

Supplementary Information

Upcycling of spent graphite and iron housing from waste lithium-ion batteries for fabricating cost-effective high-capacity anode

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Supporting Figures and Tables

S1.1. Degree of degradation of pristine and spent graphite

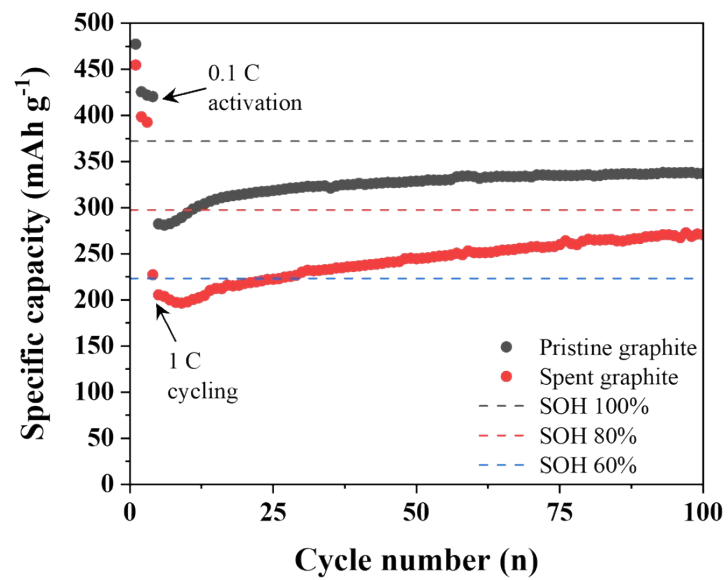


Figure S1. Discharge profile of coin type pristine graphite (PG) and spent graphite (SG) half-cell.

S1.2. Iron housing of LIBs

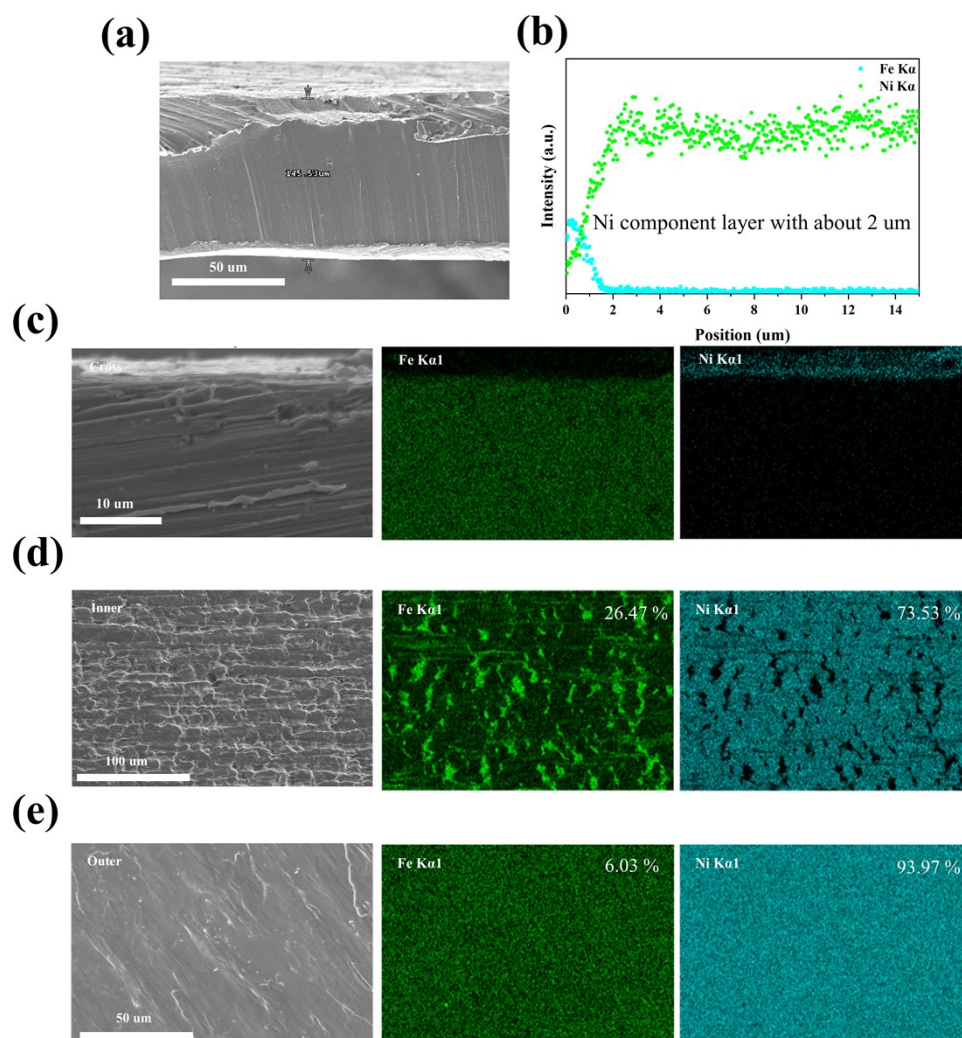


Figure S2. (a) Cross-sectional SEM image of the iron housing and (b) corresponding EDX line profile. SEM and corresponding EDX mapping images of (c) cross-, (d) inner-, and (e) outer-sections of the iron housing.

S1.3. Morphological characterization of F-SGO before heat treatment

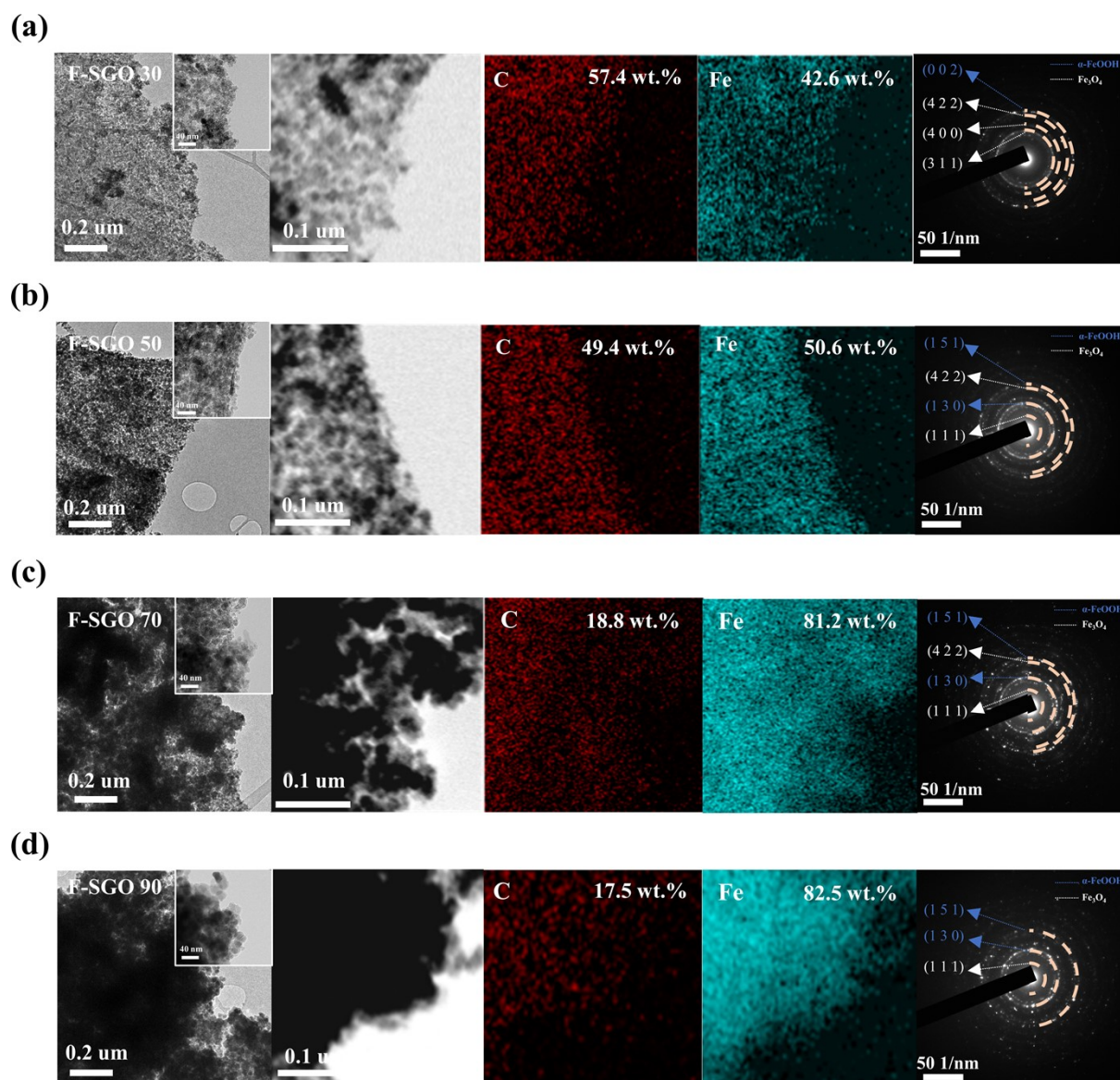


Figure S3. HR-TEM and corresponding EDX mapping images and SAED patterns of (a) F-SGO 30, (b) F-SGO 50, (c) F-SGO 70, and (d) F-SGO 90.

Table S1. Element composition of F-SGO 30 detected by TEM-EDX.

Element	Weight%	Atomic%
C	33.01	53.93
O	25.68	31.50
Na	0.00	0.00
S	0.11	0.07
Cl	0.07	0.04
K	0.00	0.00
Mn	0.38	0.14
Fe	40.76	14.32
Ni	0.00	0.00
Total	100.00	100.00

S1.4. Physical characterizations of F-SGO before and after heat treatment

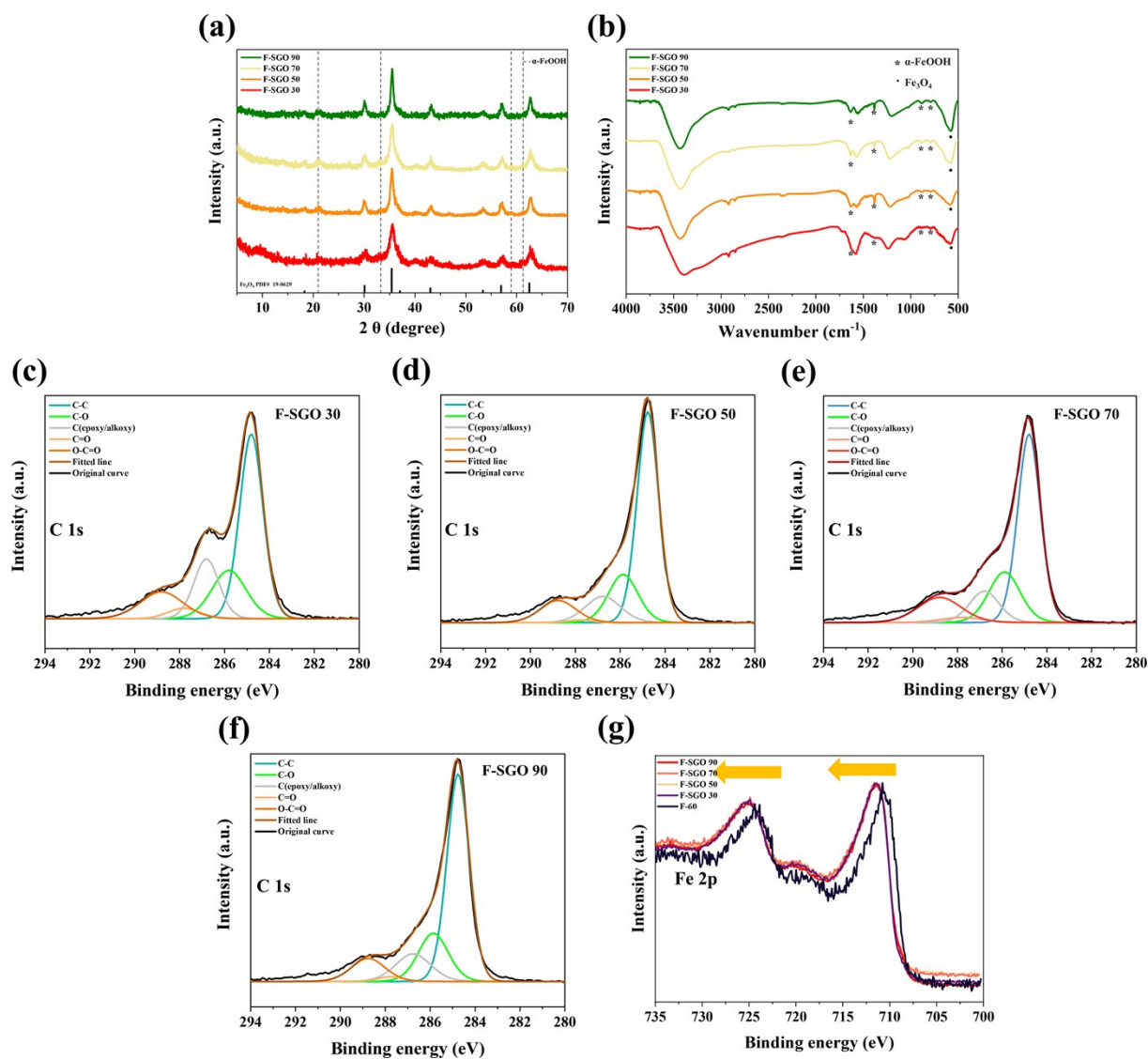


Figure S4. (a) XRD patterns, (b) FT-IR spectra, and C 1s XPS analysis of (c) F-SGO 30, (d) F-SGO 50, (e) F-SGO 70, and (f) F-SGO 90. (g) Fe 2p XPS profiles of F-SGO30, 50, 70, and 90.

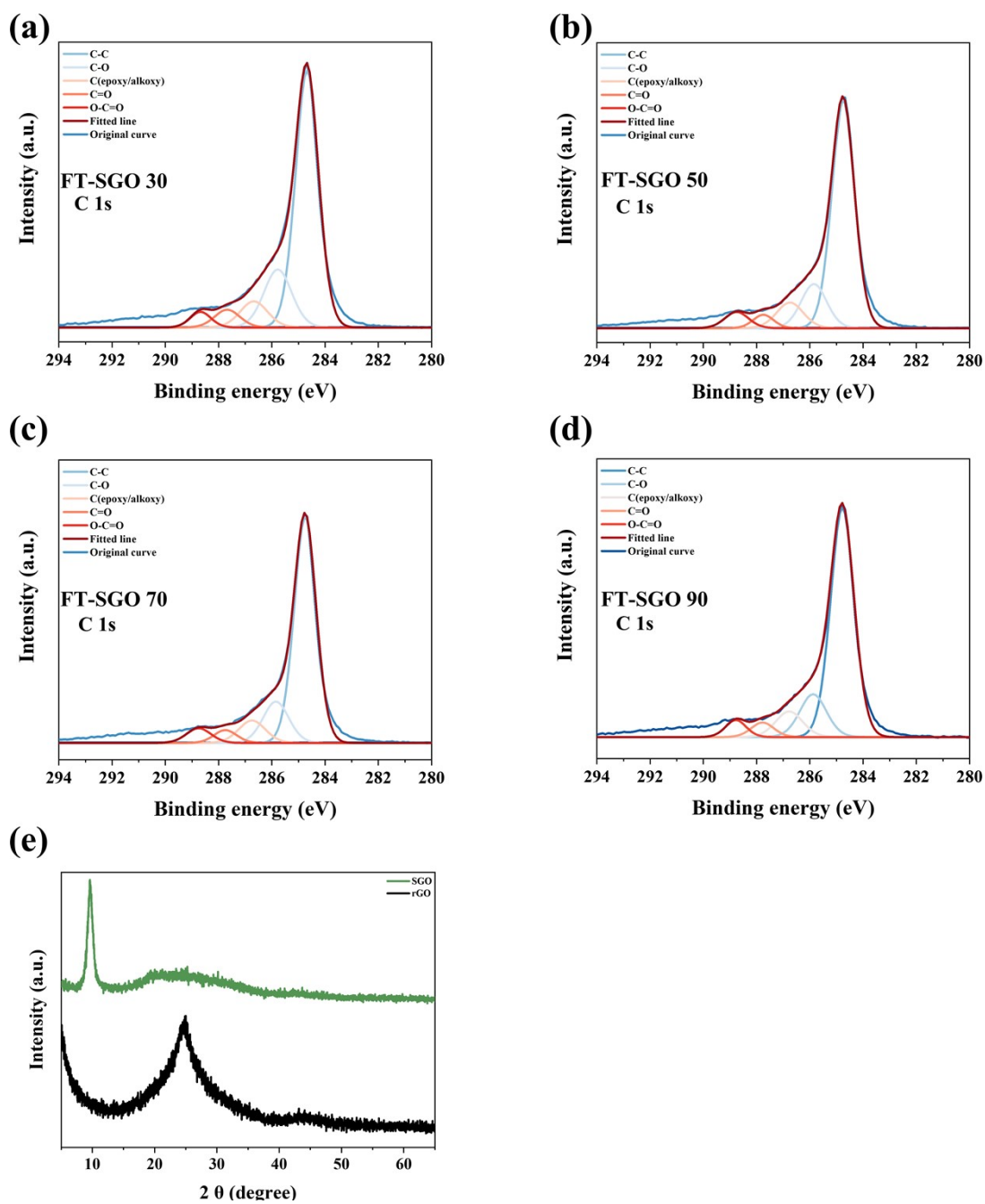


Figure S5. C 1s XPS analysis of (a) FT-SGO 30, (b) FT-SGO 50, (c) FT-SGO 70, and (d) FT-SGO 90. (e) XRD patterns of SGO (green) and rGO (black).

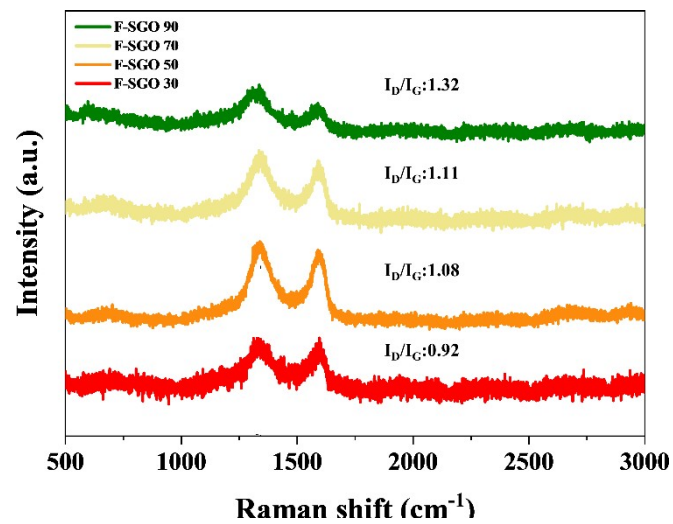


Figure S6. Raman spectra of F-SGO at different reaction times.

S1.5. Volume expansion of anode before cycling and after 100th cycle

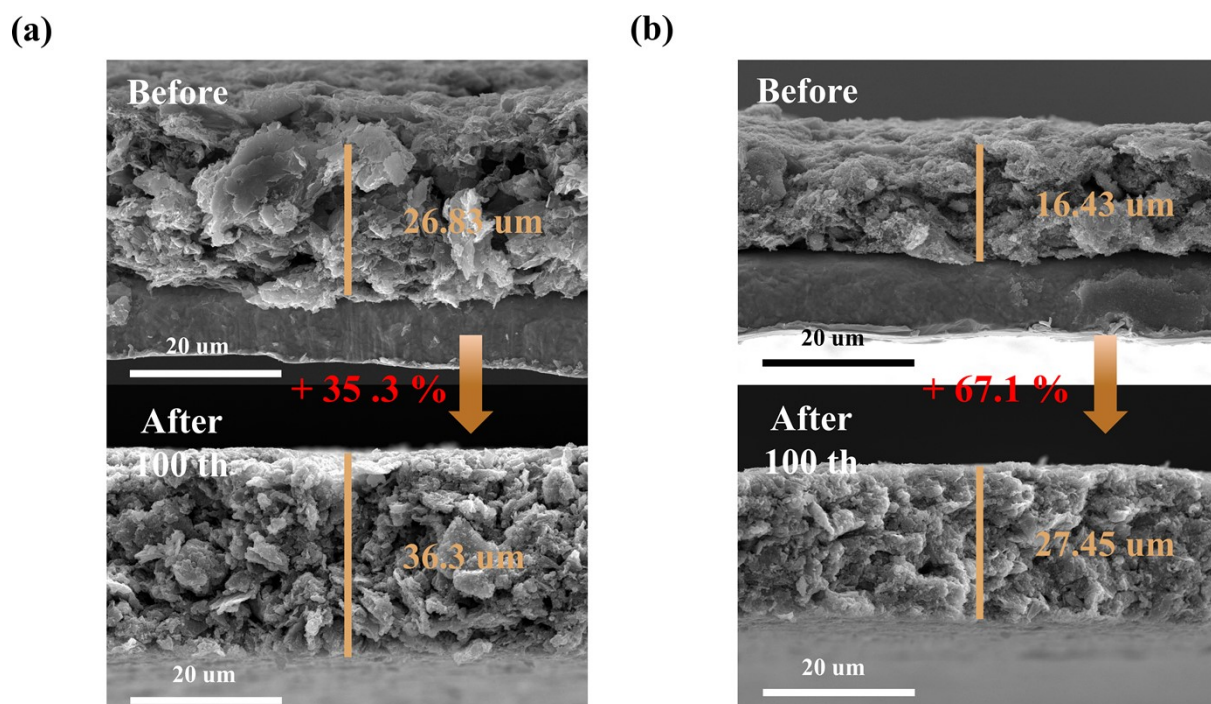


Figure S7. SEM images before and after 100th cycle at a current density of 1 A g^{-1} for (a) FT-SGO 30 and (b) FT-SGO 90.

S1.6. Physical characterizations of FT-SGO

Table S2. Zeta potentials of FT and GO

Zeta potential (mV)				
	1st	2nd	3rd	Average
FT	55.09	54.77	52.98	54.28
SGO	-40.37	-38.58	-41.09	-40.01

Table S3. Electrochemical impedance spectroscopy (EIS) data for FT-SGO anode at different reaction times

		6030_450	6050_450	6070_450	6090_450
R_s(Ω)	0 th	4.58	5.29	6.853	6.49
R_{ct}(R) (Ω)	0 th	37.45	51.618	73.16	76.85

Table S4. Economic analysis of the upcycling process and graphite-silicon monoxide with a 9:1 weight ratio

Materials	Conditions	Unit price	Cost	Reference
H ₂ SO ₄	8.42 mL	0.01 (\$ mL ⁻¹)	0.0841 (\$)	http://www.daejungchem.co.kr/02_product/search/?search_value=7664-93-9
H ₂ O ₂	2.26 mL	0.0076 (\$ mL ⁻¹)	0.0171 (\$)	http://www.daejungchem.co.kr/02_product/search/?search_value=7664-93-9
KMnO ₄	1.6 g	0.0118 (\$ g ⁻¹)	0.0188 (\$)	http://www.daejungchem.co.kr/02_product/search/?search_value=7664-93-9
NaCl	3 g × 1 g product/0.4 g per rep	0.0144 (\$ g ⁻¹)	0.108 (\$)	https://www.junsei.co.jp/product_search/search_seihin_result_e.html
Electricity price	2.5 V × 0.18 A × 0.5 h × 1 g product/0.4 g per rep (potentiostat) + 0.164 kWh (ultrasonication) + 0.355 kWh (tube furnace)	0.1219 (\$/KW h)	0.0632 (\$)	https://doi.org/10.1016/j.jpowsour.2020.229163
Argon price	25.3 L (including purification and synthesis)	0.009 (\$ L ⁻¹)	0.227 (\$)	http://www.higas.co.kr/
Graphite anode	0.753 g	0.2378 (\$ g ⁻¹)	0.179 (\$)	https://www.sigmaaldrich.com/KR/ko/product/aldrich/907154
SiO anode	0.247 g	6.593 (\$ g ⁻¹)	1.628 (\$)	https://www.sigmaaldrich.com/KR/ko/product/aldrich/336823

* All price calculations are based on the retail price and 1 g of product.

* Most chemical prices are in Korean Won and all costs are converted to US Dollars using the exchange rate as of February 10, 2023.

* Theoretical capacity calculation was based on the following equation:

$$774 \text{ mAh } g^{-1} \times 0.336 + 924 \text{ mAh } g^{-1} \times 0.664 = 873.6 \text{ mAh } g^{-1}$$

- 774 and 924 $\text{mAh } g^{-1}$ represent the theoretical capacities of reduced graphene oxide and Fe_3O_4 , respectively.
- The weight ratios of reduced graphene oxide and Fe_3O_4 based on TGA were 0.336 and 0.664, respectively.