Cost-effective direct regeneration of spent NCM cathode via solvothermal regeneration process

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Figure. S1. Step-by-step pre-treatment process for recovering spent NCM cathode active materials from end-of-life battery cells.

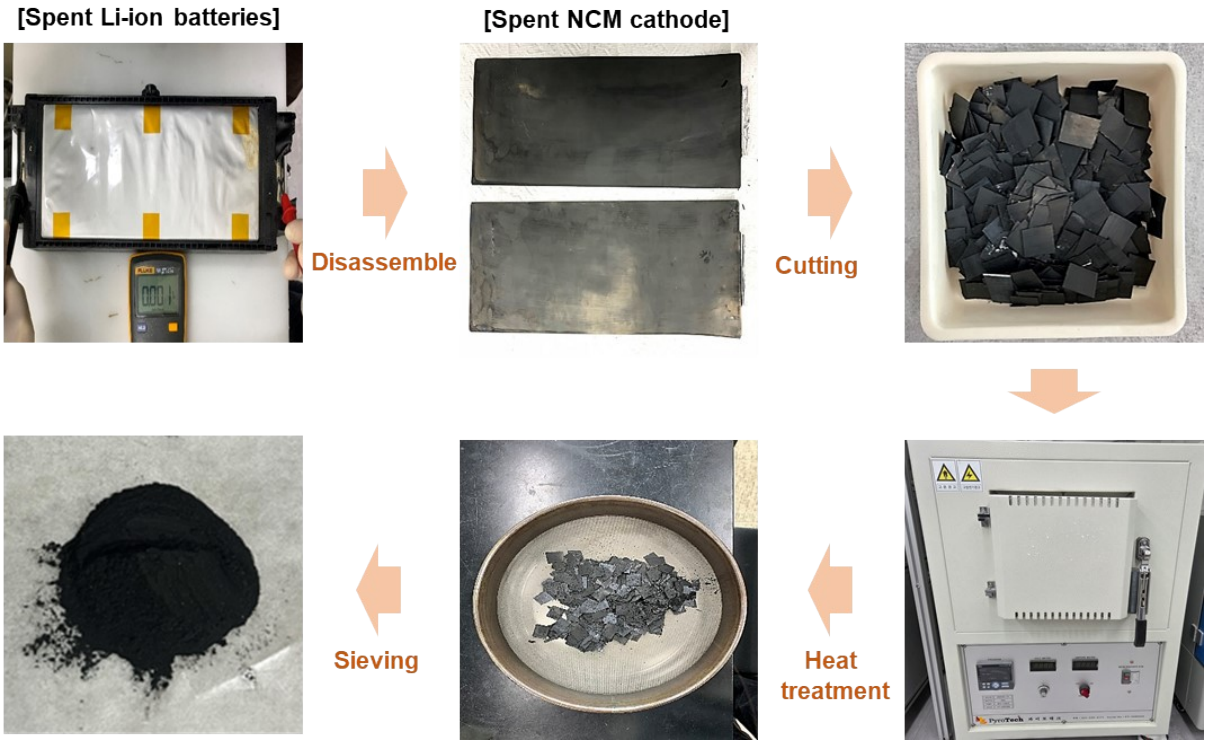

Figure. S2. Rate performance of the S-NCM, ST-NCM and commercial NCM

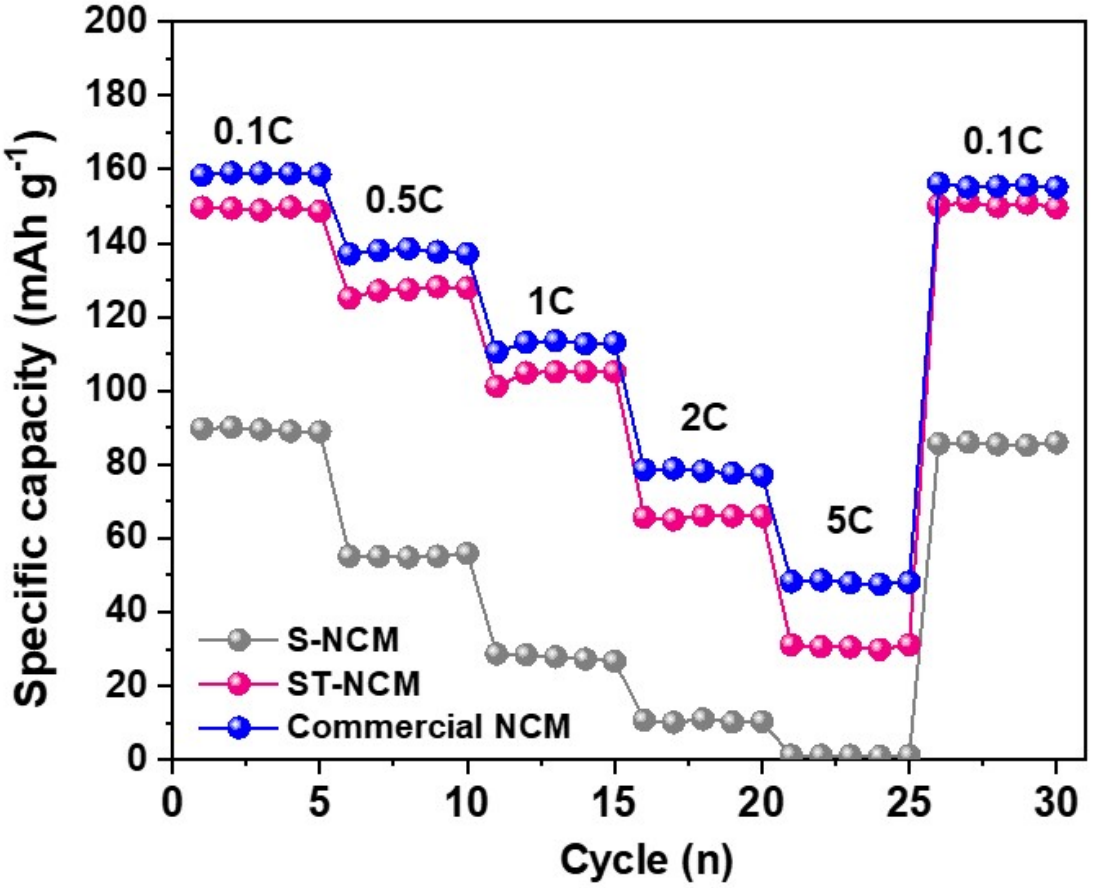

Table S1. Comparison of lithium leaching from cathode materials in water and ethanol

Solvent	Stirring time	Li (mol)	Ni (mol)	Mn (mol)	Co (mol)
Water (deionized)	10 min	0.88	0.65	0.20	0.15
	1h	0.88	0.65	0.20	0.15
	8h	0.87	0.65	0.20	0.15
	24h	0.87	0.65	0.20	0.15
Ethanol	10 min	0.98	0.65	0.20	0.15
	1h	0.98	0.65	0.20	0.15
	8h	0.98	0.65	0.20	0.15
	24h	0.97	0.65	0.20	0.15

Economic and environmental assessment

The economic and environmental impacts of the four different recycling strategies—pyrometallurgical, hydrometallurgical, hydrothermal, and solvothermal processes—were evaluated based on the following process flow assumptions. For a consistent and fair comparison, all four recycling plants were simulated with an annual processing capacity of 10,000 metric tons of spent lithium-ion battery cells.

Pyrometallurgical recycling process

In the pyrometallurgical route, spent batteries are directly introduced into a high-temperature smelter. During this stage, the organic components, including plastics and electrolytes, are combusted to provide thermal energy, while the graphite and aluminum foils act as reducing agents. Consequently, transition metals such as cobalt, nickel, copper, and iron are recovered in the form of a mixed metal matte. Meanwhile, lithium and oxidized aluminum are largely lost to the slag. To be reused for new cathode production, the recovered Co/Ni/Cu/Fe matte must undergo subsequent hydrometallurgical refining steps, including acid leaching and solvent extraction.

Hydrometallurgical recycling process

For the hydrometallurgical process, the spent batteries are first discharged, dismantled, and mechanically shredded. The shredded scrap undergoes a low-temperature calcination to eliminate the polymeric binders and residual electrolytes. After a series of physical separation steps to remove metallic scraps (aluminum, copper, steel) and plastics, the remaining black mass is thoroughly dissolved using strong acids. This leaching step is followed by sequential solvent extraction and precipitation processes to isolate individual Co, Ni, and Mn compounds, along with lithium carbonate, which serve as raw precursors for synthesizing fresh cathode materials.

Hydrothermal recycling process

In the hydrothermal direct recycling process, end-of-life batteries are similarly discharged, dismantled, and physically separated to isolate the degraded cathode powder from scrap metals, plastics, and anode materials. The harvested spent cathode is then subjected to a chemical relithiation step followed by thermal annealing to rejuvenate the material. For this assessment,

the hydrothermal relithiation was assumed to be conducted at a high temperature of 220 °C using an aqueous solution of lithium hydroxide (LiOH) as the lithium source.

Solvothermal recycling process

The proposed solvothermal recycling process shares the identical pre-treatment and physical separation procedures with the hydrothermal process. Instead of an aqueous system, the solvothermal process utilizes ethanol as the solvent and lithium acetate (LiOAc) as the lithium source

Materials input

The materials requirements for the generic pyrometallurgical and hydrometallurgical processes are obtained from EverBatt. The materials requirements for the hydrothermal and solvothermal recycling process are obtained based on our lab process. Life cycle analysis accounts for the environmental impacts of all the materials consumed in the process by capturing the environmental impacts associated with their upstream production.

Energy input

To calculate the life-cycle environmental impacts attributable to all types of energy consumed in the process, life cycle analysis considers the environmental impacts associated with upstream fuel production and electricity generation, as well as those associated with on-site fuel combustion (e.g., diesel/natural gas combustion).

Process emissions

In the life cycle analysis, we also accounted for environmental impacts associated with process emissions that are not due to fuel combustion. For the four recycling processes, process emissions include those from material combustion and thermal decomposition. The former arises from burning off materials during the recycling processes, including graphite, carbon black, electrolyte, plastics, and the binder material in the pyrometallurgical process, and electrolyte and the binder material in the hydrometallurgical and direct recycling processes. As the pyrometallurgical process involves burning various battery constituents, it leads to much higher process emissions than those from the other two recycling methods.

Total energy use

Total energy use is the cumulative energy use pertaining to the process, including fossil energy use and renewable energy use. Fossil energy use can be further broken down to that of coal, natural gas, and petroleum. GHG emissions are calculated based on 100-year global warming potentials from the fifth assessment report of the Intergovernmental Panel on Climate Change.

Table S2. Material requirements (kg) for recycling 1kg of waste batteries through various recycling technologies. (NR = Not required).

Unit : kg

	Pyrometallurgical	Hydrometallurgical	Hydrothermal	This work
Ammonium Hydroxide	NR	0.031	NR	NR
Urea	NR	NR	NR	4.00
Ethanol	NR	NR	NR	5.00
Hydrochloric Acid	0.21	0.012	NR	NR
Hydrochloric Peroxide	0.06	0.366	NR	NR
Sodium Hydroxide	NR	0.561	NR	NR
Limestone	0.30	NR	NR	NR
Sand	0.15	NR	NR	NR
Sulfuric Acid	NR	1.08	NR	NR
Soda Ash	NR	0.02	NR	NR
Lithium Hydroxide	NR	NR	0.05	NR
Lithium Acetate	NR	NR	NR	0.014
Lithium Carbonate	NR	NR	0.018	0.018
Water consumption (gal)	NR	1.00	5.00	NR

Table S3. Unit prices of recovered materials ($/kg)

Unit : $/kg

Materials	Unit Prices
Aluminum	1.30
Copper	6.60
Steel	0.30
Plastics	0.10
NMC(Recycled)	19.00
Lithium carbonate	7.90
$Ni2^{+}$ in output	11.00
$Co2^{+}$ in output	55.00
$Mn2^{+}$ in output	2.00
Electrolyte solvents	0.15
Graphite	0.28

Table S4. Comparison of energy consumption by process stage for different recycling methods.

Unit : MJ/kg

	Pyrometallurgical	**Hydrometallurgical**	**Hydrothermal**	**Solvothermal**
Pretreatment	2.0	2.0	2.1	2.1
Main process	25.0	4.0	2.5	1.5
Chemical intrinsic energy	3.5	12.0	1.7	0.2
Post-processing	5.0	10.1	5.5	6.3
Total	35.5	28.1	11.8	10.1

Table S5. Comparison of greenhouse gas emissions by source for different recycling methods.

Unit : kg/kg

	Pyrometallurgical	**Hydrometallurgical**	**Hydrothermal**	**Solvothermal**
Direct emissions	1.80	0.10	0.15	0.15
Energy use	1.60	0.90	0.75	0.65
Chemical substance	0.40	1.90	0.40	0.20
Total	3.80	2.90	1.30	1.00

Table S6. Detailed cost breakdown for different recycling methods.

Unit : $/kg

	Pyrometallurgical	**Hydrometallurgical**	**Hydrothermal**	**Solvothermal**
Materials	0.5	5.5	2.8	1.1
Energy	3.5	1.5	0.7	0.6
Labor	4.0	3.0	2.0	2.0
Capital	6.5	2.8	2.0	2.1
Total	14.5	12.8	7.50	5.8

Table S7. Detailed cost breakdown for different recycling methods.

Unit : $/kg

	Pyrometallurgical	**Hydrometallurgical**	**Hydrothermal**	**Solvothermal**
Revenue	16.3	17.0	22.0	22.0
Cost	-14.5	-12.8	-7.5	-5.8
Profit	1.8	4.2	14.5	16.2

Table S8. Quantitative comparative indicators for direct regeneration technologies

Direct regeneration method	Cathode material	Li Source	Procedure (Temp & Time)	Post-annealing (Temp & Time)	Energy consumption (Mj/kg)	Discharge capacity, retention	Ref.
This work	NCM 622	0.4M LiOAc	130℃ 4h	850℃ 4h	10.1	148.7 mAh g^{-1} (0.1C), 94.08% (100cyc)	This work
Oxalic acid regeneration	NCM 523	CH3COOLi, LiOH (3:2 molar ratio)	400℃ 4h	850℃ 4h	N/A	146.1 mAh g^{-1} (1.0C), 84.9% (100cyc)	S1
Solid state	NCM 622	Li2CO3	500℃ 5h, 800℃ 9h	-	83.2	149.7 mAh g^{-1} (1.0C), 96.3% (150cyc)	S2
Hydrothermal	NCM 111	4M LiOH	130℃ 4h	850℃ 4h	N/A	163.7 mAh g^{-1}, 85.58% (200cyc)	S3
Molten salts	NCM 523	LiOH-Li2CO3 (0.85:0.15 molar ratio)	450℃ 4h	900℃ 6h	149.3	160.1 mAh g^{-1} (0.1C), 90.33% (100cyc)	S4
Redox mediation	NCM 622	Lithiated DTBQ solution	6h	-	N/A	178.2 mAh g^{-1} (0.1C), 76.7% (100cyc)	S5

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