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Electronic Supplementary Information (ESI)

Upcycling spent medium-Ni cathodes via novel liquified salts sourcing

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Experimental Details

Material synthesis

Double-sided coated cathode sheets, composed of NCM523, carbon black and poly(vinylidene fluoride) (PVDF, as the binder), were obtained from spent batteries (cylindrical cells with a nominal capacity of 3000 mAh) with a capacity retention of approximately 80%. Subsequently, the cathode sheets were thermally treated at 400°C for 20 min to reduce the adhesivity of PVDF binder, facilitating the separation of the cathode powder from the aluminum foil. The separated cathode powder was collected and stored in the argon-filled glove box with an extremely low level of O_2/H_2O (below 0.1ppm).

To transform the deteriorated NCM523 (Li_{1-x}Ni_{0.5}Co_{0.2}Mn_{0.3}O₂) into LS-NCM, 10g of spent cathode powder was mixed with 0.078mol of LiOH·H₂O (99.9%, Sigma Aldrich), 0.116mol of LiNO₃ (99.9%, Sigma Aldrich) at the eutectic composition, and 0.163 mol of NiNO₃·6H₂O (98.5%, Sigma Aldrich) using a planetary centrifugal mixer (ARE-310, THINKY) at 2,000 rpm (around 510 g force) for 12 min. The molar ratio of transition metal to Li was set to 1:1.03. The planetary-centrifugally mixed powders were calcinated at 920°C for 2 h and then at 760°C for 8 h in flowing oxygen. For SS-NCM synthesis, the spent NCM523 cathode powder was mixed with LiOH·H₂O and NiOH (60.0-70.0%, Sigma Aldrich) by using planetarycentrifugal mixer in a 1:1.03 molar ratio of transition metal to Li for 30 min. The mixture was then subjected to calcine at 920 °C for 2 h and then at 760 °C for 8 h in an oxygen atmosphere. The venting line was securely connected outside at the opposite side of the tube furnace to exhaust the gas naturally. Using this venting line, the gas pressure of the furnace can be maintained, and gas products (e.g., toxic NO_x) are effectively removed.

Electrochemical measurements

For electrochemical measurements, the electrodes were fabricated by mixing 90 wt% of cathode active material, 5 wt% of super-P (as the conductive agent), and 5 wt% of poly(vinylidene fluoride) (PVDF, as the binder) in N-methyl-2-pyrrolidone (NMP). The slurry was coated onto aluminum foil and dried at 120°C for 2 h, followed by vacuum-drying for 10 h prior to cell assembly. All cathodes were controlled with a loading level of 12.0 ± 0.5 mg cm⁻². The prepared electrodes were assembled using 2032R coin type cell in Ar-filled glove box, with cathodes (diameter 12 mm), lithium metal foils (diameter 14 mm, thickness 1 mm) as the counter and reference electrode, respectively, and 1.15 M LiPF₆ in ethylene carbonate/ethyl methyl carbonate/diethyl carbonate with 5wt% fluoroethylene carbonate additive (EC:EMC:DEC = 3/6/1 vol% with 5% FEC; Enchem Co., Ltd) as the electrolyte. The cells were evaluated with constant current-constant voltage mode between 2.8 and 4.3/4.4/4.5 V (vs. Li/Li⁺) at 25°C. The first charge-discharge cycle (as the formation step) was conducted at 0.1 C (for SS-NCM and LS-NCM cathode, 1.0 C is defined as 200, 210, and 220 mA g⁻¹ for cut-off voltage of 4.3, 4.4 and 4.5V, respectively). After the first cycle, the cells were charged and discharged at 0.5 C/1.0 C for 100 cycles. After specific cycles, EIS measurements were conducted on cells charged to 4.3 V (vs. Li/Li⁺) from 1 mHz to 10 MHz and with AC voltage amplitude of 10 mV using Reference 3000 (Gamry Instrument). GITT measurements were carried out following the 5th and 100th

cycles of 0.5 C/1.0 C cycling between 2.8 and 4.3 V (versus Li/Li⁺) with a titration step at 0.3 C of 8 min and a relaxation step of 1 h. To evaluate the rate capability, the cells were charged at 0.5 C and discharged at rates of 0.5, 1.0, 2.0, 3.0, and 5.0 C. For full-cell tests, the upcycled cathode and graphite anode were utilized to assemble 700 mAh-scale pouch-type full-cells. The ratio of negative to positive electrode capacity (N/P ratio) was fixed at 1.07 ± 0.01 . The cathode loading level was 12.04 mg cm^{-2} on each side of the double-side coated Al foil. The anode loading level was 6.88 mg cm^{-2} on the double-side coated Cu foil. The graphite electrode density was 1.52 g cm^{-3} and the cathode density was 3.31 g cm^{-3} . The pouch-type full-cells were assembled in an argon-filled glovebox with a humidity of less than 1%. The separator and liquid electrolytes were the same as those used in coin cells. The weight of the electrolyte used in full-cells was 1.75 g, which corresponded to 2.5 g Ah^{-1} . The cycling voltage window was set at 2.8-4.30 V, and one formation cycle with three pre-cycles was executed at 0.1 C and 0.2 C, respectively, before embarking long-term cycling of 300 cycles at 1 C ($1.0 \text{ C} \approx 2.21 \text{ mA cm}^{-2}$ for full-cell). All electrochemical assessments (except for EIS) were performed using a CT2001A battery cycler (Landt Instrument).

Characterizations

The chemical compositions of prepared cathode powders were determined by an inductively coupled plasma optical emission spectrometer (ICP-OES, AVIO 550, Perkin Elmer). Phases were characterized by XRD using a parallel beam XRD instrument (X'Pert Pro MPD, Malvern Panalytical, Cu K α with a wavelength of 1.542 Å) at the Korea Basic Science Institute (KBSI) at Seoul Western Center. High-temperature XRD measurements were conducted using an Anton Paar HTK 1200N stage integrated with the diffractometer, enabling the analysis of structural changes under elevated temperatures. The crystallographic analysis was conducted by using PDXL analysis software (Rigaku). Phase identification was performed using PDXL software package, including crystallography open database (COD). The prepared materials were characterized by scanning electron microscopy (SEM, Merlin, Zeiss) equipped with energy dispersive X-ray spectroscopy (EDS, XFlash® 6130, Bruker) detector. For TEM analysis, samples were prepared by a dualbeam focused ion beam (FIB, Helios 450HP, FEI). High-resolution TEM (HR-TEM, ARM300, JEOL) was conducted under 150 and 300 keV to collect scanning transmission electron microscopy (SEM) images for atomic and structural analysis. Electron energy loss spectroscopy (EELS) and energy dispersive X-ray spectroscopy (EDX) were conducted by HR-TEM (Aztec, Oxford).

Note S1. Life cycle analysis

1. Goal and scope

The closed-loop life cycle analysis (LCA) evaluates and compares the economic and environmental impacts of recycling waste cathode materials from spent lithium-ion batteries and synthesizing new cathode materials.^{1, 2} The analysis examines critical metrics such as greenhouse gas (GHG) emissions, energy consumption, and water usage across several recycling approaches: hydrometallurgical, pyrometallurgical, and liquified-nitrates-assisted upcycling, as detailed in this study. To provide a baseline for comparison, the production of cathode materials from virgin materials is also analyzed. The scope excludes costs related to material transportation and GHG disposal. However, the analysis accounts for raw material expenses, manufacturing costs, and fixed annual capital investments. Energy costs are based on \$0.04 per kWh (1 Wh \approx 3600 J), while water costs are set at \$0.0189 per liter with wastewater discharge fees of \$0.0265 per liter. General expense (~30% of process cost) and overhead costs (~15% of manufacturing cost including resources) are also considered.

2. Scenario description, system boundaries, and inventory analysis.

Four scenarios were analyzed: (1) pyrometallurgical upcycling, (2) hydrometallurgical upcycling, (3) liquified-nitrates-assisted upcycling, and (4) virgin cathode material (Ni-rich cathode) production. Each scenario assumes the treatment of 1 kg of spent lithium-ion batteries, with material and energy flows standardized in a life cycle inventory (**Tables S7-S9**). The LCA adopts a cradle-to-gate perspective, assessment excludes the usage and disposal stages of the cathode materials, under the assumption that recycled and virgin materials exhibit similar or improved performance during use and recycling.

(Scenario 1 : Pyrometallurgical upcycling method) The process discussed here was modified based on the previous work.³ This method involves direct smelting of spent batteries after a discharge pretreatment. Smelting process operates at approximately 1873 K for 3 hours, decomposing organic impurities and reducing transition metal oxides to metallic forms. During smelting process, gas treatment systems mitigate toxic emissions. Lithium salts combine with aluminum and calcium to form slag as a byproduct (e.g. Li₂CO₃, CaAl). Lithium can be later recovered either via slag treatment or evaporating lithium during smelting.⁴ Energy consumption for smelting is based on a 36 kW pilot-scale furnace processing 174 kg of material. Subsequent acid leaching (0.86 mol L⁻¹ H₂SO₄ solution) dissolves the matte, and virgin metal sulfates are added to tailor the Ni, Mn, and Co ratios to target composition. The adjusted metal sulfate solution is utilized to synthesize Transition metal(TM)-based cathode precursor through co-precipitation process. The coprecipitation process forms transition metal hydroxides at pH 10–11 in a nitrogen atmosphere. TM-based precursors are mixed with lithium hydroxide (Li:TM = 1:1.03) and sintered at 1173 K for 2 hours and 1053 K for 10 hours in a 45 kW furnace handling 245 kg. GHG emissions and energy consumption for each step were modeled using EverBatt 2020 and GREET.

(Scenario 2 : Hydrometallurgical upcycling method) In this scenario, 1 kg of spent lithium-ion batteries undergoes a sequence of pretreatment, separation, and chemical processing to recover cathode

materials. The process begins with the discharge of spent batteries to eliminate residual charge and ensure safety during subsequent handling. Following discharge, the batteries are subjected to crushing and shredding operations to break them into smaller fragments. These steps produce a mixture of materials that includes cathode and anode powders, current collectors, binders, and electrolyte residues. The shredded fragments are calcined at 873 K for 2 hours (6 kW and a mass loading of 227 kg in pilot-scale process) to decompose organic binders, evaporate residual electrolytes, and remove impurities like solid electrolyte interface (SEI) components.³ This step also volatilizes moisture and reduces contamination in downstream processes. After calcination, mechanical separation techniques, including wet granulation, density separation, and froth flotation, isolate cathode powders. Wet granulation facilitates particle disaggregation, while density separation removes heavy components like copper and aluminum. Froth flotation uses surfactants to selectively recover cathode particles. These steps help reduce water consumption and minimize the total amount of acid required for the subsequent leaching step, thus increasing the purity of TM-based precursors. The acid amount used in the leaching process was determined by the solid mass and pulp density ($\sim 2\%$). The concentration of the acid was set as was set as 2.8 mol L^{-1} (H₂SO₄ solution) to improve leaching efficiency. The remaining processes for treating the leachate and re-synthesizing cathode materials were assumed to be similar to those in the pyrometallurgy method described in Scenario 1.

(Scenario 3: liquified-nitrates assisted upcycling method) In this scenario, the spent batteries were disassembled to collect the spent cathode electrodes after discharge pretreatment. Some work also used 1methyl-2- pyrrolidinone to soak the cathode side for ~6 hours to remove the poly(vinylidene fluoride) binder. ^{3, 5} Here, a one-step low temperature calcination at 673 K for 20 min was applied to remove the polymer binder, electrolytes, and the formed impurities during electrochemical cycling, which was mainly the organic SEI components on the cathode particles. The energy consumption for low temperature calcination was estimated based on the pilot-scale recycling process using the commercial furnace, whose temperature, power, and mass loading were ~873 K, 6 kW and 227 kg, respectively. After the pretreatment, the spent cathode powder was scrapped and dismantled from Al current collectors. And the spent cathode powder was mixed with LiOH, LiNO₃ and Ni(NO₃)₂ to achieve tailored to the desired ratio of TM (Ni, Co, Mn)with the molar ratio of Li:TM to 1.03. The mixture completely forms homogeneously liquified after 15min of planetary centrifugal mixing. The large-scale planetary centrifugal mixer is set with a power rating of 6 kW and a capacity of 300 kg. Finally, the mixture is sintered at 1173 K for 2 hr and subsequently at 1053K for 8hr to re-lithiate the cathode powder and reconstruct the microstructure. The energy consumption for high temperature calcination was estimated based on the commercial furnace, whose temperature, power, and mass loading were ~1173 K, 40 kW and 245 kg, respectively.³ The cathode powder with high-Ni concentration was regarded as the final product from liquified-nitrate assisted method.

(Scenario 4 : Virgin cathode production) This scenario assesses the production of 0.35 kg of virgin cathode materials derived from mined ores. According to the EverBatt 2020 software, producing virgin cathode materials consumes 37.45 L of water and 127.97 MJ of energy while emitting 8.92 kg of GHG. The estimated production cost is \$20.60 per kg of cathode material.



Fig. S1. Process flow for the collection of spent $\text{Li}_x \text{Ni}_{0.5} \text{Co}_{0.2} \text{Mn}_{0.3} \text{O}_2$ (NCM523, $x \approx 0.7$) cathode powder. The spent commercial cylindrical cell was disassembled, and the spent NCM523 electrodes were retrieved. The electrodes underwent a pre-heat treatment at 400 °C for 20 minutes. Following pre-heat treatment, the NCM523 cathode powder was separated from the aluminum foil using a silicon carbide blade. The separated powder was subsequently pulverized into a fine powder using a mortar, and stored after sieving for further analysis and testing.



Fig. S2. (a-c) Top-view scanning electron microscopy (SEM) images of spent NCM523 electrode. (d-f) Spent NCM523 cathode powder collected from the electrodes after pre-heat treatment at 400°C for several minutes.



Fig. S3. Deagglomeration of NCM523 secondary particle cathode after planetary centrifugal mixing of 12min. (a-f) SEM images of spent NCM523 particles and Li-/Ni-based liquified molten-salts after planetary centrifugal mixing, and (g, h) corresponding energy dispersive spectroscopy (EDS) mapping results of NCM523 powder with Li-/Ni-based liquified molten-salts



Fig. S4. Phase diagram of LiOH–LiNO₃ system (adapted from FactSage thermochemical software and databases)⁶ with melting temperature of Ni(NO₃)₂·6H₂O. The frictional forces between the mixed particles helps to reach an 'effective' temperature higher than the melting points of LiOH–LiNO₃ eutectic ($T_m = 183 \text{ °C}$) and Ni(NO₃)₂·6H₂O ($T_m = 56.7 \text{ °C}$) during planetary centrifugal mixer.



Fig. S5. (a-d) SEM images of SS-NCM and LS-NCM cathode powders. SS-NCM was synthesized from spent NCM523 powder using LiOH and NiOH as precursors. For the synthesis of LS-NCM, a eutectic mixture of LiOH–LiNO₃ and extra Ni(NO₃)₂·6H₂O was used to replenish deficient Li and increase the Ni concentration from ~0.5 to ~0.8.



Fig. S6. (a,b) Particle size distribution and surface area of each LS-NCM and SS-NCM



Fig. S7. (a-d) Transmission electron microscope (TEM) and scanning transmission electron microscope (STEM) images of cross-sectioned spent NCM523 cathode particles collected from cylindrical cell.



Fig. S8. Schematic STEM-Electron energy loss spectroscopy (EELS) scanning pathway (0 to 40 nm from outer surface) of (a) SS-NCM and (b) LS-NCM, corresponding to EELS profile data in Fig. 3(g) and (h), respectively.



Fig. S9. (a) EDS mapping results of SS-NCM, and (b, c) EDS-line scanning profile and the corresponding region in SS-NCM, showing the inhomogeneity in stoichiometric distribution on the surface of SS-NCM.



Fig. S10. (a) EDS mapping results and spectra of LS-NCM, and (b, c) EDS-line scanning profile and the corresponding region in LS-NCM, showing the stoichiometric coherence with target composition.



Fig. S11. (a) High-temperature X-ray diffraction (HT-XRD) patterns of the mixture of spent NCM523, LiOH, and Ni(OH)₂ for SS-NCM synthesis. (b) HT-XRD patterns of the mixture of spent NCM523 with a eutectic mixture of LiOH–LiNO₃ and extra Ni(NO₃)₂ for LS-NCM synthesis.



Fig. S12. Rietveld refinement of XRD patterns for (a) SS-NCM and (b) LS-NCM. Fitting details available in Table S3.



Fig. S13. (a) Charge-discharge voltage profiles of different NCM cathodes, including spent NCM523, SS-NCM, LS-NCM, single-crystalline NCM811 (MTI), and poly-crystalline NCM811 (POSCO Future M), and (b) their corresponding initial coulombic efficiency (C.E.) during the first cycle with 0.1 C charge – 0.1 C discharge within the voltage range of 2.8 - 4.3 V (vs. Li/Li⁺). Voltage profiles of (c) LS-NCM and (d)SS-NCM during 0.5C charge – 1.0 C discharge cycling test within the voltage range of 2.8 - 4.3 V (vs. Li/Li⁺). The voltage range of 2.8 - 4.3 V (vs. Li/Li⁺) for 100 cycles at 25° C (1C = 200mA g⁻¹)



Fig. S14. Charge-discharge curves of (a) LS-NCM and (b)SS-NCM during rate capability test within the voltage range of 2.8 to 4.3 V (vs. Li/Li+) at 25°C. The discharge C-rate increased from 0.5 to 5.0 C with a fixed charge C-rate of 0.5 C (1 C = 200mA g⁻¹).



Fig. S15. Galvanostatic intermittent titration technique (GITT) measurements on SS-NCM and LS-NCM after certain cycles during 0.5C/1 C cycling in **Fig. 5(d)**. The voltage profiles after 5th cycle and 100th cycle for (a) SS-NCM and (b) LS-NCM. The ohmic and non-ohmic voltage losses were separately plotted as a function of depth of discharge in (c) SS-NCM and (d) LS-NCM.



Fig. S16. Electrochemical impedance spectroscopy (EIS) measurements on (a) LS-NCM and (b) SS-NCM after 1, 50 and 100 cycles of 0.5C/1C cycling between 2.8 V and 4.3 V (vs. Li/Li⁺) at 25 °C. Insets: Magnified EIS data and equivalent circuit. Fitted results are listed in (c). R_{CT} contributes most to the impedance of cycled cathodes.



Fig. S17. Voltage profiles of LS-NCM during 0.5C/1.0C cycling test within the voltage range of 2.8 to 4.4 V (vs. Li/Li^+) for (a) and 2.8 to 4.5 V (vs. Li/Li^+) for (b) at 25°C.



Fig. S18. Electrochemical performance of SS-NCM/spherical graphite (SS-NCM/Gr) and LS-NCM/Gr fullcells tested in the voltage range of 2.8-4.3V at 25° C. (a) Initial full-cell formation step performed at 2.8-4.3V with a charge and discharge C-rate of 0.1 C. (b) Initial charge-discharge voltage profile of the Gr anode at 25° C (CC-CV mode, charge and discharge C-rate: 0.1 and 0.1C). (c) Cycle performance of the Gr anode at the voltage range of 0.005-1.5 V (vs. Li/Li+) with charge and discharge C-rate of 0.5 and 0.5C, respectively. Voltage profiles of (d) SS-NCM/Gr and (e) LS-NCM/Gr full-cells during 300 cycles (galvanostatic chargedischarge cycling was performed with C-rate of 1.0 C ≈ 2.21 mA cm⁻²).



Fig. S19. (a-d) Comparative cycling performance and average voltage retention for three cells of LS-NCM/Gr (left panels, blue) and SS-NCM/Gr (right panels, red) tested up to 300 cycles at 1.0 C in the range of 2.8-4.3 V at 25 °C (galvanostatic charge-discharge cycling was performed with C-rate of $1.0 \text{ C} \approx 2.21 \text{ mA} \text{ cm}^{-2}$).



Fig. S20. (a-e) Process cost, energy consumption, water consumption, GHG emission, and concentrated H₂SO₄ consumption in treating 1kg of spent batteries (cells). LSU : liquified-salts-assisted upcycling. Pyro : pyrometallurgical method. Hydro : hydrometallurgical method.



Fig. S21. (a) Manufacturing cost and revenue per kg of NCM811 cathode upcycled by LSU, hydro, and pyro processes. The profits from 1kg of upcycled NCM811 are USD 8.32, 5.56, and 5.81 for LSU, hydro, and pyro methods, respectively. (b) Detailed breakdown of manufacturing costs to produce 1kg of upcycled NCM811 cathode from each process. LSU : liquified-salts-assisted upcycling. Pyro : pyrometallurgical method. Hydro : hydrometallurgical method.



Fig. S22. (a) Estimated GHG emissions and total energy consumption of liquified-salts-assisted upcycling process for recycling 1 kg of spent battery cells. The results are compared with those from other direct upcycling/recycling methods in the literatures. (b) Estimated manufacturing costs and profits of liquified-salts-assisted upcycling process for 1 kg of upcycled NCM811 cathode, compared with other direct upcycling/recycling methods from the literatures. (N.P* indicates non-provided value from literature).

	M	Weight pe (mg	ercentage g ⁻¹)			
Sample	Li	Ni	Co	Mn	Al	Cu
Spent NCM523	81.1	49.8	19.2	31.0	/	/
LS-NCM	101.4	80.4	7.99	12.05	/	/
SS-NCM	102.1	80.5	7.68	12.32	/	/

Table S1. Chemical composition of spent NCM523, LS-NCM and SS-NCM measured by Inductively

 Coupled Plasma Optical Emission Spectrometry (ICP-OES)

Samula	BET surface area	Particle size distribution			
Sample	$(m^2 g^{-1})$	D ₁₀ (μm)	D ₅₀ (µm)	D ₉₀ (µm)	
LS-NCM	0.7811	2.6	5.8	10.2	
SS-NCM	1.0776	4.7	8.6	14.3	

Table S2. Particle size distributions of LS-NCM and SS-NCM.

LS-NCM	Element	Site	x	У	Ζ	Occupancy
	Li	3a	0	0	0	0.981(6)
<i>a</i> =2.874184(3) Å	Li	3b	0	0	0.5	<u>0.019(6)</u>
$R_{\rm wp} = 9.75\%$	Co	3b	0	0	0.5	0.082(4)
$R_{\rm p} = 6.57\%$	Ni	3b	0	0	0.5	0.783(3)
ICSD ID : 162291	Mn	3b	0	0	0.5	0.110(6)
	Ni	3a	0	0	0	<u>0.019(6)</u>
	0	6c	0	0	0.259152	1.0(4)
SS-NCM	Element	Site	x	у	Z	Occupancy
SS-NCM	Element Li	Site 3a	<i>x</i> 0	у 0	<i>z</i> 0	Occupancy 0.966(6)
SS-NCM	Element Li Li	Site 3a 3b	x 0 0	y 0 0	z 0 0.5	Occupancy 0.966(6) <u>0.034(6)</u>
SS-NCM a=2.85320(3) Å c=14.3420(6) Å $R_{wp}=9.34\%$	Element Li Li Co	Site 3a 3b 3b	x 0 0 0	y 0 0 0	z 0 0.5 0.5	Occupancy 0.966(6) <u>0.034(6)</u> 0.083(4)
SS-NCM a=2.85320(3) Å c=14.3420(6) Å $R_{wp}=9.34\%$ $R_p=5.96\%$ Brage peak	Element Li Li Co Ni	Site 3a 3b 3b 3b	x 0 0 0 0	y 0 0 0 0	z 0 0.5 0.5 0.5	Occupancy 0.966(6) <u>0.034(6)</u> 0.083(4) 0.771(4)
SS-NCM a=2.85320(3) Å c=14.3420(6) Å $R_{wp}=9.34\%$ $R_p=5.96\%$ Bragg peak ICSD ID : 162291	Element Li Li Co Ni Mn	Site 3a 3b 3b 3b 3b 3b	x 0 0 0 0 0 0	y 0 0 0 0 0 0	z 0 0.5 0.5 0.5 0.5	Occupancy 0.966(6) <u>0.034(6)</u> 0.083(4) 0.771(4) 0.112(4)
SS-NCM a=2.85320(3) Å c=14.3420(6) Å $R_{wp}=9.34\%$ $R_p=5.96\%$ Bragg peak ICSD ID : 162291	Element Li Li Co Ni Mn Ni	Site 3a 3b 3b 3b 3b 3b 3a	x 0 0 0 0 0 0 0 0	y 0 0 0 0 0 0 0 0	z 0 0.5 0.5 0.5 0.5 0	Occupancy 0.966(6) <u>0.034(6)</u> 0.083(4) 0.771(4) 0.112(4) <u>0.034(6)</u>

Table S3. Refined XRD data for LS-NCM and SS-NCM assuming Ni can cation-mixed with Li.

Table S4. Comparison of fresh and upcycled (recycled) Ni-rich cathode materials on the synthesis method,

 particle size, and electrochemical performances. (*AM : Active material)

Active material (Reference number)	Synthesis method	Particle size	Voltage range (vs. Li/Li ⁺)	Discharge capacity @ 1 st cycle (mAh g ⁻¹)	Capacity retention	Electrode loading and *AM ratio in electrode
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 11)	Commercial	2-3µm	3.0-4.3V	180	79.6% after 200 cycles (0.1C)	• 10 mg cm ⁻² • 80% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 12)	CATL (China)	2-3µm	2.8-4.3V	195	90.9% after 50 cycles (0.2C)	• 10 mg cm ⁻² • 80% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 13)	Commercial	2-3µm	3.0-4.3V	192	89.0% after 100 cycles (1.0C)	• 26.4 mg cm ⁻² • 96% CAM
Li1.0Ni0.8Co0.1Mn0.1O2 (Ref. 14)	Co-precipitation High-temperature synthesis	2-3µm	2.7-4.3V	186	85.0% after 100 cycles (0.5C)	• 4-5 mg cm ⁻² • 90% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 15)	Commercial	3-6µm	2.8-4.3V	184	86.5% after 200 cycles (1.0C)	• 3.75mg cm ⁻² • 80% CAM
Li _{1.0} Ni _{0.88} Co _{0.09} Al _{0.03} O ₂ (Ref. 16)	Co-precipitation High-temperature synthesis	3-6µm	3.0-4.3V	185	85.0% after 100 cycles (0.2C)	 12 mg cm⁻² 92% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 17)	Commercial	~3 µm	3.0-4.3V	180	77.4% after 200 cycles (0.5C)	• 7 mg cm ⁻² • 90% CAM
$\begin{array}{c} \text{Li}_{1.0}\text{Ni}_{0.83}\text{Co}_{0.11}\text{Mn}_{0.06}\text{O}_2\\ \text{(Ref. 18)} \end{array}$	Co-precipitation High-temperature synthesis	1-4 μm	2.75-4.4V	191	84.5% after 150 cycles (1.0C)	• 8.5 mg cm ⁻² • 89% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 19)	Co-precipitation High-temperature synthesis	2-3µm	2.5-4.4V	210	92.6% after 100 cycles (0.33C)	• 3 mg cm ⁻² • 80% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 20)	Co-precipitation High-temperature synthesis	2-5 μm	2.8-4.5V	190	58.7% after 400 cycles (1.0C)	• 4 mg cm ⁻² • 80% CAM
LiNi _{0.8} Mn _{0.12} Co _{0.08} O ₂ (Ref. 7)	Direct upcycling	1-2 μm	2.8-4.3V	175	85.0% after 200 cycles (1.0C)	• 2~3mg cm ⁻² • 90% CAM
Li _{1.13} Ni _{0.88} Co _{0.095} Al _{0.025} O ₂ (Ref. 21)	Direct upcycling	1-2 μm	3.0-4.3V	200	93.3% after 100 cycles (1.0C)	• 15 mg cm ⁻² • 90% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 22)	Direct upcycling	~1µm	2.7-4.3V	192	88.7% after 100 cycles (0.2C)	• 3 mg cm ⁻² • 80% CAM
Li _{1.0} Ni _{0.8} Co _{0.1} Mn _{0.1} O ₂ (Ref. 23)	Direct regeneration	12~10 μm	3.0-4.3V	198	85.5 % after 50 cycles (0.5C)	• 4 mg cm ⁻² • 80% CAM
Li1.0Ni0.8Co0.1Mn0.1O2 (Ref. 24)	Direct regeneration	8~10 μm	2.7-4.3V	155	63.5 % after 300 cycles (3.0C)	• Unknown • 90% CAM
			2.8-4.3V	178	94.1% after 100 cycles (1.0C)	• ~12 mg cm ⁻² • 90% CAM
Our work	Liquified-salts- assisted upcycling	3-5µm	2.8-4.4V	193	91.7% after 100 cycles (1.0C)	• ~12 mg cm ⁻² • 90% CAM
			2.8-4.5V	201	90.4% after 100 cycles (1.0C)	• ~12 mg cm ⁻² • 90% CAM

Full-cell specifications						
Electrodes	Cathode	Anode				
Materials	LS-NCM / SS-NCM	Spherical graphite (Gr)				
Cell dimension	72.5 mm*55 mm	75.5 mm*58mm				
Composition	Active material (AM) : CM : Bi = 94 : 3 : 3 (CM: Super P, Bi: polyvinylidene fluoride, PVDF)	AM : CB : CMC : SBR = 97.0 : 0.5 : 1.1 : 1.4 (CB: carbon black, CMC: carboxymethyl cellulose, SBR: styrene butadiene rubber)				
One-side Loading level (mg cm ⁻²)	12.04 ± 0.4	6.88 ± 0.4				
Two-side Loading level (mg cm ⁻²)	24.10 ± 0.5	13.75 ± 0.5				
Electrode thickness (µm)	87 ± 4 (Al foil thickness of 15µm included)	101.5 ± 1 (Cu foil thickness of 10μm included)				
Electrode density (g cm ⁻³)	3.31 ± 0.02	1.52 ± 0.02				
Stacking	4	5				
Negative/positive (N/P) ratio	1.07 ±	0.01				
Separator thickness (µm)	16	Ĵ				
Amount of electrolyte	1.75 g (~2.	5g Ah^{-1})				
Formation step testing condition	Voltage range: 2.8 – 4.3 V Charge: 0.1C (CC) – 0.05C (CV) / Discharge: 0.1C (CC) Charge/discharge current density: 1C≈2.21 mA cm ⁻²					
Cycling test condition	Voltage range: $2.8 - 4.3$ V Charge: $1.0C$ (CC) $- 0.05C$ (CV) / Discharge: $1.0C$ (CC) Charge/discharge current density: $1C\approx 2.21$ mA cm ⁻²					

Fable S5. Full-cell specifications	(CC: constant current. CV: constant voltage.)
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Table S6. The gravimetric composition of commercial lithium-ion batteries

Materials	Ratio (wt%)	Materials	Ratio (wt%)
Cathode materials	~33.5	Graphite anode	~21.3
Conductive carbon and Binder	~5.3	Separator	~1.3
Aluminum	~5.3	Copper	~13.3
Electrolyte	~14.7	Others	~5.3

Pyrometallurgy upcycling method							
Procedures	Input	Amount	Output	Amount	Notes		
	Spent batteries	1.00 kg	Spent batteries (100% SOD)	1.00 kg	The data is estimated based on Everbatt 2020. 1 MJ electricity produces 0.12 kg GHG and		
Discharging and collecting	Energy	0.03 MJ	GHG	0.004 kg	consumes 0.53 L water. 1 kg of U.S. conventional diesel produces 37.9 MJ energy. ~5		
	Water	0.52 L	/	/	wt% NaCl solution is used for the discharging process. [Ref. 3]		
	Spent batteries (100% SOD)	1.00 kg	Matte (Co, Ni, Mn, Cu)	0.338 kg	The purpose is to reduce the transition metals and remove all the impurities. The temperature		
Smelting +	Energy	2.42 MJ	GHG	1.569 kg	is~1873 K for 3 h. The byproducts include the slag		
Gas treatment + Water-leaching	Water	1.81 L	Slag residue (Li ₂ CO ₃ , CaAl)	0.263kg	with AI, Ca, and LI, which requires the post-treatment to recycle the Li salts. The ratio of		
	Slag formation reagent	0.22 kg	/	/	is set 2:1. [Ref. 25,26]		
	Matte (Co, Ni, Mn, Cu)	1.00 kg	Fine metal particles	1.00 kg			
Granulator	Energy	0.007 MJ	GHG	0.001 kg	[Ref. 26]		
	Water	0.004 L	/	/			
	Fine metal particles	1.00 kg	Leachate	~20 kg			
	Energy	0.11 MJ	GHG	0.013 kg	The average pulp density is ~5%		
Acid leaching	Water	11.4 L	Cu compounds	0.41 kg	H ₂ SO ₄ for the pyrometallurgical method. The density of 10 M H ₂ SO ₄ is ~ 1.54 g cm ⁻³		
	10 M H ₂ SO ₄ solution	3.04 kg	/	/	[Ref. 27]		
	NiSO ₄ · 6H ₂ O	4.56 kg	/	/			

 Table S7. Life cycle inventory of pyrometallurgy upcycling method.

	Leachate	1.00 kg	Transition metal hydroxide	0.108 kg	The pH is adjusted to 10-11. The
Co precipitation	Energy	0.61MJ	GHG	0.07 kg	details to adjust the pH to precipitate hydroxide precursor is omitted here for the
Co-precipitation	NaOH	0.046 kg	/	/	convenience of calculation since the amount of reagent (NH4OH) is too small.
	Water	0.32 L	/	/	[Ref. 28]
High- temperature resynthesize	Transition metal hydroxide	1.00 kg	Resynthesized cathode	1.10 kg	
	LiOH·H ₂ O	0.466 kg	GHG	1.62 kg	The molar ratio of Li: TM=1.03 and the mixture is calcined at 723
	Energy	13.53 MJ	/	/	K for 5 h and 1123 K for 14 h. [Ref. 28]
	Water	7.17 L	/	/	

Hydrometallurgy upcycling method							
Procedures	Input	Amount	Output	Amount	Notes		
	Spent batteries	1.00 kg	Spent batteries (100% SOD)	1.00 kg	The data is estimated based on Everbatt 2020. 1 MJ electricity produces 0.12 kg GHG and		
Discharging and collecting	Energy	0.03 MJ	GHG	0.004 kg	consumes 0.53 L water. 1 kg of U.S. conventional diesel produces 37.9 MJ energy. ~5		
	Water	0.52 L	/	/	wt% NaCl solution is used for the discharging process. [Ref. 3]		
	Spent batteries (100% SOD)	1.00 kg	Battery pieces	0.81 kg	The battery pieces included the spent batteries without the		
Shredding	Energy	0.38 MJ	GHG	0.046 kg	organic solvents. Recycling rate in shredding process is set to		
	Water	0.20 L	Electrolyte	0.14 kg	[Ref. 26]		
	Battery pieces	1.00 kg	Black mass	0.642 kg	The purpose is to separate the active materials with current		
Incineration + Sieving	Energy	0.31 MJ	GHG	0.626 kg	collectors, to decompose tl binder, electrolyte residu plastics, and SEI. Tl		
	Water	0.16 L	Current collector	0.218 kg	temperature is ~873 K for 2 h. [Ref. 29]		
	Black mass	1.00 kg	Granulated particles	0.99 kg	The amount of water is set to ~20		
Wet granulation	Energy	0.007 MJ	GHG	0.001 kg	wt% of the solid. Recycling rate in wet granulation is set to 99%. [Ref. 30]		
	Water	0.204 L	/	/			
	Spent active materials	1.00 kg	Spent cathode powder	0.61kg			
Froth flotation	Energy	0.67 MJ	GHG	0.087 kg	The purpose is to separate the anode materials. [Ref. 26]		
	Water	3.36 L	Spent graphite	0.39 kg			

 Table S8. Life cycle inventory of hydrometallurgy upcycling method.

	Spent cathode powder	1.00 kg	Leachate	~50 kg	
	Energy	0.11 MJ	GHG	0.065 kg	The average pulp density is ~2% and the concentration is ~3.27 M
Acid leaching	Water	25.37 L	/	/	H ₂ SO ₄ for the hydrometallurgy method. The density of 10 M
	10 M H ₂ SO ₄ solution	19.01 kg	/	/	H ₂ SO ₄ is ~1.54 g cm ⁻³ . [Ref. 31]
	$NiSO_4 \cdot 6H_2O$	4.62 kg	/	/	
	Leachate	1.00 kg	Transition metal hydroxide	0.051 kg	The pH is adjusted to 10-11. The
	Energy	0.61MJ	Li ₂ CO ₃	0.008 kg	details to adjust the pH to precipitate hydroxide precursor
Co-precipitation	NaOH	0.022 kg	GHG	0.07 kg	is omitted here for the convenience of calculation since the amount of reagent (such as NH ₄ OH) is too small. [Ref. 28]
	Na ₂ CO ₃	0.010 kg	/	/	
	Water	0.32 L	/	/	
	Transition metal hydroxide	1.00 kg	Resynthesized cathode	1.10 kg	
High-	LiOH·H ₂ O	0.466 kg	GHG	1.62 kg	The molar ratio of Li: TM=1.03 and the mixture is calcined at 723
resynthesize	Energy	13.53 MJ	/	/	K for 5 h and 1123 K for 14 h. [Ref. 28]
	Water	7.17 L	/	/	

Liquified-salts-assisted upcycling method										
Procedures	Input	Amount	Output	Amount	Notes					
Discharge + collecting	Spent batteries	1.00 kg	Spent batteries (100% SOD)	1.00 kg	1 MJ electricity produces 0.13 kg GHG and 0.67 L water. 1 kg diesel produces 45.6 MJ energy. The data is estimated based on Everbatt 2020. ~5 wt% NaCl solution is used for the discharging process. [Ref. 3]					
	Energy	0.03 MJ	GHG	0.004 kg						
	Water	0.52 L	/	/						
Disassembly + Low-temperature calcination + Scrapping	Spent batteries (100% SOD)	1.00 kg	Cathode material	0.34 kg	The purpose of low-temperature calcination is to decompose the binder, electrolyte residue, and SEI. The temperature is ~623 K for 10min. Manual disassembly is considered to separate the spent active materials. The dismantling step can be achieved by a commercial core drill with a silicon carbide blade, which can reduce the manual disassembly cost. [Ref. 7, 32, 33]					
	Energy	0.40 MJ	Al foil	0.05 kg						
	Water	0.21 L	Anode electrode	0.37 kg						
	/	/	Electrolyte	0.15 kg						
	/	/	Separator and others	0.07 kg						
	/	/	GHG	0.090 kg						
Planetary centrifugal mixing + High- temperature resynthesize	Spent cathode powder	0.34 kg	Upcycled cathode	1.0 kg	Conduct planetary centrifugal mixing for 15min on the powder mixture to form homogeneous liquified mixture of spent cathode powder and molten- salts. The energy requirement for planetary centrifugal mixer is 1.33MJ kg ⁻¹ . Then, the powder					
	LiOH·H ₂ O	0.120 kg	GHG	1.73 kg						
	LiNO ₃ ·H ₂ O	0.296 kg	/	/						
	Ni(NO ₃) ₂ ·6H ₂ O	1.630 kg	/	/						
	Energy	14.33 MJ	/	/	mixture is calcined at 1193 K for 2 h and subsequently at 1053K for 8h.					
	Water	7.59 L	/	/						

 Table S9. Life cycle inventory of liquified-salts-assisted upcycling method.

	Cost	Requirements f	Dof					
	(unit)	Pyro	Hydro	LSI	Kel.			
Energy	0.111 (USD MJ ⁻¹)	16.49 MJ	21.44 MJ	4.94 MJ	Ref. 26			
Water	0.018 (USD L ⁻¹)	13.58 L	21.41 L	3.31 L	Ref. 26			
Wastewater Treatment	0.027 (USD L ⁻¹)	5.84 L	10.74 L	0.36 L	Ref. 26			
Spent batteries (with NCM523)	3.2 (USD kg ⁻¹)	1 kg	1 kg	1 kg	Ref. 34			
Concentrated H ₂ SO ₄	0.12 (USD L ⁻¹)	1.03 L	5.97 L	/	Ref. 26			
NiSO4 · 6H2O	3.2 (USD kg ⁻¹)	1.54 kg	1.45 kg	/	Ref. 26 Ref. 35			
Ni(NO ₃) ₂ · 6H ₂ O	3.82 (USD kg ⁻¹)	/	/	1.63 kg	Ref. 26 Ref. 35			
LiOH · H ₂ O	9.50 (USD kg ⁻¹)	0.34 kg	0.37 kg	0.12 kg	Ref. 26 Ref. 36			
LiNO ₃ · H ₂ O	13.78 (USD kg ⁻¹)	/	/	0.296 kg	Ref. 26 Ref. 36			
Total cost in upcycling 1kg of spent batteries	(USD kg cell ⁻¹)	\$ 13.73	\$ 15.17	\$ 15.27	Ref. 26			
	Products of upcycling process from medium-Ni to Ni-rich cathode							
Amount of upcycled NCM811 Cathode from each process	/	0.8 kg	0.88 kg	1.00 kg	Ref. 26			
Total manufacturing cost to produce 1kg of upcycled NCM811	26.0 (USD kg ⁻¹)	\$ 20.41	\$ 20.75	\$ 17.74	Ref. 26			

Table S10. Required resources and products for upcycling of spent battery

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