Electronic Supplementary Information

Flash recovery of lithium from spent anode graphite by carbonthermal shock and water leaching

Duanmei Song, Beikai Zhang, Hongbiao Du, Jing Wu, Jiadong Yu*, Jinhui Li

† State Key Joint Laboratory Environmental Simulation & Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

Text S1

Pretreatment: Adequate charge/discharge cycling of the battery is related to the lithium content in the anode graphite. After a continuous charging and discharging process at a discharge rate of 1C [1], the capacity of the NCM battery was severely degraded until more than 20% capacity degradation [2]. To prevent deflagration and explosion caused by the incomplete discharge, the decommissioned NCM battery was immersed in a 5% NaCl aqueous solution for 24 h to ensure complete discharge of any remaining charge and natural drying.[3] Then, the NCM battery was manually disassembled and separated within a glovebox to cathode, anode, and separator. The anode sheet was used as raw materials for experiments. For the electrolyte remaining on the anode sheet, we did not consider its removal. Because the lithium in the electrolyte ($LiPF_6$, $LiClO_4$) is valuable, it can be recovered with the lithium in graphite by CTS and water leaching to recover the lithium resource sufficiently. $LiPF_6$ thermally decomposes at high temperatures to produce LiF, as shown in Eq. S1. [4] LiF can be reacted with the medium (Fig. S2) to recover lithium by water leaching. $LiClO_4$ begins to decompose at about 400°C, producing oxygen and water-soluble LiCl [5], as shown in Equation S2. The fluorine gas generated from the pyrolysis of electrolytes can also be absorbed by our auxiliary medium without causing pollution.

$$LiPF_6 \xrightarrow{pyrolysis} LiF + PF_5 \tag{S1}$$

$$LiClO_4 \xrightarrow{pyrolysis} LiCl + 2O_2 \tag{S2}$$

Text S2

We did not consider the effect of the L/S ratio during ultrasonication, which is consistent with the studies of Subramanian et al and Liu et al [6-8]. We had the following considerations in mind. Acid leaching of lithium requires investigation of the S/L parameter because the solid-liquid ratio of the acid is related to the H^+ concentration gradient and viscosity. In contrast, when using water leaching for lithium extraction is not related to the above, the only concern is the solubility. According to the solubility parameter, the salts Li_2CO_3 (1.29 g/100 mL) and LiF (0.134 g/100 mL) are soluble in water at 25℃, and the solubility increases with decreasing temperature.[9, 10] Theoretically, at room temperature, an aqueous solution of 25 mL can dissolve at least 0.3225 g of Li_2CO_3 and 0.0268 g of LiF. The 1 cm^2 anode piece used in our CTS experiments was about 0.025 g. After CTS and ultrasonic aqueous leaching for 10 s, based on the ICP measured lithium concentration and the range of concentration of the standard curve, we chose 25 mL as the calibration volume. Even if a 1 cm^2 anode piece consists entirely of Li_2CO_3 (0.025 g), 25 mL of water can completely dissolve 10 anode pieces. The anode sheet is mainly composed of copper foil and graphite with low lithium content. Depending on the charging and discharging state of the battery, the lithium content of the anode piece ranges from 0.5% to 10%. In other words, 25 mL of water is enough to completely dissolve the lithium in hundreds of anode pieces. Therefore, based on the above solubility characteristics, for the recovery of lithium anode, water is a suitable leaching agent for Li recovery, and there is no need to consider the effect of the L/S ratio of water on the lithium leaching rate. The leaching rate of lithium we observed is trustworthy.

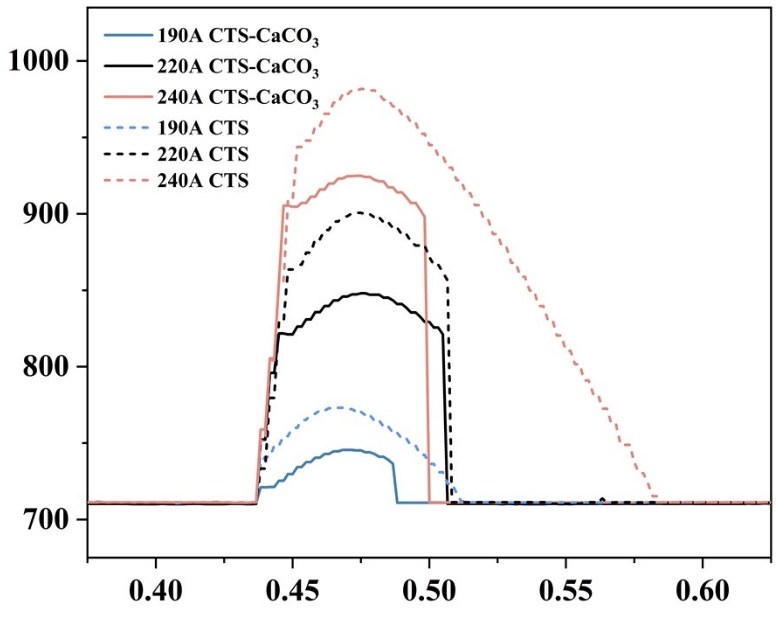

Fig. S1 Time-temperature relationship of heating procedure in direct CTS and with added $CaCO_3$ medium conditions (fast mode, time parameter set to 5s).

Table S1 Study on the recovery of lithium from anodes

No.	Methods	Li leaching rate	Reference
1	Dismantling, Calcination (500°C, 1 h), Leaching with HCl (S/L 1:50 g/mL, 80°C, 90 min)	99.4 wt%	[11]
2	Dismantling, Crushing, Sieving, Leaching with H_2SO_4 (S/L 60 g/L, 45°C, 40 min)	98.5 wt%	[12]
3	Dismantling, Leaching with trifluoroacetic acid (S/L 60 g/L, 40°C, 30 min)	1.08% Li,100% Cu foil recovery rate	[13]
4	Dismantling, Calcination (600°C, 4 h), Leaching with sulfaminic acid (S/L 5 g/L, 40°C, 45 min)	97.2%	[14]
5	Dismantling, Calcination (400°C, 2 h)+Alkaline roasting (350°C, 2 h), Water leaching	~100%	[8]
6	Dismantling, Calcination (450°C, 1.5 h + 500°C, 1 h), Leaching with citric acid (S/L 1:50 g/mL, 90°C, 50 min)	97.6 wt%	[15]
7	Dismantling, CTS (800-900°C, 15-20s), Water leaching (ultrasound 10s)	99.5% Li，~100% Cu foil recovery rate	This work

Table S2 Comparison of conventional heating and CTS method

Methods	Li recovery rate	Time	Energy consumption	Environmental Impact
Traditional thermal furnace	~98%	1 h	~2×10^6 kWh/ton	High
CTS (this work)	99%	15 s	~9.5×10^3 kWh/ton	Low

Lithium leaching rate data from traditional method derived from Table S1 literature analysis. The weights of the samples were set at 1.0 g. The powers of the thermal furnaces were set at 2000 W. The maximum energy consumption of the CTSW method was calculated as: E=UIt/m (U: the highest voltage in the heating process, 6 V; I: the current, 95 A; t: the heating time, 15 s; m: the sample mass, 0.25 g).

Table S3 Current-temperature relationship with and without the auxiliary medium

	190A	**220A**	**240A**
1-CTS without medium	755℃	881℃	1049℃
1-CTS with $CaCO_3$ medium	713℃	834℃	935℃
2-CTS without medium	773℃	901℃	982℃
2- CTS with $CaCO_3$ medium	745℃	848℃	925℃

Table S4 Physicochemical data of metal oxides/hydroxides/carbonates (K, Na, Ca, Mg, Al, Fe)

Chemical formula	**Melting point (℃)**	**Boiling point (℃)**
$CaCO_3$	1339	~
$Ca(OH)_2$	580	2850
CaO	2570	2850
Na_2CO_3	851	1600
NaOH	318	1388
K_2CO_3	891	1689
K_2O	490	691
KOH	360	1327
Fe_2O_3	1565	3200
Fe_3O_4	1565	2861
$Fe(OH)_3$	1565	3000
Al_2O_3	2054	3000
$Al(OH)_3$	300	2850

Note: K_2CO_3 and Na_2CO_3 undergo carbon thermal shock for 5 s at 800℃, presenting a molten state, which is detrimental to the separation of the anode sheet and the medium, and thus, they are excluded.

Table S5 Data on the effect of adding different media on lithium leaching rate and separation rate

Added media	**No. of experiments**	**Li leaching rate under different mediums**	**Separation rate under different mediums**
No medium	1	31.52	68.93
	2	34.13	69.99
	3	33.01	70.40
	Standard deviation	1.26	0.80
$CaCO_3$	1	72.11	93.21
	2	74.94	95.12
	3	73.30	94.54
	Standard deviation	1.51	1.04
$Ca(OH)_2$	1	28.96	87.53
	2	32.22	88.53
	3	31.62	88.41
	Standard deviation	1.64	0.55
CaO	1	33.02	98.04
	2	33.98	100.96
	3	34.61	101.04
	Standard deviation	0.81	1.73
Fe_2O_3	1	45.96	88.52
	2	47.95	91.97
	3	47.41	87.18
	Standard deviation	1.03	2.49
Fe_3O_4	1	40.95	99.52
	2	43.08	99.93
	3	39.69	100.64
	Standard deviation	1.67	0.56
$Fe(OH)_3$	1	33.28	89.53
	2	32.05	91.94
	3	32.96	92.04
	Standard deviation	0.67	1.42
Al_2O_3	1	55.00	98.99
	2	49.96	99.32
	3	54.04	100.11
	Standard deviation	2.64	0.51

Table S6 Data on the lithium leaching rate under different experimental conditions

		Fe_2O_3				Al_2O_3				$CaCO_3$			
	Different conditions	1	2	3	Standard deviation	1	2	3	Standard deviation	1	2	3	Standard deviation
Li leaching rate under different temperatures	750℃	20.04	22.97	21.92	1.52	26.96	33.03	30.00	3.00	50.99	55.01	57.78	3.41
	800℃	21.96	26.02	26.04	2.32	32.01	36.96	36.72	2.81	60.00	65.05	63.17	2.53
	850℃	26.93	32.04	28.41	2.58	22.97	28.97	28.34	3.29	47.98	52.03	51.71	2.23
	900℃	40.05	43.96	43.61	2.21	31.03	38.02	38.86	4.31	43.02	49.99	53.90	5.52
	950℃	23.95	19.03	24.31	2.98	19.97	22.96	20.50	1.61	20.95	25.00	23.13	2.00
Li leaching rate under different holding times	Quick model-5 s	18.03	21.05	18.91	1.54	16.00	18.04	18.64	1.38	60.04	62.95	61.00	1.53
	5 s	21.95	22.96	22.03	0.57	16.99	18.94	19.14	1.20	73.20	75.01	70.96	2.00
	10 s	17.03	20.05	19.97	1.72	32.03	34.05	35.56	1.78	92.03	94.50	95.34	1.74
	15 s	7.96	13.02	9.96	2.52	21.95	27.02	24.78	2.51	96.95	99.75	101.78	2.40
	20 s	37.01	38.95	38.43	1.03	52.03	54.95	52.31	1.65	55.01	57.96	56.00	1.53
Li leaching rate under different mass ratios (Media: Anode sheet)	1:1	28.52	31.02	29.31	1.28	27.50	29.03	27.93	0.77	80.05	83.95	82.02	2.00
	2:1	35.01	37.04	35.24	1.09	57.95	59.01	57.69	0.68	97.97	100.02	100.58	1.35
	3:1	57.96	58.97	58.01	0.57	42.03	49.98	49.35	2.59	58.05	61.95	61.81	2.26
	4:1	50.50	51.99	51.17	0.75	99.01	100.04	101.00	1.00	68.88	71.03	70.03	1.00
	5:1	61.05	63.04	62.10	1.00	96.48	97.01	98.16	0.85	77.73	76.89	75.11	1.41
Li leaching rate under different shock times	1 time	55.00	59.03	58.78	2.25	98.03	99.50	99.93	1.01	99.32	99.84	99.48	0.25
	2 times	71.99	75.50	75.41	2.00	49.95	55.02	52.23	2.50	95.00	93.04	94.49	1.04
	3 times	68.98	71.03	70.88	1.12	60.01	61.96	60.44	1.05	63.50	65.01	64.82	0.82

Table S7 First ionization energy of metallic elements and lattice energy of the corresponding metal fluorides

Element	Ionization energy (eV)	Compound	Lattice energy (kJ/mol)
Li	5.3917	LiF	1030
Ca	6.1132	CaF_2	2640
Al	5.9858	AlF_3	5924
Fe	7.9025	FeF_3	5870
Na	5.1391	NaF	910

Note: NIST: Atomic Spectra Database - Ionization Energies Form.

Table S8 Contents of different elements in raw materials (wt.%), calcined products, and calcined mediums

	C	O	Li	Cu	F
Raw material (SG)	64.4	16.11	6.63	3.65	9.21
Calcined graphite-no medium	81.61	9.19	5.48	1.72	2
Calcined graphite-$CaCO_3$	75	14.16	8.11	1.35	1.39
Calcined graphite-Al_2O_3	73.79	16.98	6.92	1.78	2.04
Calcined graphite-Fe_2O_3	81.62	8.37	8.04	1.01	0.97
Calcined medium-$CaCO_3$	42.15	32.95	–	–	11.23
Calcined medium-Al_2O_3	12.85	49.52	–	–	4.18
Calcined medium-Fe_2O_3	19.22	60.90	–	–	2.26

Table S9 Gas products from CTS anode sheet

Peak No.	RT (Min)	Area Percentage (%)	Compounds
1	1.479	1.16	Carbon dioxide
2	4.225	1.01	Cyclopropyl carbinol
3	5.182	0.97	1,2-Benzenediol, O-(3-Cyclopentylpropionyl)-O'-(1-naphthoyl)-
4	13.543	27.14	1,3-Dioxolan-2-one
5	13.724	8.15	Phosphonic acid, (p-hydroxyphenyl)-
6	15.309	4.23	2-Ethyl-1-hexanol
7	15.571	0.71	2-Pyrrolidinone, 1-methyl-
8	19.803	0.63	5-Octadecene, (E)-
9	19.976	1.45	3-Ethyl-6-trifluoroacetoxyoctane
10	20.090	1.69	Dichloroacetic acid, 6-ethyl-3-octyl ester
11	20.208	1.25	1-Hexanol, 3,5,5-trimethyl-
12	20.281	1.79	Formic acid, 2-propylpentyl ester
13	20.363	1.63	5-Tetradecene, (E)-
14	20.511	2.13	2-Hexadecene, 3,7,11,15-tetramethyl-, [R-[R*,R*-(E)]]-
15	20.799	0.68	7-Tetradecanol
16	20.872	0.75	5-Dodecanol
17	20.959	1.36	3,7-Dimethyloctyl acetate
18	21.312	0.97	7-Tridecanol
19	28.287	22.86	Pentanoic acid, 2,2,4-trimethyl-3-carboxyisopropyl, isobutyl ester
20	33.217	0.77	m-Terphenyl

Table S10 Gas products from CTS anode sheet with $CaCO_3$

Peak No.	RT (Min)	Area Percentage (%)	Compounds
1	4.247	1.65	Cyclopropyl carbinol
2	5.664	1.08	Tetrahydrofuran
3	13.679	10.76	Phosphonic acid, (p-hydroxyphenyl)-
4	15.303	5.98	2-Ethyl-1-hexanol
5	17.833	0.99	Hexanoic acid, 2-ethyl-
6	19.362	0.96	Ethylene diacrylate
7	19.801	0.98	5-Octadecene, (E)-
8	19.793	2.2	3-Ethyl-6-trifluoroacetoxyoctane
9	20.056	1.18	Ethanone, 1-(1-methylcyclopentyl)-
10	20.089	1.3	3-Cyclopropylcarboxy-6-ethyldecane
11	20.207	1.6	1-Hexanol, 3,5,5-trimethyl-
12	20.280	2.69	2-Propyl-1-Pentanol, trifluoroacetate
13	20.361	2.38	5-Tetradecene, (E)-
14	20.509	3.2	1-Octanol, 2,7-dimethyl-
15	20.797	1.06	7-Tetradecanol
16	20.870	1.15	Cyclopropanecarboxylic acid,3-methylbutyl ester
17	20.957	2.11	3,7-Dimethyloctyl acetate
18	21.313	1.73	Cyclohexane, 1-methyl-3-propyl-
19	28.285	24.38	Pentanoic acid, 2,2,4-trimethyl-3-carboxyisopropyl, isobutyl ester
20	31.128	1.39	Benzoic acid, dec-2-yl ester

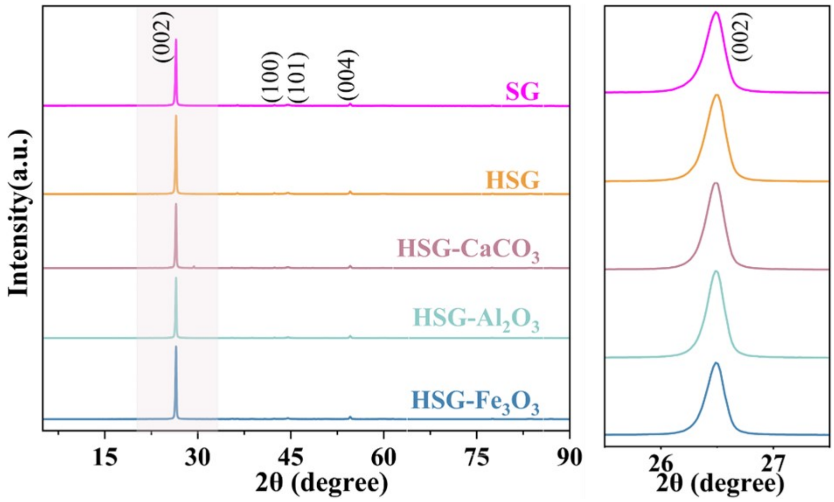

Fig. S2 XRD patterns of spent graphite, HSG without medium, and HSG samples with different mediums.

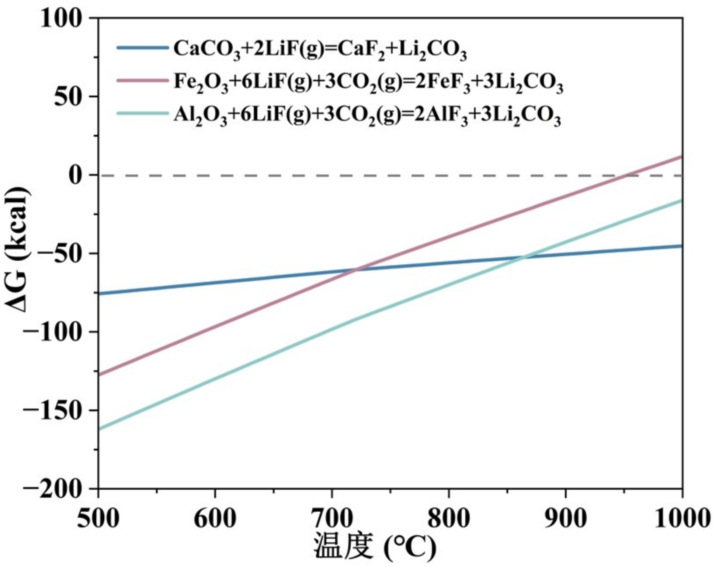

Fig. S3 Gibbs free energy calculation for possible reactions.

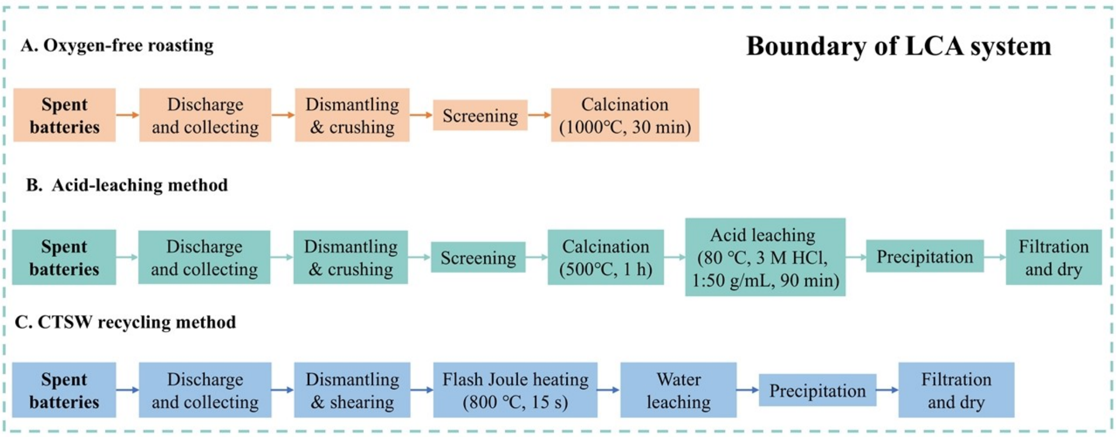

Fig. S4 The boundary of the system.

Process	Input	Amount	Unit	Output	Amount	Unit	Type	Flow
Discharge	Spent batteries	1	kg	Discharged battery	1	kg		
	NaCl	0.026	kg	NaCl	0.026	kg		
	H_2O	0.52	kg	H_2O	0.52	kg		
	Electricity	0.08	kWh					
Dismantling & Crushing	Discharged battery	1	kg	Broken anode products	0.20	kg		
	Electricity	0.1	kWh					
Screening	Broken anode products	0.20	kg	Overflow products (coarse Cu)	0.04	kg	product	sale
	Electricity	1	kWh	Underflow products	0.16	kg		
Calcination	Underflow products	0.16	kg	Calcined graphite	0.12	kg		
	Electricity	64	kWh	Exhaust gas	26.54	kg	waste	
	N_2	26.07	kg					
Gas treatment	Exhaust gas	26.54	kg	Clean gas	26.13	kg		to air
	Electricity	0.12	kWh	Ca $(OH)_2$ (aq)	0.7105	kg		circulate
	H_2O	0.30	kg					
	Ca $(OH)_2$	0.0005	kg					

Table S11 Data inventory for the oxygen-free roasting method

Note: N_2 flow rate was based on 50 mL/min.

Table S12 Data inventory of the acid leaching method

Process	Input	Amount	Unit	Output	Amount	Unit	Type	Flow
Discharge	Spent batteries	1	kg	Discharged battery	1	kg		
	NaCl	0.026	kg	NaCl	0.026	kg		
	H_2O	0.52	kg	H_2O	0.52	kg		
	Electricity	0.08	kWh					
Dismantling & Crushing	Discharged battery	1	kg	Broken anode products	0.20	kg		
	Electricity	0.1	kWh					
Screening	Broken anode products	0.20	kg	Overflow products	0.04	kg	product	sale
	Electricity	1	kWh	Underflow products	0.16	kg		
Calcination	Underflow products	0.16	kg	Calcined graphite	0.146	kg		
	Electricity	60	kWh	Exhaust gas	12.33	kg		
	N_2	10.6	kg					
Gas treatment	Exhaust gas	12.33	kg	Clean gas	10.8	kg		to air
	Electricity	0.45	kWh	$Ca(OH)_2$ (aq)	2.62	kg		circulate
	H_2O	1.09	kg					
	$Ca(OH)_2$	0.0018	kg					
Acid leaching	Calcined graphite	0.146	kg	Graphite residue	0.1453	kg		
	HCl	1.825	kg	Filtrate	7.3007	kg		
	Electricity	27.9	kWh					
	H_2O	5.475	kg					
Precipitation	Filtrate	7.3007	kg	Li_2CO_3 (1)	0.0033	kg		
	Na_2CO_3	0.021	kg	Wastewater	7.3724	kg		
	Electricity	18.6	kWh					
	H_2O	0.05	kg					
	NaOH	0.004	kg					
Filtration & Dry	H_2O	0.0024	kg					
	Li_2CO_3 (1)	0.0033	kg	Li_2CO_3 (2)	0.0032	kg	product	sale
	Electricity	19.24	kWh					

Note: High-speed multifunctional grinder (SL-350), power 1.4 kW; vibrating sieve

(BO-300SY), power 0.25 kW; N_2 flow rate calculated as 50 mL/min, muffle furnace heating rate at 10℃/min, power 60 kW; mechanical stirrer power 18.6 kW; Na_2CO_3 addition ratio/theoretical ratio = 4, lithium precipitation rate calculated at 90%; circulating water vacuum pump 0.18 kW; oven power 1.6 kW.

Process	**Input**	**Amount**	**Unit**	**Output**	**Amount**	**Unit**	**Type**	**Flow**
Discharge	Spent batteries	1	kg	Discharged battery	1	kg		
	NaCl	0.026	kg	NaCl	0.026	kg		
	H_2O	0.52	kg	H_2O	0.52	kg		
	Electricity	0.08	kWh					
Dismantling & shearing	Discharged battery	1	kg	Anode sheet	0.20	kg		
	Electricity	0.005	kWh					
CTS	Anode sheet	0.20	kg	Calcined anode sheet	0.20	kg		
	$CaCO_3$	0.40	kg	$CaCO_3$	0.40	kg	waste	circulate
	Electricity (800℃, 15 s)	9.5	kWh	Exhaust gas	0.25	kg	waste	to air
	N_2	4	kg					
Water leaching	Calcined anode sheet	0.20	kg	Graphite residue	0.1652	kg		
	Electricity (Ultrasonic 10s)	0.001	kWh	Cu	0.034	kg	product	sale
	Water	6	kg	Filtrate	6.0008	kg		
Precipitation	Filtrate	6.0008	kg	Li_2CO_3 (1)	0.0038	kg		
	Na_2CO_3	0.024	kg	Wastewater	6.077	kg		
	Electricity (95℃,1 h)	18.6	kWh					
	H_2O	0.056	kg					
Filtration & Dry	H_2O	0.0028	kg	Li_2CO_3 (2)	0.0037	kg	product	sale
	Li_2CO_3 (1)	0.0038	kg					
	Electricity (105℃,12 h)	19.24	kWh					

Table S13 Data inventory of the CTSW method

Note: N_2 flow rate was calculated as 20 mL/min; mechanical stirrer power 18.6 kW; ultrasonic cleaner 0.5 kW; Joule heat heating device 2.85 kW (voltage 30V, current 95A, temperature 800℃, insulation for 15s), for pyrolysis of 0.25 g per cycle; Na_2CO_3 addition ratio/theoretical ratio = 4, lithium precipitation rate calculated at 90%; circulating water vacuum pump 0.18 kW; oven power 1.6 kW.

Table S14 Environmental impact analysis results comparison of different processes

	GWP 100 (kg CO_2 eq.)	ADP elements (kg Sb eq.)	ADP fossil (MJ)	AP	EP	FAETP	HTP	MAETP	ODP	POCP	TETP
A. Oxygen-free roasting	1.20E-11	-1.79E-12	9.93E-12	4.05E-12	6.54E-13	1.81E-13	8.28E-12	1.97E-10	7.94E-18	3.32E-12	2.52E-13
B. Acid-leaching	2.26E-11	-1.60E-12	1.88E-11	7.63E-12	1.23E-12	3.52E-13	2.01E-11	3.73E-10	1.53E-17	6.23E-12	5.36E-13
C. CTSW	8.27E-12	-1.52E-12	6.78E-12	2.79E-12	4.53E-13	1.20E-13	4.90E-12	1.07E-10	5.48E-18	2.29E-12	1.62E-13

Table S15 Environmental impact analysis results for the oxygen-free roasting by steps

	Discharge and collecting	Dismantling & Crushing	Screening	Calcination	Gas treatment	Graphite recovery	Cu recovery	Total
GWP 100	1.47E-14	1.75E-14	1.75E-13	1.19E-11	2.12E-14	-8.96E-14	-3.62E-14	1.20E-11
ADP elements	7.77E-15	7.63E-17	7.63E-16	5.21E-14	1.60E-16	-5.42E-16	-1.85E-12	-1.79E-12
ADP fossil	1.25E-14	1.46E-14	1.46E-13	9.94E-12	1.77E-14	-1.69E-13	-3.39E-14	9.93E-12
AP	5.04E-15	5.96E-15	5.96E-14	4.05E-12	7.19E-15	-1.43E-14	-6.21E-14	4.05E-12
EP	8.69E-16	9.54E-16	9.54E-15	6.48E-13	1.16E-15	-1.49E-15	-4.54E-15	6.54E-13
FAETP	2.70E-16	2.81E-16	2.81E-15	1.91E-13	3.46E-16	-7.23E-15	-6.25E-15	1.81E-13
HTP	1.61E-14	1.96E-14	1.96E-13	1.33E-11	2.37E-14	-3.01E-14	-5.29E-12	8.28E-12
MAETP	2.37E-13	2.90E-13	2.90E-12	1.97E-10	3.50E-13	-4.19E-13	-3.19E-12	1.97E-10
ODP	9.61E-21	1.17E-20	1.17E-19	7.95E-18	1.41E-20	-2.44E-20	-1.32E-19	7.94E-18
POCP	4.09E-15	4.85E-15	4.85E-14	3.29E-12	5.85E-15	-1.14E-14	-2.33E-14	3.32E-12
TETP	4.27E-16	4.69E-16	4.69E-15	3.18E-13	5.84E-16	-4.12E-15	-6.85E-14	2.52E-13

Table S16 Environmental impact analysis results for the acid leaching method by steps

	Discharge and collecting	Dismantling & Crushing	Screening	Calcination	Gas treatment	Acid leaching	Precipitation	Filtration & Dry	Li_2CO_3 recovery	Graphite recovery	Cu recovery	Total
GWP 100	1.47E-14	1.75E-14	1.75E-13	1.08E-11	7.96E-14	5.02E-12	3.26E-12	3.37E-12	-2.82E-15	-1.08E-13	-3.62E-14	2.26E-11
ADP elements	7.77E-15	7.63E-17	7.63E-16	4.71E-14	5.92E-16	1.58E-13	2.47E-14	1.47E-14	-3.10E-15	-6.56E-16	-1.85E-12	-1.60E-12
ADP fossil	1.25E-14	1.46E-14	1.46E-13	9.02E-12	6.65E-14	4.26E-12	2.73E-12	2.82E-12	-2.80E-15	-2.05E-13	-3.39E-14	1.88E-11
AP	5.04E-15	5.96E-15	5.96E-14	3.67E-12	2.70E-14	1.69E-12	1.11E-12	1.15E-12	-1.30E-15	-1.74E-14	-6.21E-14	7.63E-12
EP	8.69E-16	9.54E-16	9.54E-15	5.87E-13	4.34E-15	2.75E-13	1.78E-13	1.83E-13	-3.74E-16	-1.81E-15	-4.54E-15	1.23E-12
FAETP	2.70E-16	2.81E-16	2.81E-15	1.73E-13	1.30E-15	8.33E-14	5.23E-14	5.40E-14	-3.78E-17	-8.75E-15	-6.25E-15	3.52E-13
HTP	1.61E-14	1.96E-14	1.96E-13	1.21E-11	8.89E-14	5.54E-12	3.66E-12	3.78E-12	-7.13E-16	-3.64E-14	-5.29E-12	2.01E-11
MAETP	2.37E-13	2.90E-13	2.90E-12	1.79E-10	1.31E-12	8.27E-11	5.40E-11	5.59E-11	-9.47E-15	-5.08E-13	-3.19E-12	3.73E-10
ODP	9.61E-21	1.17E-20	1.17E-19	7.21E-18	5.30E-20	3.59E-18	2.18E-18	2.25E-18	-5.68E-22	-2.96E-20	-1.32E-19	1.53E-17
POCP	4.09E-15	4.85E-15	4.85E-14	2.99E-12	2.19E-14	1.37E-12	9.03E-13	9.33E-13	-6.49E-16	-1.38E-14	-2.33E-14	6.23E-12
TETP	4.27E-16	4.69E-16	4.69E-15	2.89E-13	2.19E-15	1.35E-13	8.73E-14	9.02E-14	-6.13E-17	-4.99E-15	-6.85E-14	5.36E-13

Table S17 Environmental impact analysis results for the CTSW method by steps

	Discharge and collecting	Dismantling & Shearing	CTS	Water leaching	Precipitation	Filtration & Dry	Li_2CO_3 recovery	Graphite recovery	Cu recovery	Total
GWP 100	1.47E-14	8.76E-16	1.77E-12	2.70E-15	3.26E-12	3.37E-12	-3.26E-15	-1.23E-13	-3.08E-14	8.27E-12
ADP elements	7.77E-15	3.82E-18	7.77E-15	1.37E-15	2.51E-14	1.47E-14	-3.59E-15	-7.46E-16	-1.58E-12	-1.52E-12
ADP fossil	1.25E-14	7.32E-16	1.48E-12	2.60E-15	2.73E-12	2.82E-12	-3.24E-15	-2.33E-13	-2.88E-14	6.78E-12
AP	5.04E-15	2.98E-16	6.02E-13	8.49E-16	1.11E-12	1.15E-12	-1.51E-15	-1.97E-14	-5.28E-14	2.79E-12
EP	8.69E-16	4.77E-17	9.64E-14	2.70E-16	1.79E-13	1.83E-13	-4.32E-16	-2.06E-15	-3.86E-15	4.53E-13
FAETP	2.70E-16	1.40E-17	2.84E-14	1.72E-16	5.23E-14	5.40E-14	-4.37E-17	-9.95E-15	-5.31E-15	1.20E-13
HTP	1.61E-14	9.82E-16	1.99E-12	2.71E-15	3.66E-12	3.78E-12	-8.24E-16	-4.14E-14	-4.50E-12	4.90E-12
MAETP	2.37E-13	1.45E-14	2.94E-11	3.67E-14	5.40E-11	5.59E-11	-1.09E-14	-5.77E-13	-2.71E-12	1.36E-10
ODP	9.61E-21	5.85E-22	1.19E-18	1.43E-21	2.18E-18	2.25E-18	-6.57E-22	-3.36E-20	-1.12E-19	5.48E-18
POCP	4.09E-15	2.42E-16	4.90E-13	6.76E-16	9.04E-13	9.33E-13	-7.51E-16	-1.57E-14	-1.98E-14	2.29E-12
TETP	4.27E-16	2.34E-17	4.75E-14	2.73E-16	8.73E-14	9.02E-14	-7.09E-17	-5.68E-15	-5.82E-14	1.62E-13

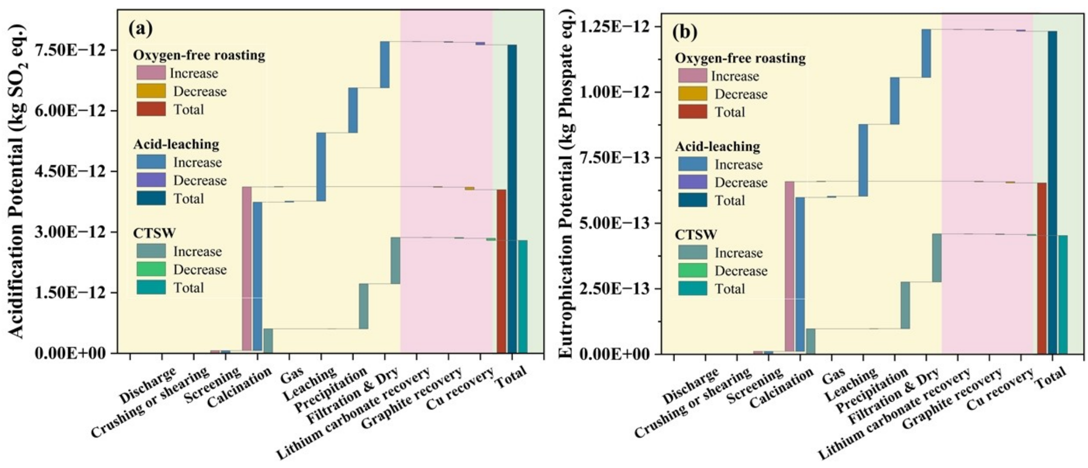

Fig. S5 Comparison of environmental impact between CTS, acid leaching, and conventional roasting process for utilizing spent graphite based on life cycle assessment (a. acidification potential; b. eutrophication potential)

This study adopted the TEA-LCA coupling method to conduct a techno-economic analysis of the innovative approach. The economic costs and quality parameters of each process are shown in the table below.

Table S18 Cost evaluation of various methods

Impact Category	Oxygen-free roasting		Acid leaching		CTSW	
	Dosage (kg or kWh)	Cost (CNY)	Dosage (kg or kWh)	Cost (CNY)	Dosage (kg or kWh)	Cost (CNY)
NaCl	0.026	1.3	0.026	1.3	0.026	1.3
N_2	26.07	52.14	10.6	21.2	4	8
HCl			1.825	65.7		
Water	0.82	0.0034	7.1374	0.0293	6.5788	0.0270
Na_2CO_3			0.021	0.924	0.024	1.056
Ca $(OH)_2$	0.0005	0.027	0.0018	0.0972		
NaOH			0.004	0.136		
$CaCO_3$					0.4	34.4
SUM of Materials		53.4704		89.3865		44.783
Discharge	0.08	0.12	0.08	0.12	0.08	0.12
Crushing & Shearing	0.1	0.15	0.1	0.15	0.005	0.0075
Screening	1	1.5	1	1.5		

Calcination	64	96	60	90	9.5	14.25
Gas treatment	0.12	0.18	0.45	0.675		
Leaching			27.9	41.85	0.001	0.0015
Precipitation			18.6	27.9	18.6	27.9
Filtration and Dry			19.24	28.86	19.24	28.86
SUM of Energy		97.95		191.055		71.139
Cu	0.04	2.242	0.04	2.242	0.034	2.04
Graphite	0.12	5.1	0.1453	6.1752	0.1652	7.021
Li_2CO_3			0.0032	0.389	0.0038	0.463
SUM of Revenue		7.342		8.8062		9.524
SUM		144.078		271.635		106.398

Note: The consumed energy was assumed to be from electricity.

In this study, raw material costs were based on commercial product prices, including NaCl (50 CNY/kg)、N_2 (2 CNY/L)、HCl (36 CNY/L)、Na_2CO_3 (44 CNY /kg)、NaOH (34 CNY/kg)、Ca $(OH)_2$ (54 CNY/kg)、$CaCO_3$ (86 CNY/kg)、water (0.0041 CNY/L)、coarse copper (56.05 CNY/kg,[16])、Cu sheet (60 CNY/kg)、graphite (42.5 CNY/kg,[16])、Li_2CO_3 (121.86 CNY/kg). Some price data was sourced from EverBatt 2023, reagent prices are sourced from the China National Pharmaceutical Reagent website. Energy consumption costs were based on the industrial electricity price in China (Beijing), which is 1.5 CNY/kWh. Operating costs are calculated as the sum of material and energy costs, excluding labor costs. Notably, this study calculated only the costs and benefits of anode recovery, excluding the cathode, separator, and other components. Moreover, since the mass of functional unit products was low while equipment operating costs were high, the cost calculation results in generally low returns.

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