Supplementary Information

Regeneration of Spent Graphite via TEATFB-Assisted Surface Cleaning and Defect Healing

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† Lili Li, Taiyu Jin and Xiaofeng Liu contributed equally to this paper.

This supporting information contains:

- Fig. S1-S21
- Table S1-S9

Supporting Figures:

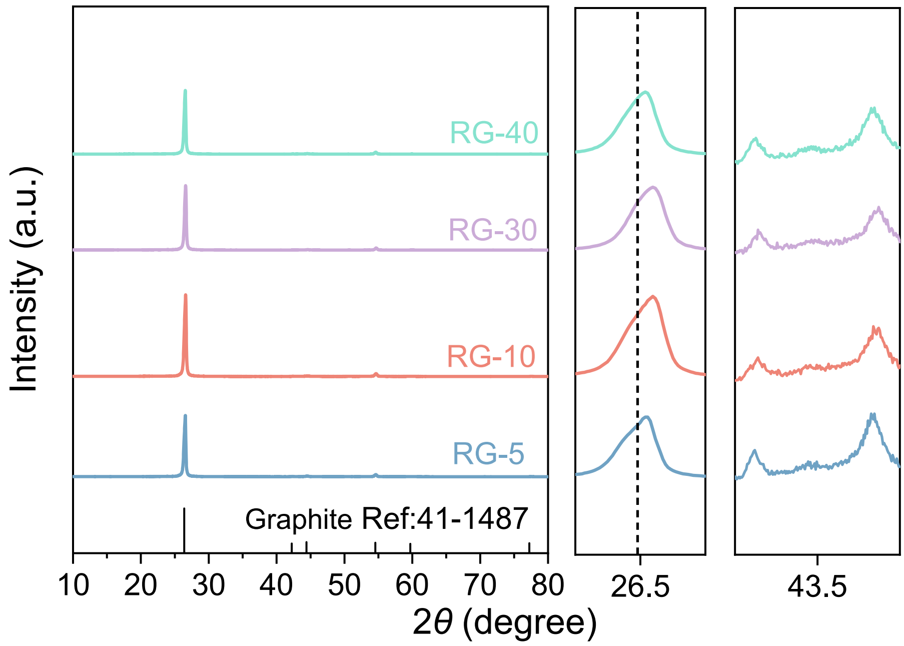

Fig. S1 XRD Patterns of Additional Samples.

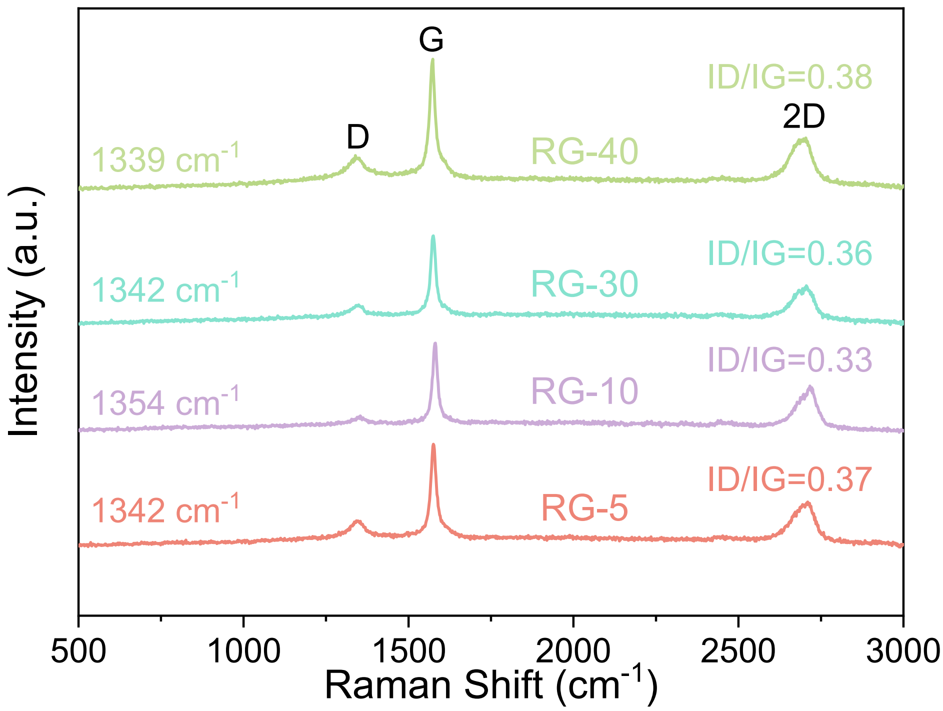

Fig. S2 Raman spectra of Additional Samples.

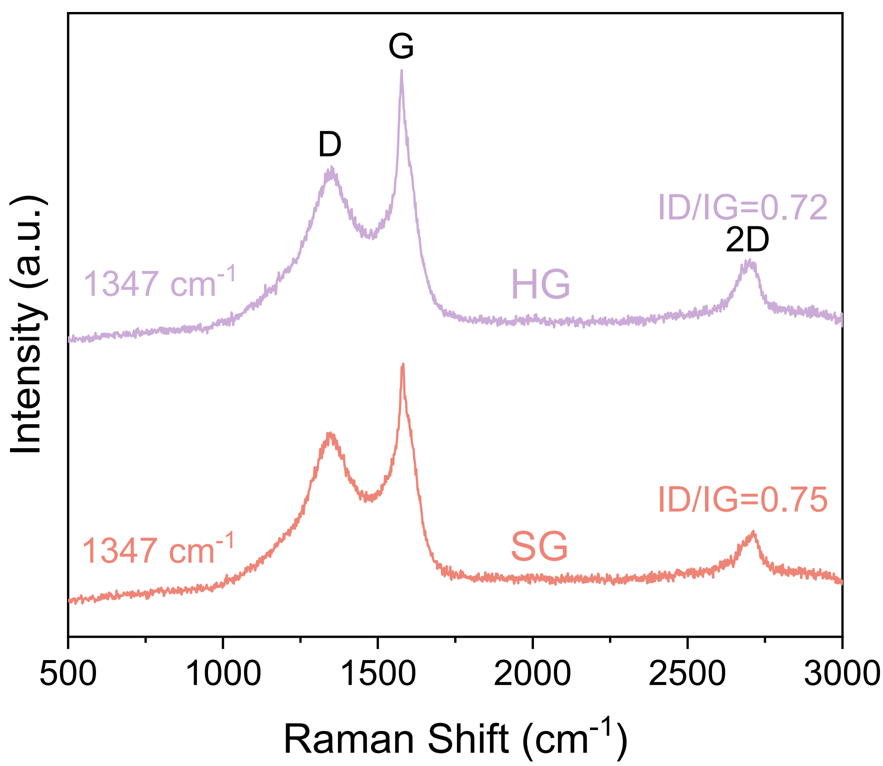

Fig. S3 Raman spectra of HG and SG.

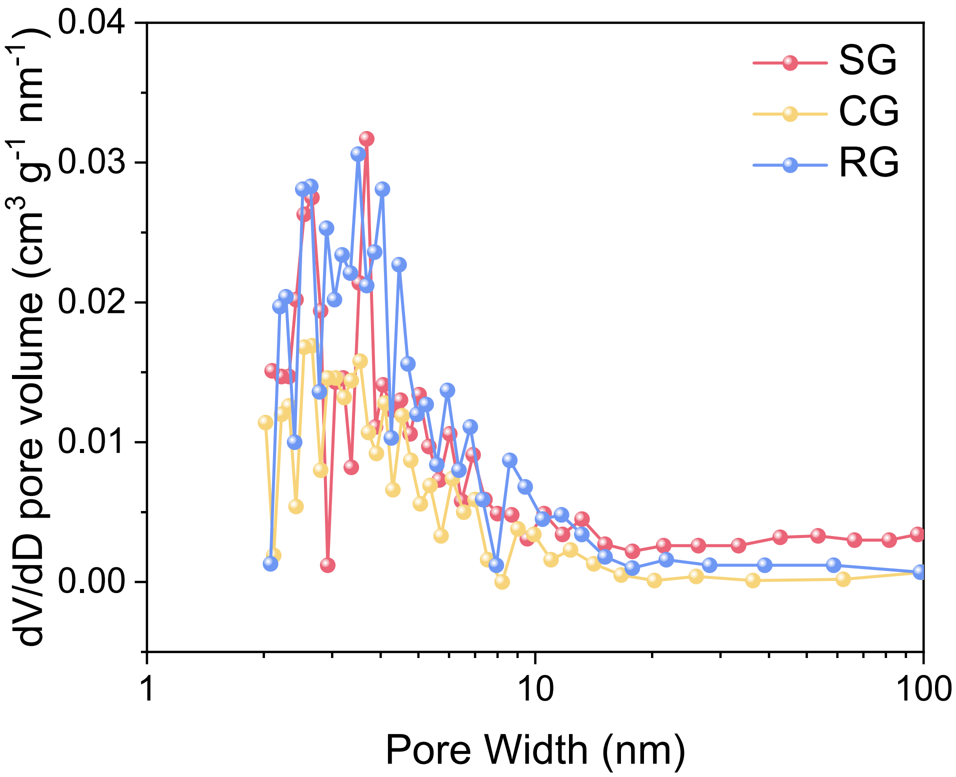

Fig. S4 Pore size distribution of SG, CG, and RG samples measured by BET.

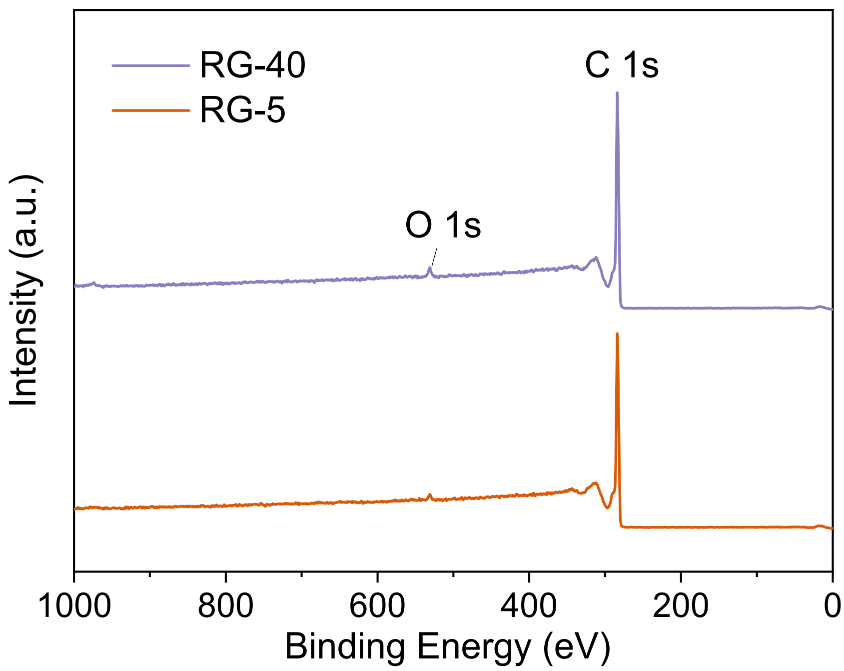

Fig. S5 XPS full survey spectra of RG-5 and RG-40 samples.

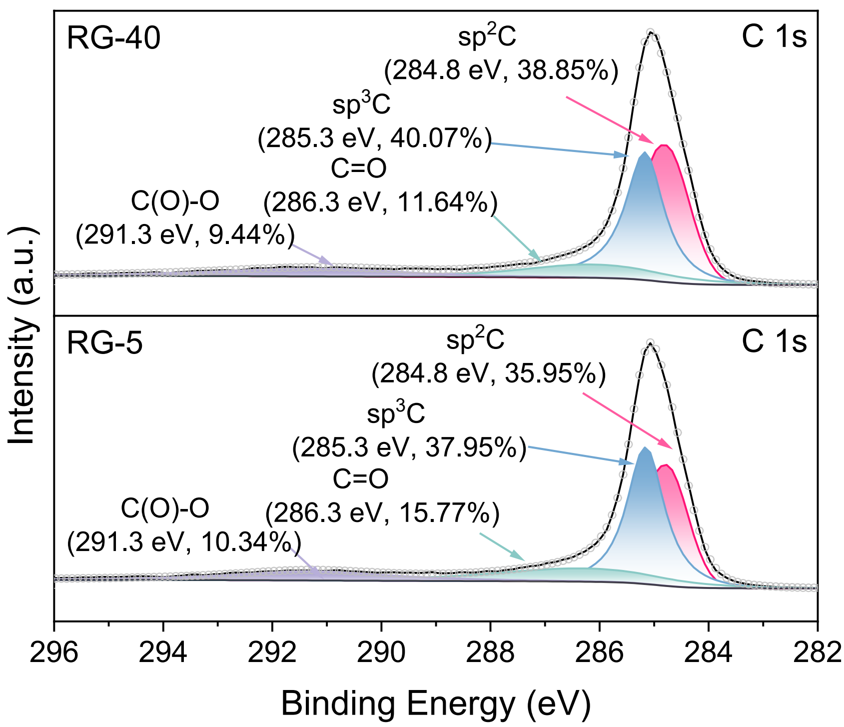

Fig. S6 High-resolution C 1s XPS spectra of RG-5 and RG-40 samples.

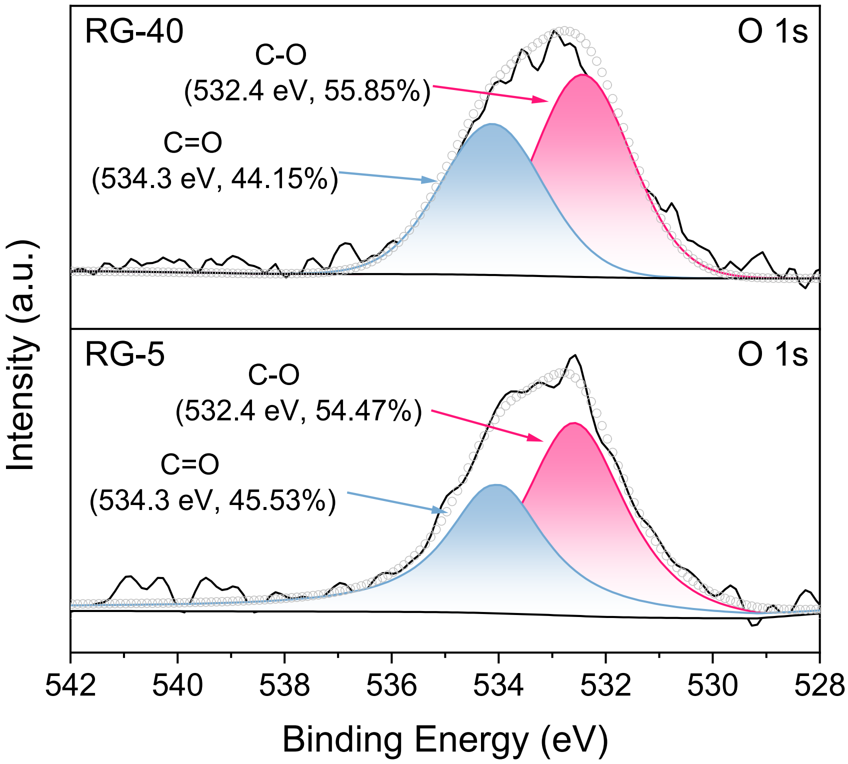

Fig. S7 High-resolution O- 1s XPS spectra of RG-5 and RG-40 samples.

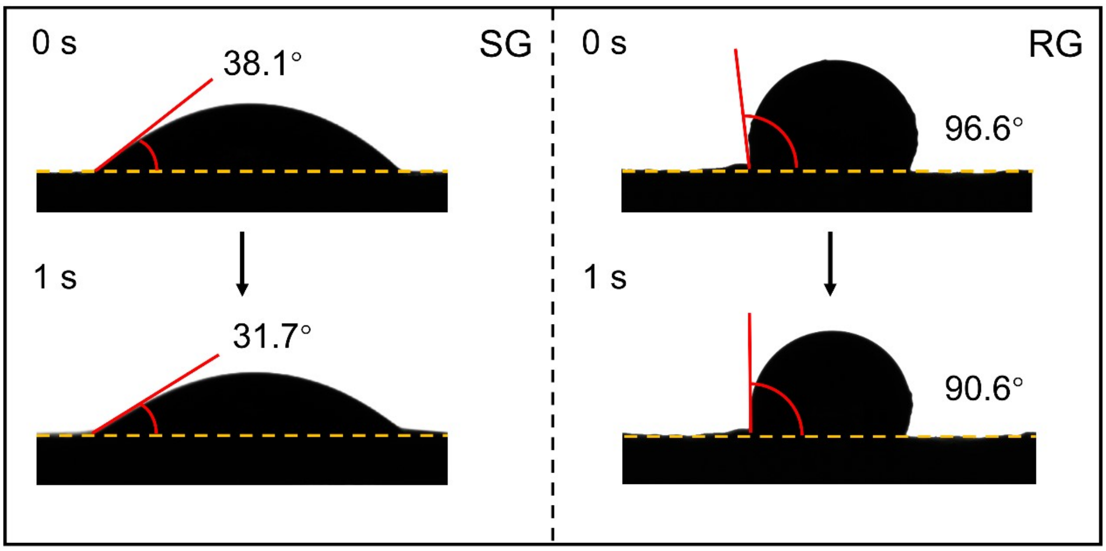

Fig. S8 Water contact angle measurements of SG and RG.

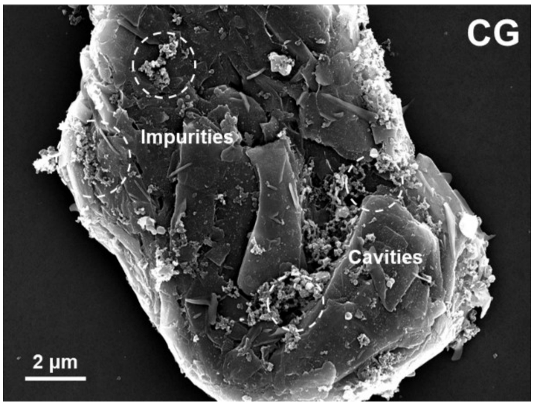

Fig. S9 SEM images of HG.

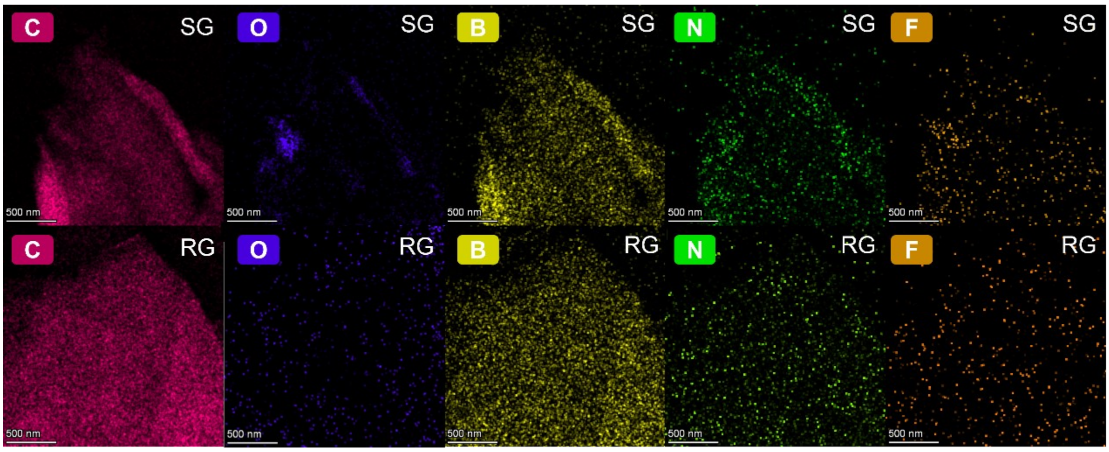

Fig. S10 Comparison of HRTEM-EDX elemental mapping of SG and RG samples.

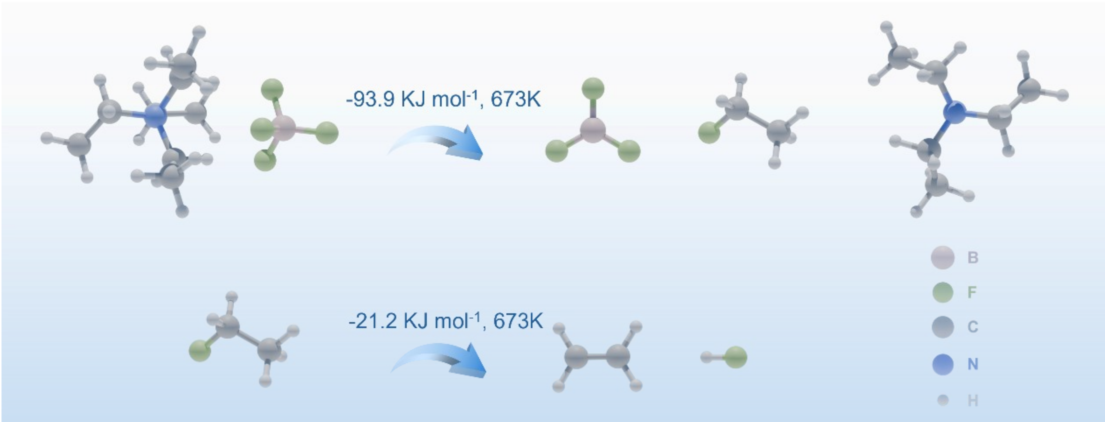

Fig. S11 Proposed decomposition pathways of TEATFB at 673 K and their associated Gibbs free energy changes.

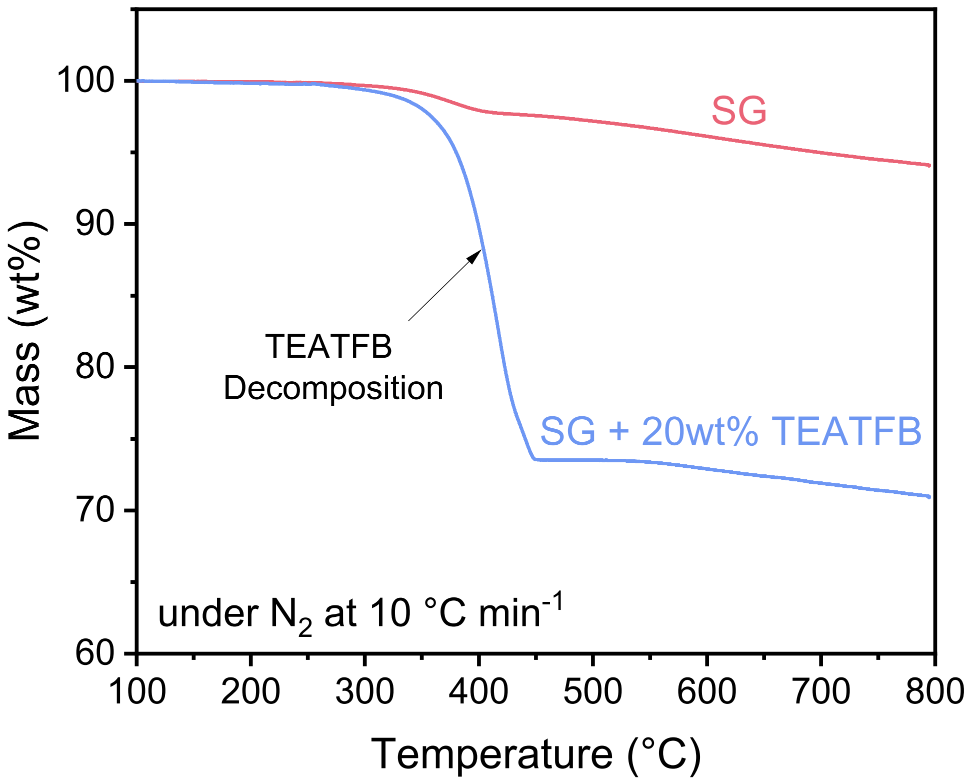

Fig. S12 TGA curves of SG and SG + 20 wt% TEATFB under N_2.

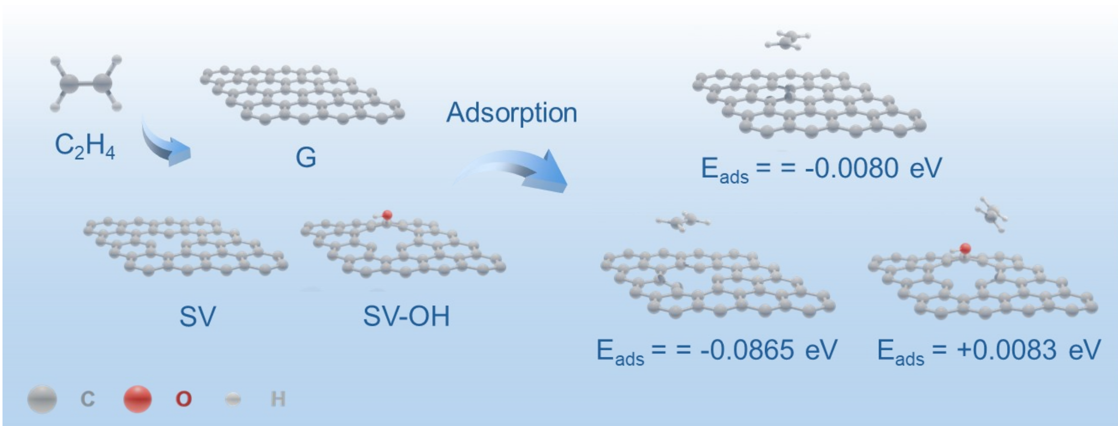

Fig. S13 Comparison of adsorption energies of C_2H_4 on pristine graphene (G), single-vacancy defective graphene (SV), and OH-passivated single-vacancy graphene (SV–OH).

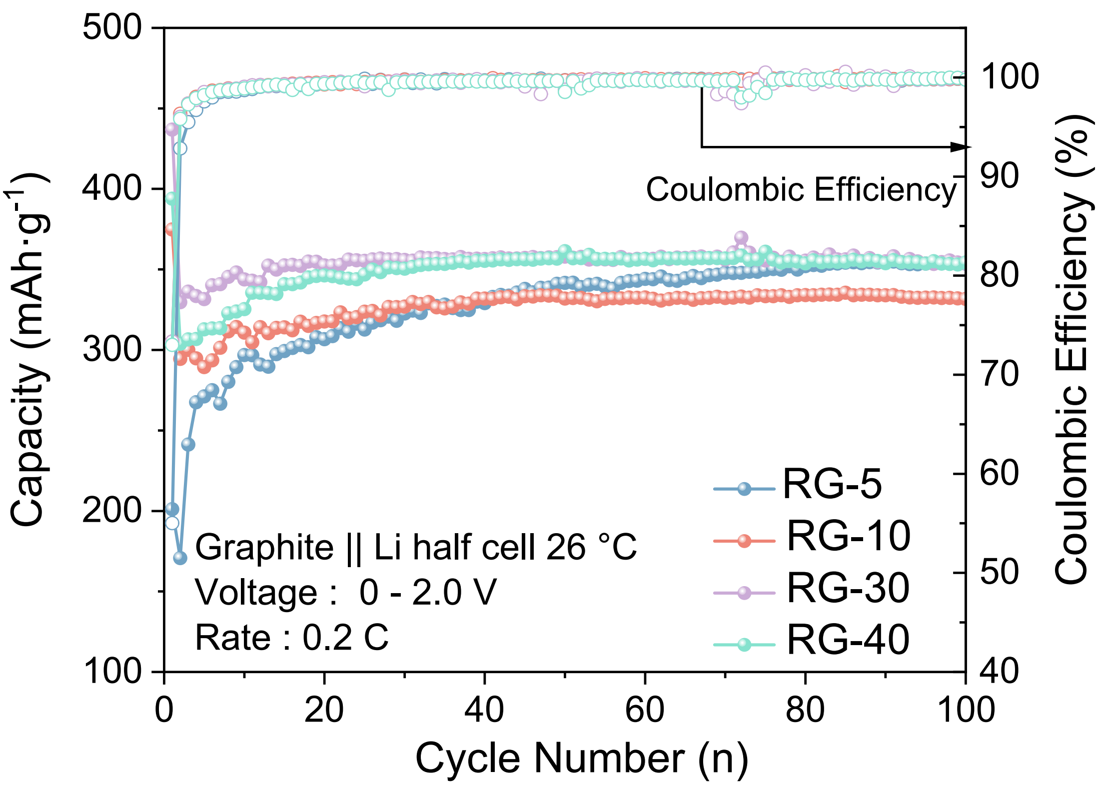

Fig. S14 Cycling performance of other samples at 0.2 C.

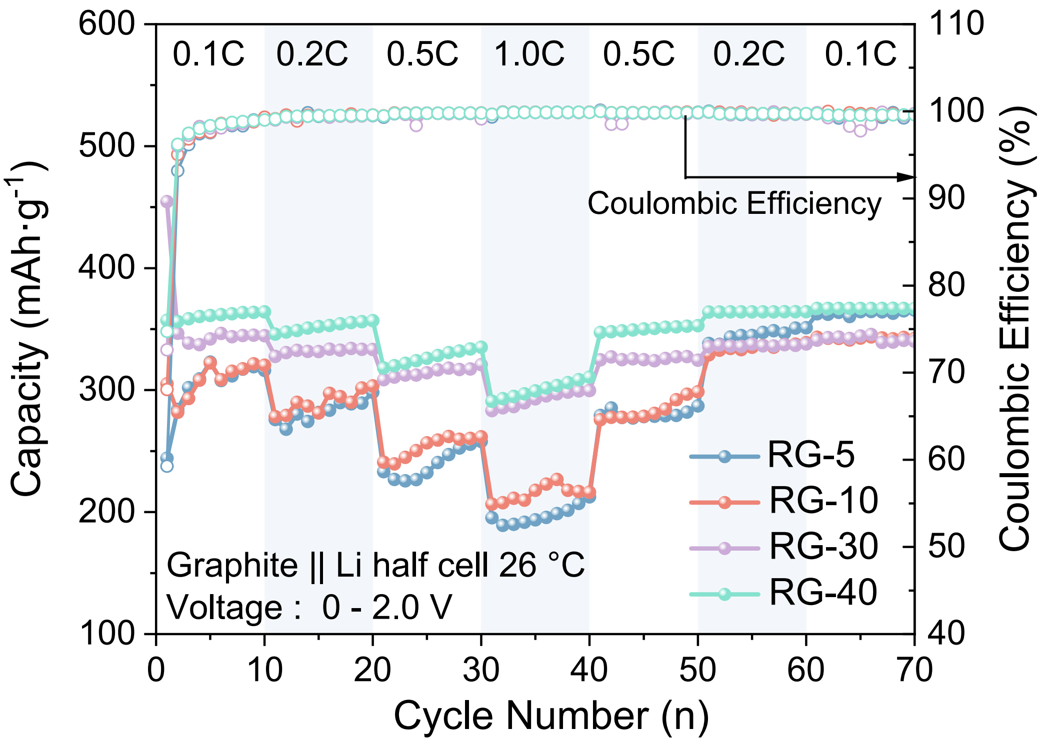

Fig. S15 Electrochemical rate performance of different samples.

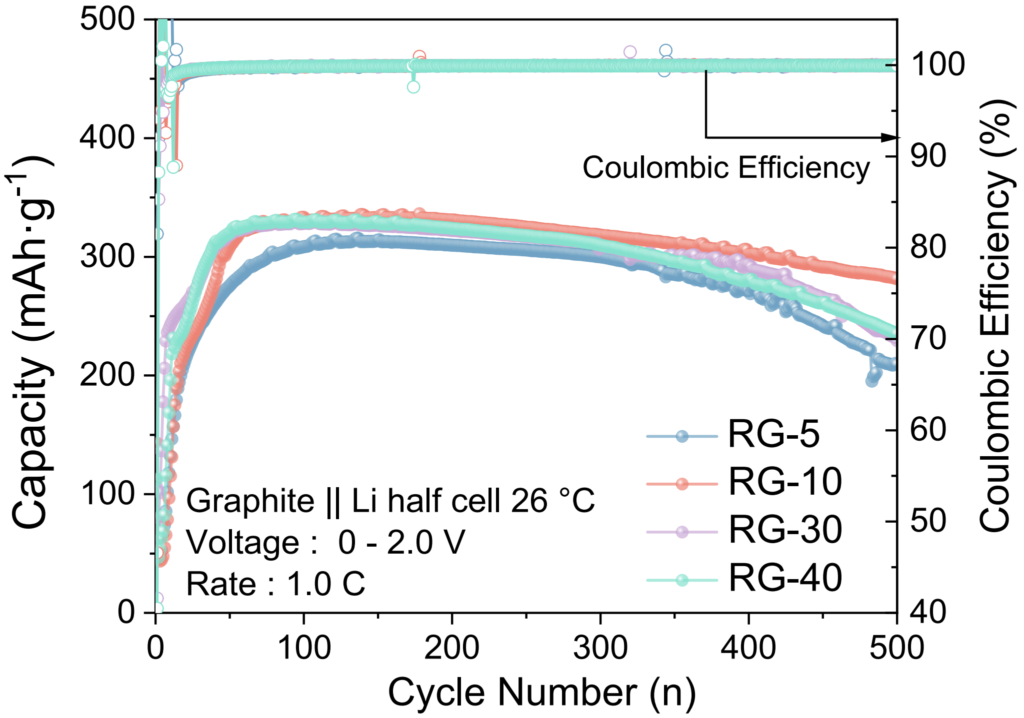

Fig. S16 Cycling performance of other samples at 1.0 C.

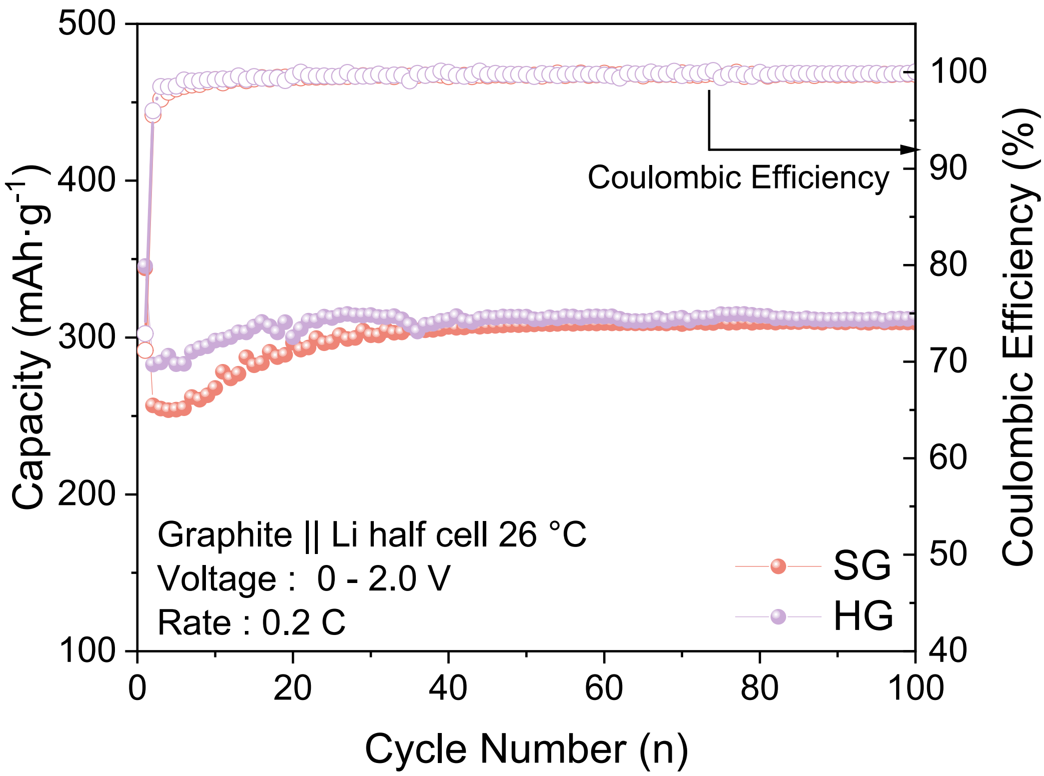

Fig. S17 Cycling performance of HG and SG at 0.2 C.

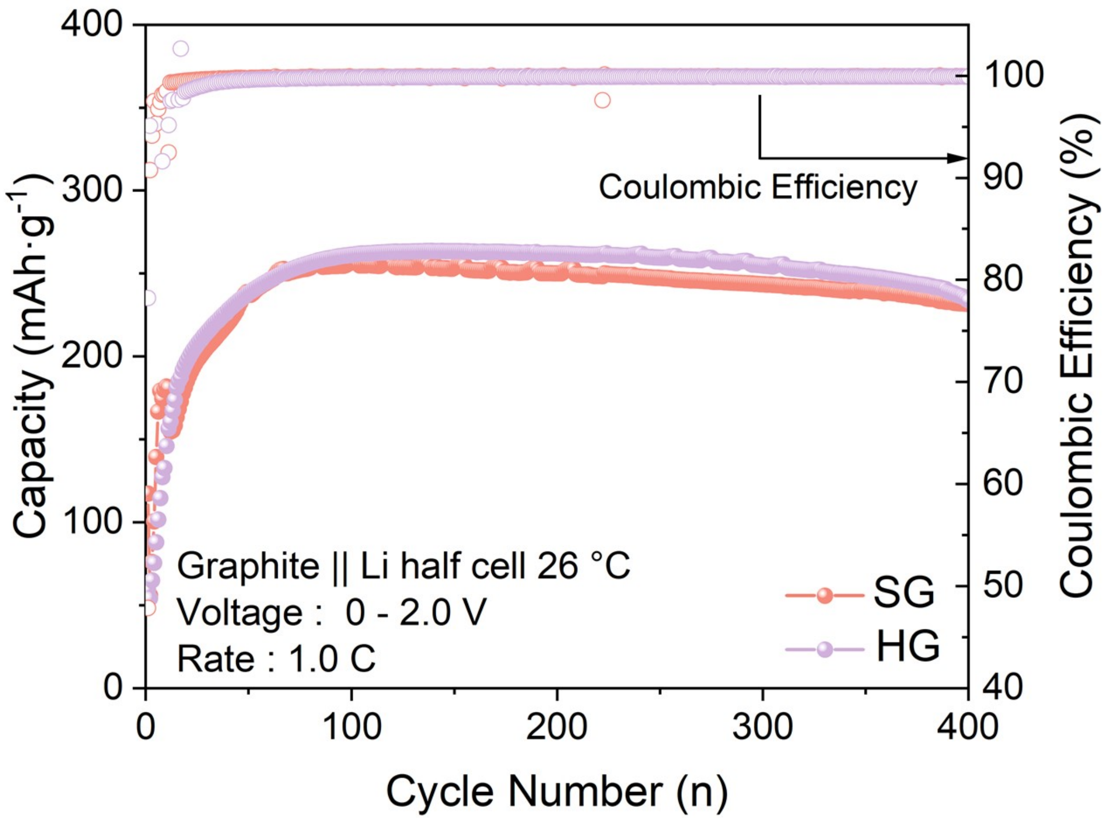

Fig. S18 Cycling performance of HG and SG at 1.0 C.

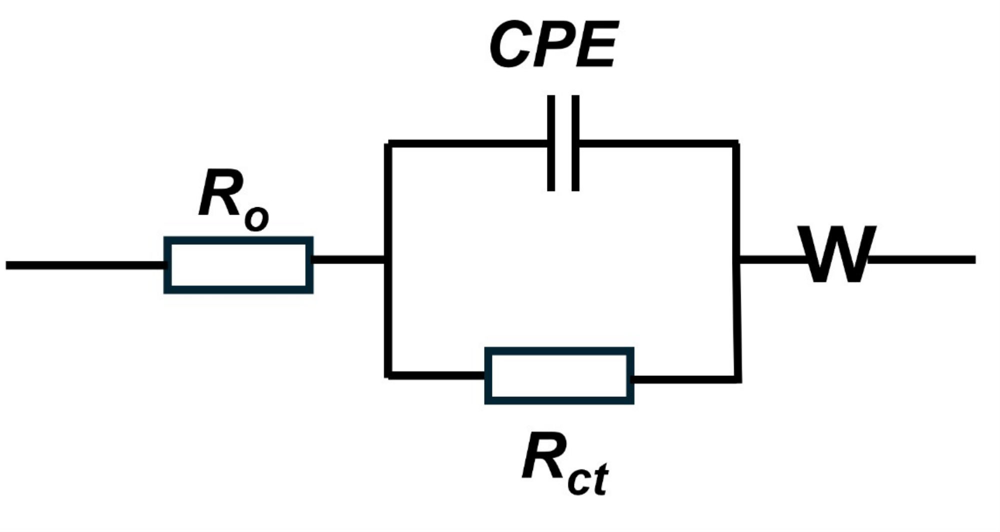

Fig. S19 Equivalent circuit model used for EIS fitting.

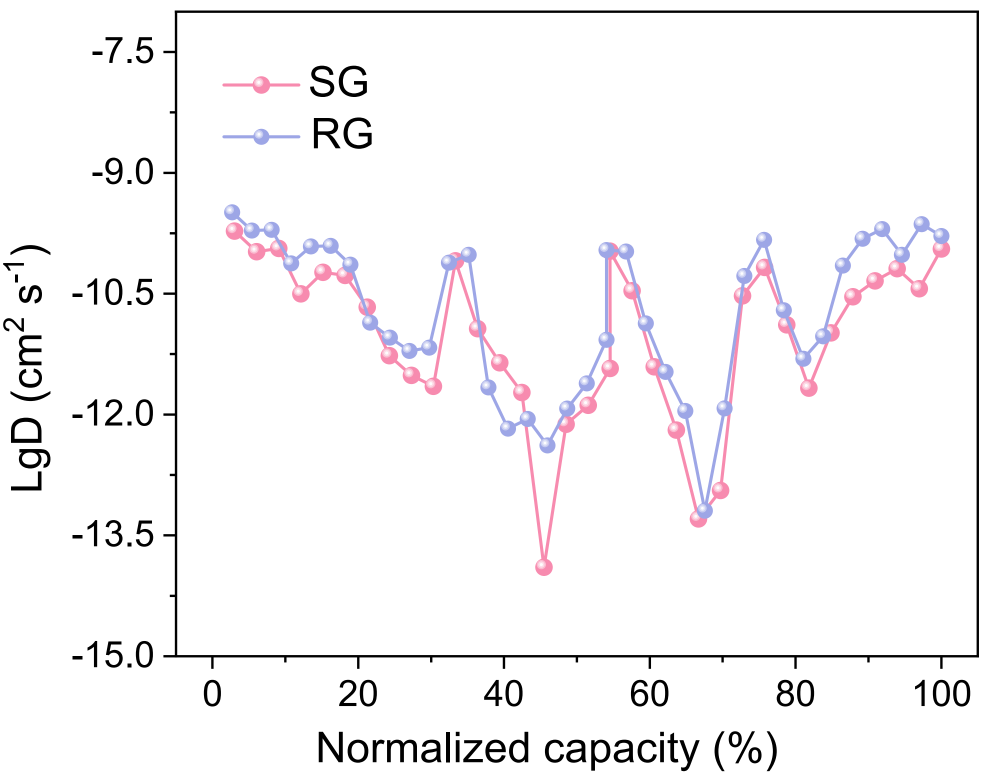

Fig. S20 Li^+ diffusion coefficients of SG and RG from GITT.

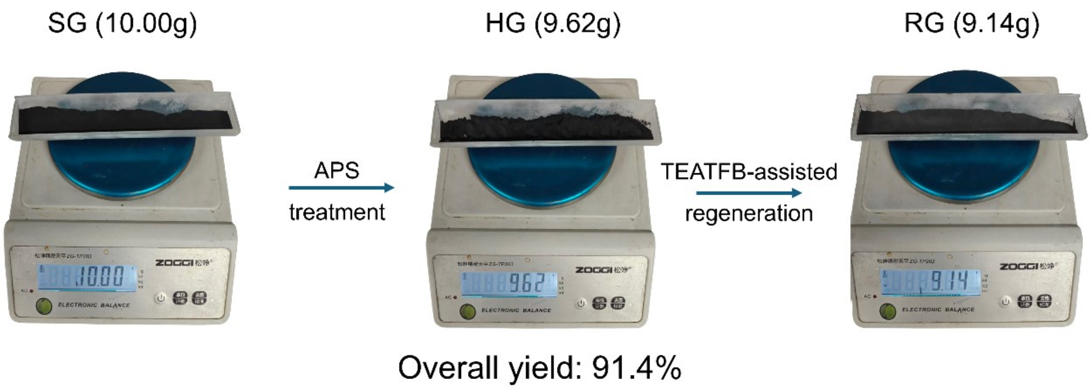

Fig. S21 The mass change and yield of graphite during the regeneration process.

Supporting Tables:

Table S1 XRD parameters of each sample on the (002) plane

Sample	2θ(°)	d (Å)	Peak Height
SG	26.478	3.3635	43747
CG	26.483	3.3629	49993
RG-5	26.555	3.3539	56510
RG-10	26.595	3.3489	75329
RG-20 (RG)	26.538	3.3561	70586
RG-30	26.596	3.3489	59491
RG-40	26.537	3.3562	58753

Table S2 ICP analysis of elemental impurities in SG, HG, and RG samples.

Sample	Cu	Co	Fe	Li	Mn	Ni	P
SG	5.67	<LOD	26.33	128.33	0.33	1.00	487.67
HG	<LOD	<LOD	3.00	30.00	<LOD	0.33	114.67
RG	<LOD	<LOD	0.33	7.00	<LOD	<LOD	25.00

Table S3 Thermodynamic parameters for Reaction 1 (TEATFB → BF_3 + C_2H_5F + NEt_3).

T / K	ΔH_1 / kJ·mol^{-1}	ΔG_1 / kJ·mol^{-1}
298	138.0	33.3
673	127.5	-93.9
1073	115.2	-222.3

Table S4 Thermodynamic parameters for Reaction 2 ($C_2H_5F \rightarrow HF + C_2H_4$).

T / K	ΔH_2 / kJ·mol^{-1}	ΔG_2 / kJ·mol^{-1}
298	67.1	28.7
673	69.5	-21.2
1073	67.6	-74.7

Table S5 EIS fitting parameters of fresh SG, CG, and RG cells.

Parameter	SG	CG	RG
R_s	1.748	2.25	1.502
R_{ct}	72.92	67.78	62.41

Table S6 EIS fitting parameters of fresh SG, CG, and RG cells.

Sample	Parameter	Fresh	5th	50th	100th
SG	R_s	1.748	2.219	3.278	2.029
	R_{ct}	72.92	58.18	34.13	30.64
RG	R_s	1.502	1.947	2.483	1.709
	R_{ct}	62.41	46.16	35.99	28.78

Details of economic and environment analysis

Table S7 The cost analysis of hydrometallurgical recycling.

	Dosage	Unit	Unit price		Cost ($)	Data source
Spent graphite	1.25	kg	0.2	$/kg	0.25	MS
Energy	48.8	MJ	0.03	$/MJ	1.464	BD
Water	3.1	gal	0.0025	$/gal	0.008	BD
Ar	3.5	L	18	$/40L	1.575	B2B
		Total ($/kg)			3.297	

Notes: The first heating is a calcination/smelting step at ~1773 K for 4 h under Ar. The second heating is used for carbonization and graphitization at ~2773 K for 4 h; electricity demand is ~39.0 $MJ\cdot kg^{-1}$ feed (≈10.83 $kWh\cdot kg^{-1}$ feed). Process water is ~2.5 $gal\cdot kg^{-1}$ feed for off-gas/dust scrubbing.

MS: https://www.mysteel.net/. B2B: https://b2b.baidu.com/.

BD: https://www.baidu.com/. SMM: https://www.smm.cn/.

Table S8 The cost analysis of synthetic graphite

	Dosage	Unit	Unit price		Cost ($)	Data source
Needle coke	0.9	kg	0.7	$/kg	0.63	SMM
Tar pitch	0.2	kg	0.35	$/kg	0.07	SMM
Energy	69.6	MJ	0.03	$/MJ	2.088	BD
Water	2.8	gal	0.0025	$/gal	0.010	BD
Ar	3.1	L	18	$/40L	1.395	B2B
		Total ($/kg)			4.193	

Notes: Artificial graphite is produced from petroleum or needle coke and coal tar pitch via mixing and granulation, baking at 900 °C, coking at 1300 °C, and graphitization at 2800 °C under argon. Argon consumption is 0.005 $kg\cdot kg^{-1}$ feed. Electricity demand is 63.1 $MJ\cdot kg^{-1}$ feed. Process water is 2.5 $gal\cdot kg^{-1}$ feed. The product yield is 0.907 kg of artificial graphite per kg of feed.

Table S9 The cost analysis of regenerated graphite

	Dosage	Unit	Unit price		Cost ($)	Data source
Spent graphite	1.12	kg	0.2	$/kg	0.224	SMM
Energy	10.1	MJ	0.03	$/MJ	0.303	BD
Water	0.74	gal	0.0025	$/gal	0.002	BD
APS	0.26	kg	0.42	$/kg	0.109	B2B
TEATFB	0.21	kg	1.39	$/kg	0.292	B2B
Ar	1.9	L	18	$/40L	0.855	B2B
	Total ($/kg)				1.785	

Notes: Recycled graphite production process. A mild pre-oxidation is conducted by soaking in 0.5 M ammonium persulfate at a liquid-to-solid ratio of 2.0 $L\cdot kg^{-1}$ followed by a 0.5 $L\cdot kg^{-1}$ rinse. The powder is then mixed with tetraethylammonium tetrafluoroborate at 20 wt% relative to graphite and heat-treated at 800 °C for 2 h under argon. Argon consumption is 0.003 $kg\cdot kg^{-1}$ feed. Electricity demand is 9.0 $MJ\cdot kg^{-1}$ feed. Process water is 0.66 $gal\cdot kg^{-1}$ feed.

Reference:

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