Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

Regeneration of anode materials from complex graphite residue in spent lithium-ion battery recycling process

Table S1 Element composition of GR by XRF

Element	Cu	Al	Ca	Fe	Ni	Co	Mn	Li	Si	Ti	Zr
Content (wt.%)	0.01	0.37	0.76	0.08	0.12	0.34	2.78	0.02	1.01	0.05	0.19

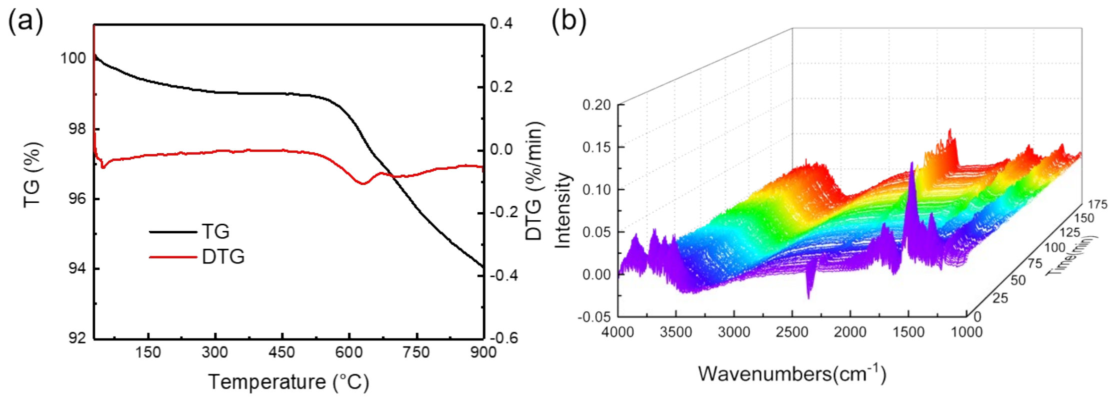

Figure S1 (a) Thermogravimetry (TG) and differential thermal analysis (DTA) of GR in argon; (b) Thermogravimetric infrared of GR in argon.

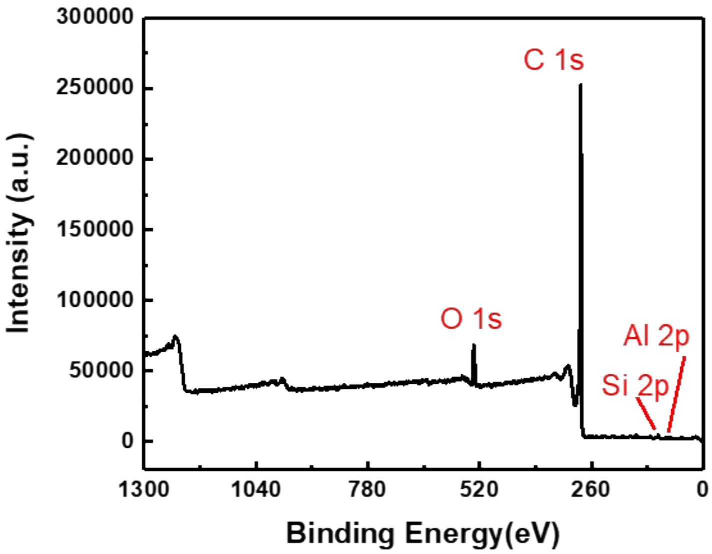

Figure S2 XPS spectra of GR.

The Environment-economic impacts of the process are calculated from the end of the pretreatment.

Table S2 Environmental-economic relevant data of the proposed process in this work

Technology	Raw material	CO_2 (g)	NO_x (g)	SO_x (g)	Total energy (MJ)	Water (L)	Profit ($)
Leaching & Extraction (solid-to-liquid ratio10g/L)	Spent LIBs 1kg	102.76	0.80	5.38	0.60	10.32	-2.21
Alkali swelling	Graphite 0.2kg	111.00	0.11	0.06	10.99	1.23	
Hydrochloric acid leaching	Graphite 0.2kg	368.60	0.32	0.17	6.50	2.88	
Restore structure	Graphite 0.2kg	-	-	-	27.00	-	
Product							+4.71
Total		582.37	1.22	5.61	45.09	14.44	+2.50

1、 Leaching & Extraction：The raw material 1kg spent LIBs, solid-liquid ratio set to 10g/L, requires 10L of 1mol/L sulfuric acid solution. The CO_2, NO_x, SO_x, energy and water consumed from the process and the preparation of the raw material are available by searching in the GREET software, as shown in Table S2 (GREET is a traditional multidimensional model that provides a comprehensive, life-cycle-based approach to compare the energy use and emissions of conventional and advanced vehicle technologies. It includes Vehicle-Cycle Model (evaluates the energy and emission effects associated with vehicle material recovery and production, vehicle component fabrication, vehicle assembly, and vehicle disposal/recycling)).

2、 Alkali swelling：Set each 1kg spent LIBs contains 20% of anode graphite, so set the raw material for this step is 0.2kg graphite, according to the addition of 10wt.% NaOH in this paper, 0.06kg of sodium hydroxide solid is needed, and at the same time, high temperature roasting at 500°C for 3h is needed, and the search in GREET software shows the process and the raw material preparation generated CO_2, NO_x, SO_x, the energy and water consumed, as shown in Table S2.

3、 Hydrochloric acid leaching：The material loss in the swelling process is small and negligible relative to the total amount, so the raw material is still set to 0.2kg of graphite for this step, and the solid-liquid ratio is set to 10g/L, requiring 2L of 1mol/L hydrochloric acid solution, which can be looked up in the GREET software to know the process and the raw material preparation of CO_2, NO_x, SO_x, energy and water consumed, as shown in Table S2.

4、 Restore structure：The material loss in the leaching process is small and negligible compared to the total amount, so the raw material is still set at 0.2 kg of graphite for this step, which needs to be heated at 2600°C for 2 h. The energy consumed in the process can be found by looking up in the GREET software, as shown in Table S2.

5、 The total energy consumed by the above process is combined and converted into electrical energy consumption, and the energy consumption cost is calculated according to the Chinese commercial electricity consumption of 0.15$ / (kW h), and the total expenditure is calculated by combining the cost of reagents used (query in GREET software).

6、 Based on the ratio of each component in the LIB, Li 1.9%; Co 3%; Ni 12.1% and graphite anode material 20%, the yield of the product obtained (price per kg of material from GREET software) is calculated and the results are shown in the Product row in Table S2.

7、 Finally, the above components are summed to obtain the total gas emissions, energy and water consumption, and total benefits, as shown in Table S2.

Table S3 Environmental-economic relevant data of Partially-recovery technology

Technology	Raw material	CO_2 (g)	NO_x (g)	SO_x (g)	Total energy (MJ)	Water (L)	Profit ($)
Leaching & Extraction (solid-to-liquid ratio10g/L)	Spent LIBs 1kg	102.76	0.80	5.38	0.60	10.32	-1.68
Burning	Graphite 0.2kg	822.19	2.80	2.22	36.00	4.74	
Product							+3.97
Total		924.95	3.60	7.60	36.60	15.06	+2.29

1、 Leaching & Extraction：Same as Note 1 of Table S2.

2、 Burning：It is set that each 1kg of spent LIBs contains 20% of anode graphite, so the raw material for this step is set to be 0.2kg of graphite, and heated at 750°C in air for 3h. The CO_2, NO_x, SO_x, energy consumed, and water produced by this process can be found by searching in the GREET software, as shown in Table S3.

3、 The total energy consumed by the above process is combined and converted into electrical energy consumption, and the energy consumption cost is calculated according to the Chinese commercial electricity consumption of 0.15$ / (kW h), and the total expenditure is calculated by combining the cost of reagents used (query in GREET software).

4、 Based on the ratio of each component in the LIB, Li 1.9%; Co 3%; Ni 12.1%, the yield of the product obtained (price per kg of material from GREET software) is calculated and the results are shown in the Product row in Table S3.

5、Finally, the above components are summed to obtain the total gas emissions, energy and water consumption, and total benefits, as shown in Table S3.

Table S4 Environmental-economic relevant data of Separation-recovery technology

Technolog y	Raw material	CO_2 (g)	NO_x (g)	SO_x (g)	Total energy (MJ)	Water (L)	Profit ($)
Separation	Spent LIBs 1kg	-	-	-	36.00	10.00	-2.02
Purification	cathode materials 0.35kg	151.47	0.31	2.68	0.30	5.16	
Purification	Anode material 0.35kg	351.30	0.49	3.70	0.30	5.16	
Restore structure	Graphite 0.2kg	-	-	-	27.00	-	
Product							+3.94
Total		502.77	0.80	6.38	63.60	20.32	+1.92

1、Separation：Set each 1kg spent LIBs contains 35% of cathode material, 20% of anode graphite and 15% of copper foil, and 30% of other (including battery shell, electrolyte, etc.), the above materials are separated by flotation, and the energy and water consumed by the process can be found by searching in GREET software, as shown in Table S4.

2、Purification：The purification of 0.35 kg of cathode material obtained after the above flotation, with the solid-liquid ratio set at 10 g/L, required 3.5 L of 1 mol/L sulfuric acid solution, 0.2 L of hydrogen peroxide solution, and heating at 80°C for 2 h. The process and the raw material preparation generated CO_2, NO_x, SO_x, energy and water consumed can be found by looking up in the GREET software, as shown in Table S4.

3、Purification：The 0.35 kg of anode material (including graphite and copper foil) obtained after the above flotation was purified, and the solid-liquid ratio was set to 10 g/L, requiring 3.5 L of 1 mol/L sulfuric acid solution, 0.2 L of hydrogen peroxide solution, and heating at 80°C for 2 h. The process and the raw material preparation generated CO_2, NO_x, SO_x, energy and water consumption can be found by searching in the GREET software, as shown in Table S4.

4、Restore structure：Same as Note 4 of Table S2.

5、The total energy consumed by the above process is combined and converted into electrical energy consumption, and the energy consumption cost is calculated according to the Chinese commercial electricity consumption of 0.15$ / (kW h), and the total expenditure is calculated by combining the cost of reagents used (query in GREET software).

6、Based on the ratio of each component in the LIB, Li 1.9%; Co 3%; Ni 12.1% and graphite anode material 20%, the yield of the product obtained (price per kg of material from GREET software) is calculated and the results are shown in the Product row in Table S4.

7、Finally, the above components are summed to obtain the total gas emissions, energy and water consumption, and total benefits, as shown in Table S4.