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Review

Value recovery from spent lithium-ion batteries: A review on technologies, environmental impacts, economics, and supply chain

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Supplementary

Table S1. Optimal leaching conditions derived from design of experiments using mineral acids.

Reference	Cathode material	Initial chemical composition	Leaching agent	Reductant	Optimal condition	Leaching efficiency	DOE method
[61]	$LiCoO_2$ + $LiNiO_2$ + $Li_4Mn_5O_{12}$ + $Li_{0.9}MnO_2$	Li: 7.09%, Co: 30.98%, Ni: 9.09%, Mn: 4.96%, Al: 0.61%, Cu: 0.19%, Fe: 0.91%	Sulfuric acid	Fe^{2+} from iron scrap dissolution	H_2SO_4: 1 mol/L, Iron scrap: 25 g/L, S/L ratio: 10 g/L, Temperature: 70° C, Leaching time: 150 min, Agitation rate: 500 rpm	Li: 90.5%, Co: 99.1%, Ni: 94.9%	Response surface methodology
[24]	Hybrid powders of $LiFePO_4$ and $LiMn_2O_4$	Li: 43.5%, Mn: 33.45%, Fe: 17.18%, P: 9.36%	Hydrochloric acid and hydrogen peroxide	N/A	HCl: 6.5 mol L^{-1}, H_2O_2: 15% vol%, S/L ratio: 1:5, Temperature: 60°C, Leaching time: 2.0 h, Agitation rate: 1000 rpm	Li: 80.93%, Mn: 81.02%, Fe: 85.40%	Fractional factorial design
[115]	$LiCoO_2$	Li: 6.7%, Co: 51.4%, Ni: 7.0%, Mn: 6.0%	Sulfuric acid and vitamin C	N/A	H_2SO_4: 3 mol/L, Vitamin C: 0.56 mol/L, S/L ratio: 15 $g{\cdot}L^{-1}$, Temperature: 70°C	Co: 95.5%	Fractional factorial design
[62]	$LiCoO_2$	Co: 73.67%, Ni: 8.66%, Mn: 8.73%, Cu: 1.46%	Sulfuric acid	Hydrogen peroxide	H_2SO_4: 3.0 mol/L, H_2O_2: 1.6 mL/g, L/S ratio: 7:1, Temperature: 70°C, Leaching time: 2.5 h	Not specified	Fractional factorial design

[116]	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	Not specified	Sulfuric acid	Hydrogen peroxide	H_2SO_4: 1.5076 mol/L, H_2O_2: 8 vol%, S/L: 30 g/L, Temperature: 25℃, Leaching time: 60 min	Li: 80.2%, Co: 93.2%, Ni: 91.5%, Mn: 91.5%	Central composite face-centered design (CCFD)
[117]	$LiNi_{0.4}Co_{0.3}Mn_{0.3}O_2$	Li: 72.0%, Co: 31.2%, Ni: 42.5%, Mn: 15.2%	Sulfuric acid	30% Zinc	**Pretreatment stage:** Cathode material to Zn powder mass ratio of 7:3, Rotational speed: 500 rpm, Milling time: 2 h, Ball-powder mass ratio:19:1 **Leaching condition:** Acid concentration: 1.5 mol/L, L/S ratio: 50 mL/g, Temperature: 50℃, Leaching time: 15 min	Li : 99.9%, Co: 94.3%, Ni: 96.2%, Mn: 91.0%	Fractional factorial design
[63]	$LiNi_{0.15}Mn_{0.15}Co_{0.70}O_2$	Li: 5.79%, Co: 35.8%, Ni: 8.77%, Mn: 8.11%, Cu: 0.05%, Al: 0.24%	Sulfuric acid	Hydrogen peroxide	H_2SO_4: 1.0 M H_2O_2: 0.62 wt.%, L/S ratio: 25.8 mL/g, Temperature: 51℃, Leaching time: 60 min	~100% recovery of Li, Ni, Mn, and Co	Response Surface Methodology
[118]	$LiCoO_2$	Li: 6.69%, Co: 58.11%, Ni: 0.39%, Mn: 0.14%, O: 34.55%	Phosphoric acid (also precipitation agent)	Hydrogen peroxide	H_3PO_4: 0.7 mol/L, H_2O_2: 4 vol%, L/S ratio: 20 mL/g, Temperature: 40℃, Leaching time: 60 min,	Li: 99.9%, Co: 99.7%	Fractional factorial design

Table S2. Optimal leaching conditions derived from the design of experiment using organic acids.

Reference	Cathode material	Initial chemical composition	Leaching agent	Reductant	Optimal condition	Leaching efficiency	DOE method
[60]	$LiCoO_2$	Li: 3.27% Co: 27.64% Ni: 1.04% Cu: 3.87% Al: 6.36%	Citric acid	Hydrogen peroxide	Citric acid: 2 M, Hydrogen peroxide: 1.25 vol.%, S/L: 30 gL^{-1}, Temperature: 60°C, Leaching time: 5 h	Li: 99.80%, Co: 96.46%	Response surface methodology
[64]	$LiCoO_2$	Li: 6.94% Co: 60.16% Cu: 0.026% Al: 0.020% Fe: 0.030%	Oxalic acid (Also used as precipitating reagent)	N/A	$H_2C_2O_4$: 1M S/L ratio: 15 gL^{-1}, Temperature: 95°C, Retention time: 150 min, Rotation rate: 400 rpm	Li: 98%, Co: 97%	Fractional factorial design
[65]	$LiCo_{1/3}Ni_{1/3}Mn_{1/3}O_2$	Li: 6.79% Co: 17.68% Ni: 17.58% Mn: 16.46%	Lactic Acid ($C_3H_6O_3$)	Hydrogen peroxide	$C_3H_6O_3$: 1.5 $molL^{-1}$, H_2O_2: 0.5 vol%, S/L ratio: 20 gL^{-1}, Temperature: 70°C, Leaching time: 20 min	Li: 97.7%, Co: 98.9%, Ni: 98.2%, Mn: 98.4%	Fractional factorial design
[119]	$LiNi_{0.05}Mn_{0.05}Co_{0.9}O_2$	Li: 9.73% Co: 28.82% Ni: 33.91% Mn: 24.39% Cu: 0.11% Al: 1.48%	Citric acid	Hydrogen peroxide	Citric acid: 1.5M, DL-malic acid: 1M, H_2O_2 content: 2 vol%, Temperature: 95°C, Leaching time: 30 min	**Citric Acid**: Li: 97%, Co: 95%, Ni: 99%	Full factorial design
[120]	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	N/A	Malonic acid	Hydrogen peroxide	Malonic acid: 1.5 mol L^{-1}, H_2O_2: 0.5 vol%, S/L ratio: 20 gL^{-1}, Temperature: 70°C, Leaching time: 20 min	Li: 95.74%, Co: 98.06%, Ni: 98.27%, Mn: 98.54%	Fractional factorial design

[66]	$LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	N/A	Acetic acid or Maleic acid	Hydrogen peroxide	**Maleic acid:** Maleic acid: 2 M, H_2O_2: 4 vol%, Temperature: 70°C, Leaching time: 60 min **Acetic acid**: Acid concentration: 1 M, H_2O_2 volume: 6 vol%, Temperature: 70°C, Leaching time: 60 min	**Maleic acid:** Li: 99.45%, Co: 98.45%, Ni: 98.58%, Mn: 98.16% **Acetic acid**: Li: 98.83%, Co: 97.85%, Ni: 97.93%, Mn: 97.74%	Fractional factorial design
[69]	$LiCoO_2$ and $Li_2CoMn_3O_8$	Li: 5.1% Co: 26.3% Mn: 28.2%	Acetic acid	Hydrogen peroxide	Acetic acid: 3 M, H_2O_2: 7.5 vol%, Pulp density: 20 g/L, Temperature: 70°C, Leaching time: 40 min	Li: 99.9%, Co: 98.7%, Mn: 99.5%	Fractional factorial design
[67]	$LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$	Li: 6.64% Co: 12.1% Ni: 35.1% Mn: 11.2% Cu: 0.0141% Al: 0.0313% Fe: 0.04%	DL-malic acid	Hydrogen peroxide	DL-malic acid: 1.0 mol/L, H_2O_2: 4 vol%, Pulp density: 5 g/L, Ultrasound power: 90 W, Temperature: 80°C, Leaching time: 30 min	Li: 98%, Co: 97.6%, Ni: 97.8%, Mn: 97.3%	Fractional factorial design
[70]	$LiCoO_2$ and $Li_{0.45}Ni_{1.025}O_2$	N/A	Lemon Juice	Hydrogen peroxide	Lemon juice: 57.8% (v/v), H_2O_2: 8.07% (v/v), S/L ratio: 0.98% (w/v)	Li: 100%, Co: 96%, Ni: 96%	Response surface methodology
[121]	$LiCoO_2$	Li: 4.35% Co: 32.35% Ni: 0.98% Cu: 2.04% Al: 0.20%	Lactic Acid	Hydrogen peroxide	Lactic acid: 1.52 M, H_2O_2: 4.84 vol%, S/L ratio: 16.3 g.L^{-1}, Temperature: 79°C, Leaching time: 2 h	Li: 100%, Co: 97.36%	Response surface methodology

[122]	$LiCoO_2$	N/A	Maleic acid	$SnCl_2$	Maleic acid: 1 mol L^{-1}, SnCl2: 0.3 mol L^{-1}, S/L ratio: 20 gL^{-1}, Temperature: 60°C, Leaching time: 40 min,	Li: 98.67%, Co: 97.59%	Not clearly specified

Table S3. Battery and waste battery regulation proposal by European Commission in 2020 [103].

Measures	**Option 2. medium level of ambition**	**Option 3. high level of ambition**	**Option 4. very high level of ambition**
1. Classification and definition	New category for EV batteries Weight limit of 5 kg to differentiate portable from industrial batteries	New calculation methodology for collection rates of portable hatteries based on batteries available for collection	/
2. Second-life of industrial batteries	At the end of the first life, used batteries are considered waste (except for reuse). Repurposing is considered a waste treatment operation. Repurposed (second life) batteries are considered as new products which have to comply with the product requirements when they are placed on the market	At the end of the first life, used batteries are not waste. Repurposed (second life) batteries are considered as new products which have to comply with the product requirements when they are placed on the market	Mandatory second life readiness
3. Collection rate for portable batteries	65% collection target in 2025	70% collection target in 2030	75% collection target 2025
4. Collection rate for automotive and industrial batteries	New reporting system for automotive, EV and industrial batteries	Collection target for batteries powering light transport vehicles.	Explicit collection target for industrial, EV and automotive batteries
5. Recycling efficiencies and recovery materials	Lithium- ion batteries and Co, Ni, Li, Cu: Recycling efficiency lithium-ion batteries: 65% by 2025 Material recovery rates for Co, Ni, Li, Cu: resp. 90% 90% 35% and 90% in 2025 Lead-acid batteries and lead: Recycling efficiency lead-acid batteries: 75% by 2025	Lithium-ion batteries and Co, Ni, Li, Cu: Recycling efficiency lithium-ion batteries: 70% by 2030 Material recovery rates for Co, Ni, Li, Cu: resp. 95% 95% 70% and 95% in 2030 Lead-acid batteries and lead:	/

	Material recovery for lead: 90% in 2025	Recycling efficiency lead-acid batteries: 80% by 2030 Material recovery for lead: 95% by 2030	
6. Carbon footprint for industrial and EV batteries	Mandatory carbon footprint declaration	Carbon footprint performance classes and maximum carbon thresholds for batteries as a condition for placement on the market	/
7. Performance and durability of rechargeable industrial and EV batteries	Information requirements performance and durability	Minimum performance and durability requirements for industrial batteries as a condition for placement on the market	
8. Non-rechargeable portable batteries	Technical parameters for performance and durability of portable primary batteries	Phase out of portable primary batteries of general use	Total phase out of primary batteries
9. Recycled content in industrial, EV and automotive batteries	Mandatory declaration of levels of recycled content, in 2025	Mandatory levels of recycled content, in 2030 and 2035	/
10. Extended producer responsibility	Clear specifications for extended producer responsibility obligations for industrial batteries. Minimum standards for PROs	/	/
11. Design requirements for portable batteries	Strengthened obligation removability	New obligation on replaceability	Requirement interoperability
12. Provision of information	Provision of basic information (as labels, technical documentation or online). Provision of more specific information to end-users and economic operators (with selective access)	Setting up an electronic information exchange system for batteries and a passport scheme (for industrial and electric vehicle batteries only)	/
13. Supply-chain due diligence for raw materials in industrial and EV batteries	Voluntary supply-chain due diligence	Mandatory supply chain due diligence	/