Supplementary Material

Direct regeneration of spent graphite anode material via a simple thermal treatment method

Xiaoxue Li [a], Baoyu Wu [a], Hao Sun [a], Kai Zhu* [a], Yinyi Gao [a], Tianzeng Bao [b], Hongbin Wu [b], and Dianxue Cao* [a]

[a]Key Laboratory of Superlight Materials and Surface Technology of Ministry of Education, College of Materials Science and Chemical Engineering 1, Harbin
Engineering University, Harbin 150001,(P. R. China)

[b]Hunan Hongshan New Energy Technology Co.2, Ltd, Henglongqiao Town, Heshan District, Yiyang City, Hunan Province, 413000 (P. R. China)

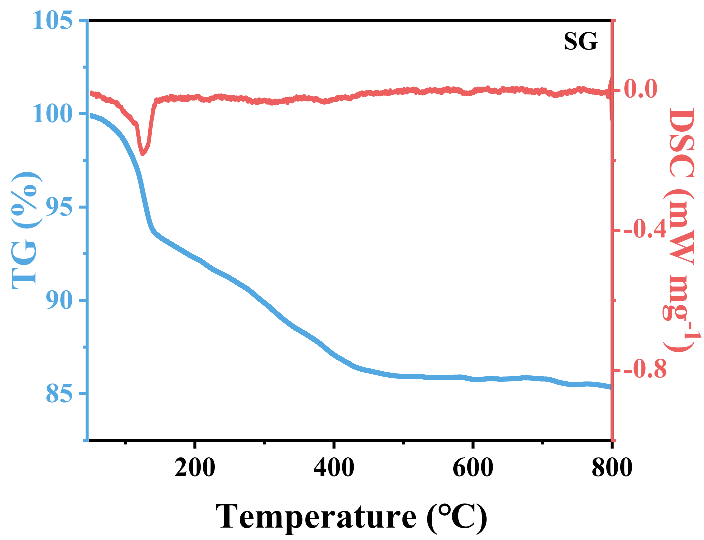
*Corresponding authors.

E-mail: caodianxue@hrbeu.edu.cn
E-mail: kzhu@hrbeu.edu.cn

Supplementary electrochemical measurements

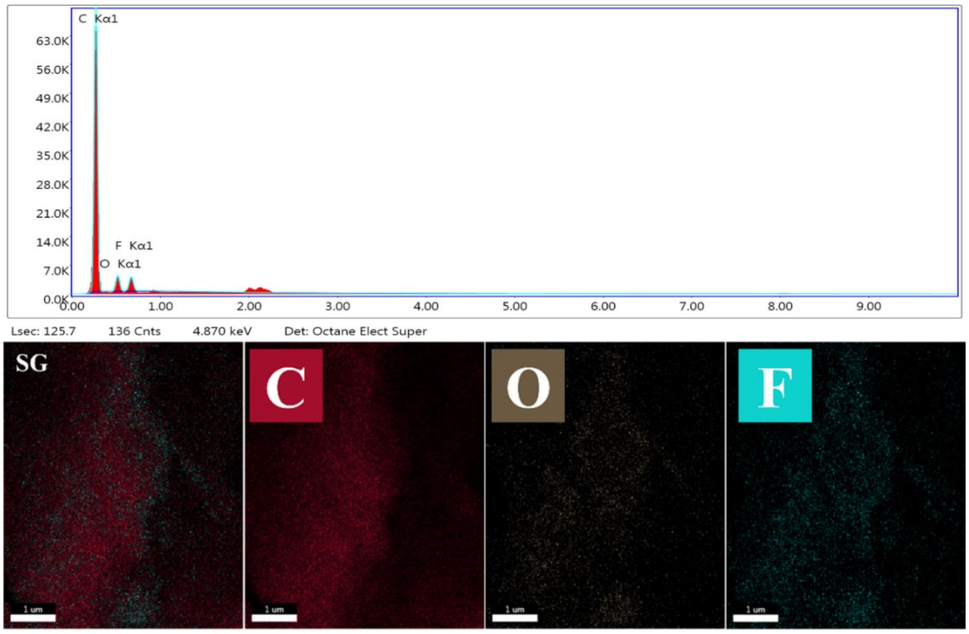
"A well-mixed slurry is applied to the copper foil with a mass loading of the active material of approximately 0.9 mg, and the mass loading of the active material does not fluctuate up or down by more than 0.1 mg."

"Placing coin cells in a temperature-regulated incubator at 26°C during testing, helps minimize variations arising from fluctuations in ambient temperature."

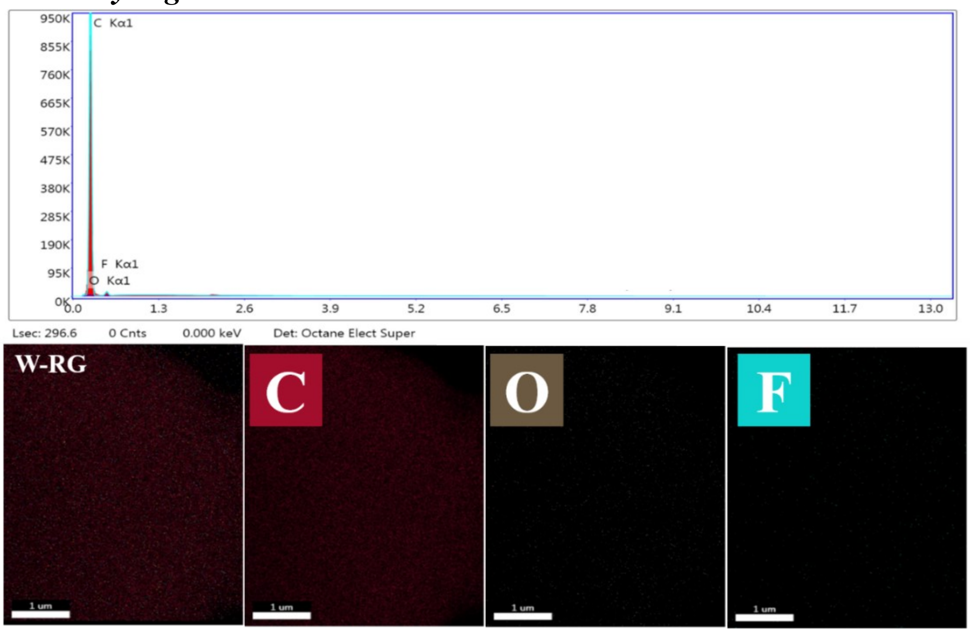


Supplementary Fig S1: thermogravimetric analysis (TGA) of the spent graphite (SG)

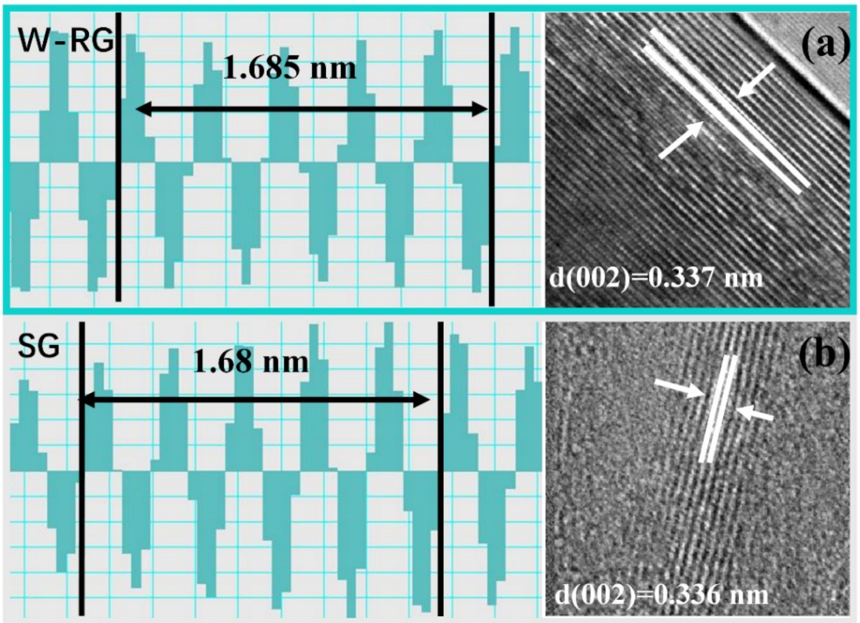
Supplementary Fig S2: comparison of graphite copper foil stripped by heat treatment



Supplementary Fig S3: SG-EDX

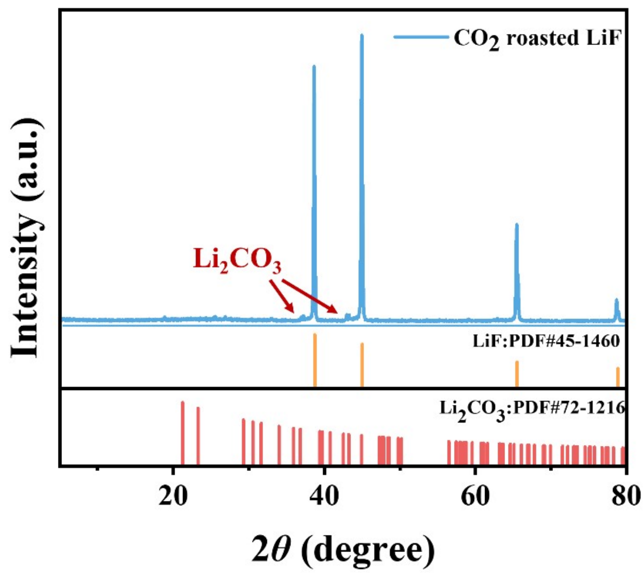


Supplementary Fig S4: W-RG-EDX

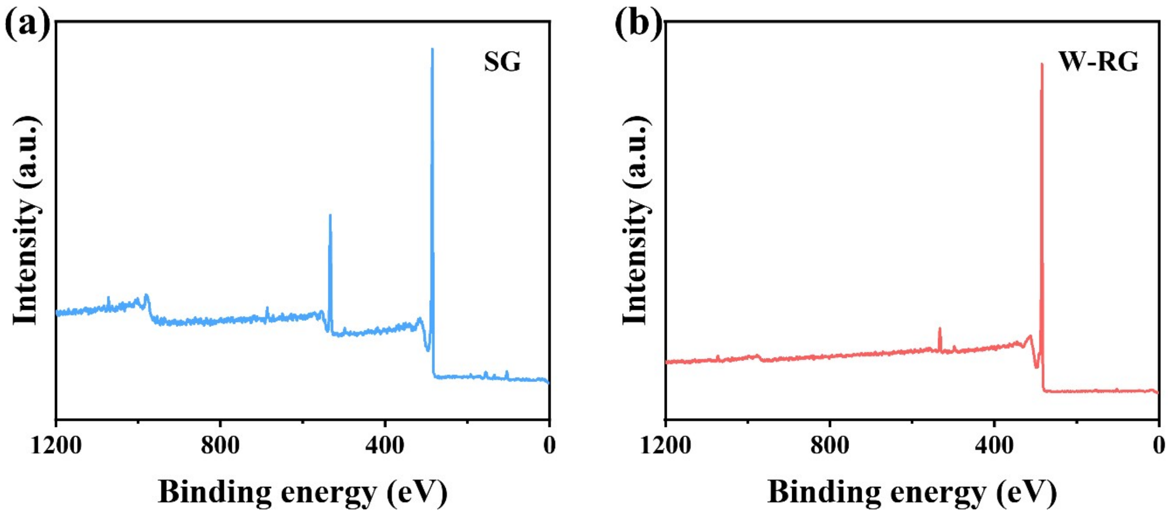


Supplementary Fig S5: line profiles of (a) W-RG, and (b) SG

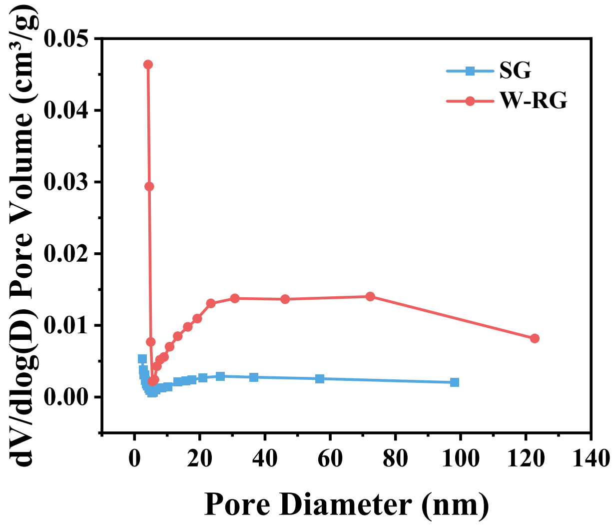
Supplementary Fig S6: white crystals from evaporation of washing solution.



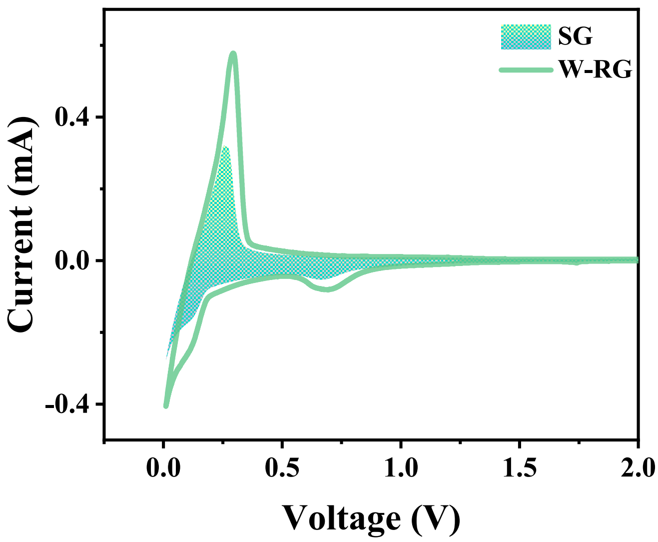
Supplementary Fig S7: XRD of LiF roasted with CO_2.



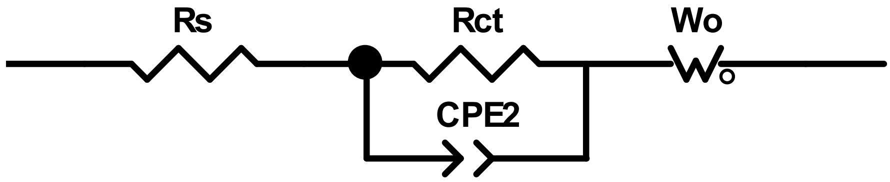
Supplementary Fig S8: XPS survey scan of (a) SG, and (b) W-RG.



Supplementary Fig S9: Cumulative pore volume as a function of pore size, all based on BJH theory.



Supplementary Fig S10: Cumulative pore volume as a function of pore size, all based on BJH theory.



Supplementary Fig S11: Equivalent circuit models for W-RG, SG and CG

***Supplementary Table S1*：**

The metal impurity content of each sample.

Metal(ppm) / Sample	Li	Fe	Cu
SG	85.51	27.16	34.69
	93.6	30.98	38.75
	87.64	31.54	31.52
WG	38.46	9.516	10.83
	34.91	9.156	9.826
	40.2	9.173	11.51
N-RG	28.43	8.93867	8.402
	24.92	8.76717	8.74
	20.84	8.59567	8.082
W-RG	0.2736	3.82	4.882
	0.3461	3.597	4.018
	0.2208	2.835	3.597

***Supplementary Table S2*：**

The electrochemical properties of graphite recovered using different methods are demonstrated.

Recycling methods	Initial coulombic efficiency (%)	Discharge specific capacity (mAh g^{-1})	Author
Bituminous coating [S1]		394 (1C)	YiHua Xiao
Carbon modification [S2]	82.47	263.38(0.5C)	Yongzhi Chen
Water treatment [S3]	75.90	345(0.2C)	Huirong Wang

Calcinedacid leaching [S4]		370(0.1C)	Dan Yang
Structural reconstruction [S5]	92.8%	147.26(50 mA g^{-1})	Kui Liu

Supplementary Table S3：

Table S3. The parameters of the fitted circuit data for each sample.

Sample	**R_S**	**Rct**	**CPE_2**	**W_O**
SG	20.16	135.62	3.8099 E-5	516.5
W-RG	2.091	84.56	1.6629E-5	204.8
CG	2.002	82.4	1.5978E-5	213.3

[S1]Y Xiao, J Li, W Huang, et al. Green & efficient regeneration of graphite anode from spent lithium ion batteries enabled by asphalt coating[J]. Journal of Materials Science: Materials in Electronics. 2022, 33(21): 16740-16752.

[S2]Y Chen, X Wen, X Zhang, et al. Effect of carbon modification on the structure and electrochemical properties of recycled graphite anode materials[J]. Journal of Materials Science: Materials in Electronics. 2023, 34(20): 1518.

[S3] H Wang, Y Huang, C Huang, et al. Reclaiming graphite from spent lithium ion batteries ecologically and economically[J]. Electrochimica Acta. 2019, 313: 423-431.

[S4] D Yang, Y Yang, H Du, et al. An efficient recycling strategy to eliminate the residual “impurities” while heal the damaged structure of spent graphite anodes[J]. Green Energy & Environment. 2022:

[S5] K Liu, S Yang, L Luo, et al. From spent graphite to recycle graphite anode for high-performance lithium ion batteries and sodium ion batteries[J]. Electrochimica Acta. 2020, 356: 136856.