

Research article

Towards a circular economy in lithium ion battery recycling by integrating microbial processes with electrowinning and precipitation for sustainable metal recovery

Mahsa Baniyadi^{a,b,*}, Kumar Upvan^{a,c}, Fatemeh Pourhossein^a, John E. Graves^c, Egle Latvyte^c, Sebastien Farnaud^{a,**}

^a Research Centre for Health & Life Sciences, Coventry University, Coventry, UK

^b Faculty of Engineering and Applied Sciences, Cranfield University, UK

^c Research Centre for Manufacturing and Materials, Coventry University, Coventry, UK

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ABSTRACT

With increased use of Lithium-Ion Batteries (LIBs) and the scarcity of some of their components, their recycling and the recovery of their metals have become essential. In this work, an indirect bioleaching process was designed to solubilise metals from LIB black mass using biogenic acid generated in a stirred tank bioreactor. The biogenic acid was used in addition to H₂O₂ as a reductant for improved solubilisation, and influential factors including pulp density, temperature, and concentration of H₂O₂ were optimised. The best results were achieved at 55 °C, with a pulp density of 7.5% (w/v) and 0.5% (v/v) H₂O₂, which resulted in 82% Li, 32% Ni, 24% Co and 21% Mn solubilisation in 5 min of the process. However, over time transition metals in the leachate did not remain in solution, due to their adsorption onto the carbon content of the black mass. To selectively recover solubilised Co, Ni, Mn, and Li from the leachate, a combined process of electrowinning and precipitation was applied to the leachate, leading to the successful electroplating of Co, Ni and Mn with 100%, 100% and 97.2% of solubilised metals respectively, while 40% of the Li was recovered by precipitation following the addition of sodium carbonate. These results constitute a promising step toward closing the loop for the sustainable selective recovery of critical metals used in LIB manufacturing and suggest the next targets to improved bioleaching efficiency.

1. Introduction

In recent years, the use of portable electronic devices, such as mobile phones, laptops, and cameras powered by lithium-ion batteries (LIBs) has increased sharply (Miao et al., 2022). The demand for LIBs has been further increased by the growing popularity of electric vehicles, which rely on these batteries to generate and store energy, marking a shift away from traditional fossil fuels. Several metals used in these batteries are classified as critical materials so that sustainable recycling of these high value metals, such as lithium (Li), cobalt (Co) and nickel (Ni), is essential to reduce the supply risk in the future (Miao et al., 2022; Roy et al., 2021). In addition to environmental concerns and carbon footprint of their mining extraction, the supply risk of these metals also includes geopolitical risks of raw material resources (Woeste et al., 2024).

Conventional methods for metal recovery from electronic waste, such as pyrometallurgy and hydrometallurgy, often pose sustainability challenges. In pyrometallurgical processes for Li recovery, a portion of Li is typically lost in the slag (Alipanah et al., 2023; Roy et al., 2021). Although hydrometallurgy is currently the most widely adopted method for commercial metal recovery from LIBs, it relies heavily on the use of strong chemical acids for leaching, followed by the purification and extraction of the metals from the leachate. This approach generates acid waste and large volumes of contaminated wastewater, leading to significant environmental management challenges (Moazzam et al., 2021). These challenges, including the production of secondary waste and high operational costs, highlight the urgent need for more efficient, eco-friendly, and sustainable recovery processes.

Bioleaching, which relies on the solubilisation of metals with the

* Corresponding author. Research Centre for Health & Life Sciences, Coventry University, Coventry, UK.

** Corresponding author.

E-mail address: mahsa.baniyadi@cranfield.ac.uk (M. Baniyadi).

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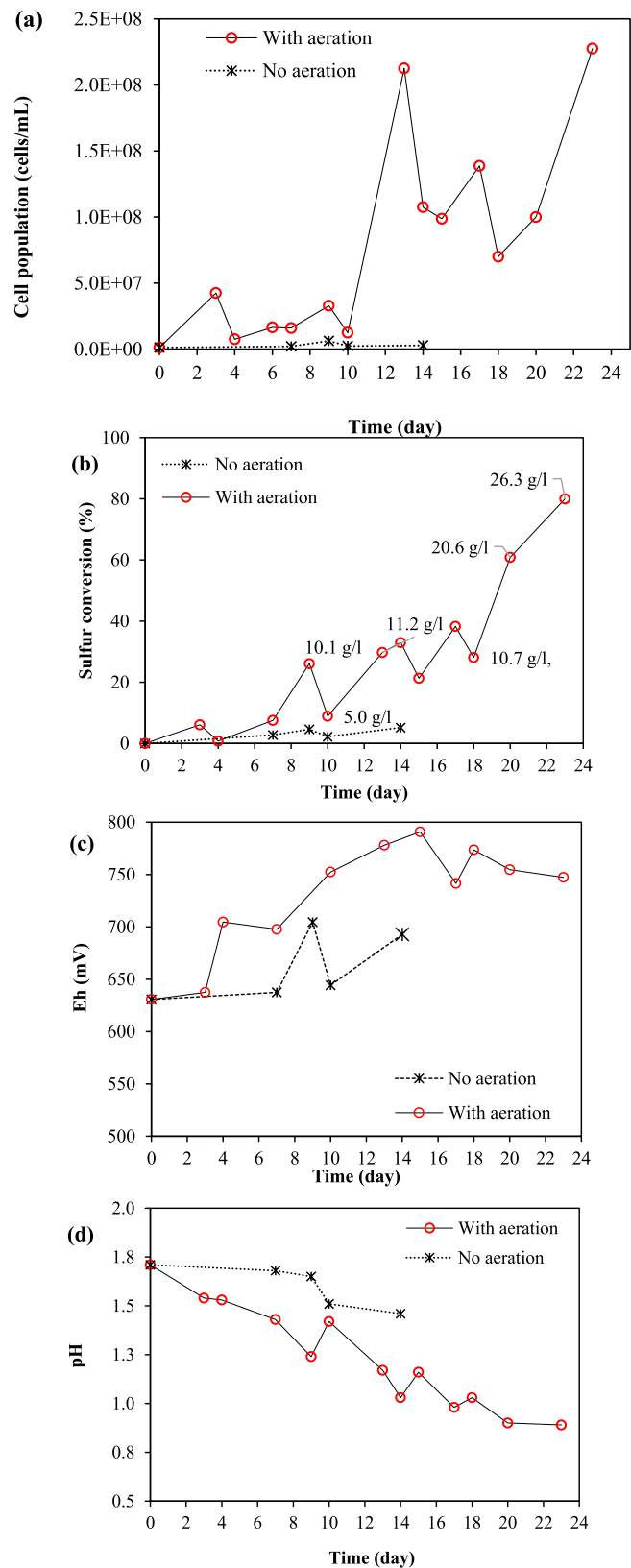


Fig. 1. Growth and activity of *A. thiooxidans* in stirred tank bioreactor a) cell count; b) sulfur conversion (number labels are sulfur concentration); c) Eh and d) pH.

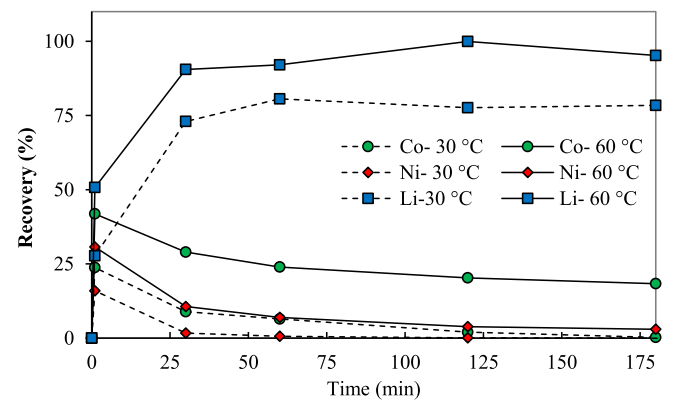


Fig. 2. Impact of temperature on metal dissolution at pulp density of 6% (w/v) and 4% (v/v) H_2O_2 .

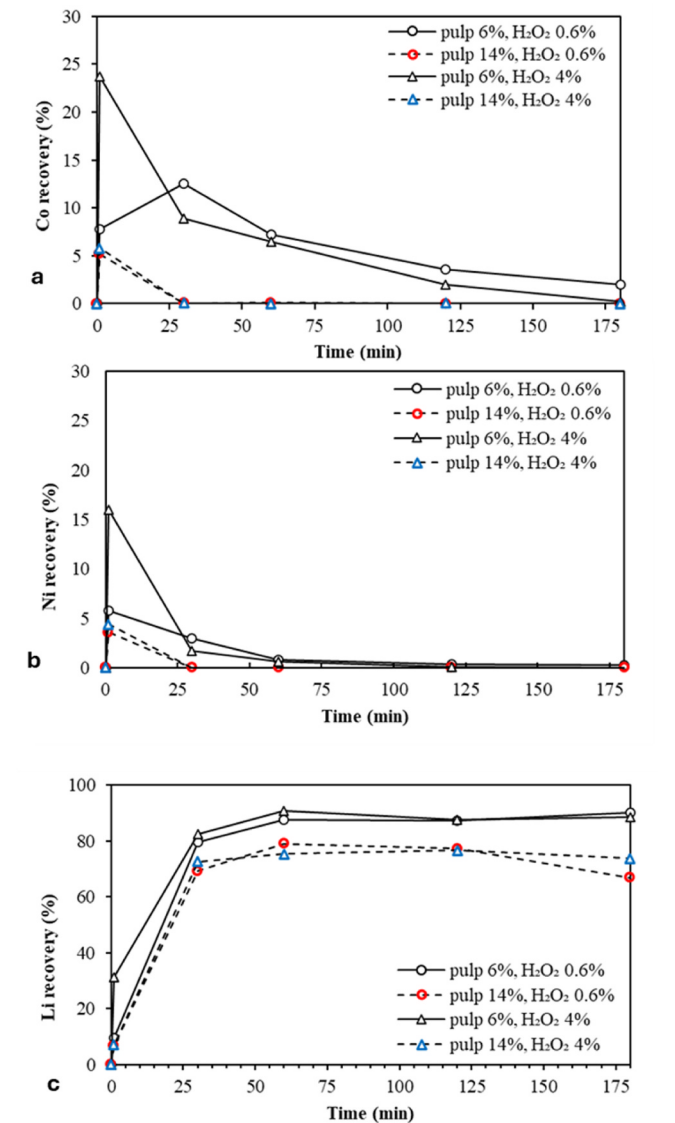


Fig. 3. Impact of pulp density and H_2O_2 concentration on metal recovery a) Co; b) Ni; and c) Li at 45 °C.

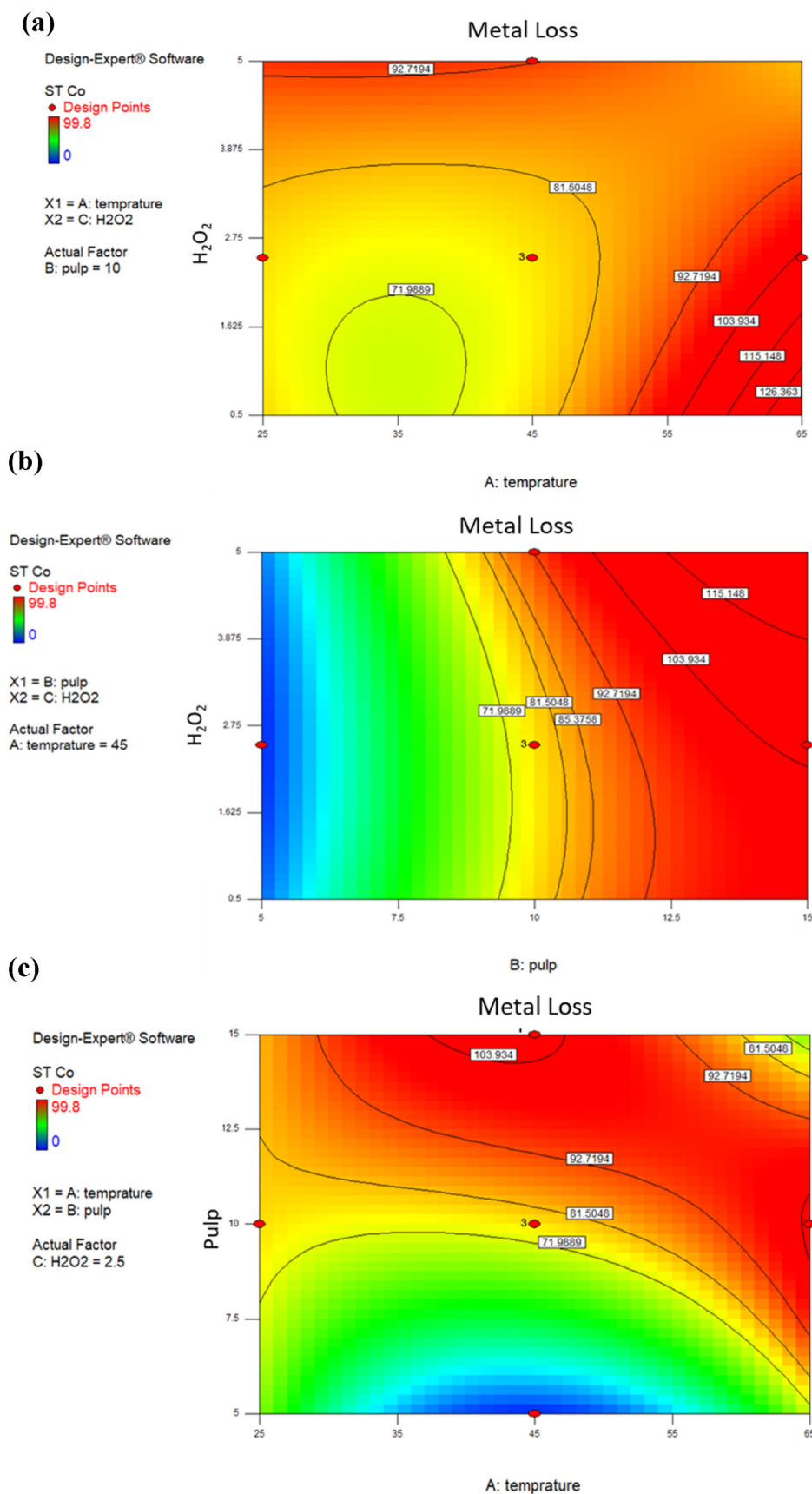


Fig. 4. Impact of influential parameters on metal loss a) temperature and H₂O₂ at pulp density 10% (w/v); b) pulp and H₂O₂ at temperature 45 ° C; c) temperature and pulp density at 2.5% (v/v) H₂O₂.

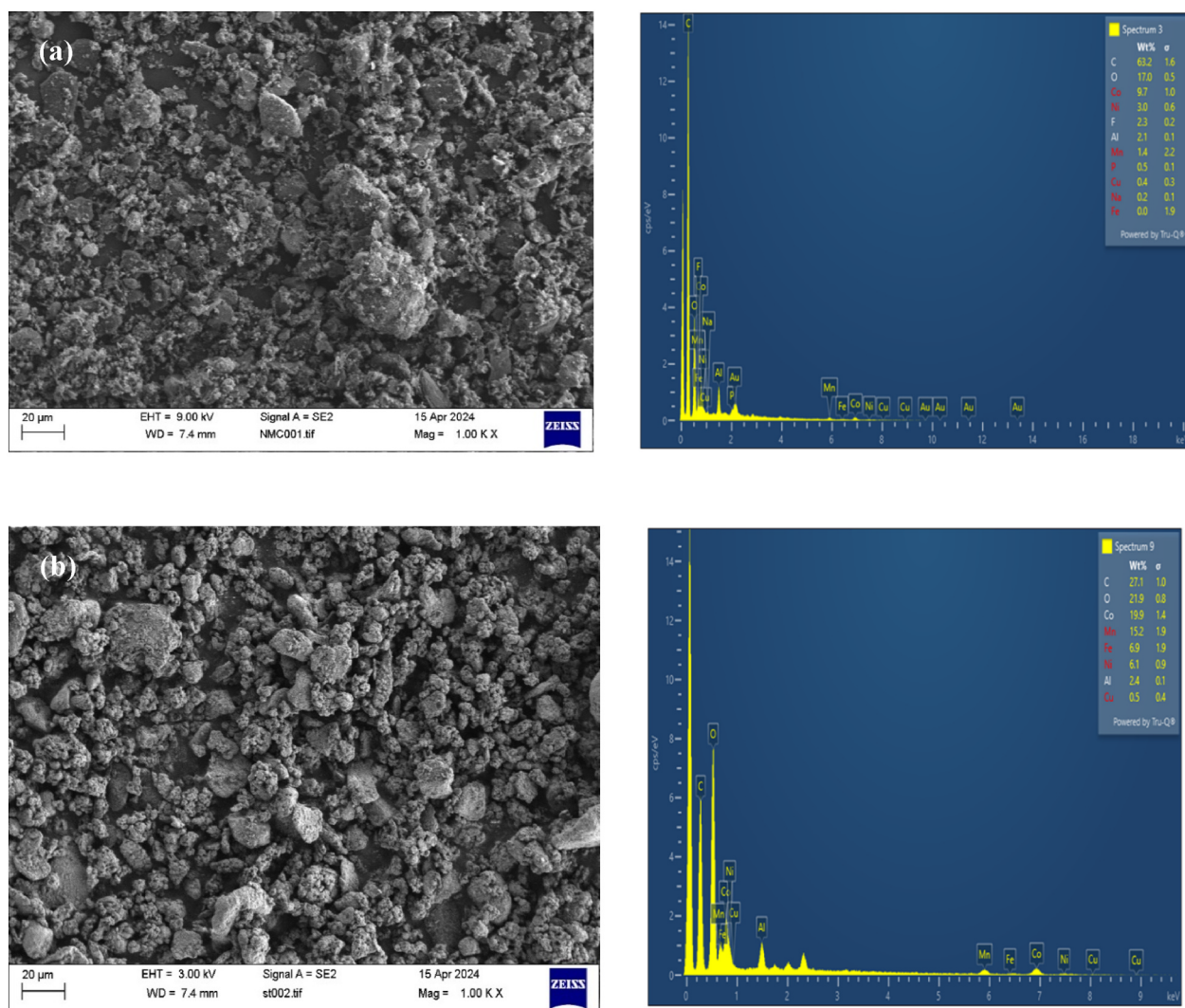


Fig. 5. Surface morphology and EDS analysis of black mass a) before and b) after indirect bioleaching at optimum conditions.

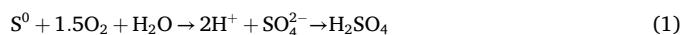
help of microorganisms, initially gained attention for the extraction of ores in the mining industry, and more recently for the recovery of metals from electronic waste in urban mining (Baniasadi et al., 2019). Several advantages can be found in using bioleaching, also called biohydrometallurgy, including simplicity of operation, lower emission of toxic gases and lower energy requirement. However, challenges such as incomplete metal recovery, slow kinetics and instability in the process efficiency have been mentioned in the literature and remain a current focus for researchers (Roy et al., 2021; Moazzam et al., 2021).

Metal extraction from ternary NMC (nickel-manganese-cobalt oxide) lithium-ion batteries, symbolised as $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$, is a challenge due to the difficulty of dissolution of high valent metals. However, this can be addressed by the addition of oxidising or reducing agents to the acidic leaching agent (Contestabile et al., 2002). In hydrometallurgy, and biohydrometallurgy using biogenic acid, protons react with oxygen in the layered structure of the batteries that contains the metals (Ni, Co, Mn) (Ning et al., 2024). Addition of different oxidising or reducing agents such as NH_4Cl , formic acid, and HCl has been investigated by other researchers (Gao et al., 2018; Lv et al., 2020; Contestabile et al., 2002; Olubambi et al., 2006) and has been reported to be an effective agent to improve the process kinetics in acidic leaching, while using less acid (Mukai et al., 2024; Viecei et al., 2023; Zhao et al., 2023).

One of the drawbacks of the direct bioleaching process, where bacterial cells are in direct contact with the metals, is the potential inhibitory effect of electronic waste on bacterial growth and activity. For LIBs,

their alkaline nature inhibits the growth of the bacteria, which are acidophilic in conventional bioleaching applications. The application of indirect bioleaching, which eliminates direct contact between bacteria and metals, has shown greater promise compared to direct bioleaching. Another positive aspect of indirect bioleaching processes is the reuse of the generated acid for bacterial growth, thereby creating a closed-loop system. This approach enables operation across a broader range of pH, temperatures, and pulp densities without adversely affecting bacterial activity (Heydarian et al., 2018). A variety of bacterial species and fungi have been reported to be effective in producing reagents suitable for metal dissolution from solid waste (Heydarian et al., 2018; Bahaloo-Horeh et al., 2018). Chemolithotrophic acidophiles are the most studied bacteria for research and applications in bioleaching (Baniasadi et al., 2019; Roy et al., 2021; Heydarian et al., 2018), with the most commonly used acidophilic species being the mesophilic *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans* with optimal growth at around 30 °C.

Chemolithotrophic sulfur oxidising bacteria grow by oxidising elemental sulfur to sulfuric acid (reaction (1)) (Baniasadi et al., 2019) through the biochemical pathway that is shown in reaction (2) (Pourhossein and Mousavi, 2022):



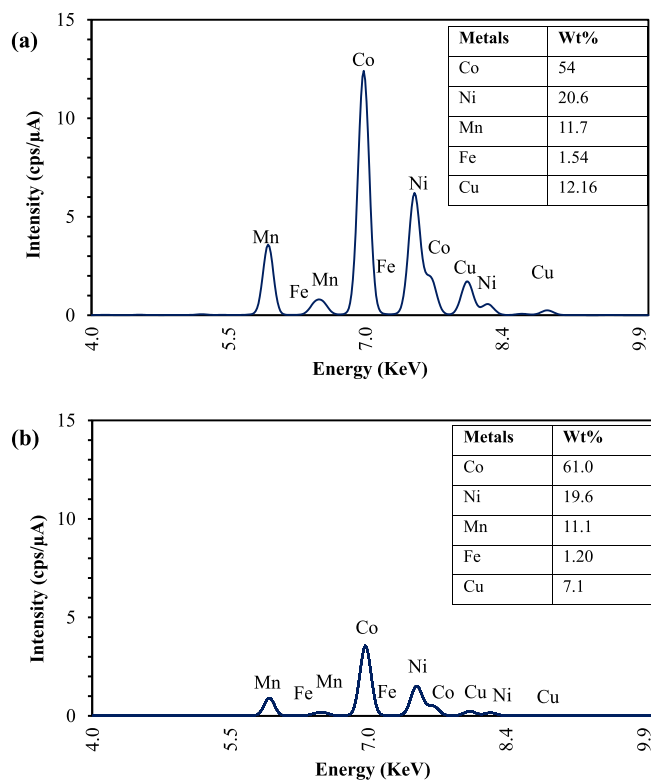


Fig. 6. XRF analysis of black mass (a) before bioleaching and (b) after bioleaching.

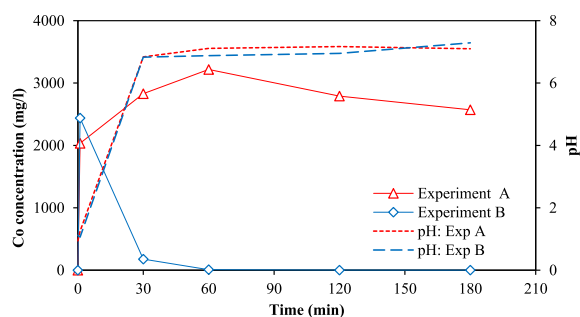
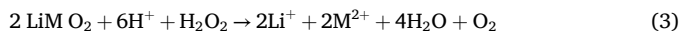


Fig. 7. Metal loss and pH change vs. time.

The addition of H_2O_2 as a reducing agent has been reported to improve the kinetics of metal dissolution in acid leaching of mineral ores (Olubambi et al., 2006). In the study by Mukai et al. (2024) benefits of acid leaching of NMC black mass in the presence of H_2O_2 have been demonstrated. They investigated the dissolution mechanisms of NMC in HNO_3 , with and without H_2O_2 . Without H_2O_2 , delithiated NMC remained after 24 h, whereas with H_2O_2 , complete dissolution occurred within 3 h. A typical reaction during acid leaching in the presence of H_2O_2 is described as reaction (3) where M represents Ni, Co and Mn (Mukai et al., 2024):



In this work, indirect bioleaching was applied, where biogenic sulfuric acid is produced using *A. thiooxidans* in a bioreactor. This process typically utilizes elemental sulfur or sulfide minerals, which can originate from waste streams or as by-products of other processes. Although the comprehensive comparison between bioleaching and using commercial sulfuric acid would require a detailed Life Cycle Assessment (LCA), biological route can offer lower carbon footprint and low-energy alternative than conventional industrial production of sulfuric acid,

which relies on fossil fuel-derived SO_2 and involves high-temperature oxidation (Kremser et al., 2022). Additionally, bio-production can occur on-site, minimising transportation and handling hazards associated with concentrated chemical acids use. While requirements for bacterial growth need to be considered, these requirements are often preferable compared to the large-scale chemical acid production's environmental and safety costs. Moreover, bacterial cultures can be maintained continuously in bioreactors, providing a sustainable source of acid over long periods, when system works in continuous closed loop, same as the system suggested in the work of Baniasadi et al. (2021). The biogenic acid was used in combination with H_2O_2 to bioleach NMC black mass within a specific range of pulp density, temperature and reductant (H_2O_2) concentration. Following metal recovery, the biogenic acid was reused as part of a closed-loop system. Experimental design included RSM (Response Surface Methodology) optimisation procedures for the settings of factorial variables, to minimise the number of tests and optimise the conditions. The results were used to mathematically optimise the process with the best operational conditions to achieve maximum metal recovery and minimum metal loss for high pulp densities.

In this study, in addition to the application of bioleaching as a greener approach than conventional chemical acid leaching, selective recovery of Co, Ni, Mn and Li from bioleachate is also addressed. In the work of Bertuol et al. (2016) electrowinning was applied to the leachate obtained from supercritical CO_2 leaching of lithium cobalt oxide material, in the absence of Ni, and the product analysis showed +99% Co purity. In that work electrowinning was performed in a two-compartment cell separated by membrane, to avoid anodic and cathodic reactions and avoiding sharp pH changes.

The combination of sulfuric acid and H_2O_2 has been used previously for dissolution of metals from NMC (Fan et al., 2021; Roshanfar et al., 2024). Investigation of acid leaching and H_2O_2 for metal recovery from LFP black mass of LIBs was studied by Zhao et al. (2023). However, to our knowledge, the use of H_2O_2 in presence of high pulp density of NMC black mass, as in the present study, has not been investigated, and the process bottlenecks are yet unknown.

While bioleaching has been extensively explored by our group and others as a green method for dissolving metals from solid waste, the development of a closed-loop system with selective separation of valuable metals from solution remains largely underexplored. This gap hinders the efficient reintegration of recovered metals into industrial processes, limiting the advancement of a true circular economy. In this study, this challenge will be addressed by integrating complementary techniques such as electrowinning and precipitation, with the bioleaching of metals from NMC, highlighting their combined potential to enhance the sustainability and efficiency of metal recovery processes.

2. Materials and methods

2.1. Black mass as electronic waste

The material used in this work was black mass from nickel-manganese-cobalt oxide (NMC) LIBs, which had been provided by Saubermacher Dienstleistungs AG, Austria, containing Al: 52.6, Co: 145.0, Li: 27.6, Mn: 41.7, Ni: 58.6 g/kg. The material was supplied in form of black mass and was reported to contain 31.2% total carbon and 22.3% bound carbon in the total solid.

2.2. Production of biogenic sulfuric acid

Biogenic sulfuric acid was produced in stirred-tank bioreactor inoculated with 2% freshly grown *Acidithiobacillus thiooxidans* (*A. thiooxidans*), which was obtained from the Acidophile Laboratory Bangor University, Wales, UK. The bioreactor was a 2.3 L vessel equipped with a temperature, pH and oxygen flow controlling system (Fermac 200 system; Electrolab, UK). The vessel and bioreactor

Table 1Analysis of variance (ANOVA) for influential factors (temperature, pulp density and H₂O₂) on metal recovery and precipitation.

Source of variation	SS	df	MS	F value	P-value	Significant	Model
ANOVA for Li recovery							
Model	1641.7	9	182.4	6.2	0.0125	Yes	Quadratic
A-temperature	351.5	1	351.5	12.0	0.0106		
B-pulp	996.2	1	996.2	33.9	0.0006		
C-H ₂ O ₂	17.1	1	17.1	0.6	0.4709		
ANOVA for Ni recovery							
Model	757.8	6	126.3	13.0	0.0003	Yes	2 Factor Interaction
A-temperature	125.9	1	125.9	13.0	0.0048		
B-pulp	374.0	1	374.0	38.6	<0.0001		
C-H ₂ O ₂	122.7	1	122.7	12.7	0.0052		
ANOVA for Co recovery							
Model	1916.7	9	213.0	25.0	0.0002	Yes	Quadratic
A-temperature	188.0	1	188.0	22.0	0.0022		
B-pulp	1120.4	1	1120.4	131.3	<0.0001		
C-H ₂ O ₂	155.0	1	155.0	18.2	0.0037		
ANOVA for metal loss							
Model	12,127.3	13	932.9	9.6	0.0438	Yes	Cubic
A-temperature	380.9	1	380.9	3.9	0.1421		
B-pulp	4980.0	1	4980.0	51.2	0.0056		
C-H ₂ O ₂	78.1	1	78.1	0.8	0.4360		

Table 2

Optimisation target and response values.

Parameters	Target	Value
Temperature (°C)	In range	55
Pulp% (w/v)	Max	7.5
H ₂ O ₂ % (v/v)	In range	4.4
Li recovery (%)	Max	82
Ni recovery (%)	Max	32
Co recovery (%)	Max	24
Precipitation (%)	Min	54

compartments were sterilised at 120 °C for 20 min in autoclave and then basal salt-trace media (Wakeman et al., 2008) at pH 2 was added together with 20 g of elemental sulfur. The system was aerated using a compressor with a rate of 1 l/min through flowmeter and air filter. The temperature was set to optimal temperature for *A. thiooxidans* as a mesophilic bacterium at 30 °C, and pH variations were constantly monitored. Stirring rate was fixed at 100 rpm during the process. Daily sampling and analysis were performed and oxidation reduction potential (ORP), concentration of sulfate ions and cell population were measured. After reaching stationary phase and satisfactory conditions including suitable pH, the bacterial cells were excluded were excluded from the solution by centrifugation.

2.3. RSM-based indirect bioleaching optimisation

To achieve maximum recovery of metals, three factors, including pulp density (5–15% w/v), temperature (30–60 °C) and a reductant (H₂O₂) concentration (0–5% v/v) were optimised by Central Composite Design-Response Surface Methodology (CCD-RSM). According to CCD, the total number of experiments was calculated as shown in Eq. (1).

$$\text{Number of Experiment} = 2^k + 2K + n_0 \quad (1a)$$

Where 2k is the factorial run, 2^K is the axial run, n₀ is the number of central point replications, and K depicts the number of independent variables.

In this study, pulp density, temperature and H₂O₂ concentration were considered as independent variables (factors), with Li, Co, and Ni leaching, as well as Co loss considered as response variables. The 18 experimental trials were conducted in shaker incubator at 180 rpm initially for 2 h as outlined by Design-Expert Software Version 7. Data collected were analysed to model the interactions and relationships between factors. The significance of the interactions between variables and responses was assessed. Model quality and statistical significance

were represented by the coefficient of determination (R²) and F-test, as calculated in the same software.

The results were used for optimisation to target maximising metal recoveries, minimising metal loss and working at the highest pulp density, all of which are important parameters in industrial applications. Metal loss was quantified by considering the percentage of Co concentration reduced in the solution after 1 h, in comparison to the maximum Co concentration of the solution during the process, and used for mathematical modelling. Since the dissolution trends of Co and Mn are comparable, Mn recovery was not considered for optimisation purposes. Due to the similar behaviour at optimum condition, where Co dissolution is maximised, and metal loss is minimised, Mn recovery is also at the optimum value.

Confirmation tests were carried out under statistically determined optimal conditions to validate the model against experimental results. Additional chemical acid leaching experiment, using chemical sulfuric acid with identical concentration to the biogenic sulfuric acid, was also conducted, both with and without H₂O₂, to compare the results of chemical acid leaching with bioleaching.

2.4. Metal recovery from the bioleachate

Electrowinning was used to extract Co and Ni in their elemental forms. For this purpose, two different cathode materials were tested, which were (i) stainless steel, area 4 cm² and thickness 0.1 cm (316 sheet, RS component), and (ii) carbon felt, area 4 cm² and thickness 0.635 cm (99%; Thermo Scientific Alfa Aesar, UK). The anode, for both setups was a mixed metal oxide (IrO₂-Ta₂O₅) titanium mesh (De Nora, Italy) with same dimension as the cathode (4 cm²).

Initially, a mimic electrolyte solution was used to optimise the electrowinning conditions. The synthetic electrolyte solution consisted of a basal salt-trace media solution, into which sulfate salts of Co, Mn, and Ni were added in the same ratio as in the optimum bioleachate, but at a concentration ten times higher.

The pH was adjusted to 4 using 1M NaHCO₃ solution. The applied current density was 5 mA/cm². Agitation was applied (magnetic stirrer set at 200 rpm) to avoid mass transport limitations from convection processes by enhancing ion transport to the cathode. One side of the cathode was covered with plating tape. The anode and cathode were weighed before and after the process to calculate the deposited mass. The foil plated on stainless steel cathode was removed using a thin scraper. Current efficiency was calculated by dividing the actual mass by the theoretical mass of metals to be plated based on Faraday's Law (Eq. (2)):

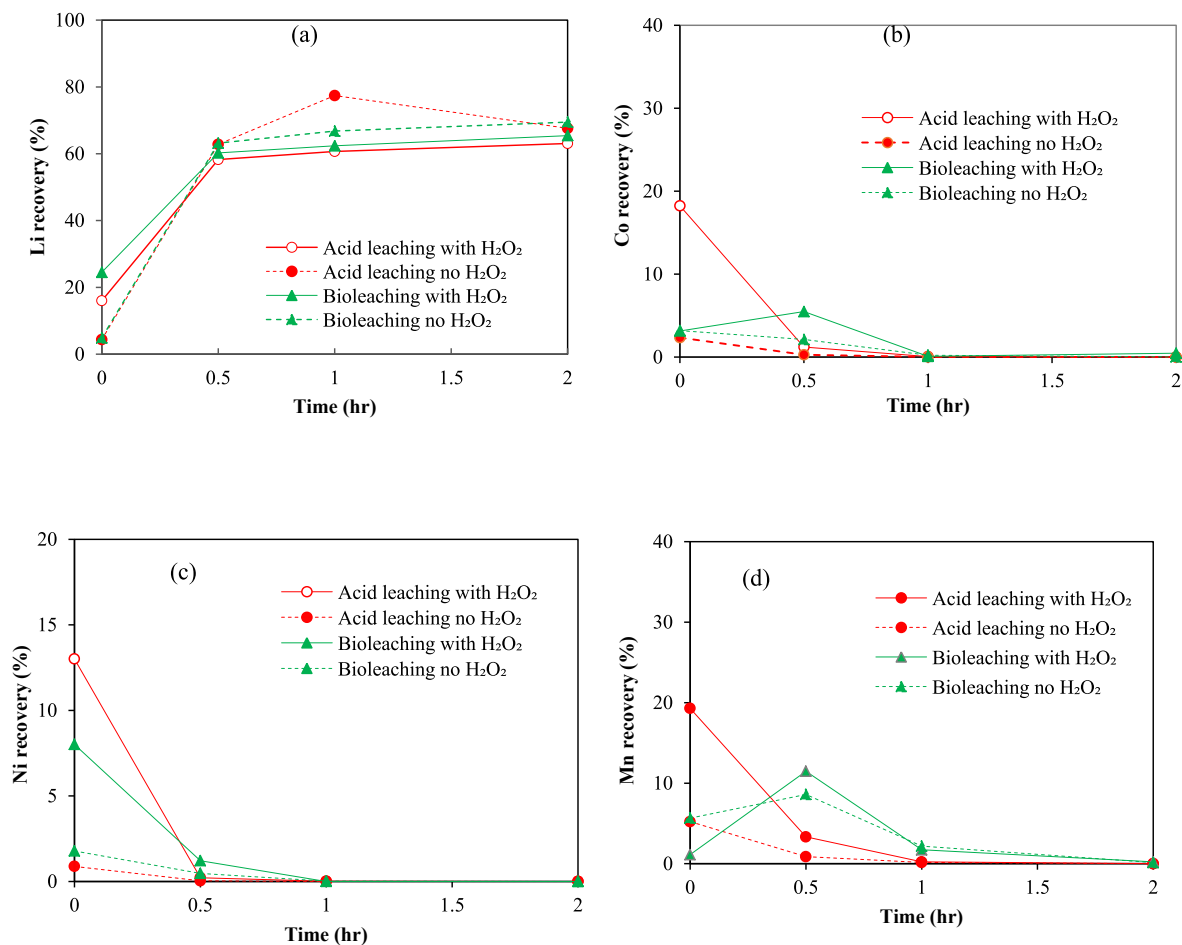


Fig. 8. Comparison of acid leaching and bioleaching in the presence and absence of reducing agent performed a) Li, b) Co, c) Ni and d) Mn at optimal conditions.

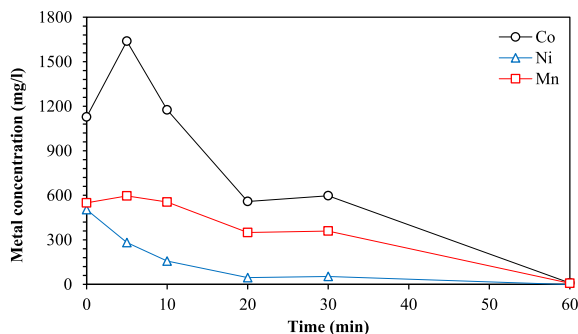


Fig. 9. Metal concentration in bioleaching solution over time.

$$w = ItM/zF$$

(2a)

In Eq. (2), w is the mass of metal plated, I is the set current (A), t is electro-winning time (s), M is the atomic weight (Ni: 58.69 u; Co: 58.93 u), F is the Faraday constant and z is the number of electrons.

The bioleaching experiment was performed with cathode surface of 10 cm^2 with the opposite surface taped, at a current density of 5 mA/cm^2 with the addition of 14 g/L boric acid to the bioleachate. Boric acid is present in the solution as a borate ion and non-ionized boric acid, which regulates the chemical reaction during the plating process. Additionally, boric acid improves the surface morphology of plating and increases the current efficiency (Lu et al., 2010). The setup was operated at room temperature. The products (Ni, Co and Mn) obtained in the form of foil or powder, were analysed with XRF (NEX DE, Rigaku, UK). Following

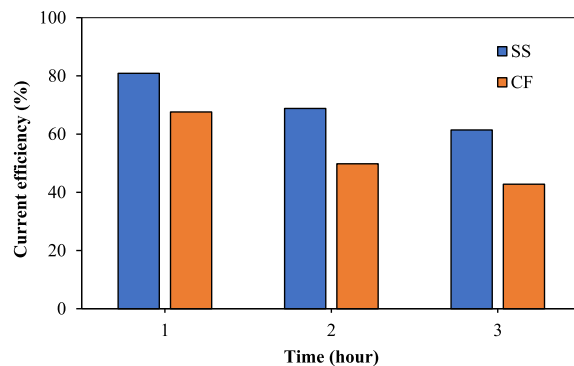


Fig. 10. A plot of current efficiency vs. time at 5 mA/cm^2 for stainless steel and Carbon Felt cathodes from a synthetic solution of Co, Mn and Ni.

electrowinning, precipitation experiment was performed using a saturated solution of Na_2CO_3 (Thermo fisher), where the solution was heated to 90°C at a constant stirring of 120 rpm on a magnetic stirring. Na_2CO_3 was added to the solution till the pH of 10.5 was reached. Li obtained after dissolution in acid was analysed with Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES, Perkin Elmer Optima 8300).

2.5. Analytical methods

Cell growth and cell count were monitored by using a Thoma Chamber and phase contrast microscope. A Thermo Scientific Sure-Flow Combi Redox/ORP electrode filled with Ag/AgCl solution, calibrated

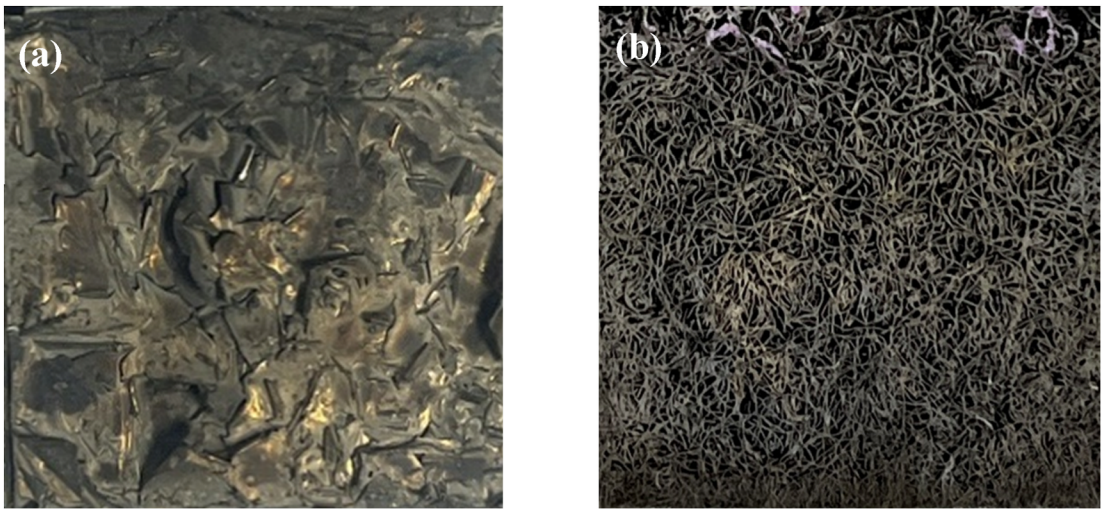


Fig. 11. Foil deposited on (a) stainless steel and (b) deposition on carbon felt cathodes from a synthetic solution of nickel, cobalt and manganese at 5 mA/cm².

Table 3
Metal composition of bioleachate, left over solution after electrowinning and after Li extraction at pH 10.5

	Metal concentration (mg/l)				
	Co	Ni	Mn	Cu	Li
Raw bioleachate	863.3	148.7	375.7	0.7	1551.3
Solution after EW	1.1	0.9	10.5	0.0	1529.1
After Li precipitation	0.0	0.0	0.0	0.0	916.6



Fig. 12. Image of bioleaching solution before (left) and after (right) metals recovery by electrowinning and precipitation.

daily using Thermo Scientific ORP standard was used for the Redox measurements.

Concentration of sulfate ions was measured as suggested by Pourhossein and Mousavi (2022), which is based on reaction between sulfate and BaCl₂, where the absorbance of the milky solution is measured with a spectrophotometer at 420 nm, after suitable amount of dilution. Percentage of converted sulfur can then be calculated as:

$$\text{Sulfur conversion\%} = \frac{\text{Sulfate concentration in sample} \left(\frac{\text{g}}{\text{l}} \right) / \text{Sulfate MW} \times \text{Sulfur MW}}{\text{Initial Sulfur concentration} \left(\frac{\text{g}}{\text{l}} \right)} \times 100\%$$

(3a)

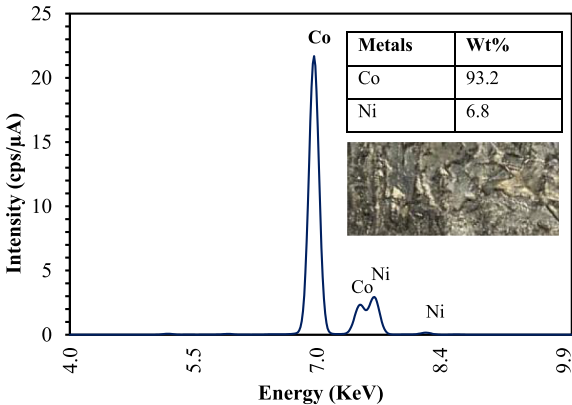


Fig. 13. Product analysis of foil plated on cathode surface (Co and Ni) at 5 mA/cm².

The concentration of the metals (Li, Co, Ni, Cu, Al, Mn) were determined after filtration using ICP-OES, calibrated using standard solutions obtained from Merck (TraceCERT®). This calibration was carried out using diluted standard solutions and five-point standard in the range of 10–100 mg/l. The correlation coefficient for all metals were higher than 0.9997. Metal recovery can be calculated in comparison to the initial concentration of metals in the sample.

High-resolution energy dispersive X-ray fluorescence (ED-XRF) spectroscopy was used to analyse the black mass before and after treatment, and solid products obtained by electrowinning and precipitation.

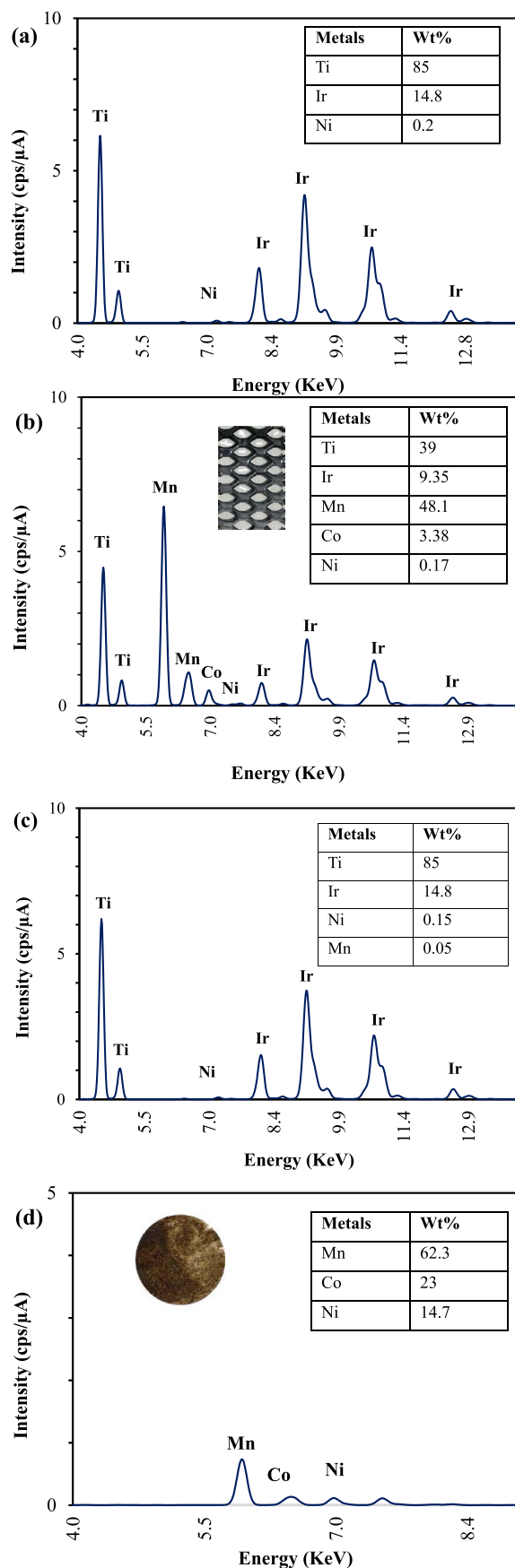


Fig. 14. XRF analysis of (a) mixed metal oxide anode as received, (b) mixed metal oxide anode plated with MnO₂, (c) mixed metal oxide anode cleaned and (d) Mn(OH)₂ precipitate.

3. Results and discussion

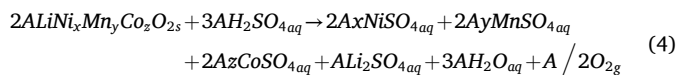
3.1. Production of biogenic acid and impact of aeration

Acidithiobacillus thiooxidans is a chemolithoautotroph that requires oxygen to oxidize sulfur. As shown in Fig. 1, which illustrates the effect of aeration on biogenic acid production, since *A. thiooxidans* is aerobic, no growth or activity could be observed without air supply. However, with oxygen supply, *A. thiooxidans* growth becomes noticeable, leading to an increase in biomass concentration (Brierley and Brierley, 2001). The bacteria entered a lag phase that lasted around 8–10 days, after which cell population significantly increased to 2×10^8 , while without oxygen, cell concentration remained very low. Sulfur conversion and pH reduction also became more pronounced; as seen in Fig. 1(b) and (d), approximately 80% of sulfur was converted into sulfuric acid by the end of the process, with the pH dropping significantly to 0.9 after 20 days. Low Eh values in the absence of aeration (Fig. 1(c)) also confirms poor growth of bacteria, while suitable aeration supports sulfur oxidation, producing sulfuric acid, which is essential for maintaining an acidic environment that promotes *Acidithiobacillus* activity (Namgung and Song, 2015). The large fluctuation in Fig. 1(a) results from the increased effects of biological and physicochemical parameters under aerated conditions in the bioreactor. Continuous aeration not only enhances microbial activity, but also creates fluctuating oxygen levels and localized stress, leading to alternating growth and cell population. Additionally, nutrient depletion (sulfur) along with the temporary accumulation of inhibitory intermediates can affect the microbial growth. Cell aggregation on the surface of sulfur and uneven sampling from the solution further contributes to apparent variations in cell counts. Together, these factors cause dynamic population shifts and transient instability, resulting in the observed variations in cell population with considerable experimental error range. Efficient aeration also helps preventing the buildup of toxic by-products, such as ferrous iron and sulfur compounds, which could otherwise inhibit bacterial function, therefore allowing for sustained bacterial activity. The biogenic acid was collected at pH 0.9 and used for indirect bioleaching of NMC powder.

3.2. Impact of test parameters variation on metal recovery

The impact of temperature on metal recovery can be observed in Fig. 2 at a constant pulp density of 6% (w/v) with 4% (v/v) H₂O₂. An increase in system temperature, which increases the reaction rate, improves acid dissolution mechanism and increases metal solubility, due to the higher number of activated molecules and better diffusion of ions (Fan et al., 2021). Li as monovalent metal can be dissolved better than higher valance transition metals Co and Ni, which benefit from the addition of H₂O₂ as a reductant. In sulfuric acid leaching, the main processes are acid dissolution and redox reactions, which are both affected by temperature variation. Since the reaction between lithium oxide and sulfuric acid to produce lithium sulfate is endothermic, increased temperature enhances lithium dissolution, resulting in higher recovery rates. As shown in Fig. 2, increasing the temperature from 30 °C to 60 °C raised Li recovery to 100%. Notably, Co and Ni dissolved rapidly within the first 10 min, after which their leaching rate slowed down considerably. At 60 °C, Li dissolution reached approximately 100%, while only a slight increase in Co and Ni extraction was noted. This outcome aligns with the metal arrangement in the cathode material, where cobalt leaching primarily involves the reduction of cobalt oxides. Co(III) oxides are converted to soluble Co(II) forms at elevated temperatures, accelerating Co dissolution. The higher temperature also enhances the redox reaction rates required for dissolving cobalt compounds. Studies on NMC show that Co and Ni leaching efficiency improved from 30% to 40% when the temperature increased from 30 °C to 80 °C with 1 M H₂SO₄ (Partinen et al., 2023). Different mechanism of dissolution of Li and transition metals can be explained as mentioned in

reaction (4)–(5) (Vieceli et al., 2023):



The effect of pulp density on metal recovery at constant levels of 0.6% and 4% (v/v) H_2O_2 is indicated in Fig. 3(a)–(c). As expected, by increasing pulp density, metal recovery decreases as more metal atoms compete for a given amount of protons available in the system, and a decrease in available surface area per volume of the leaching agent (Fan et al., 2021; Roshanfar et al., 2024). As a result, the proton concentration was limited to dissolve all the metals at a high pulp density of NMC powder.

According to Fig. 3(a) and (b), H_2O_2 increases the Co and Ni dissolution drastically especially in the first 10 min of the process. However, as the process continues, diminution of solubilised Ni and Co, considered as loss, can be observed. By increasing the H_2O_2 concentration from 0.6 to 4% (v/v), Co and Ni loss occurs even earlier (less than 10 min) in the process. Therefore, H_2O_2 functions as a reductant and significantly shortening the leaching time to just 10 min. A similar trend was observed in the study by Zhao et al. (2023), where a higher concentration of H_2O_2 was reported to cause metal loss earlier in the process. It can be observed that when the concentration of H_2O_2 increased to 2.5% (v/v) the Co and Ni recovery significantly decreased.

As can be seen in Fig. 3(c), Li dissolution is not impacted by increasing H_2O_2 concentration from 0.6% to 4% (v/v). The results indicate that the Li concentration in the leaching solution stabilizes after 1 h without any metal loss. NMC structure is composed of layered lattices of transition metals (Ni, Mn, Co) bound with oxygen, with Li ions occupying the spaces between these layers. This arrangement allows Li ions to dissolve more readily during leaching through a deintercalation mechanism, while H_2O_2 does not significantly affect the Li dissolution. In contrast, transition metals like Co and Mn tend to stay within the lattice structure (Partinen et al., 2023). To achieve higher dissolution rates of Co and Ni, a reducing agent is usually suitable, as promoted by H_2O_2 which increases the dissolution rate. Whereas Li is dissolved mostly by proton attack, Ni and Co dissolution benefits from prior reaction with H_2O_2 to be dissolved. As observed, Li dissolution does not improve further, after 1 h of the process, therefore, continuing the process further than that point is unnecessary. Zhao et al. (2023) suggested this as a selective Li recovery technique. However, as Ni and Co are valuable critical metals, and cannot be considered as impurities, their loss from the solution needs to be investigated and prevented. Based on similar results achieved in the work of Bertuol et al. (2016), it was suggested that as H_2O_2 degrades during the process, and that transition metals are displaced by more reactive metals such as Li.

3.3. Impact of influential parameters on transition metal loss

As mentioned in section 3.2, some of the leached metals do not remain in solution in the bioleachate during the bioleaching experiments. To quantify metal loss, Eq. (4) was used. The impact of H_2O_2 , temperature and pulp density on metal loss was considered as can be seen in Fig. (4). Metal loss increases with increase of temperature and H_2O_2 concentration, so that both factors impacting negatively on Co bioleaching after the first hour of the process. Therefore, process conditions with temperature higher than 45 °C and H_2O_2 concentration

higher than 2.5% (v/v) are not recommended due to high amount of Co loss. Based on Fig. 4(b), Co loss also significantly increases at higher pulp density. When pulp density exceeds 10% (w/v), over 50% of Co is lost in low or high concentration of H_2O_2 at 45 °C. This limits the bioleaching at higher pulps. Fig. 4(c) shows that at low pulp density, metal loss is quite low for the whole temperature range and 2.5% (v/v) H_2O_2 .

$$\text{Metal loss \%} = \frac{\text{MaxCo concentration achieved} - \text{Co concentration at 1 hr}}{\text{MaxCo concentration achieved}} \times 100\% \quad (4a)$$

Study of high pulp densities of black mass is not commonly available in scientific literature, with most of studies using active cathode materials (Dutta et al., 2018; Fan et al., 2021). Zhao et al. (2023) observed a similar pattern for loss of transition metals during bioleaching of Lithium Iron Phosphate (LFP) black mass and suggested it could be due to either hydrolysis reaction of ions and formation of precipitates, or to the adsorption of metals on carbons available in LIB anode. This is considered in this study since solid material used in our work is a black mass of LIB that contains around 30% total carbon.

SEM analysis of black mass before and after indirect bioleaching process (Fig. 5) showed that small particles disappeared while the edges of the larger particles were corroded and became sharp after bioleaching. However, XRF results (Fig. 6) before and after bioleaching show that the relative proportions (or elemental composition ratios) of transition metals remained essentially unchanged. Therefore, the adsorption of these metals on anode carbon surface in black mass powder can be considered, as observed in the work of previous researchers (Zhao et al., 2023).

To our knowledge, metal loss during acid leaching has not been reported in any previous work on NMC, although none of these used black mass. The metal loss observed in the present study using black mass is suggested to be due to the adsorption of metals onto carbon and is reminiscent of the previously reported use of biochar or activated carbon as an effective adsorbent of heavy metals in wastewater treatment due to its large surface area and suitable surface functional groups (Khanzada et al., 2024).

Oxidised graphite, available from anode surface of black mass, is in single (C-O) and double (C=O) carbon oxygen bond. Transition metals (M) after reaction with sulfuric acid and H_2O_2 are available as M^{2+} , MOH^+ , $M(OH)_2$, M^{3+} , $M(OH)^{2+}$, $M(OH)_2^+$, $M(OH)_3$. These forms of transition metals react with carbon-oxygen bonds to form (C-O)₂-M, (C-O)₂-MOH, (C-O)₂-M(OH)₂²⁻, (C-O)₂-M⁺, (C-O)₂-MOH, (C-O)₂-M(OH)₂, (C-O)₂-M(OH)₂²⁻ respectively (Corapcioglu and Huang, 1987). Analysis of functional groups before and after treatment of black mass in the work of Zhao et al. (2023) confirmed the above-mentioned reactions.

Another hypothesis is that the metal loss takes place due to the alkaline condition of the medium. The alkaline nature of NMC black mass, increases the pH of medium directly at the beginning of the process. However, in Fig. (7) two sets of experimental conditions are compared, with similar pH = 7, but where metal loss occurs only in one test. These compared experiments are Experiment A) at 45 °C, 5% (w/v) pulp density and 2.5% (v/v) H_2O_2 and Experiment B) at 60 °C, 6% (w/v) pulp and 0.6% (v/v) H_2O_2 . It can be observed that whereas Experiment A is stable, Experiment B is not, despite similar pH variation during both experiments. The major difference in these two experiments is the concentration of hydrogen peroxide. However, based on Fig. 4(a) and (b), H_2O_2 is not responsible for metal loss in this range. Temperature

differences could be responsible for the differences, since Experiment A is performed at much lower temperature than Experiment B. It was previously reported that temperature can significantly impact removal of Co and Ni from wastewater using activated carbon. This is due to several reasons which are: 1) amplification of metal molecules diffusion in internal pores of activated carbon at elevated temperatures, 2) increased interaction of metals and active sites of carbon surface, 3) opening of pores at higher temperatures and enhanced penetration of metals in carbon pores (Olupot et al., 2024).

3.4. Statistical analysis

For statistical analysis, analysis of variance (ANOVA) with $\alpha = 0.05$ was performed as can be seen in Table 1. All four responses were significant, with F-test and p-values below 0.05. Based on the F-values for all four responses, it can be observed that pulp density is the most influential factor on the responses. Regarding metal loss, these results are in agreement with previous results of adsorption of metals on carbon surface, which confirmed increased metal removal by increase of pulp density, which creates more active sites for metals to bind and higher probability of ion adsorption (Olupot et al., 2024). For Li, as can be seen by F-values, the effects of addition of reducing agent is not very significant and does not improve the results.

3.5. Bioleaching vs. chemical acid leaching, with and without reducing agent

The results obtained from DoE and the mathematical models were used for optimisation by calculating the desirability of the experiments based on defined targets that can be seen in Table 2. Apart from maximising recoveries and minimising metal loss, the target was working at maximum pulp density. This is beneficial for industrial application as can drastically decrease the operational cost by possibility of processing larger amount of waste in each run. The predicted values for bioleaching and actual values (after confirmation test) are shown in Table 2.

At optimal conditions, comparison of chemical acid leaching and bioleaching with and without reductant was performed, the results of which can be seen in Fig. (8). While no difference was observed between chemical leaching and bioleaching for Li dissolution (Fig. 8(a)), Co, Ni and Mn leaching is improved with chemical acid leaching. In bioleaching, dissolved Ni and Co are more stable in comparison to chemical acid leaching. The result is also confirmation that Li recovery is not significantly impacted with addition of H_2O_2 as a reductant, in acid leaching addition of reducing agent has even negative impact for dissolution of Li. However, H_2O_2 acts as a reductant for Ni, Co and Mn, during acid leaching and bioleaching. The difference in dissolution mechanism of Li with Ni, Co and Mn is due to the high valent of the transition metals, which are highly stable transition metal oxide, which is facilitated with H_2O_2 (Ning et al., 2024). Less dissolution with H_2O_2 can be due to the fact that in presence of reductant, some fractions of protons have been reacted with Ni, Co and Mn rather than Li. Similar dissolution trend of Co and Mn, is confirmed in Fig. (8(b)) and Fig. (8(c)).

As can be observed, in Fig. (8), metal loss took place very early during the process. Therefore, the impact of time was also studied on bioleaching with reducing agent, in shorter intervals to study the kinetic of the process. Fig. (9) shows, how Co and Ni concentration change during the process. It can be observed that the maximum Co, Ni and Mn concentration can be achieved after 5 min. Such a short processing time by addition of H_2O_2 was also reported by Bertuol et al. (2016) using supercritical CO_2 . Based on Fig. (9), bioleaching targets were met in as fast as 5 min during the process although not 100% recovery is achieved. This would suggest that processing the waste powder in a system such as fixed bed column where waste will be constantly exposed to a high flow rate of fresh biogenic acid would be an ideal system to achieve both fast dissolution and reducing the risk of metal loss. It can be observed that

after 20 min, the rate of metal loss from solution decreases. This may suggest that at this point most of available active surface on carbon anode are occupied.

3.6. Electrowinning and precipitation results

To select the optimal cathode material, two types of cathodes; stainless steel and carbon felt; were tested using a solution that was ten times more concentrated than the bioleachate, containing Co, Mn, and Ni in the same ratio. This solution also contained acidophilic basal salt and trace elements. An operating current density of 5 mA/cm^2 was applied to the electrowinning cell, and Faradaic (current) efficiencies were monitored and analysed over time (Fig. 10).

As expected, during the process current efficiency drops as the metal concentration in the leachate decreases. Plated mass on the cathode falls further below the theoretical values. As the electrowinning process continues, metal concentrations in the solution, become lower, and mass transfer of ions to the electrode is more challenging, and metal deposition is competing with other parasitic reactions (Baniasadi et al., 2021). Stainless steel exhibited better current efficiency compared to carbon felt. Given the final product of the cathode materials, stainless steel is more practical, as the alloy can be recovered through mechanical scraping. In contrast, carbon felt requires smelting in a furnace (Pakostova et al., 2024). Therefore, stainless steel as cathode was used further in this study. Metal deposition on the cathode material can be seen in Fig. 11.

In the next stage, the obtained optimum parameters of electrowinning were applied on actual bioleachate, which has metal concentration mentioned in Table 3. After 4 h, a foil was obtained on cathode surface and the weight of anode increased, which confirms deposition of metals on the surface of both electrodes. The current efficiency of the cathode process was 90% at 5 mA/cm^2 over the first hour. The left-over solution was tested with ICP-OES after electrowinning, and compared with the original bioleachate it shows significant recovery of Co, Ni and Mn. As expected, because Li has a very negative reduction potential, the Li concentration was unaffected by electrowinning therefore, sodium carbonate was added to the left-over solution which resulted in significant precipitation of Li_2CO_3 . Fig. 12 indicated the bioleaching solution before and after metals recovery.

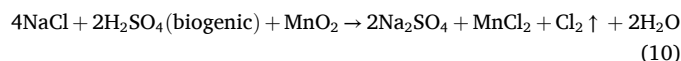
3.7. Electrowinning product analysis

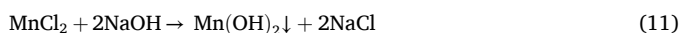
The presence of Co and Ni in the foil deposited on the cathode was confirmed by XRF analysis (Fig. 13). The deposit consisted of 93% Co and 7% Ni.

XRF analysis of the anode material, showed the presence of Mn and Co as can be observed in Fig. (14). MnO_2 plating on anode was explained by Ghaemi and Binder (2002) by reactions (6)–(9).



To separate MnO_2 from the mixed metal oxide substrate, the anode was dipped in biogenic acid with addition of 1M sodium chloride and the solution was kept undisturbed for 24 h. Later sodium hydroxide in the same solution was added to initiate precipitation through following reactions (reaction (10)–(11)). The obtained powder, collected by filtration, was later analysed by XRF to confirm the presence of Mn (Fig. 11(d)).





The presence of Co on the anode is not usual and is being deposited as an oxide. It has the property to lose electron in presence of iridium which is abundantly present in the mix metal oxide anode and forms Co_xO_y (Tae et al., 2015). Obtained Li_2CO_3 powder, which is highly demanded in energy industry, was analysed after digestion in acid with ICP-OES which confirmed the powder to be 40% Li_2CO_3 .

4. Conclusion

In this work, the recovery of metals from NMC black mass of LIBs was performed at high pulp density, using biogenic sulfuric acid, with the addition of H_2O_2 as a reductant to improve dissolution of transition metals Co, Mn and Ni. The addition of H_2O_2 proved to improve dissolution of these transition metals and reaction kinetics to result in optimal condition in the short time of 5 min. However, early in the process, some of the dissolved transition metals did not remain in solution, and the concentration of solubilised metals diminished with time. Parameters that affected the metal solubilisation were studied, and the loss of solubilised metals is suggested to be due to the adsorption of these metals to the surface of graphite present in black mass. The rapid efficacy of bioleaching demonstrates the benefit of bioleaching to extract the metal, but the relatively low percentage of metals solubilised suggest that the process should be improved. The analysis of these results suggests that the preliminary removal of carbon binders could improve the process. The bioleachate solution obtained for optimal conditions was used for electrowinning and Co and Ni were successfully plated and recovered on the surface of the cathode. MnO_2 was harvested on the anode surface to be later washed off by biogenic sulfuric acid and precipitated as Mn(OH)_2 . Li was kept in the solution to be recovered by precipitation as lithium carbonate.

The results of this work demonstrate the potential use of bioleaching to extract metals from LIBs black mass, and the benefit addition of H_2O_2 , while identifying the challenges to keep metals solubilised in chemical and biogenic acid leaching. This work also demonstrates the benefit of combining bioleaching with electrowinning and precipitation for selective metal recovery from LIBs towards in a closed-loop system essential in circular economy.

CRedit authorship contribution statement

Mahsa Baniyadi: Writing – original draft, Software, Project administration, Methodology, Investigation, Data curation, Conceptualization. **Kumar Upvan:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Fatemeh Pourhossein:** Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Data curation, Conceptualization. **John E. Graves:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Egle Latvyte:** Methodology, Data curation. **Sebastien Farnaud:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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Baniasadi, Mahsa; Upvan, Kumar; Pourhossein, Fatemeh; Graves, John E.; Latvyte, Egle ...

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