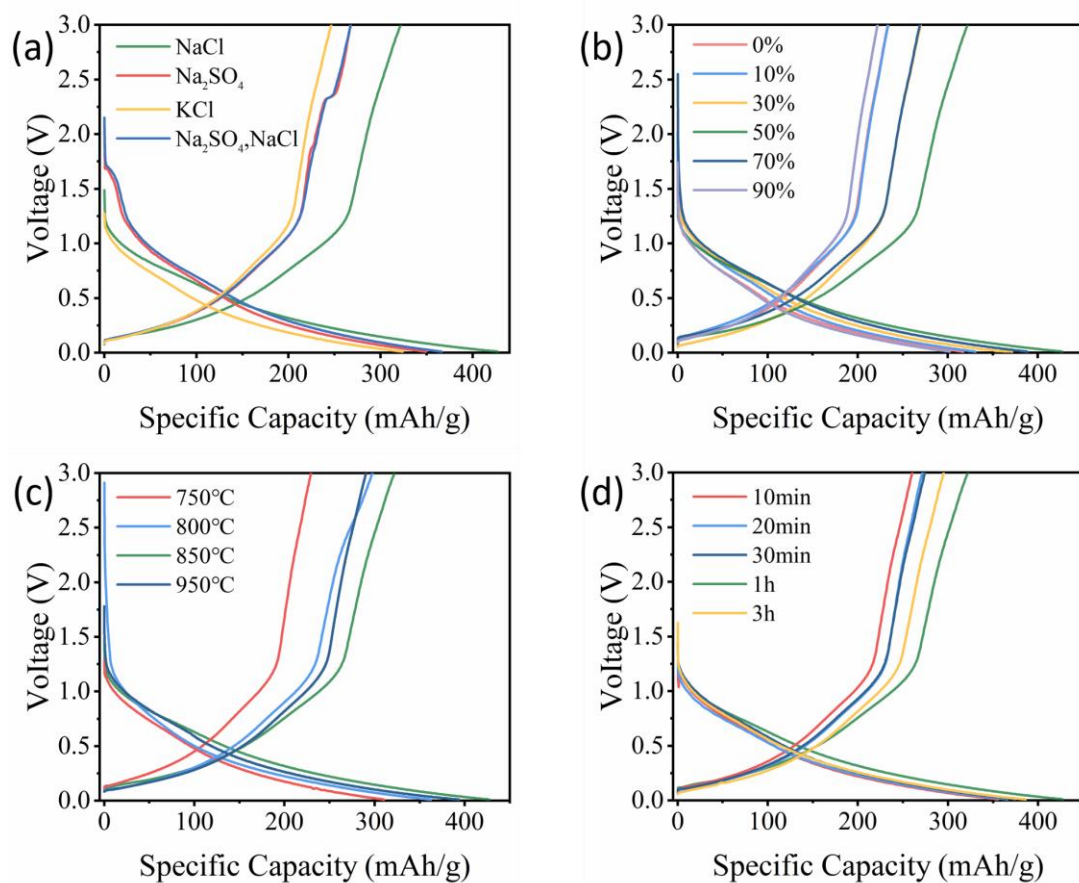


Fig. S7 Impact of different conditions on charge-discharge specific capacity: (a) types of inorganic salts (NaCl , Na_2SO_4 , KCl , $\text{Na}_2\text{SO}_4+\text{NaCl}$), (b) proportion of inorganic salts, (c) temperature, (d) time.

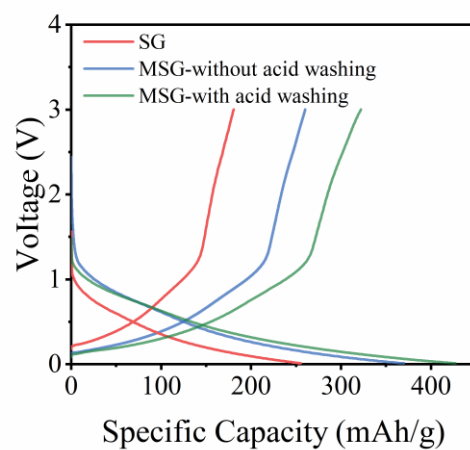


Fig. S8 The initial cycle charge–discharge profile of SG and MSG with/without acid washing.

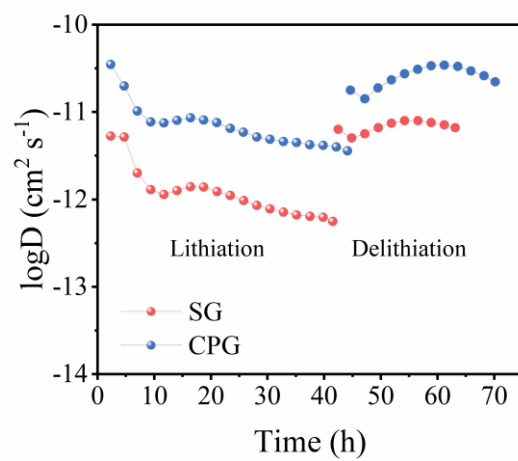


Fig. S9 Variation curve of Li⁺ diffusion coefficient of SG and CPG with time.

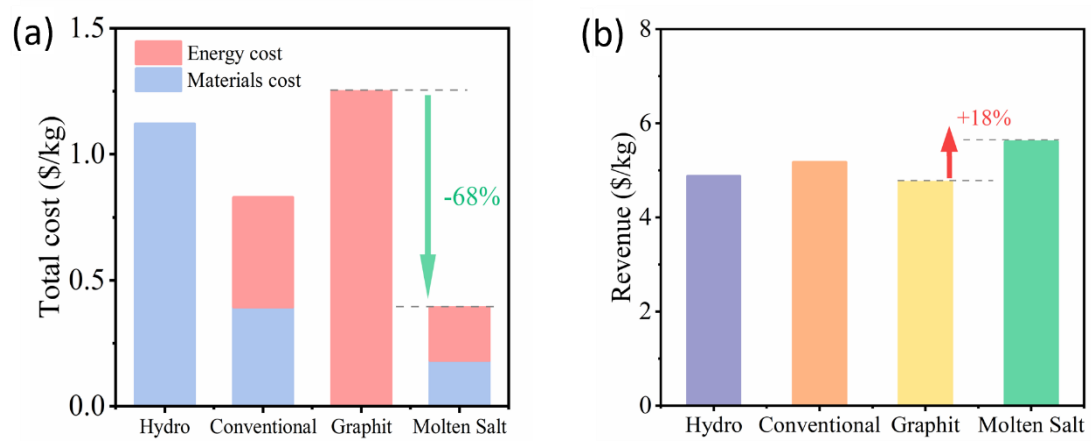


Fig. S10 (a) Total cost and (b) Revenue for regenerating 1 kg of graphite.

Table S1 ICP–OES test results of SG,CPG and MSG. (mg/kg)

Sample	Al	Fe	Li	Mn	Cu	Co	Ni	Total content
SG	337.58	170.64	1.12	2.91	0.96	5.18	3.27	521.66
CPG	23.67	70.97	0.34	6.98	0.33	0.39	2.21	104.89
MSG	30.81	49.80	0.22	1.46	0.44	0.03	1.52	84.28

Table S2 Electrochemical performance of regenerated graphite at high rates.

Sample	Method	Electrochemical performance	Ref.
B-Graphite	Boric acid leaching	140 mAh/g (1 C, 1st)	[1]
LG	Citric acid leaching	174 mAh/g (2 C, 1st)	[2]
RG	Roasting at 900 °C for 4 h	245.8mAh/g (1 C, 500th)	[3]
SCC	Roasting at 1600 °C for 1 h	235.0 mAh/g (1 C, 500th)	[4]
SCC	Roasting at 2600 °C for 0.5 h	263.0 mAh/g (1C, 300th)	[5]
PGC	Fluorination roasting at 200 °C for 1 h	140.2 mAh/g (1C, 100 th)	[6]
RG	Electrolysis method	125 mAh/g (2C, 100 th)	[7]
MSG	Molten salt roasting at 850 °C for 1 h	201 mAh/g (2C, 1000 th)	This work

Table S3 Total cost and metal recovery for recycling graphite.

Various methods	Materials		Total cost (\$/kg)	Metal recovery (%)
	cost (\$/kg)	Energy cost (\$/kg)		
Hydrometallurgy	1.12	0	1.12	99.9
Conventional method	0.394	0.435	0.829	79.9
Graphitization (2600-3300 °C)	0	1.249	1.249	0
Molten Salt	0.182	0.209	0.391	83.8

2. References

- 1 B. Markey, M. Zhang, I. Robb, P. Xu, H. Gao, D. Zhang, J. Holoubek, D. Xia, Y. Zhao, J. Guo, M. Cai, Y. S. Meng and Z. Chen, *J. Electrochem. Soc.*, 2020, **167**, 160511.
- 2 J. Yang, E. Fan, J. Lin, F. Arshad, X. Zhang, H. Wang, F. Wu, R. Chen and L. Li, *ACS Appl. Energy Mater.*, 2021, **4**, 6261–6268.
- 3 Q. Chen, L. Huang, J. Liu, Y. Luo and Y. Chen, *Carbon*, 2022, **189**, 293–304.
- 4 K. Yang, P. Gong, Z. Tian, Y. Lai and J. Li, *Journal of Cleaner Production*, 2020, **261**, 121090.
- 5 K. Yang, Z. Zhao, X. Xin, Z. Tian, K. Peng and Y. Lai, *Journal of the Taiwan Institute of Chemical Engineers*, 2019, **104**, 201–209.
- 6 X. Zhu, J. Xiao, Q. Mao, Z. Zhang, Z. You, L. Tang and Q. Zhong, *Chemical Engineering Journal*, 2022, **430**, 132703.
- 7 N. Cao, Y. Zhang, L. Chen, W. Chu, Y. Huang, Y. Jia and M. Wang, *Journal of Power Sources*, 2021, **483**, 229163.