Efficient and Selective Recovery of Spent NCM Cathode Materials: A Green Deep Eutectic Solvent Process via Synergistic Acidity-Viscosity Regulation

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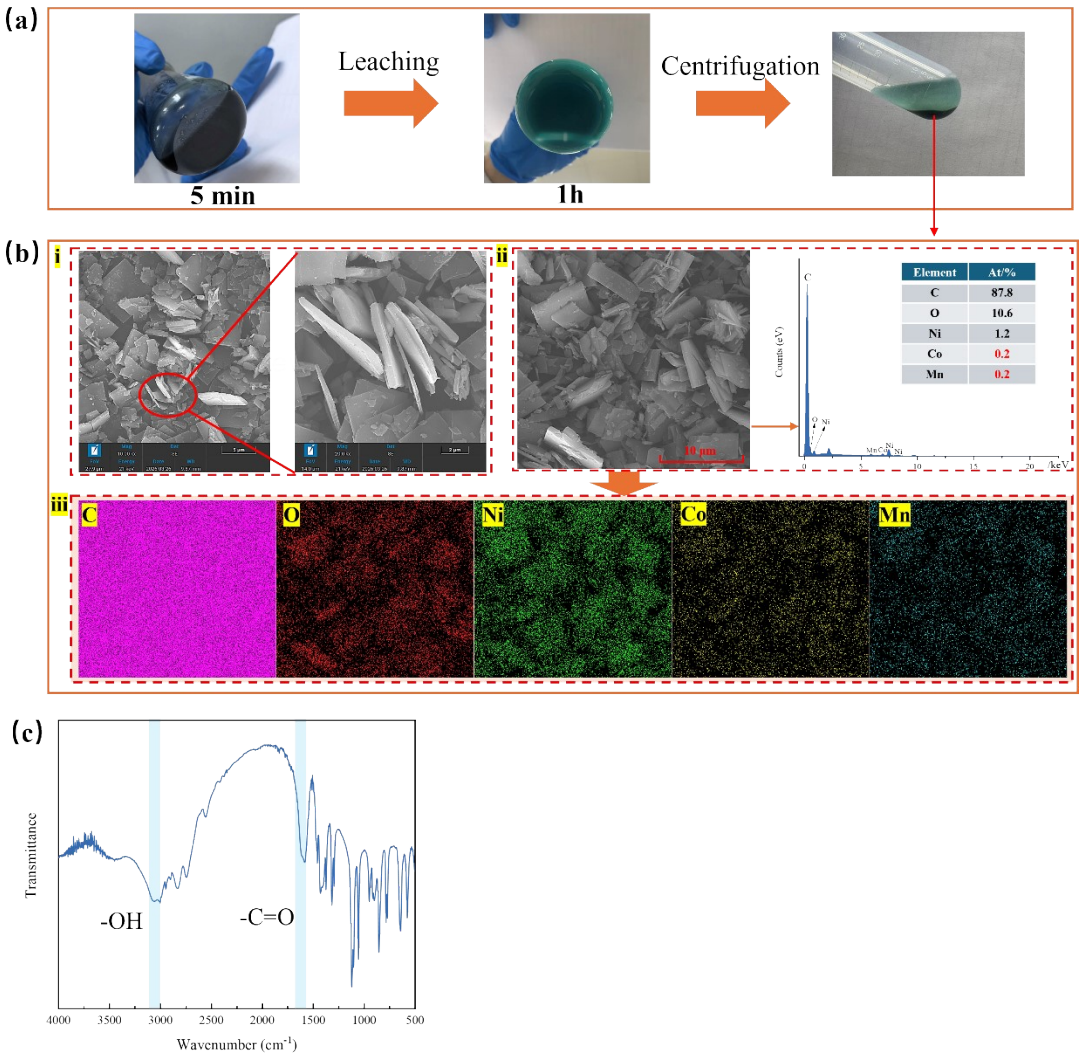

Figure S1. Characterization of the solid residue obtained after leaching at 140 °C for 1 h: (a) Photographs of the leachate at 5 min and 1 h, and the leachate at 1 h after centrifugation; (b) SEM image and EDS elemental mapping of the precipitate; (c) FTIR spectrum of the precipitate.

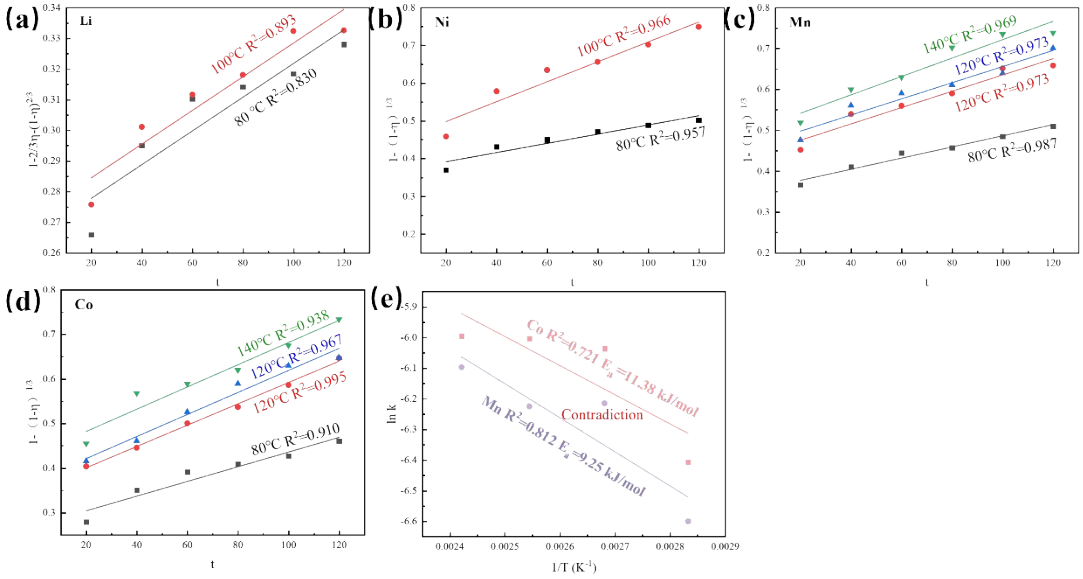

Figure S2. Leaching kinetics based on the shrinking core model (ash-layer diffusion control) for (a) Li, (b) Ni, (c) Mn, (d) Co, and their corresponding Arrhenius plots.

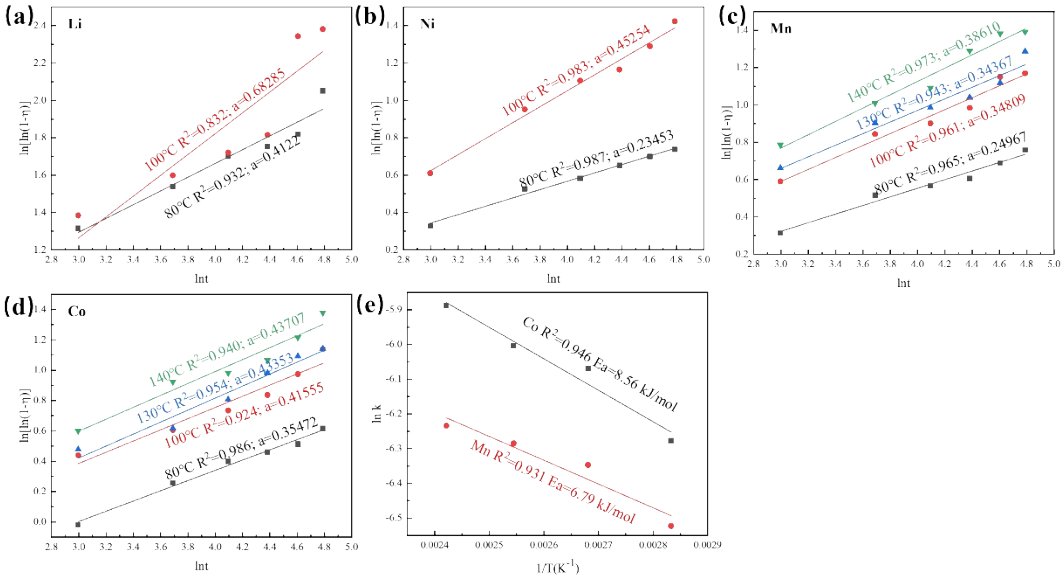

Figure S3. Leaching kinetics based on the Avrami–Erofeyev model for (a) Li, (b) Ni, (c) Mn, (d) Co, and their corresponding Arrhenius plots.

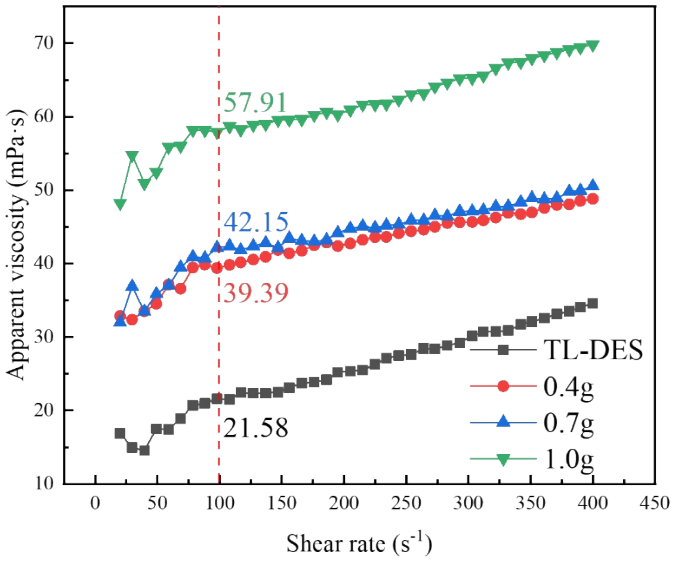

Figure S4. Viscosity of TL-DES leachate as a function of cathode material loading measured at 25 ℃ after leaching at 100 ℃for 2 h.

Figure S5. State and UV-Vis spectroscopic analysis of the leachate. (a) Photographic comparison of the original leachate and the diluted leachate; (b) UV-Vis absorption spectrum of the original leachate.

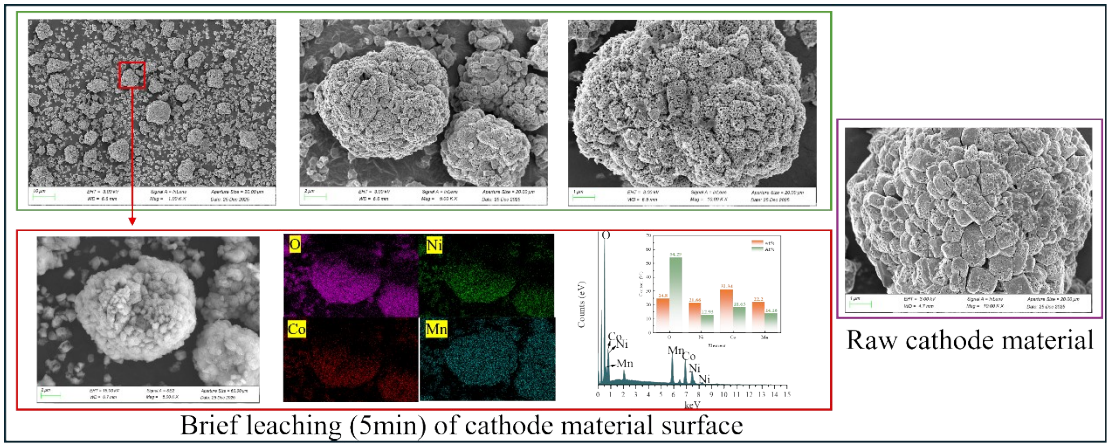

Figure S6. Different regions morphology of the cathode surface after brief leaching (5min).

Figure S7. Immediate formation of a light pink precipitate upon the addition of ethanol to the leachate.

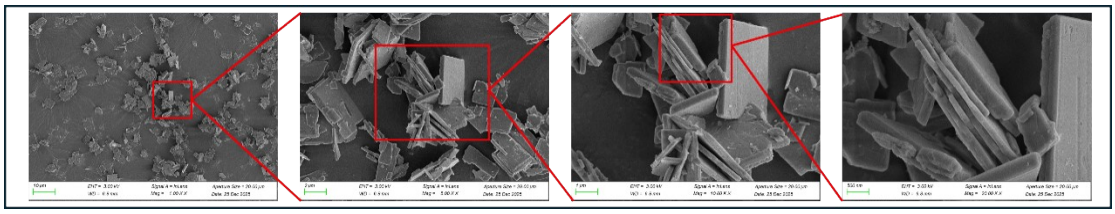

Figure S8. Different regions morphology of the precipitates.

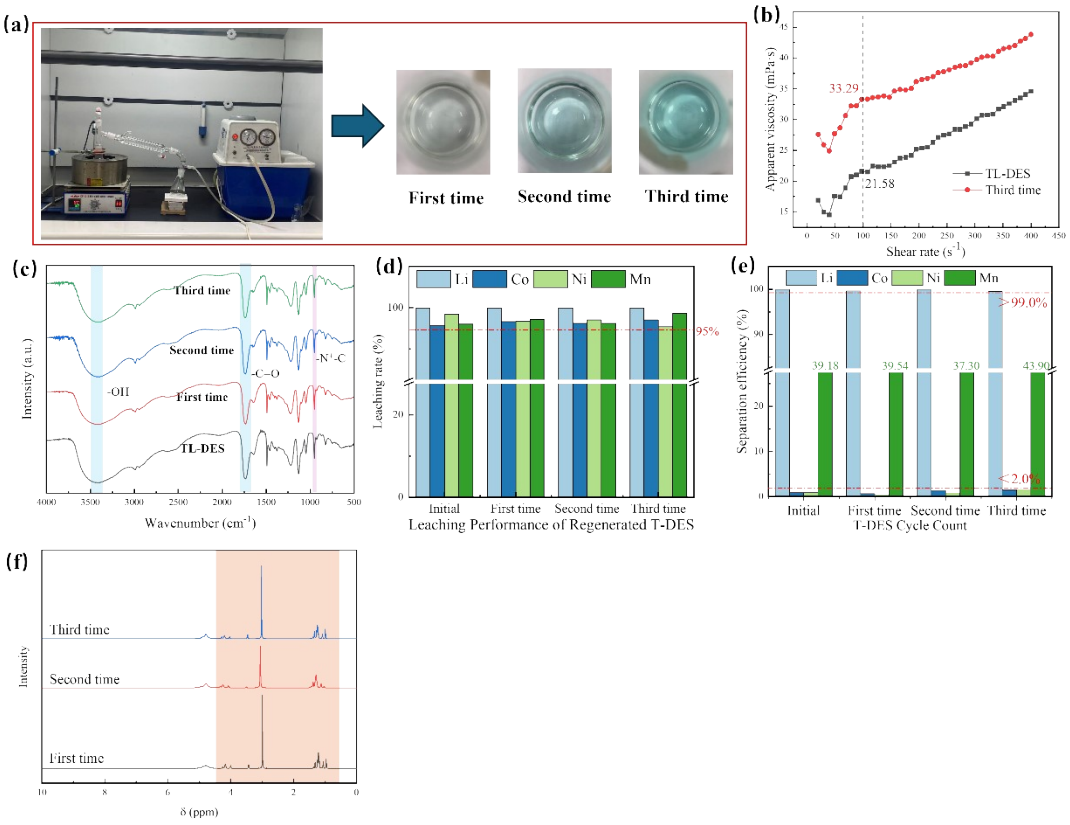

Figure S9. Recyclability study of the TL-DES system. (a) Process flowchart for DES regeneration; (b) Viscosity comparison of fresh DES and DES after three cycles; (c) FTIR spectra of fresh DES and regenerated DES; (d) Leaching efficiencies of Li, Ni, Co, and Mn over three consecutive cycles; (e) Selective separation efficiencies of metals after each cycle; (f) ^{1}H NMR spectra of fresh DES and regenerated DES.

Table S1. Elemental Analysis of NCM Raw Materials.

Element	Li	Co	Ni	Mn
Content/%	4.123	24.544	25.382	22.930

Table S2. Kinetic parameters obtained from the Avrami–Erofeyev model for the leaching of Li, Ni, Mn, and Co in TL-DES at different temperatures.

Element	Temperature (℃)	R^2	a	Interpretation
Li	100	0.832	0.683	Mixed control
Li	80	0.932	0.412	Diffusion control
Ni	100	0.983	0.453	Diffusion control
Ni	80	0.987	0.235	Diffusion control
Mn	140	0.973	0.386	Diffusion control
Mn	130	0.943	0.344	Diffusion control
Mn	100	0.961	0.348	Diffusion control
Mn	80	0.965	0.250	Diffusion control
Co	140	0.940	0.437	Diffusion control
Co	130	0.954	0.434	Diffusion control
Co	100	0.924	0.416	Diffusion control
Co	80	0.986	0.355	Diffusion control

Table S3. Ethanol recovery efficiencies over three regeneration cycles.

Number of cycles	Anti-solvent - Ethanol		
	Dosage/g	Recovery amount/g	Recovery rate/%
First	29.96	28.241	94.262
Second	31.12	29.881	96.018
Third	32.128	29.554	91.988

Table S4. Costs percentage of Hydrometallurgy.

Project	Percentage/%
Annualized capital cost	5.40
Other fixed costs	1.03
Plant overhead	0.00
Maintenance	0.92
Labor	0.34
Other variable costs	1.84
Utilities	0.46
Materials	92.31

Table S5. The treatment of battery components by different recycling methods.

Handling methods	Pyrometallurgy	Direct	This work
Active cathode materials	Recycle	Recycle	Recycle
Graphite	Burn for energy	Recycle	Recycle
Copper	Recycle	Recycle	Landfill
Aluminum	Landfill	Recycle	Landfill
Plastics	Burn for energy	Landfill	Landfill
Electrolyte: solvents	Burn for energy	Intermediate	Intermediate
Carbon black	Burn for energy	Recycle	Landfill
Binder	Burn for energy	Landfill	Landfill

Table S6. Environmental impacts of different recycling methods.

	Pyrometallurgy	Direct	This work
Energy use in MJ per kg feedstock processed			
Total Energy	**8.48**	**15.638**	**7.220**
Total Emissions in g per kg feedstock processed			
VOC	0.091	0.245	0.129
CO	0.355	0.861	0.429
NOx	0.957	2.220	1.614
PM10	0.117	0.287	0.116
PM2.5	0.066	0.208	0.094
SOx	1.509	1.591	6.597
BC	0.015	0.035	0.022
OC	0.011	0.050	0.032
CH_4	1.026	2.007	1.005
N_2O	0.016	0.018	0.013
CO_2	2,051	1,223	484
CO_2 (w/ C in VOC & CO)	2,052	1,225	485
GHGs	**2,087**	**1,290**	**519**

Table S7. The cost in different recycling methods ($/kg).

	Pyrometallurgy	Direct	This work
Annualized capital cost	1.50	0.87	0.47
Materials	6.24	7.86	8.04
Labor	0.03	0.03	0.03
Utilities	0.08	0.02	0.04
Maintenance	0.14	0.14	0.08
Other variable costs	0.13	0.16	0.16
Other fixed costs	0.09	0.09	0.09
Total	8.21	8.90	8.71

Table S8. The product recycling value in different recycling methods ($/kg).

	Pyrometallurgy	Direct	This work
Li^+ in product	/	/	2.06
Co^{2+} in product	6.12	/	6.19
Ni^{2+} in product	2.04	/	2.07
Mn^{2+} in product	/		0.37
Graphite	/	/	0.06

Copper metal	0.07	/	/
NCM	/	12.42	/
Total	8.23	12.42	10.75