Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

Low-carbon Recycling of Spent Lithium Iron Phosphate Batteries *via* a Hydro-oxygen Repair Route

Kang Liu [a,e], Junxiong Wang [b], Mengmeng Wang [a,e], Qiaozhi Zhang [a], Yang Cao [a], Longbin Huang [c], Marjorie Valix [d], Daniel C. W. Tsang [a,e]*

[a] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China

[b] Frontiers Science Center for Transformative Molecules, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

[c] Ecological Engineering of Mine Wastes, Sustainable Minerals Institute, The University of Queensland, Brisbane, Qld 4072, Australia

[d] School of Chemical and Biomolecular Engineering, University of Sydney, New South Wales 2006, Australia

[e] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, China

* Corresponding author: dan.tsang@polyu.edu.hk

Supporting Information

Table of Content

Figure S1

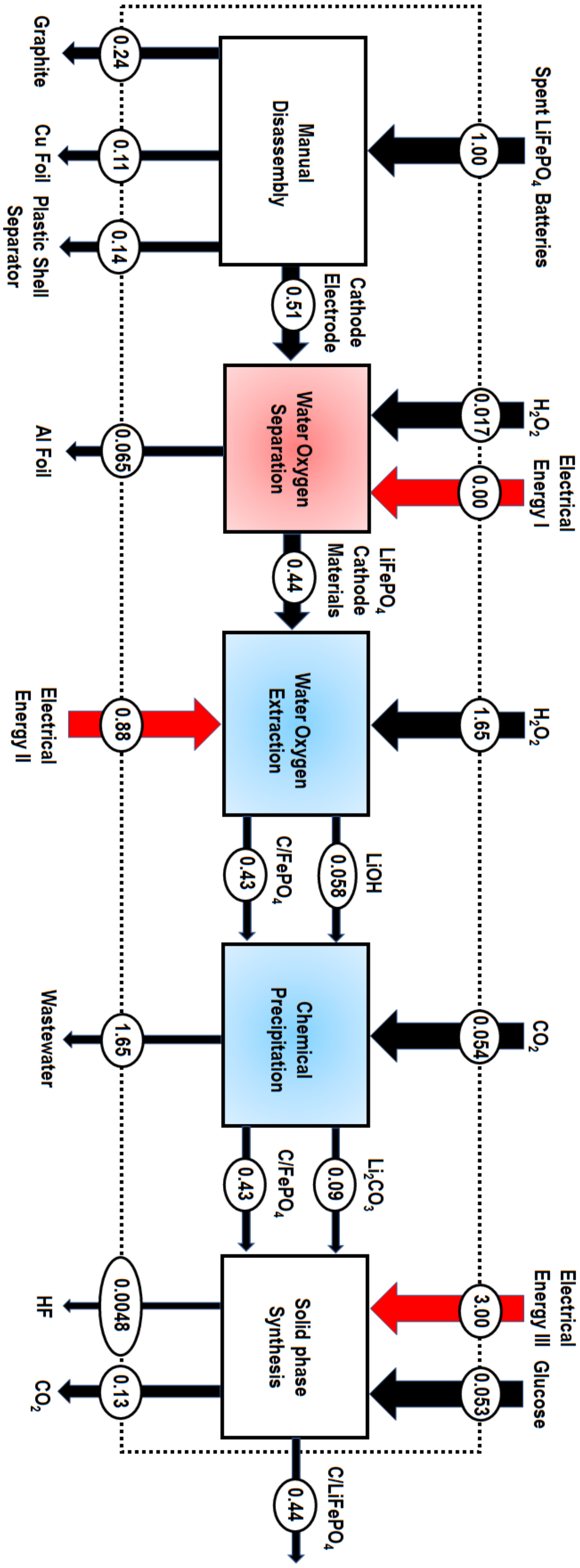

Figure S1 Life cycle boundaries for HOR route

Figure S2

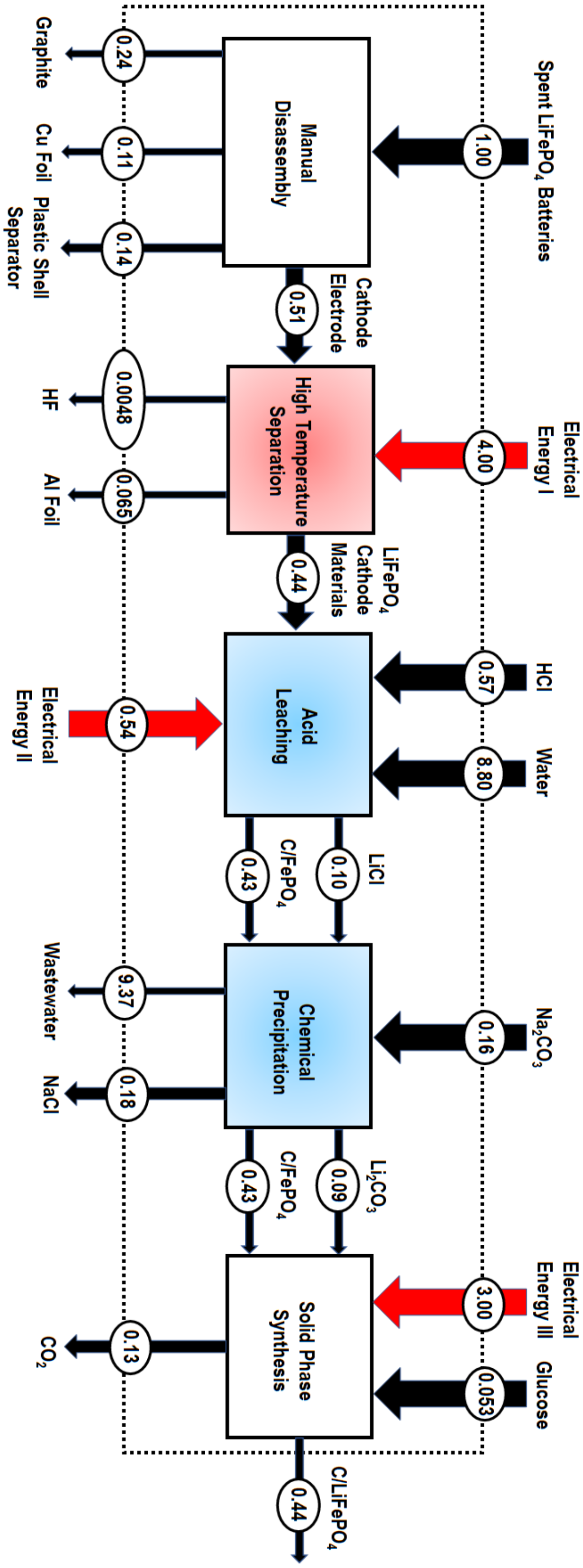

Figure S2 Life cycle boundaries for pyrometallurgy & hydrometallurgy route

Figure S3

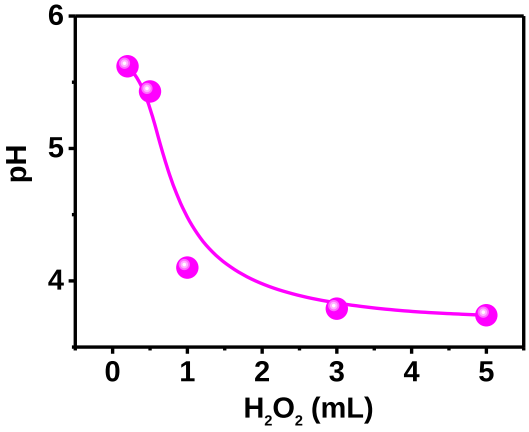

Figure S3 pH of different concentrations of hydro-oxygen solution.

Figure S4

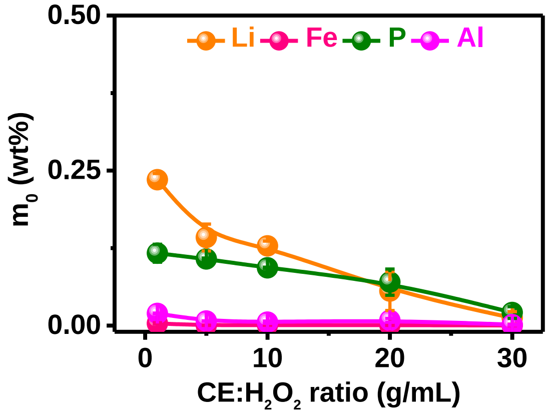

Figure S4 Effects of CE:H_2O_2 ratio on the leaching percentages of elements during the hydro-oxygen separation (time of 0.25 min).

Figure S5

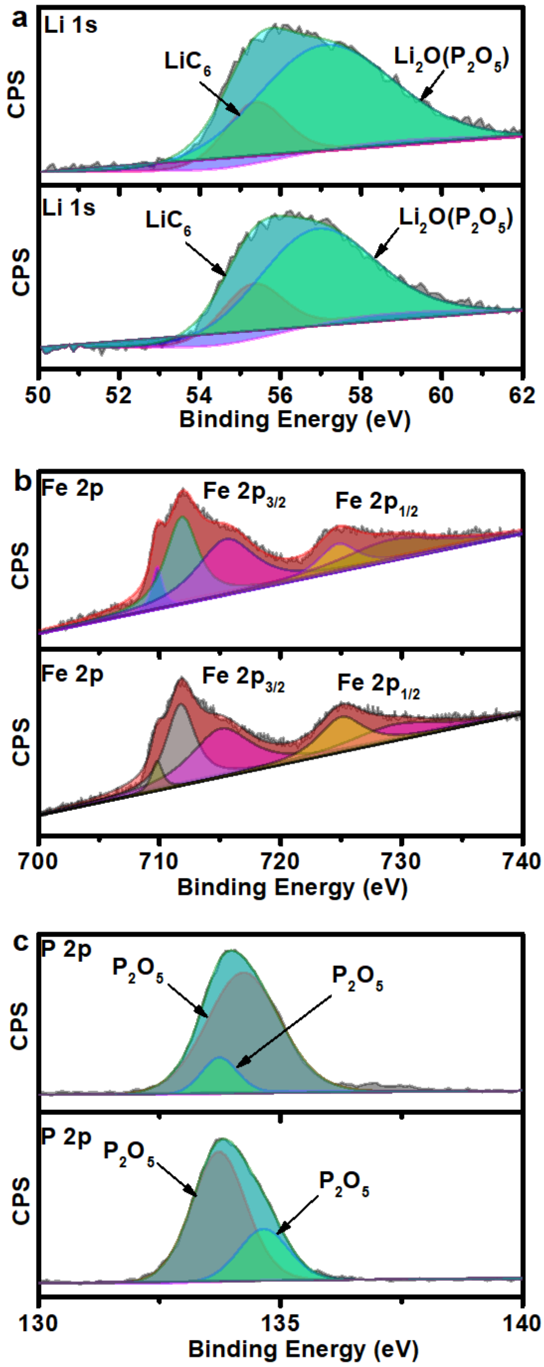

Figure S5 XPS high-resolution energy spectra of (a) Li 1s, (b) Fe 2p, (c) P 2p of CE (up) and LFP (down) cathode materials.

Figure S6

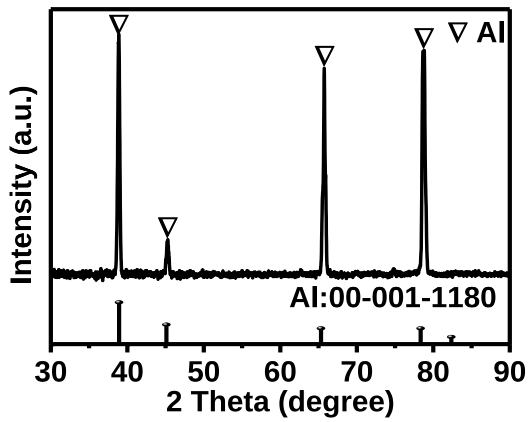

Figure S6 XRD patterns of the peeled aluminum foil products

Figure S7

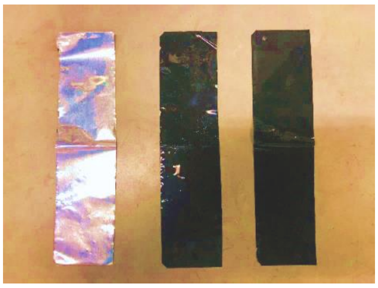

Figure S7 Physical image of cathode materials and aluminum foil after hydro-oxygen separation.

Figure S8

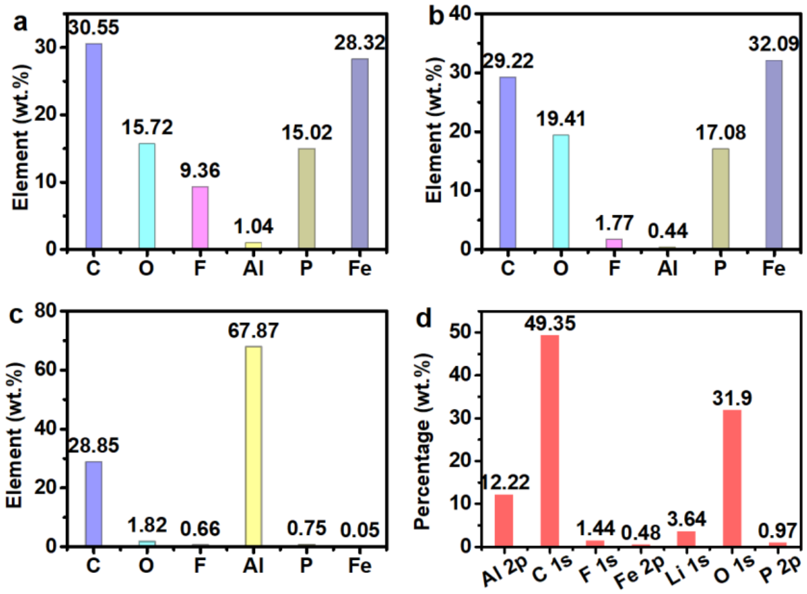

Figure S8 EDAX results of different products (a) cathode electrode, (b) cathode materials, (c) aluminum foil; (d) XPS results of aluminum foil.

Figure S9

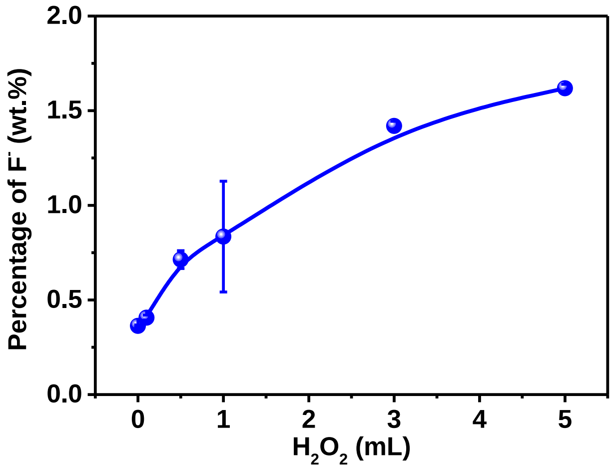

Figure S9 The leaching percentage of fluoride ions in hydro-oxygen solutions with different H_2O_2 concentrations.

Figure S10

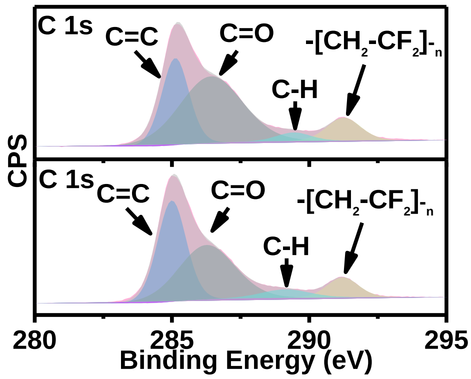

Figure S10 High-resolution C 1s spectra of cathode materials (a) before and (b) after hydro-oxygen separation.

(C=C: 284.8 eV, C=O: 286.3 eV, C-H: 289.2 eV, and -[CH_2-CF_2]-: 291.2 eV)

Figure S11

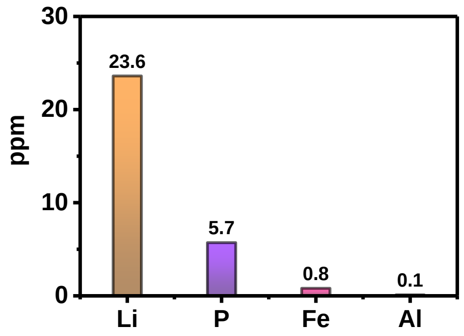

Figure S11 The concentrations of elements in the leachate (conditions: S/L (g/mL) ratio of 1:37.5, the mechanochemical time of 10 min, room temperature, and the rotational speed of 1000 rpm).

Figure S12

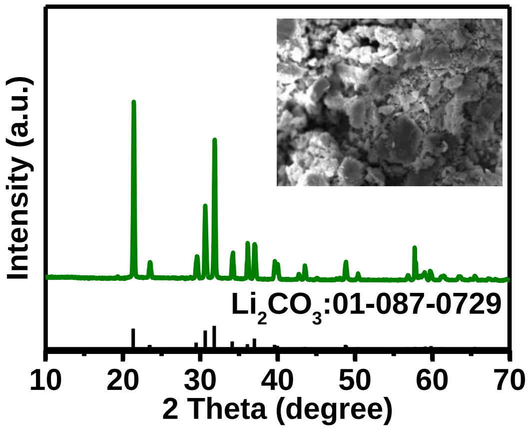

Figure S12 The XRD pattern and SEM image of the obtained Li_2CO_3 product

Figure S13

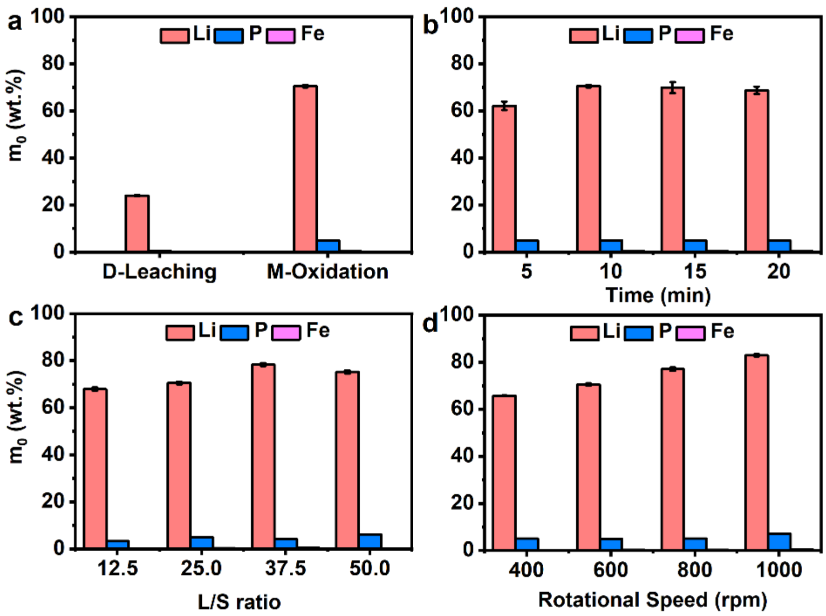

Figure S13 (a) the oxidative extraction efficiencies of Li in different scenarios (I) Direct leaching of cathode material piece; (II) Li extraction by mechanochemical oxidation (M-oxidation); different extraction parameters: (b) time, (c) S/L (g/mL) ratio, and (d) rotational speed, on Li extraction efficiencies.

Conditions:

(a) Direct Leaching: time of 10 min, S/L (g/mL) ratio of 1:25;

Mechano-Oxidation: time of 10 min, S/L (g/mL) ratio of 1:25, rotational speed of 600 rpm;

(b) rotational speed of 400 rpm, S/L (g/mL) ratio of 1:25;

(c) time of 10 min, rotational speed of 400 rpm;

(d) time of 10 min, S/L (g/mL) ratio of 1:25.

Compared with direct leaching, the S-LFP sheet can be hermetically broken by the destructive action of grinding balls in the mechanochemical process, thus significantly improving the extraction efficiency of Li (Figure S13a). Extending the time was not obvious for the extraction of Li, because oxidative extraction was a relatively fast process (Figure S13b). Increasing the S/L (g/mL) ratio from 1:25 to 1:37.5 slightly improved the oxidative extraction efficiency of Li (Figure S13c). The improvement of the Li extraction efficiency by the rotational speed was extremely obvious (Figure S13d). When the rotational speed was 400 rpm, the extraction efficiency of Li was 65.8 wt.%, and when the rotational speed was 1000 rpm, the extraction efficiency of Li was 83.0 wt.%. The optimal parameters for initial leaching of Li were the S/L (g/mL) ratio of 1:37.5, the mechanochemical time of 10 min, and the rotational speed of 1000 rpm, with a highest Li extraction percentage of 83.0 wt.%. The remaining Li in the residue can be further extracted by performing secondary leaching. Finally, we will obtain Li containing leaching solution and Li-free $FePO_4$ residue.

Figure S14

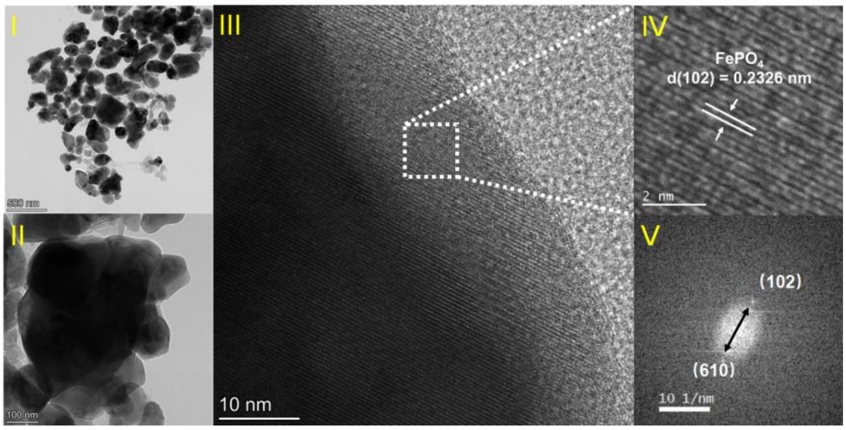

Figure S14 TEM, HR-TEM, and FFT results of $FePO_4$

(I) TEM:500 nm scale, (II) TEM:100 nm scale, (III) HRTEM: 10 nm scale, (IV) HRTEM: 2 nm, and (V) FFT results.

Under the observation of TEM, the $FePO_4$ presented irregular round particles (Figure 14 I). Further magnification image revealed that the irregular particles clustered together (Figure 14 II). The $FePO_4$ crystal has a clear lattice spacing with a distance of 0.2326 nm, fitting with the spacing of the (102) planes by $FePO_4$ (JCPDS card No. 83-2092) (Figure 14 III and IV). The FFT result (Figure 14 V) shows that the nano-single crystal character of the $FePO_4$ cathode material and the (102) crystal plane can be seen.

Figure S15

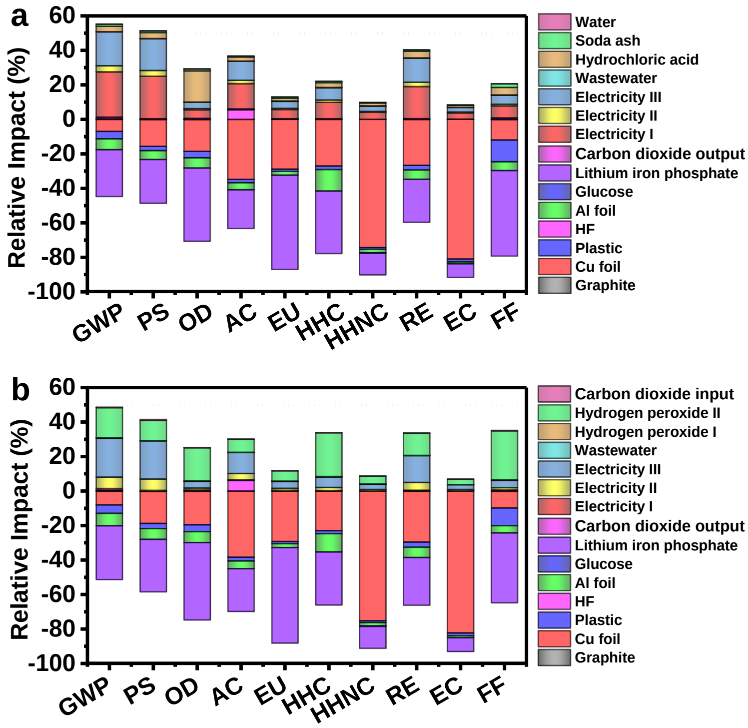

Figure S15 Contribution analysis of each material and energy consumption on the selected indicators: (a) pyro & hydro, and (b) HOR. Negative values indicate benefits (reduction of impacts due to recovered materials), positive values are environmental impacts (due to inputs for the recycling process).

In the pyro & hydro process, the positive values of GWP and PS were greater than 50%, which means that the pyro & hydro process will have a negative impact on the global environment in terms of carbon footprint and acid rain formation. Overall, within the selected indicators, the positive values of HOR were all less than 50%, which means that the designed process can reduce the environmental impacts on the ecological environment to varying degrees (Figure S15).

Table S1 Disassembly data of spent $LiFePO_4$ battery

Composition		Weight (g)
Cathode electrode		274.21
	Aluminum foil	35.57
	Cathode materials	238.64
Anode electrode		190.80
	Copper foil	60.11
	Graphite	130.69
Soft shell (Plastic)		22.10
Separator Plastic (Plastic)		56.16
Sum		543.27

Table S2 Element proportion in $LiFePO_4$ cathode materials

Element	Percentage (wt.%)
Li	3.34
Fe	33.72
P	17.76
C	7.07
Al	0.03

Table S3 Life cycle inventory for recycling 1.0 kg spent $LiFePO_4$ battery (HOR)

Items	Items	Amount	Unit	Corresponding LCI	Database	Comment
Input	Electricity I	0.00	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
Input	Electricity II	0.88	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
Input	Electricity III	3.00	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
Input	Spent $LiFePO_4$ battery	1.00	kg	/	/	/
Input	H_2O_2 I	0.017	kg	Hydrogen peroxide, without water, in 50% solution state {RoW}| market for hydrogen peroxide	Ecoinvent 3	
Input	H_2O_2 II	1.65	kg	Hydrogen peroxide, without water, in 50% solution state {RoW}| market for hydrogen peroxide	Ecoinvent 3	
Input	Glucose	0.0053	kg	Glucose {GLO}| market for glucose | APOS, U	Ecoinvent 3	
Input	CO_2	0.0054	kg	Liquid {Row}| market for | APOS, U	Ecoinvent 3	
Output	Graphite	0.24	kg	Graphite {GLO}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
Output	Al foil	0.065	kg	Aluminium collector foil, for Li-ion battery {GLO}| market for aluminium collector foil, for Li-ion battery | APOS, U	Ecoinvent 3	Avoided product/ Co-product
Output	Cu foil	0.11	kg	Copper collector	Ecoinvent 3	Avoided

				foil, for Li-ion battery {GLO}| market for copper collector foil, for Li-ion battery | APOS, U		product/ Co-product
	Plastic (Shell&Separator)	0.14	kg	Battery separator {GLO}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	$LiFePO_4$ cathode materials	0.44	kg	Self-modeling	Ref. 1	Intermediate product
	$FePO_4$	0.39	kg	Self-modeling	Ref. 2	Avoided product/ Co-product
	HF	0.0048	kg	Air emission	Ecoinvent 3	Avoided product/ Co-product
	CO_2	0.13	kg	Liquid {Row}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Wastewater	1.65	kg	Wastewater, average {RoW}| market for wastewater, average | APOS, U	Ecoinvent 3	Avoided product/ Co-product

Table S4 Life cycle inventory for recycling 1.0 kg spent $LiFePO_4$ battery (Pyro & Hydro)

Items	Items	Amount	Unit	Corresponding LCI	Database	Comment
Input	Electricity I	4.00	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
	Electricity II	0.54	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
	Electricity III	3.00	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	
	Spent $LiFePO_4$ battery	1.00	kg	/	/	/
	HCl	0.57	kg	Hydrochloric acid, without water, in 30% solution state {RoW}| market for | APOS, U	Ecoinvent 3	
	Na_2CO_3	0.16	kg	Soda ash, dense {GLO}| market for | APOS, U	Ecoinvent 3	
	Glucose	0.0053	kg	Glucose {GLO}| market for glucose | APOS, U	Ecoinvent 3	
	Water	8.80	kg	Water, harvested from rainwater {GLO}| market for water, harvested from rainwater | APOS, U	Ecoinvent 3	
Output	Graphite	0.24	kg	Graphite {GLO}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Al foil	0.065	kg	Aluminium collector foil, for Li-ion battery {GLO}| market for aluminium	Ecoinvent 3	Avoided product/ Co-product

				collector foil, for Li-ion battery | APOS, U		
	Cu foil	0.11	kg	Copper collector foil, for Li-ion battery {GLO}| market for copper collector foil, for Li-ion battery | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Plastic (Shell&Separator)	0.14	kg	Battery separator {GLO}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	$LiFePO_4$ cathode materials	0.44	kg	Self-modeling	Ref. 1	Intermediate product
	$FePO_4$	0.39	kg	Self-modeling	Ref. 2	Avoided product/ Co-product
	HF	0.0048	kg	Air emission	Ecoinvent 3	Avoided product/ Co-product
	NaCl	0.18	kg	Municipal solid waste {RoW}| treatment of, sanitary landfill | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	CO_2	0.13	kg	Liquid {Row}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Wastewater	9.37	kg	Wastewater, average {RoW}| market for wastewater, average | APOS, U	Ecoinvent 3	Avoided product/ Co-product

Table S5 LCA data source for recycling 1.0 kg spent $LiFePO_4$ battery (HOR)

Impact category	OD	GWP	PS	AC	Eu	HHC	HHNC	RE	EC	FF
Unit	kg CFC-11 eq	kg CO_2 eq	kg O_3 eq	kg SO_2 eq	kg N eq	CTUh	CTUh	kg PM2.5 eq	CTUe	MJ surplus
Total	-5E-07	-4E-01	-2E-01	-5E-02	-1E-01	-8E-07	-1E-05	-6E-03	-1E+03	-4E+00
Graphite	-2E-09	-2E-02	-2E-03	-1E-04	-4E-05	-2E-09	-4E-09	-2E-05	-2E-01	-2E-02
Cu foil	-2E-07	-1E+00	-2E-01	-5E-02	-4E-02	-6E-07	-1E-05	-5E-03	-1E+03	-1E+00
Plastic (Shell&Separator)	-4E-08	-7E-01	-3E-02	-3E-03	-2E-03	-4E-08	-2E-07	-5E-04	-3E+01	-1E+00
HF	0E+00	0E+00	0E+00	8E-03	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00
Hydrogen peroxide I	2E-09	2E-02	1E-03	1E-04	8E-05	7E-09	8E-09	2E-05	5E-01	4E-02
Al foil	-7E-08	-1E+00	-6E-02	-6E-03	-3E-03	-3E-07	-3E-07	-1E-03	-2E+01	-6E-01
Hydrogen peroxide II	2E-07	2E+00	1E-01	1E-02	8E-03	6E-07	8E-07	2E-03	5E+01	4E+00
Electricity II	1E-08	9E-01	7E-02	5E-03	1E-03	5E-08	2E-07	8E-04	1E+01	2E-01
Carbon dioxide	2E-09	4E-02	6E-03	1E-04	1E-04	3E-09	1E-08	4E-05	7E-01	4E-02
Wastewater	7E-11	9E-04	8E-05	8E-06	5E-05	5E-10	5E-09	2E-06	3E-02	8E-04
Electricity III	4E-08	3E+00	2E-01	2E-02	5E-03	2E-07	5E-07	3E-03	4E+01	6E-01
Glucose	6E-09	7E-02	4E-03	6E-04	4E-04	8E-09	-5E-08	7E-05	2E+00	9E-02
Lithium iron phosphate	-5E-07	-4E+00	-3E-01	-3E-02	-7E-02	-8E-07	-2E-06	-5E-03	-1E+02	-5E+00
CO_2 emission	0E+00	1E-01	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00

Table S6 LCA data source for recycling 1.0 kg spent $LiFePO_4$ battery (Pyro & Hydro)

Impact category	OD	GWP	PS	AC	Eu	HHC	HHNC	RE	EC	FF
Unit	kg CFC-11 eq	kg CO_2 eq	kg O_3 eq	kg SO_2 eq	kg N eq	CTUh	CTUh	kg PM2.5 eq	CTUe	MJ surplus
Total	-5E-07	2E+00	3E-02	-4E-02	-9E-02	-1E-06	-1E-05	-4E-03	-1E+03	-6E+00
Graphite	-2E-09	-2E-02	-2E-03	-1E-04	-4E-05	-2E-09	-4E-09	-2E-05	-2E-01	-2E-02
Cu foil	-2E-07	-1E+00	-2E-01	-5E-02	-4E-02	-6E-07	-1E-05	-5E-03	-1E+03	-1E+00
Plastic (Shell&Separator)	-4E-08	-7E-01	-3E-02	-3E-03	-2E-03	-4E-08	-2E-07	-5E-04	-3E+01	-1E+00
HF	0E+00	0E+00	0E+00	8E-03	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00
Al foil	-7E-08	-1E+00	-6E-02	-6E-03	-3E-03	-3E-07	-3E-07	-1E-03	-2E+01	-6E-01
Electricity I	6E-08	4E+00	3E-01	2E-02	7E-03	2E-07	7E-07	4E-03	6E+01	8E-01
Electricity II	8E-09	6E-01	4E-02	3E-03	9E-04	3E-08	9E-08	5E-04	7E+00	1E-01
Hydrochloric acid	2E-07	5E-01	4E-02	3E-03	2E-03	6E-08	3E-07	7E-04	2E+01	5E-01
Water	1E-09	3E-03	2E-04	2E-05	1E-05	5E-10	2E-09	3E-06	1E-01	3E-03
Soda ash	1E-08	2E-01	1E-02	8E-04	1E-03	2E-08	6E-08	2E-04	4E+00	2E-01
Wastewater	4E-10	5E-03	5E-04	5E-05	3E-04	3E-09	3E-08	9E-06	2E-01	5E-03
Sodium chloride emission to water	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00
Electricity III	4E-08	3E+00	2E-01	2E-02	5E-03	2E-07	5E-07	3E-03	4E+01	6E-01
Glucose	6E-09	7E-02	4E-03	6E-04	4E-04	8E-09	-5E-08	7E-05	2E+00	9E-02
CO_2 emission	0E+00	1E-01	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00
Lithium iron phosphate	-5E-07	-4E+00	-3E-01	-3E-02	-7E-02	-8E-07	-2E-06	-5E-03	-1E+02	-5E+00

Table S7 Economic analysis data for recycling 1.0 kg spent $LiFePO_4$ battery

	Item	Pyro & Hydro				HOR		
		Unit	Amount	Unit price	Cost (USD)	Amount	Unit price	Cost (USD)
Raw materials	Power consumption							
	Electricity I	kW·h	4.00 kW·h	1.12 USD/kW·h	2.46 USD	0.00 kW·h	1.12 USD/kW·h	0.00 USD
	Electricity II	kW·h	0.54 kW·h	1.12 USD/kW·h	0.61 USD	0.88 kW·h	1.12 USD/kW·h	1.26 USD
	Electricity III	kW·h	3.00 kW·h	1.12 USD/kW·h	3.36 USD	3.00 kW·h	1.12 USD/kW·h	3.36 USD
	Raw material input							
	Spent Lithium Iron Phosphate Batteries	kg	1.00 kg	793.65 USD/t	0.79 USD	1.00 kg	793.65 USD/t	0.79 USD
	LFP cathode materials	kg	0.44 kg	—	—	0.44 kg	—	—
	Hydrochloric acid	kg	0.57 kg	53.97 USD/t	0.031 USD	—	—	—
	Hydrogen peroxide I	kg	—	—	—	0.017 kg	123.81 USD/t	0.002 USD
	Hydrogen peroxide II	kg	—	—	—	1.65 kg	123.81 USD/t	0.20 USD
	Sodium carbonate	kg	0.16 kg	238.09 USD/t	0.038 USD	—	—	—
	CO_2	kg	—	—	—	0.054 kg	—	—
	Glucose	kg	0.053 kg	444.44 USD/t	0.024 USD	0.053 kg	444.44 USD/t	0.024 USD
	Water	kg	8.80 kg	0.54 USD/t	0.0048 USD	—	—	—
	Primary product output							
	Al foil	kg	0.065 kg	4444.42 USD/t	0.29 USD	0.065 kg	4444.42 USD/t	0.29 USD
	Copper foil	kg	0.11 kg	8730.23 USD/t	0.96 USD	0.11 kg	8730.23 USD/t	0.96 USD
	Plastic (Shell + Separator)	kg	0.14 kg	1587.31 USD/t	0.22 USD	0.14 kg	1587.31 USD/t	0.22 USD
	Graphite	kg	0.24 kg	—	—	0.24 kg	—	—
Direct air emissions	Secondary product output							
	LFP cathode materials	kg	0.44 kg	63.0 USD/kg	27.72 USD	0.44 kg	63.0 USD/kg	27.72 USD
	NaCl	kg	0.18 kg	—	—	—	—	—
	Hydrogen fluoride	kg	0.0048 kg	—	—	0.0048 kg	—	—
	CO_2	kg	0.13 kg	—	—	0.13 kg	—	—
	Wastewater	kg	9.37 kg	—	—	1.65 kg	—	—
SUM					21.87 USD			23.55 USD

Note: The yellow color block represents the input benefit.

The blue color block represents the output benefit.

The green color block represents the surplus profit.

Text S1 Electrochemical test steps

The S-LFP, R-LFP, and commercial LFP (named C-LFP) were mixed with carbon blacks (Denka Black Li-400) in a mortar by manual grinding for 30 min. Polyvinylidene fluoride (PVDF) dissolved in N-methylpyrrolidone was then added to the mixture for another 30 mins of stirring. The mass ratio of LFP, carbon blacks, and PVDF was controlled at 8:1:1. The obtained slurry was coated on Al foil with a mass loading of approximately 5 mg/cm^2. The coated Al foil was subsequently dried under a vacuum at 80 °C for 12 h. The dried plate was then cut into small round pieces with a diameter of 12 mm to assemble CR2032 coin cells. Metallic Li chips were selected as the anode, and separators were purchased from Celgard. Lithium hexafluorophosphate ($LiPF_6$) dissolved in ethylene carbonate/dimethyl carbonate with a volume ratio of 50/50 was utilized as the electrolyte. The sealed coin cells were tested via a Lanhe battery testing system at room temperature. The rate capabilities were conducted at 0.1, 0.2, 0.5, and 1.0 C, and the charging cutoff voltage was set to 4.0 V (versus Li/Li^+). The cycling performances at charging cutoff voltages of 4.0 V (versus Li/Li^+) were all tested at a current of 0.5 C. [1]

[1]*Wang, Junxiong, et al. "Direct conversion of degraded $LiCoO_2$ cathode materials into high-performance $LiCoO_2$: A closed-loop green recycling strategy for spent lithium-ion batteries." Energy Storage Materials 45 (2022): 768-776.*

Text S2 Calculation processes of LCA results

The calculation of LCA used the recycling of 1.0 kg of spent $LiFePO_4$ batteries as the functional unit. The dismantling data of the spent $LiFePO_4$ batteries are shown in Table S1 and S2. In this study, the industrialized recycling base of spent $LiFePO_4$ batteries is in Shenzhen, Guangdong Province, near Hong Kong, China. Therefore, the prices of water and electricity in this study were calculated according to Shenzhen City, Guangdong Province. Currently, the price of industrial electricity in Shenzhen is about 0.12 USD/kW·h. The price of industrial water is about 0.54 USD/t.

I Hydro-oxygen Repair Process

(a) power consumption

I Mechanochemical Extraction

The planetary ball-milling device used was rated at 0.75 kW·h. When the rotary speed was 1000 rpm, the power of the planetary ball-milling device was 0.75 kW·h. The planetary ball-milling device was equipped with four 1.0 L agate ball mill tanks. The liquid loading capacity of each tank is designed to be 0.60 L. Therefore, 440.0 g of $LiFePO_4$ cathode material powder obtained by dismantling 1.0 kg of spent $LiFePO_4$ battery required 7 runs of the ball-milling device. The run time of the mechanochemical reaction was set to 10.0 min. The total reaction time was 100.0 min (1.67 h). The total energy consumption was 0.88 kW·h.

II Solid Phase Synthesis

Atmosphere furnace used in the sintering process of lithium iron phosphate battery was rated at 3.0 kW·h. When the operating temperature was 700 °C, the operating power of the tube furnace was 1.5 kW·h. In this solid synthesis, the running time was 10.0 h. The amount of solid sample that can be sintered in the atmosphere furnace was about 2.5 kg each time. Therefore, the energy consumption was 3.0 kW·h.

(b) Reagent consumption

The consumption of H_2O_2 was calculated according to the optimal conditions of the HOR experiment. Carbon dioxide acts as a chemical reagent for precipitating LiOH. In actual production, industrial waste gas will be used as the carbon source of CO_2. The precipitation equation of Li_2CO_3 was as follows:

$$2LiOH + CO_2(g) \rightarrow Li_2CO_3\downarrow + H_2O \quad \text{(Eq.S1)}$$

Δ Prices of recycled products such as aluminum foil, copper foil, plastic, $FePO_4$, and Li_2CO_3 were calculated from the current domestic market price in China.

Δ After conducting F tracing analysis, it has been concluded that the organic binder PVDF will enter the recrystallization stage of LFP. For further information regarding the LCA boundaries, please consult Figures S1 and S2.

II Pyrometallurgy + Hydrometallurgy

(a) power consumption

I High Temperature Separation:

The muffle furnace used in this study was rated at 3.00 kW·h. When the operating temperature was 500 °C, the operating power of the muffle furnace was 1.50 kW·h. The heating rate of the muffle furnace was 5 °C/min, and the operation time and holding time were 100.0 min and 60.0 min, respectively. 510.0 g of $LiFePO_4$ cathode electrode obtained by dismantling 1.0 kg of spent $LiFePO_4$ battery required 1 runs of the muffle furnace. The total reaction time was 160.0 min (2.67 h). The total energy consumption was 4.0 kW·h.

II Acid Leaching

The power of the magnetic stirring device we adopted was 0.25 kW·h. When the stirring speed was 200 rpm, the operating power of the magnetic stirring device was 0.06 kW·h. For a magnetic stirring device, the volume of the reaction solution per treatment was 1.0 L. 440.0 g of $LiFePO_4$ cathode material powder obtained by dismantling 1.0 kg of spent $LiFePO_4$ battery required 9 runs of the magnetic stirring device. The run time of the leaching reaction was set to 60.0 min. The total reaction time was 60.0 min (1.0 h). The total energy consumption was 0.54 kW·h.

III Solid Phase Synthesis

Atmosphere furnace used in the sintering of LFP battery was rated at 3.0 kW·h. When the operating temperature was 700 °C, the operating power of the atmosphere furnace was 1.5 kW·h. In this solid synthesis, the running time was 10.0 h. The amount of solid sample that can be sintered in the atmosphere furnace was about 2.5 kg each time. Therefore, the energy consumption was 3.0 kW·h.

(b) Reagent consumption

The liquid-solid ratio in the acid leaching process was calculated as 20:1.

The usage amount of HCl was calculated according to the following equation:

$$LiFePO_4 + 2H^+ + 2Cl^- \rightarrow [LiCl_2]^- + Fe^{2+} + [H_2PO_4]^- \quad \text{(Eq.S2)}$$

In general, the molar ratio of HCl and $LiFePO_4$ was 2:1. Therefore, in this study, the consumption of HCl was 0.57 kg. The consumption of the precipitant Na_2CO_3 was calculated as 1.2 times the molar ratio of Li. It can be known by calculation that the amount of Na_2CO_3 used was 0.16 kg, and the NaCl produced was 0.18 kg.

For the price of R-LFP, we refer to Saibo Electrochemical Materials (https://saibo-elect-material.taobao.com/?spm=2013.1.0.0.786b51119pQ1hM).