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ADVANCES IN LITHIUM RECOVERY FROM PRIMARY AND SECONDARY SOURCES

Recycling of Li-Ion Batteries: Recovery of Critical Metals by Hydrometallurgy

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In this study, we aimed to study acid leaching and solvent extraction for recycling of NMC-type batteries (622—pouch; 631—prismatic). Physical structures have no impact on our physical process (milling and sieving). In acid leaching, the addition of reducing agents was unnecessary because of the presence of Al foil. Up to 30% of acid excess is required for complete cathode material leaching, a crucial process conclusion that varies with different black mass compositions. This calculation was made based on the amount of metals in the black mass per acid. Al and Cu were removed by precipitation. In solvent extraction experiments, Cyanex 272 has higher selectivity than D2EHPA and separates 100% of Mn/Co with the co-extraction of Li/Ni (20%). Products of the process were Mn oxide, Ni hydroxide, and Li carbonate. We clearly observed that precipitation of Mn before solvent extraction steps is necessary for Co recovery from the recycling of NMC batteries because of the co-extraction and subsequent losses of Co, as clearly illustrated in the process flowchart. Our conclusions are in accordance with previous studies reported in the literature. Separation of metals in the leach solution has higher efficiencies without the presence of Mn ions (in NCA-type processing or after Mn removal by ozone). In the presence of Mn in the leach solution, low recovery efficiencies were obtained for Li (48.0%), Ni (62.8%), and Co (58.6%). Our conclusions and aim are to demonstrate that traditional approaches from extractive metallurgy are not suitable for the ongoing challenge in recycling and waste valorization.

INTRODUCTION

Li-ion batteries are important for the transportation sector's low-carbon future,[1] and they are classified based on physical structure and cathode material. The module cells of the battery can be cylindrical, prismatic, and pouch, and their main differences are the organization of internal materials (cathode, anode, electrolytes, and polymeric separator) and composition of external structure (cylindrical and prismatic = usually Al alloys, pouch = Al foils with plastic).[2] Beyond the physical structure, the main difference (e.g., efficiency and cost) is the cathode composition. The market is mostly focused on NMC ($LiNiMnCoO_2$ - ∼ 60%) and LFP ($LiFePO_4$ - ∼ 40%) batteries.[3–6]

There are different proportions of Ni, Mn, and Co in NMC batteries: NMC 111 (Ni = 1, Co = 1, and Mn = 1), NMC 532, NMC 622, NMC 811, and most recently NMC 955 (Ni = 9, Co = 0.5 and Mn = 0.5).[7,8] However, there are several concerns regarding the impact of mining to obtain these metals for Li-ion battery production to meet the growing demand. Although mining and extractive metallurgy have

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negative impacts on the environment and society, these activities are crucial to meeting the energy transition. For this reason, we must find alternatives to metal supply, and recycling has been shown to be a more low-environmental and greener process than primary extraction,[1,9–11] including for domestic supply.[12] Recycling reduces the demand for mining and reduces environmental and social impacts, and high recovery rates (80-95% at least) are needed for sustainability and a circular economy.[13,14]

Hydrometallurgy is an alternative for a low-energy intensive process and recovery of pure products (including Li).[6,15–19] After discharging and a milling/sieving process[20] to obtain a cathode-rich material (also known as black mass), the acid leaching dissolves the metals into the acid solution, separating them from the graphite. Among the acids, H_2SO_4 is reported as the most feasible among inorganic acids (economically and environmentally), while organic acids have been reported as well as greener alternatives[21–24] in specific cases. The challenge in acid leaching is determining the suitable process parameters for the extraction of different black mass compositions.

Furthermore, the leach solution contains all metals from the cathode with a few impurities (e.g., Al, Cu, and Fe), including Li, Ni, Co, and Mn as the main elements (for NMC batteries).[24] The state-of-the-art separation of these elements includes solvent extraction, ion exchange resins, and precipitation, where solvent extraction is the most studied because of the selectivity and application in mining processes.[25,26] However, the literature reports losses of valuable metals in solvent extraction separation steps.[27–29]

Despite the well-known impact of solvent extraction, the literature has continuously reported the applicability of solvent extraction for the separation of metals after acid leaching of NMC batteries. Solvent extraction has demonstrated high separation efficiency for NCA-based black mass,[30] but the presence of Mn in the leach solution results in losses of valuable metals by co-extraction.[27] Our previous studies have demonstrated that oxidative precipitation (ozone) is technically feasible for Mn precipitation, including in NMC recycling[31]; on the other hand, the impacts of Mn ions on the solvent extraction path and their comparison have yet to be reported.

Our study's main question is whether solvent extraction is a suitable technique for NMC battery recycling. The impact of NMC-type in acid leaching and the effectiveness of the solvent extraction technique in the separation of Li, Ni, Co, and Mn were studied. Two different batteries (pouch and prismatic) were milled to prepare a black mass material for acid leaching. Acid leaching experiments were carried out with H_2SO_4 and $C_6H_8O_7$ (citric acid) for comparison in terms of costs and efficiency. D2EHPA (di-(2-ethylhexyl)-phosphoric acid) and Cyanex 272 (bis(2,4,4-trimethylpentyl)phosphinic acid) were tested for the separation of metallic ions.

This study reveals that NMC-type leaching parameters are related to the amount of metals in the black mass per acid in reaction (g of black mass per mL of leach solution). This finding is crucial in recycling flexibility for different feedstock compositions. Although similar studies have been reported in the literature,the solvent extraction technique has been shown to be unfeasible for a selective separation because of Co losses throughout the process. Selective separation will be reached under selective removal of Mn by other techniques such as oxidation precipitation.[31] Conclusions are reached based on Mn presence compared to leach solutions with the absence of Mn ions.[30,32] Our work meets the sustainable development and sustainable mining goals for critical minerals supply.[12,33–37]

MATERIALS AND METHODS

Characterization

Two different NMC batteries were used in this study—pouch and prismatic— received from the automotive sector's original equipment manufacturing (OEM). The main goal of this section was to provide complete composition information. For characterization, the modules were discharged and connected in an electrical resistance furnace until the voltage reached 0 V (after 24 h). Then, the cells were manually dismantled and separated into cathode, anode, polymeric separator, and case and dried at room temperature for 48 h in a fume hood.[38] Chemical characterization of cathode and anode was carried out by aqua-regia digestion at 50°C for 24 h and analyzed after dilutions with 3% HNO_3 in Flame Atomic Absorption Spectroscopy (FAAS, Shimadzu AA-7000).

Anode and cathode materials were analyzed by X-ray diffraction equipment (XRD, Rigaku MiniFlex 300) Cu Kα radiation (λ = 1.5418 Å) at 30 kV and 10 mA in the 2θ sweep range from 15 to 85° with a scanning time of 0.5°/min. Scanning electron microscope coupled with energy-dispersive spectroscopy (SEM-EDS, Phenon ProX) analysis was performed for cathode and anode after washing with ethanol 70% and drying at 60°C for 24 h. Total organic carbon determination (TOC, Eltra CS2000) and moisture content analysis (at 100°C for 2 h) were also performed for cathode and anode samples. Those characterizations were also performed for leaching and solvent extraction experiments as necessary.

Fourier transform infrared spectroscopy with an ATR sampler (FTIR, Bruker Tensor 27) and differential scanning calorimetry (DSC, Nietzsche DSC200 F3Maia) were used for polymeric separator characterization.

Hydrometallurgical Processing

The cells (pouch and prismatic) were first discharged using physical resistance until 0 V as previously reported by our group.[30,38,39] The discharged cells were milled in a knife mill using a 9.4-mm grid and dried at 60°C to remove the polymeric fraction and case from the cells and obtain the black mass concentrated in cathode material for leaching. At this stage, volatile electrolytes can be recovered.[40,41] The dried material was sieved for the polymeric separator and case removal. The pass material was milled with a 2-mm grid and then sieved to remove the remaining polymeric and case.

Leaching experiments aimed at comparing the acid leaching efficiency for different NMC black mass materials and determining the acid excess. Experiments were carried out in a glass reactor under heating and magnetic stirring. A thermometer and condenser (water at 5°C) were coupled to the reactor. Leaching agents H_2SO_4 and $C_6H_8O_7$ were compared. Although the literature reports that organic acid leaching has a higher carbon footprint than H_2SO_4,[42] its production from the bioprocess would make CO_2 emissions decrease substantially,[23,43] which motivates this study. Parameters such as solid-liquid ratio, temperature, acid concentration, and time were evaluated. Leaching efficiency was determined by mass balance.

Before solvent extraction experiments, Al and Cu were removed by precipitation. Solvent extraction experiments were carried out with Cyanex 272 (bis(2,4,4-trimethylpentyl) phosphinic acid) and D2EHPA (bis(2-ethylhexyl)phosphoric acid) diluted in kerosene. These extractants were chosen because of their selectivity for transition metals such as Mn, Co, and Ni in sulfate solution and reported for application in research and industries.[25–27]

Experiments were carried out in batches under magnetic stirring and heating, and the effects of pH, temperature, extractant concentration, and aqueous/organic phase were evaluated. Stripping was carried out in different H_2SO_4 concentrations or water and aqueous/organic ratios. Considerations regarding solvent extraction for NMC battery recycling were made based on a flowchart from the experimental data.

RESULTS AND DISCUSSION

Battery Cell Characterization and Physical Processing

A pack of pouch batteries and prismatic battery cells were received from the automotive sector, and these cells were discharged for characterization to provide complete information regarding the chemical and physical composition. Details of the characterization are depicted in Supplementary Materials. For pouch batteries, the pack was discharged and then dismantled for cell release (Fig. S1), where several electronic parts were separated (Fig. S1a) as printed circuit boards and Cu cables. Pouch batteries were mainly composed of the anode (37.5%) and cathode (31.8%), while the external structure represented a small portion of the cell (7.0%) made of plastic and Al foil. Ni (6.1%), Co (2.4%), Mn (2.1%), and Li (1.6%) were the metals present in the cathode as NMC 622 (Fig. S1e). Carbon represents 16.0% of the battery cell (anode fraction). The polymeric separator is a polypropylene (Fig. S1f and Fig. S1g).

Cells of a prismatic battery were received (no pack dismantling was necessary). The external structure, which accounts for 15.2%, is made of Al alloy to ensure physical resistance.[44] Anode (31.0%) and cathode (40.5%) are the main fractions in the prismatic cell. Metal content from the cathode was higher in prismatic than in pouch cells, with Ni (11.2%), Co (2.9%), Mn (2.6%), and Li (6.0%) as NMC 631 cathode (Fig. S2d). Lower amounts of Al and Cu foils as electron collectors and polymeric separators were verified in prismatic batteries (Fig. S2e) in contrast to pouch batteries (Fig. S1d).[2] Polymeric separators were identified as high-density polyethylene (PDPE) (Fig. S2e, f).

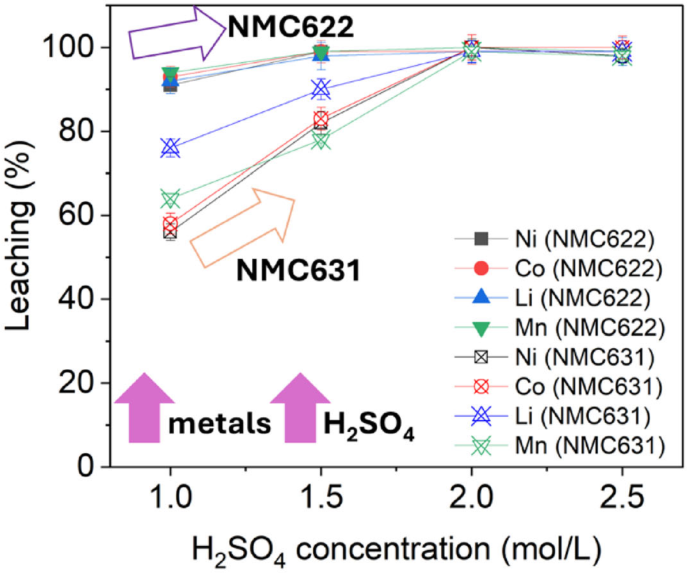
The physical treatment procedure was proposed by our group and depicted in detail by Guimarães et al. for NMC-cylindrical batteries,[38] LCO-pouch batteries,[39] and NCA-cylindrical batteries.[30] The milling followed by sieving steps separated all polymeric fractions (separator) and external structure (Table S1) producing a highly concentrated material in the cathode and anode (known as black mass). The advantage of this procedure is the low-energy-intensive requirement and purity of non-degraded fractions (e.g., polymers), making their further recycling possible. As noted in Table I, the concentration of cathode metals increased after the physical process. Al and Cu content decreased because of the sieving step as the electron collectors were easily separated. Black mass compositions for both NMC batteries were similar, demonstrating that the procedure is flexible for different battery structures and cathode types. The materials reported in Table I were used for leaching experiments.

Leaching of Spent NMC After Physical Processing

In acid leaching, the literature reports the need for a reducing agent (e.g., H_2O_2) for Co-based cathode leaching due to the strong bonding between oxygen and cobalt entities (O–Co–O) and the presence of + 3 oxidation state (Co(III)), whose reduction for Co(II) is required for the reaction.[45,46] However, the electron collector presented in the black mass after physical processing can be used as a reducing agent.[20,24,30,31,38,39,47,48] According to economic and environmental feasibility, the impact of H_2O_2 addition is higher than the use of Al foils as

Table I. Characterization of resulting material (black mass) after physically treating pouch and prismatic cells

Elements	Pouch black mass (%)	Prismatic black mass (%)
Ni	11.8	15.6
Co	3.6	3.7
Li	2.6	3.5
Mn	3.4	7.0
Al	6.9	5.0
Cu	10.7	8.2
C	32.1	38.0



Acid availability for cathode leaching (stoichiometric)

Concentration (mol/L)	Pouch black mass	Prismatic black mass
1.0	0.03%	-37.8%
1.5	33.4%	8.1%
2.0	50.0%	31.1%
2.5	60.0%	44.9%

Acid availability for Al foil leaching (stoichiometric)

Concentration (mol/L)	Pouch Al foil	Prismatic Al foil
1.0	-39.3%	-1.0%
1.5	7.1%	32.7%
2.0	30.3%	49.5%
2.5	44.3%	59.6%

Fig. 1. Comparison of leaching efficiency under the effect of (H_2SO_4) acid concentration for leaching of NMC622 and NMC631 black mass at 90°C for 90 min and solid-liquid ratio 1/5.

a reducing agent, representing up to 15% of the costs for the leaching reaction.[14,30]

The leaching of black mass (NMC622 and NMC631) reached 100% by H_2SO_4 solution. However, the main difference between the materials (black mass) is the amounts of metals (Table 1) impacting the process parameters. Leaching of NMC622 black mass reached $>$ 90% at 1.0 mol/L H_2SO_4 solution, while NMC622 black mass reached $>$ 60% under the same conditions (Fig. 1). Our findings demonstrated that the solid-liquid ratio should consider the amount of cathode in the black mass per acid for reaction (g of black mass per mL of leach solution, instead of the common solid-liquid ratio of 1/10 as 1 part of solid per 10 parts of solution).

Equations 1 and 2 depict the leaching reaction of cathode materials (NMC622 and NMC631, respectively), and Eqs. 3 and 4 show the reaction of Al foil and Cu foil by H_2SO_4 solution, respectively. The leaching of NMC-type cathode reaches 100% only with 30% of acid excess (Fig. 1). For example, for NMC622, at 1.0 mol/L, leaching efficiency achieved 90%, where the stoichiometric reaction demonstrates an excess of 0.03%, while at 1.5 mol/L, it achieved 100% with an excess of 33.4%. For NMC631, on the other hand, leaching efficiency was achieved with an acid concentration of 2.0 mol/L with 31.1% acid excess (Figs. S3 and S4b). Our conclusions will be determinant for the hydrometallurgical processing of Li-ion batteries of different NMC materials.

$$\begin{aligned} &LiNi_{0.8}Mn_{0.2}Co_{0.2}O_2 + 1.5H_2SO_4 \\ &\quad \rightleftharpoons 0.6Ni_2SO_4(aq) + 0.2\ CoSO_4(aq) \\ &\qquad + 0.2\ MnSO_4(aq) + 0.5Li_2SO_4(aq) + 1.5H_2O(l) \\ &\qquad + 0.25O_2(g) \end{aligned} \tag{1}$$

$$\begin{aligned}&LiNi_{0.8}Mn_{0.3}Co_{0.1}O_2 + 1.5H_2SO_4\\&\rightleftharpoons 0.6Ni_2SO_4(aq) + 0.1\ CoSO_4(aq)\\&\quad + 0.3\ MnSO_4(aq) + 0.5Li_2SO_4(aq) + 1.5H_2O(l)\\&\quad + 0.25O_2(g)\end{aligned} \tag{2}$$

$$2Al_{(s)} + 3H_2SO_{4(aq)} \rightarrow Al_2(SO_4)_{3(aq)} + 3H_{2(g)} \tag{3}$$

$$Cu_{(s)} + H_2SO_{4(aq)} \rightarrow CuSO_{4(aq)} + H_{2(g)} \tag{4}$$

The leaching reaches the plateau in 90 min.[30,38,39] The temperature has a linear effect on leaching, and higher efficiencies were observed for different reasons: (1) Li leaching is easier than Ni, Co, and Mn, being possible to be partially dissolved by water;[49] (2) electrolyte is a soluble source of Li that is not evaporated and then dissolves quickly in acid conditions.[41] At 90°C, the cathode leaching reaches 100% by H_2SO_4 acid concentration for 90 min and solid-liquid ratio 1/5. Kinetics and thermodynamics are not affected by acid concentration and solid-liquid ratio.[30,50]

Citric acid ($C_6H_8O_7$) was evaluated as a potential leaching agent to decrease the carbon footprint on recycling.[23,42,43] Although using H_2O_2 is also unnecessary as a reducing agent (Eqs. 3 and 4), the leaching reaction generates CO_2 (Eq. 5), which must be considered for future environmental impact analysis.[42,51] $C_6H_8O_7$ acid dissociation is lower than H_2SO_4 ($H_2SO_4 = 1.0 \times 10^2$; $HSO_4^{-1} = 1.0 \times 10^{-2}$, $C_6H_8O_7 = 7.4 \times 10^{-4}$, $C_6H_7O_7^{-1} = 1.7 \times 10^{-5}$)[46,52] although acid excess was based on a stoichiometric reaction. It results in lower leaching efficiencies, increasing the acid concentration (Fig. S5a) and solid-liquid ratio (Fig. S5b).

Organic complexes are formed in the leaching liquor (Eq. 5), which also reduces the need for reducing agents for leaching.[53] The leaching reaction also reaches equilibrium in 90 min (Fig. S5c). $C_6H_8O_7$ leaching is highly affected by the temperature increase (Fig. S5d), probably due to the dissolution of organic complexes formed and faster acid dissociation as the temperature increases.[46,54–56]

$$\begin{aligned}&18\ LiNi_{0.8}Mn_{0.3}Co_{0.1}O_2 + 19C_6H_8O_7\\&\rightleftharpoons 3.6Ni_3(C_2H_5O_7)_2(aq) + 0.6Co_3(C_2H_5O_7)_2(aq)\\&\quad + 1.8Mn_3(C_2H_5O_7)_2(aq) + 6Li_3C_2H_5O_7(aq)\\&\quad + 31H_2O(l) + 6CO_2(g)\end{aligned} \tag{5}$$

Inorganic acid costs (H_2SO_4) are lower than those for organic acid ($C_6H_8O_7$). $C_6H_8O_7$ represents 2.35-fold compared to H_2SO_4: 69.3 USD and 29.8 USD per kg of black mass, respectively (data from laballey.com, accessed in August 2024, for comparison among the acids). In addition, current processes have a lower environmental impact on H_2SO_4 production than inorganic acids. Compared to $C_6H_8O_7$, the global warming impact and respiratory effects can be 50-fold compared to H_2SO_4,[42] which would be changed if a bioprocess replaces the current technology for acid production[57–59] or if organic wastewater is used as a leaching agent.[22,53,60] Table II depicts the leaching efficiencies of metals present in the black mass and their concentration in the sulfuric solution (1.0 mol/L H_2SO_4 solution at 90°C for 90 min and solid-liquid ratio 1/5). This leaching liquor was used for solvent extraction experiments.

Solvent Extraction Separation

In this section, the focus was separation of the valuable metals (Li, Ni, Co, and Mn) from the leach liquor of NMC631-type batteries. Before solvent extraction experiments, Al and Cu were removed from the solution by precipitation using NaOH. Co-precipitation of valuable metals occurs and is impacted by the temperature. Losses of Ni can be reduced by increasing the temperature from 25°C (Fig. S6) to 80°C.[30] Stirring, time, and precipitant agent (e.g., carbonates) impact the co-precipitation of valuable metals, and their analysis is suggested for future studies.

Extractants such as D2EHPA and Cyanex 272 have been reported for the separation of metals in battery recycling. In NMC processing, D2EHPA has been reported as highly selective for Mn over Li, Ni, and Co.[27,61–63] Indeed, its selectivity was observed (Fig. 2 and Table III) as well as high co-extraction of other metals. Meshram et al. (2022) reported that Mn and Al can be extracted together. increasing the efficiency proportional to the pH (1–4), while extraction of Cu started at the same pH but with lower efficiency (90% for Al/Mn and 70% for Cu at pH 3.0); co-extraction, on the other hand, started at pH 2 (~1%) to pH 6 (50%).[61] In this study, D2EHPA reaches maximum Mn selectivity at pH 3.2 and 60°C and aqueous-organic ratio 1/1 (Mn/Li = 2.65; Mn/Co = 1.56; Mn/Li = 2.57), and the selectivity decreases as the pH increases. Furthermore, the selectivity increased from 1.8 to 2.6 (Mn/(Li, Ni or Co)) as the temperature increased, in accordance with previous reports.[27]

Table II. Leaching efficiency and concentration of metals from NMC631 black mass by 1.0 mol/L H_2SO_4 solution at 90°C for 90 min and solid-liquid ratio 1/5

Elements	Leaching (%)	Concentration (g/L)
Ni	96.9 ± 2.5	17.2
Co	97.7 ± 2.3	4.5
Li	98.4 ± 2.9	3.8
Mn	96.3 ± 1.5	7.1
Cu	43.1 ± 2.0	4.5
Al	60.3 ± 0.5	3.9

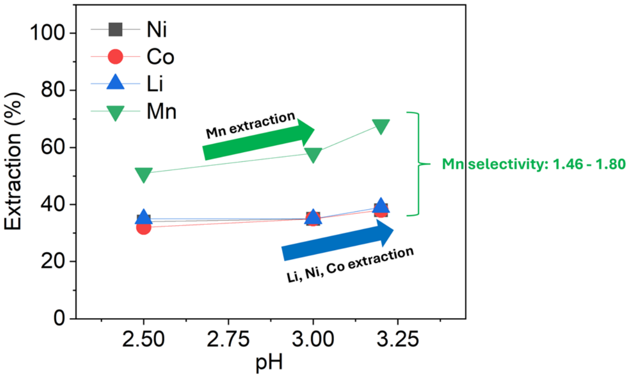


Fig. 2. Extraction percentage of Li, Ni, Co, and Mn varying the pH (2.5–3.5) by D2EHPA (10% v/v in kerosene) at 25°C and aqueous-organic ratio 1/1.

Table III. Extraction percentage of Li, Ni, Co, and Mn varying the temperature (25°C and 60°C) and aqueous-organic ratio (5% and 10%). Effect of temperature: D2EHPA (10% v/v in kerosene) at pH 3.2 and aqueous-organic ratio 1/1. Effect of aqueous-organic ratio: D2EHPA (kerosene) at pH 3.2, 60°C and aqueous-organic ratio 1/1

	Extraction (%)			
Elements	**25°C**	**60°C**	**5% v/v**	**10% v/v**
Ni	38	34	33	34
Co	38	58	37	58
Li	39	35	33	33
Mn	68	90	51	90

Solvent extraction separation requires several extraction and stripping steps in different reactors and tanks, resulting in several streams containing valuable metals being separated, and losses (mostly Co) are observed.[61–64] For this reason, past proposals for first Mn removal using selective precipitation (e.g., oxidation) are crucial to avoid Co losses in the process.[31]

Cyanex 272 extractant is widely reported for Ni/Co separation.[65] In this study, the extraction was more selective for Mn (Eq. 6), and the selectivity for Mn achieved 4.0 (Mn/(Li or Ni)) at pH 5.5, 60°C, and aqueous-organic ratio 1/3 (Table IV), while the Mn/Co ratio was 1.0 (Fig. 3a). Operational parameters (pH, aqueous-organic ratio, temperature, and concentration) increased Mn and Co selectivity (Mn > Co). The lowest co-extraction of Li and Ni was 20% (Fig. 3b). In the presence of Li, Ni, and Co (absence of Mn), Cyanex 272 demonstrated high selectivity for Co over Li and Ni.[30,61]

Table IV. Extraction percentage of Li, Ni, Co, and Mn varying the temperature (25°C and 60°C) and aqueous-organic ratio (5% and 10%). Effect of temperature: Cyanex 272 (10%v/v in kerosene) at pH 5.5 and aqueous-organic ratio 1/1. Effect of aqueous-organic ratio: Cyanex 272 (in kerosene) at pH 5.5, 60°C and aqueous-organic ratio 1/3

	Extraction (%)			
Elements	**25°C**	**60°C**	**5% v/v**	**10% v/v**
Ni	34	38	25	26
Co	49	66	53	96
Li	31	39	25	25
Mn	78	84	87	100

Extraction of Mn and Co reached similar values by Cyanex 272, while co-extraction of Ni and Li reached around 25%. The stripping in acidic conditions (Eq. 6) increases as the acid concentration and organic-aqueous ratio increase. Stripping by water and dilute acid solutions is usually reported as a washing step to remove contaminants from the organic phase.[63,66] Two steps of stripping are required for the extraction of Mn and Co with minor co-extraction of Ni and Li. Furthermore, the strong acid solution can be used for stripping of remaining Li and Ni from the organic phase of the mix with stream-rich after the extraction reaction (Fig. 3).[67]

$$(\text{Co or Mn})^{+2}{}_{(\text{aq})} + 2(\text{HX})_{2(\text{org})} \leftrightarrow (\text{Co or Mn})\text{X}_2 2\text{HX}_{(\text{org})} + 2\text{H}^{+}{}_{(\text{aq})} \qquad (6)$$

Precipitation

Two different solutions were obtained after solvent extraction separation: Li- and Ni-rich solutions after extraction (Fig. 3) and Co and Mn after stripping (Fig. 4). There is an opportunity for Mn precipitation under oxidation reaction (Fig. 5a, b), and NaOCl was tested in this study as ozone is already reported in the literature.[31,68] Commercial NaOCl solutions are available at 10-15% concentration, and the increase of solution:NaOCl ratio promoted the precipitation of Mn and Co proportionally (Fig. 5c). The Mn/Co ratio remained the same as the oxidant agent solution increased because of the effect on redox potential (Eq. 7). As the precipitation occurs, protons are released into the solution, impacting the pH and resulting in a more selective precipitation (Fig. 5a). Co and Ni precipitation is constant after 30 min, and Mn

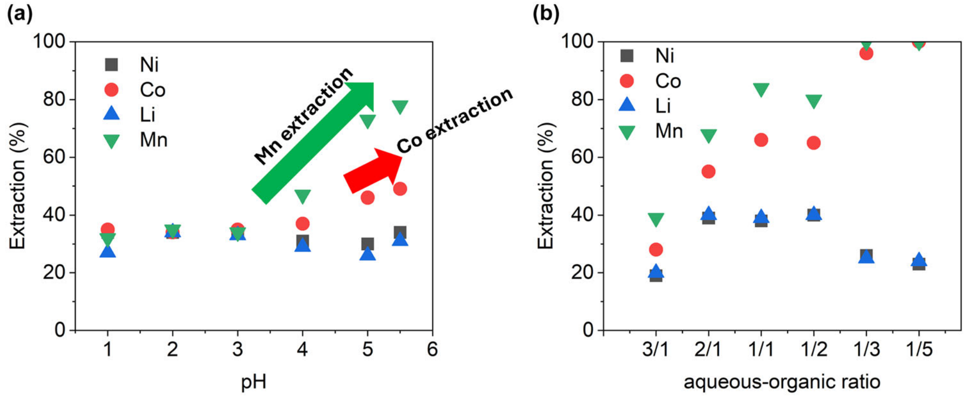


Fig. 3. Extraction percentage of Li, Ni, Co, and Mn varying the (a) pH (2.5–3.5) and (b) aqueous-organic ratio by Cyanex 272. Effect of pH: Cyanex 272 (10% v/v in kerosene) at 25°C and aqueous-organic ratio 1/1. Effect of aqueous-organic ratio: Cyanex 272 (10% v/v in kerosene) at pH 5.5 and 60°C.

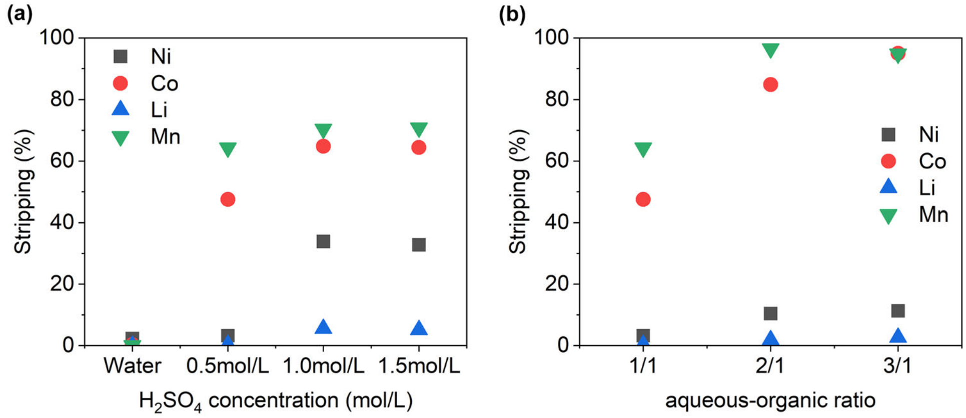


Fig. 4. Stripping percentage of Li, Ni, Co, and Mn varying (a) acid concentration and (b) aqueous-organic ratio after extraction by Cyanex 272 (10% v/v in kerosene) at pH 5.5, 60°C, and aqueous-organic ratio 1/3. Effect of acid concentration: at 25°C and aqueous-organic ratio 1/1. Effect of aqueous-organic ratio: 25°C and 0.5 mol/L H_2SO_4.

precipitation reaches the plateau after 240 min for solution:NaOCl ratio 2:1 (Mn/Co = 5) or 30 min at 1:1 (Mn/Co = 2.5). High solution:NaOCl ratio results in a high redox potential over the red highlighted area (Fig. 5a, b), resulting in more precipitation of Co. The solid obtained is Mn oxide (Fig. S7). Co can be further precipitated as carbonate, hydroxide, or oxalate, but its recovery efficiency is low and is further presented in the flowchart proposed.

$$\mathrm{Mn}^{+2}{}_{(aq)} + \mathrm{NaOCl}_{(l)} + \mathrm{H_2O}_{(l)} \rightarrow \mathrm{Mn}O_{2(s)} + \mathrm{NaCl}_{(aq)} + \mathrm{H}^{+}{}_{(aq)} \quad (7)$$

The efficient recovery of metals from cathode material is highly impacted by the presence of Mn. The selective removal of Mn using oxidant precipitation[31,68] or other new approaches to be developed is required unless NMC batteries are replaced in the market (e.g., by NCA or LFP).[4] For instance, in the NCA recycling process where Mn is not present in the cathode material, minor losses of valuable metals are identified in solvent extraction separation; if Mn is removed from the leach solution in NMC recycling, a great advantage is provided. Our previous study demonstrated that selective precipitation of Mn by ozone is not affected by the presence of Al or Cu ions.[31,68]

The Ni and Li solution stream was further treated by alkali precipitation. Ni precipitates at pH 8 and Li at pH 11.[69] Ni precipitation by NaOH increases over time until 420 min, while Li remained in solution (Fig. 6a). Hydroxides form strong bonds with metallic ions in solution and also trap part of the solution, causing co-precipitation.[70–75] The solid phase was washed with ultrapure water, reducing losses of Li from 30 to 6%. The calcinated solid is composed of Ni oxide (Fig. 6b).

Li was precipitated by carbonate (Na_2CO_3) at pH 11 and 100°C. However, using NaOH and Na_2CO_3 led to many Na ions in the solution (Fig. 6c), resulting in its co-precipitation along with Li. The

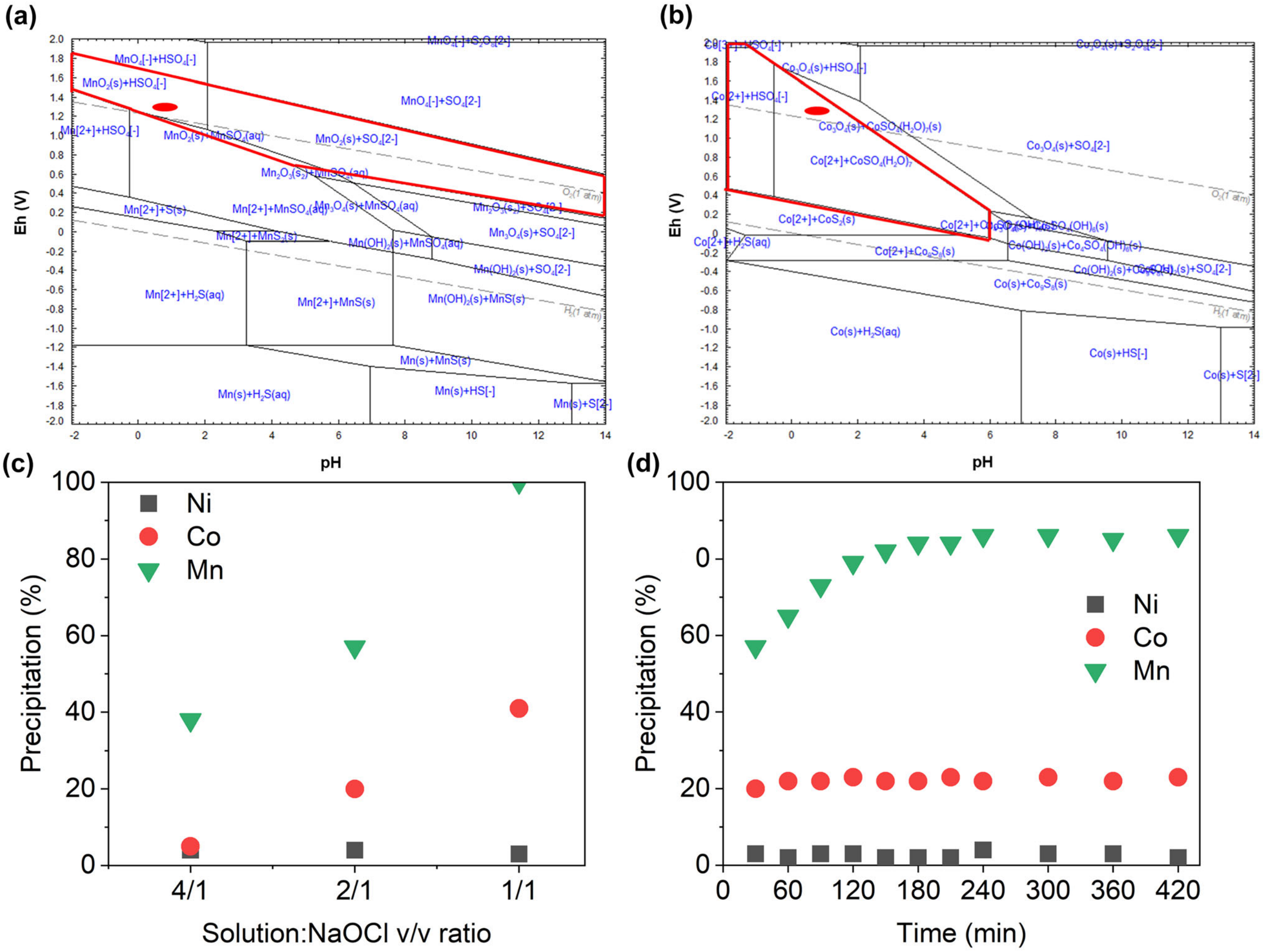


Fig. 5. Pourbaix diagram for (a) Mn and (b) Co at 25°C in the same concentration as the solutions and highlighted in red the area for Mn precipitation. Precipitation percentage of Ni, Co, and Mn at 25°C and pH 0.5 (c) varying the solution:NaOCl ratio for 30 min and (d) over time with a solution:NaOCl ratio 2:1.

solid phase was washed after filtration, and Na ions were removed, increasing Li carbonate purity to up to 99.9% (Fig. 6d).

Analysis of the Flowchart for NMC Battery Recycling Using Solvent Extraction

The main concern of NMC recycling using solvent extraction is the Co losses through the process because of Mn (Table S6). The flowchart proposed is depicted in Fig. 7 with a mass balance considering 100 kg of NMC cells (Tables V and VI). Co content in the leach solution in stream 5 (2.9 kg) is further obtained in stream 15 (1.7 kg, 58.6% recovery). Co losses in Al and Cu precipitation represent 13.7%, which can be decreased to 8% if the reaction is performed at 80°C instead of room temperature (25°C).[30] However, precipitation by NaOCl resulted in high losses of Co (20.7%), which would be avoided if ozone were used for Mn removal.[31,68,76,77] More steps for solvent extraction will increase losses of Co (Fig. 3). As observed, the main drawback for Co recovery is the solvent extraction steps where Co ions are lost in extraction/stripping stages, as clearly shown in the flowchart process (Fig. 7).

Mn recovery was not affected by solvent extraction separation (77%), where the main losses were in Al/Cu precipitation (5.6%) and Mn precipitation (13.0%). Ni losses were observed in solvent extraction separation steps reaching low recovery efficiency (62.8%). At the solvent extraction step, Ni losses were 25.5% (streams 7-9), and it could be avoided by recirculating the aqueous phase into the

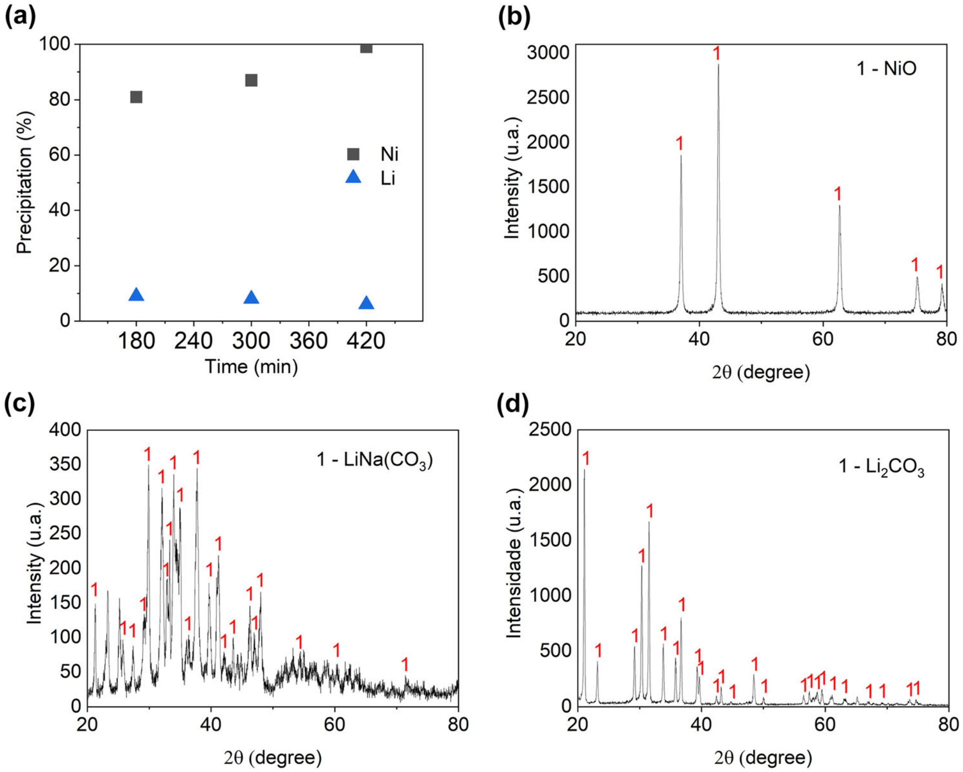


Fig. 6. (a) Precipitation efficiency of Ni and Li over time at pH 8.0 and 25°C and (b) XRD of the precipitate after reaction with NaOH; (c) XRD of the precipitation by Na_2CO_3 at pH 11.0 and 100°C for 40 min; solid phase after water with ultrapure water at pH 7.0 and 70°C.

process (stream 9, before Ni precipitation). Li losses over separation steps were also highly affected (48%).

Optimizations in lab and pilot process parameters will not have a greater impact on increasing the recovery efficiencies.[78–81] The absence of Mn will increase the metal recovery to 80-85% for Ni and Co and 92% for Li.[30] From our results, it is clear that solvent extraction is not feasible under the presence of Mn. Beyond that, sustainability through NMC battery recycling will be achieved if material recovery is $>$ 90% in an economic way.

CONCLUSION

The present study aimed at the process proposed for NMC battery recycling by hydrometallurgy. No difference was observed in physical processing for different cell structures (prismatic and pouch). About 30% of acid excess is needed for complete cathode leaching. Al foil was used as a reducing agent in the reaction, avoiding the addition of H_2O_2, reducing environmental impact and economic costs. The presence of Mn reduces the recovery rates of Li, Ni, and Co because of co-extraction in solvent extraction steps, as our flowchart has demonstrated. Removal of Mn from the leach solution before solvent extraction is the best option for recycling NMC batteries. Based on our previous studies, we propose using ozone for selective Mn precipitation, allowing the recovery of Co. Future markets will demonstrate more interest in Fe-rich batteries; however, NMC batteries will represent a great share until 2050 at least. Therefore, our conclusions are crucial for the best decision-making in battery recycling, working toward a circular economy.

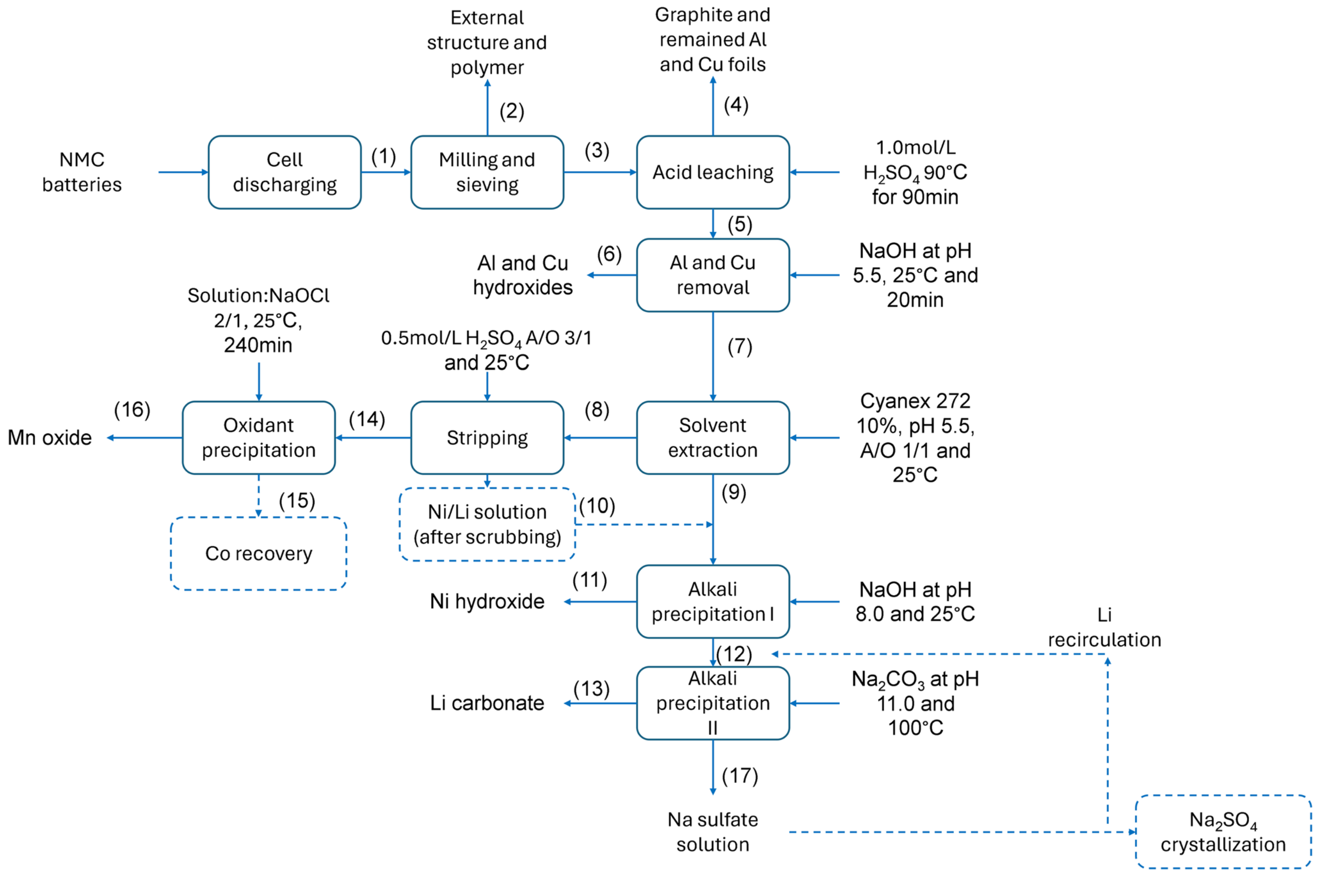


Fig. 7. Flowchart proposed for NMC recycling using acid leaching followed by solvent extraction and precipitation steps (100 kg of feed cell batteries). Dashed lines indicate potential possibilities that were beyond the scope of this study.

Table V. Elementary composition of the streams (1-8) of the flowchart proposed for recycling of NMC batteries (Fig. 7)

Elements	Stream 1 (kg)	Stream 2 (kg)	Stream 3 (kg)	Stream 4 (kg)	Stream 5 (kg)	Stream 6 (kg)	Stream 7 (kg)	Stream 8 (kg)
Ni	13.5	1.4	12.1	0.0	12.1	1.9	10.2	2.6
Li	3.1	0.4	2.7	0.0	2.7	0.1	2.6	0.7
Mn	7.2	1.8	5.4	0.0	5.4	0.3	5.1	5.1
Co	3.5	0.6	2.9	0.0	2.9	0.4	2.5	2.4
Al	3.6	0.0	3.6	1.4	2.2	2.2	0.0	0.0
Cu	7.8	1.4	6.4	3.6	2.8	2.8	0.0	0.0

Table VI. Elementary composition of the streams (9-17) of the flowchart proposed for recycling of NMC batteries (Fig. 7)

Elements	Stream 9 (kg)	Stream 10 (kg)	Stream 11 (kg)	Stream 12 (kg)	Stream 13 (kg)	Stream 14 (kg)	Stream 15 (kg)	Stream 16 (kg)	Stream 17 (kg)
Ni	7.6	2.3	7.6	0.0	0.0	0.3	0.3	0.0	0.0
Li	1.9	0.7	0.1	1.8	1.3	0.0	0.0	0.0	0.5
Mn	0.0	0.2	0.0	0.0	0.0	4.9	0.7	4.2	0.0
Co	0.1	0.1	0.1	0.0	0.0	2.3	1.7	0.6	0.0
Al	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cu	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

SUPPLEMENTARY INFORMATION

The online version contains supplementary material available at https://doi.org/10.1007/s11837-025-07677-5.

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CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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