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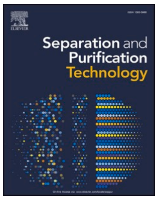

Recovery of nickel from the pregnant leach solution of spent NMC batteries using Versatic acid 10 and mixer-settler operations

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ABSTRACT

Nickel (Ni), one of the most expensive strategic metals, is frequently utilized in Li-ion batteries and other metal alloy applications due to its unique features that include corrosion resistance, high strength, storage capacity, and energy density. In this study, selective recovery of Ni from hydrometallurgical battery recycling solution of real industrial spent nickel-manganese-cobalt oxide (NMC) batteries was investigated. Versatic Acid 10 diluted in Isopar L was used as an organic extractant for the selective extraction of Ni^{2+} ions, followed by crystallization of nickel sulfate hexahydrate ($NiSO_4.6H_2O$). The kinetics of solvent extraction was studied in batch scale over a time range of 1 to 15 min at pH 6.8 ± 0.1. Results showed that the equilibrium needed for effective extraction and volumetric mass transfer coefficient could be rapidly achieved (within 3 min, 0.9 M Versatic Acid 10). The counter-current solvent extraction process was scaled up in a mixer-settler system for a pilot run using the optimized parameters established by the batch-scale experiments. Almost 100% Ni extraction was achieved through a two-stage counter-current process using 0.9 M Versatic Acid 10, with an organic-to-aqueous phase ratio (θ) of 1. The Ni loaded organic phase was subsequently stripped in two stages using 0.2 M sulfuric acid (H_2SO_4) at $\theta = 1$. $NiSO_4.6H_2O$ salt with 99.26 ± 0.01% purity was recovered from the stripped raffinate solution obtained after the mixer-settler operation via evaporative crystallization at 35 °C and a vacuum pressure of 0.1 MPa. Purity, morphology and phases of the recovered crystallized powder were analyzed with inductively coupled plasma optical emission spectrometry (ICP-OES), scanning electron microscope (SEM) and X-ray diffraction (XRD) techniques respectively. Recovered crystallized nickel sulfate was determined to have sufficient purity for use as precursor cathode active materials (pCAM) in nickel-based lithium-ion battery manufacture.

1. Introduction

Li-ion batteries (LIBs) are considered as efficient and clean energy storage devices due to their high energy density, long cycle life, high conversion efficiency and good electrochemical performance [1,2]. Consequently, the application of LIBs is widespread in diverse industries such as automobiles, electronics, large-scale energy storage devices, artificial satellites and aerospace [3]. Various metals like aluminum (Al), copper (Cu), cobalt (Co), iron (Fe), manganese (Mn) and nickel (Ni) are present in the form of current collectors, anode and cathode materials within the different chemistries of the LIBs. Therefore, recycling of the increasing level of End-of-Life (EoL) LIBs can offer a valuable secondary raw material resource of selected critical and valuable raw materials, especially as metal concentrations in LIBs waste often significantly exceeds that found in their corresponding ores [4,5].

Nickel as a strategic metal that is used in numerous applications including stainless and alloy steels, non-ferrous corrosion resistant metal alloys, high strength tools as well as lithium-ion batteries [6]. Moreover, the increased trend toward higher nickel content NMC battery chemistries-for higher energy density, high charge-discharge rates, lower oxidation potential, and improved overall energy storage capacity-further highlights the need for efficient Ni recovery processes to ensure sustainable recycling and reuse of such metals in battery production [7]. The recovery of nickel from secondary sources,

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Table 1
Literature studies on the extraction of Ni using solvent extraction (θ = Organic to Aqueous phase ratio and S = Stage).

Input	Experimental details	Key findings	Reference
Spent LIBs acidic leachates	Synergistic solvent extraction (SSE), saponified Cyanex 301 and 0–1 M P-507 HEH (*EH*)P, pH = 3–5, diluent: Sulfonated kerosene	~99% Ni^{2+} recovered with 3-stage counter-current extraction using a 1.5 Mol/L mixed extractant with 60% saponification, scrubbing: 2-stage using 0.05 Mol/L H_2SO_4 and stripping: 3-stage with 9 Mol/L HCl, at θ of 2.5	[22]
Synthetic Ni lateritic sulfuric solution	0.5 M Versatic 10 and SSE, combination of Cyanex 272, Cyanex 301, LIX 84-I, and LIX 860 N-IC	Versatic 10: 93% Ni at 6.5 pH; Versatic 10 + Cyanex 301: 99% Ni at 1.5 pH; Versatic 10 + LIX 84-I: 99% Ni at 5 pH; Versatic 10+ LIX 860 N-IC: 81% Ni at 4 pH	[23]
Sulphide Nkomati flotation concentrate	Mixer-settler, 30 vol% Versatic acid 10, pH adjustment: 400 g/L NH_4OH;	99% Ni was extracted at θ = 1, S: 5; scrubbing: 300 g/L H_2SO_4, pH = 5.9; stripping: Spent EW electrolyte, S:3	[24]
Synthetic co sulfate solution, Ni present as an impurity	SSE, dinonylnaphthalene sulfonic acid (DNNSA) and decyl 4 picolinate (4PC), 50% saponified with 10 M NaOH	Ni^{2+} was preferentially extracted over Co^{2+}. ~100% Ni extracted in 5-stage counter-current with θ = 6 at 30 °C and pH = 2.3.	[25]
Sulfate media/ Ni from ca	SSE, mixture of a versatic acid 10 and 4-(5-nonyl) pyridine, diluent xylene and C_{12} —C_{13} paraffin,	Large synergistic shifts was found for Ni, and 99% Ni extracted with 0.5 M Versatic 10 and 0.5 M 4-(5-nonyl)pyridine, θ = 1; S:3, pH = 5.8; stripping: 0.25 M H2SO4, θ = 3.33	[26]
Ni laterite sulphate solutions	Synergistic solvents Versatic Acid 10, Mextral 6103H and Aliquat 336, pH = 2.5–6	99% Ni extracted using 2 stages (S) at pH 4.5 and 5 in E1 and E2, respectively, and θ = 2.2 with 2% Mn and 1% Mg and ca as impurities	[27]
LIBs sulphate solution	60% saponified versatic 10, diluent: Kerosene, pH = 4–7.3, 30–50 vol% of versatic 10	Mixed metals >98% co, > 99% Ni, and > 97% Mn with 8% Li loss; 50 vol% Versatic 10, pH = 6, 25 °C, θ = 1, 7 min, S = 2.	[28]
Ni lateritic ore sulphate solution	SSE, 0.3–0.5 M LIX860N-IC + Versatic 10 in kerosene Exxsol D80, pH = 2–6, 40 °C,	> 99% Ni at pH = 4.5, S = 2, θ = 0.9, stripping = 50 g/L Ni + 1 M H_2SO_4 solution, S = 3, θ = 10:1	[29]
Synthetic laterite leach liquor, sulphate solution	Cyanex 272, Versatic 10 and SSE, coordination number by UV spectra, FTIR, hydration complexes by TGA	Ni and co-Versatic 10 complexes found in octahedral, co with (best separation factor) 15% Cyanex 272 + 5% Versatic 10 found in tetrahedral coordination	[30]
Synthetic sulphate solution	5–20% *v*/*v* Cyanex 272, 0–20% v/v Versatic 10 and SSE, diluent: n-heptane, *T* = 50 °C, pH = 1.5–7, θ = 1	20% v/v Cyanex 272 extracted Ni over Zn, co, cu and Mn; at pH = 3.9, however, a mixture of 20%v/v Cyanex 272 and 10% v/v Versatic 10 was found suitable to separate ca and Mg at pH =5.1	[31]
Spent LIBs acidic leachates	SSE, tung oil-based carboxylic acid (TFCA), and isonicotinic acid ester (4PC), pH = 3–6.5, scrubbing = 300 mg/L $NiSO_4$,	~97% Ni extracted at pH = 5.5, *T* = 25 °C, θ = 1, with 0.4 M TFCA and 0.8 M 4PC	[32]

specifically end-of-life lithium-ion batteries, can provide valuable raw materials for battery and other manufacturing industries while concurrently reducing the environmental impact associated with battery waste disposal. Additionally, the purity of the recovered nickel salt plays a vital role in ensuring future battery performance and maintaining a reliable supply of this strategic raw material.

Hydrometallurgical processing is required in the recycling of spent LIBs: either solely or after pyrometallurgical refining. Both hydrometallurgical and pyro-hydrometallurgical battery recycling have been reported to achieve high transition metals extraction efficiencies [8–10]. A typical hydrometallurgical process comprises three distinct parts: pretreatment, leaching and selective recovery of the targeted metals. The pretreatment process, which can include dismantling, discharging, shredding, milling, and mechanical separation, plays a key role in the production of black mass that is primarily composed of a mixture of cathode and anode active materials of spent batteries [11,12]. The black mass is then subjected to acid leaching, most typically by mineral acid like hydrochloric acid (HCl) and sulfuric acid (H_2SO_4) with or without the addition of external reducing agent (e.g., hydrogen peroxide) at 60–80 °C [13–17]. Following leaching, various unit processes, such as cementation, solvent extraction, selective precipitation, ion exchange, and electrochemical deposition, can be undertaken to purify the solution and separate and recover the valuable metals from the pregnant leach solution (PLS) of the LIBs black mass [18,19]. Of these approaches, solvent extraction methods are one of the most widely used techniques for metal separation from PLS due to their high separation efficiency, recovery, easy operation, and relatively benign experimental conditions [20,21]. There are also numerous studies in scientific literature on the extraction of Ni from Ni-rich LIBs waste PLS, synthetic solutions and primary ores by solvent extraction, some of which are summarized in Table 1.

Synergistic solvent extraction (SSE), which involves the combination of two or more organic extractants, has been studied as a promising alternative approach for enhancing the efficiency and selectivity of nickel recovery from complex leach solutions. However, the extraction of Ni from laterite and LIBs leach solution has also been extensively studied using carboxylic acid extractants, as they have been reported to provide higher solubility for Ni compared to other organophosphorus extractants [28,29,33]. As shown in Table 1, previous studies have demonstrated that Ni can be successfully extracted from primary lateritic ores using either saponified Versatic Acid 10 or solvent sublation extraction techniques, typically at pH values >5. Nevertheless, the underlying extraction rate kinetics and volumetric mass transfer coefficient—which enables extraction rate prediction, complex compound transportation, performance and optimization of process conditions during solvent extraction—has so far not been comprehensively investigated for selective Ni extraction from industrial NMC battery leachates.

In addition, selective recovery of high-purity nickel sulfate hexahydrate ($NiSO_4.6H_2O$) salts from industrially produced leach liquor from end-of-life LIBs hydrometallurgical processing has also not been

Table 2
Metal concentrations in aqueous raffinate solution used as feed to Ni extraction.

	Co	Ni	Na	Li	P
C (g/L)	0.04 ± 0.00	8.32 ± 0.05	45.44 ± 0.19	3.58 ± 0.01	0.16 ± 0.00
C (mM)	0.73 ± 0.00	140.77 ± 0.57	1976.91 ± 8.39	516.73 ± 2.07	5.19 ± 0.05

extensively addressed. Consequently, this study examines the selective extraction of Ni values from a real industrial leach solution of LIBs by solvent extraction using Versatic Acid 10 without saponification followed by selective evaporative crystallization. Solvent extraction using Versatic Acid 10, diluted in Isopar L was used to determine the kinetics of Ni extraction and volumetric mass transfer. The optimized parameters from the batch-scale experiments were applied to scale up the countercurrent solvent extraction process in a mixer-settler system. Furthermore, the resultant stripped aqueous raffinate was then subjected to evaporative crystallization to produce a high pure nickel sulfate salt, $NiSO_4.6H_2O$.

2. Materials and methods

2.1. Pretreatment processing

End-of-life (Nickel-Manganese-Cobalt oxide) NMC 111 battery packs, (provided by Volvo Cars AB, Sweden) were safely dismantled and discharged by STENA Recycling AB, Sweden. These battery packs were then crushed before undergoing mechanical separation which was carried out at Akkuser Oy, Finland. The resulting black mass, a combination of anode and cathode active material was leached at the METSO Research Centre (Pori, Finland) under previously optimized conditions (2 M H_2SO_4, S/L ratio = 200 g/L, $T = 50$ °C) [34]. After filtration, impurities in the pregnant leach solution (PLS) were removed by treatment with a stoichiometric amount of gaseous H_2S at 80 °C to eliminate Cu, followed by additions of NaOH and H_2O_2 at 80 °C at pH 4.5–4.7, to selectively precipitate Al and Fe. After the impurities removal, the selective extraction of high-purity manganese (Mn), cobalt (Co), and nickel (Ni) salt were undertaken. Extraction of Mn and Co was achieved using the organic extractants Di-(2-Ethyl Hexyl) phosphoric acid (D2EHPA) and bis(2,4,4-trimethylpentyl) phosphinic acid (Cyanex 272) diluted in Isopar L, respectively [35,36]. The elemental composition of aqueous raffinate solution after cobalt recovery is shown in Table 2 and was used as the input feed for this research. The pH, ionic strength and redox potential of the aqueous raffinate solution was 5.6 ± 0.1, 1.53 M, and 166.2 mV respectively. The limit of quantification (LOQ) for impurities such as Al, Ca, Cd, Cu, Fe, Mg, Mn, and Zn was approximately 0.1 mg/L, 0.22 mg/L, 0.04 mg/L, 0.28 mg/L, 0.04 mg/L, 0.01 mg/L, 0.01 mg/L, and 0.03 mg/L respectively.

2.2. Reagents

Organic extractant Versatic Acid 10 (Hexion, Netherlands) was diluted in Isopar L (Exxon Mobil, USA) and used during the solvent extraction of Ni - more detailed information about purity, molecular weight and density of the organic extractant are provided in the supplementary information S1. The organic extractant and diluent were used without further purification. A 10 M NaOH solution was prepared by dissolving the NaOH pellets (> 99%, EMSURE, Merck, Sweden) in ultrapure water (Milli-Q, Merck Millipore, Sweden) to adjust the pH during the extraction. Sulfuric acid (H_2SO_4) (95–98%, Sigma Aldrich, Germany) was diluted with ultrapure water to make the solution used to strip the loaded organic.

2.3. Solvent extraction

2.3.1. Batch experiment

Batch-scale experiments were initially performed to identify the suitable operating conditions such as equilibrium pH, organic concentration, phase ratio, kinetics, and number of stages for selective Ni extraction. For solvent extraction, experiments were conducted in a 100 mL polypropylene beaker with a total solution volume of 30 mL. The aqueous and organic phases were mixed with a mechanical mixer at a constant speed of 1000 rpm for 15 min to achieve equilibrium. The two phases were allowed to separate for 5–10 min, followed by visual inspection. pH adjustment during the mixing of the two phases was carried out by the addition of 10 M sodium hydroxide and concentrated H_2SO_4 (95–98%; Sigma Aldrich, Germany). A representative sample from aqueous raffinate and loaded organic was taken at equilibrium to determine the metal concentration present in both phases. In the case of the loaded organic phase, the solution was stripped with 5 M H_2SO_4 and low organic to aqueous phase ratio ($\theta = 0.125$). By varying the organic to aqueous phase ratio ($\theta = 0.1$, 0.25, 0.50, 0.75, 1, 2, 3, 5 and 10), a McCabe-Thiele (M-C-T) diagram was constructed for the extraction of Ni at constant pH. The equilibrium curve was established by plotting the metal concentrations in the organic phase against those in the aqueous phase. A vertical line, referred to as the feed line, was then used to represent the metal concentration in the aqueous feed solution. Subsequently, the operating line could be constructed by drawing a straight line with a slope corresponding to the aqueous-to-organic phase flow rate ratio (θ). This line intersects the diagram from the coordinates of the fresh aqueous input to those of the raffinate stream exiting the system. The M-C-T diagram for the stripping process was performed in small glass vials (3.5 mL) to ascertain the number of required stages for stripping, as this process typically does not require pH control. Samples were shaken in the shaking machine (IKA-Vibrax, Germany) at 1500 rpm for 30 min and then centrifuged at 5000 rpm for 5 min to ensure phase separation. All experiments were conducted at room temperature (22 ± 1 °C) and in triplicate, and results presented in this study represent an average of the measured values.

2.3.2. Data procurement

The extraction efficiency (E), distribution ratio (D), during solvent extraction and purity (P) of the crystallized salt were calculated using Eqs. (1)–(3), respectively.

$$E(\%) = \frac{[M_i] - [M_a]}{[M_i]} \times 100 \tag{1}$$

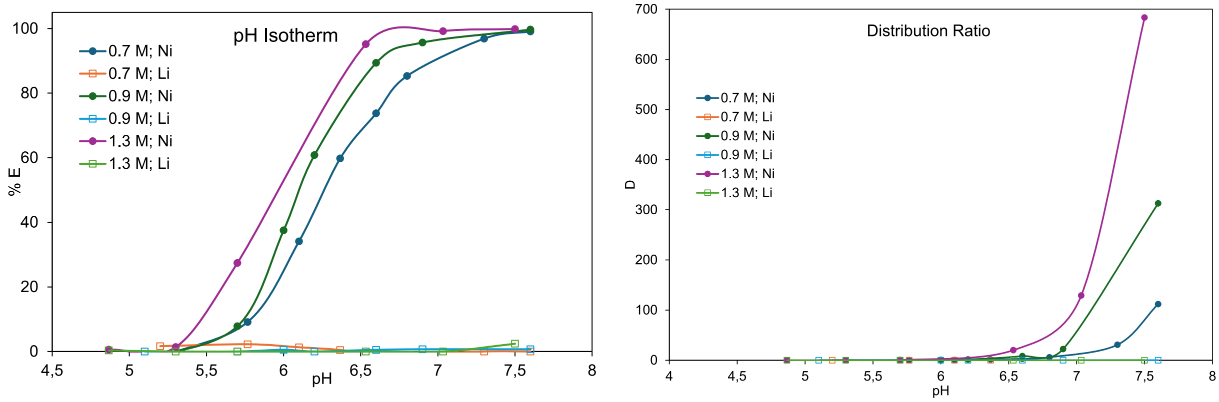
$$D = \frac{[M_{org}]}{[M_{aq}]} \tag{2}$$

$$P(\%) = \frac{[M_{Ni}]}{[M_t]} \times 100 \tag{3}$$

where, $[M_i]$, $[M_a]$, $[M_{org}]$, and $[M_{aq}]$, represents the initial metal concentration in the aqueous solution, metal concentration in aqueous raffinate after mixing and separation, concentration of metals in the loaded organic, and aqueous raffinate solution, respectively, as determined by ICP-OES. $[M_{Ni}]$, and $[M_t]$ shows the concentration of Ni only and total metal content in the solution.

2.3.3. Kinetic study

The extraction rate of Ni from the aqueous feed solution was studied by varying the contact time from 1 min to 15 min with experiments conducted in a 100 mL polypropylene beaker with $\theta = 1$ and 10 M NaOH was added to reach a desired equilibrium pH = 6.7. At 60 s intervals, a



(a)	(b)

Fig. 1. (a) pH isotherm and (b) distribution ratio for Ni extraction at various concentration of Versatic Acid 10 ($T = 25 \pm 1\ ^\circ C$, $t_{eq} = 15$ min).

fresh sample was taken and analyzed with the ICP-OES to investigate the extraction efficiency of Ni. The volumetric mass transfer coefficient during the extraction was estimated using Eq. 4 [37].

$$k = \left(\frac{E}{1-E}\right)\left(\frac{1}{t}\right)\left(\frac{V_A}{V_T}\right) \tag{4}$$

where, k represents volumetric mass transfer coefficient (sec^{-1}), E (%) extraction efficiency of metal, t is the mixing time, V_A the volume of the aqueous solution and V_T the total volume.

2.3.4. Pilot run

A total of 15 L of aqueous raffinate was obtained after the extraction of Mn and Co. Therefore, the pilot run for selective Ni extraction was conducted using a counter-current process in mixer-settler units (MEAB Metalextraktion AB, Sweden), which have an active volume of mixer and settler of 0.12 L and 0.48 L respectively. The estimated extraction and stripping stages were determined based on the M-C-T diagram studied during the lab-scale experiments. The aqueous feed and prepared organic extractant (Versatic Acid 10) were introduced into the mixing chamber using electromagnetic pumps (EW- B08TC-20EPF2, EWN-B11TCER, IWAKI, Japan) using100 strokes per minute (SPM) and a frequency of 90 Hz (Hz) for the aqueous phase and 100 SPM and 125 Hz for the organic phase, respectively to maintain the phase ratio of 1 during the process. The flow rate of both aqueous and organic phases was 15 mL/min and kept constant to maintain θ = 1. The mixer speed was controlled by a stirrer motor controller (MSU 0.5, Mymeko, Sweden) at 1000 rpm. The pH adjustment was carried out with the introduction of 10 M NaOH solution in the mixing chamber of each stage using peristaltic pumps (BT103S, Lead Fluid, China).

2.4. Evaporative crystallization

Ni-rich stripped aqueous raffinate obtained after the mixer-settler operation as the basis for Ni sulfate salt recovery was investigated. A rotary evaporator (RE150–220, Labfirst Scientific, China) of 50 L capacity was utilized to crystallize nickel sulfate hexahydrate at 45 °C and 50 mbar, with a rotating flask speed of 35 RPM [38]. The obtained salt was thoroughly washed with ethanol (95%), followed by vacuum filtration to remove any traces of liquid residues. Filtered residue composed of crystallized salt was dried in two ways, either at room temperature or at 60 °C for 5 h in a drying oven. Purity of the final product was then investigated using ICP-OES, SEM-EDS and XRD.

2.5. Methods of analysis

The concentration of metal ions presents in the aqueous, stripped aqueous, and raffinate phases was analyzed using Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Thermo Fisher Scientific, iCAP™ 6000 Series, USA). 0.5 M nitric acid (Superapur, 65%) was used to dilute the samples and Yttrium (Y) was used as an internal standard. The determination of phases in the crystallized salt was analyzed via X-ray diffraction (XRD) (Rigaku Smart Lab) using Cu-Kα radiation, in the 2 theta range of 10–80° with scanning speed of 2°/min. EVA software (Bruker AXS – DIFFRAC.EVA.Version 6.0) and JCPDS database (International Centre for Diffraction Data, ICDD, PDF-5+) were used for analytical interpretation. The structural morphology and elemental composition in weight (%) of the recovered product were also investigated using scanning electron microscopy (SEM) equipped with Energy Dispersive X-ray Spectroscopy (EDS) detector (Quanta 200 FEI-ESEM).

3. Results and discussions

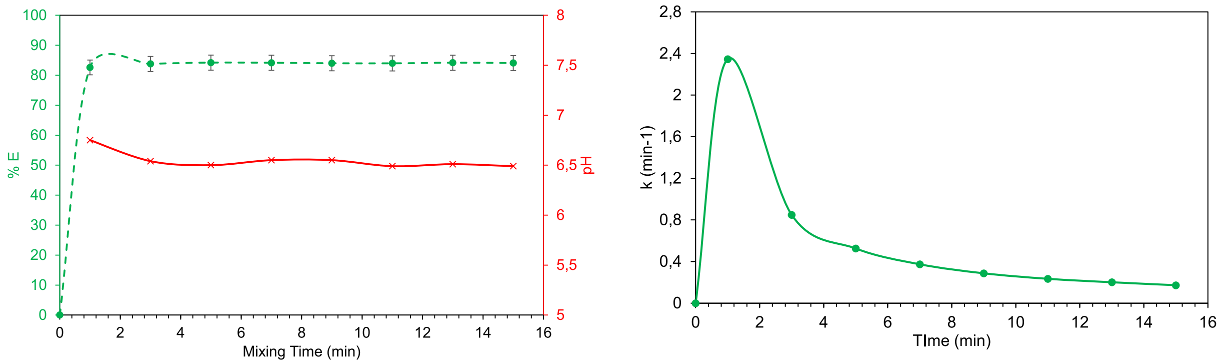
3.1. Batch scale solvent extraction

Initial laboratory batch experiments were conducted to investigate the selective extraction of Ni using varying concentrations of Versatic Acid 10 diluted in Isopar L. Versatic Acid 10 was selected as an organic extractant in this study because it provides high selectivity, chemical stability and easy downstream processing. It does not contain phosphorus like other organic extractant such as Cyanex 272, Cyanex 301 and D2EHPA etc., which simplifies its waste handling and disposal compared to phosphorus-containing compounds. The extraction mechanism of the Ni extraction with Versatic Acid 10 is expressed as shown in Eq. (5) [28,39].

$$Ni^{m+} + n(\overline{HA})_2 \rightleftarrows \overline{NiA_m(HA))_{2n-m}} + mH^+ \tag{5}$$

where $\overline{HA}$ represents the acidic extractant in the organic phase.

Versatic Acid 10 is a cationic acidic extractant that releases protons and exchanges the cations during the extraction process. The organic extractant binds the metal cations through a cation exchange reaction mechanism, which results in the formation of a nickel carboxylate complex that is soluble in the organic phase when contact between the aqueous and organic phases occurs. As the number of carboxylate ions required to form a neutral complex is lower than the coordination number of the nickel ion within the organic phase, the carboxylate complex formed by divalent nickel ions is characterized by the general stoichiometry $NiA_m(HA))_{2n-m}$, where, m molecules of the carboxylic acid act as solvating ligands [33]. According to Eq. 5, pH plays a very important role during the Ni extraction process with the carboxylic acid extractant.



(a) (b)

Fig. 2. (a) Extraction of Ni (%) and (b) mass transfer rate coefficient with mixing time.

3.1.1. *pH isotherm and distribution ratio*

Various concentrations of Versatic Acid 10 diluted in Isopar L were contacted with the aqueous solution up to 7.5 ± 0.1 pH. Three concentrations of a carboxylic extractant were investigated, selected based on the nickel concentration present in the aqueous solution and the reported metal loading efficiencies in the literature [33]. The pH isotherm and distribution ratio at different molar concentrations of Versatic Acid 10, such as 0.7 M, 0.9 M and 1.3 M, are shown in Fig. 1a and b. The extraction of metals such as Ni and Li was investigated by varying the equilibrium pH. Complete extraction of Ni was attained when the equilibrium pH $\geq$ 6.5 for all three concentrations of the organic extractant investigated. Moreover, it was experimentally observed that as the concentration of organic extractant was increased, the equilibrium pH for the complete extraction of Ni showed a reciprocal decrease. This phenomenon is attributed to the enhanced release of H^{+} ions from higher concentrations of Versatic Acid 10 upon contact with the aqueous phase, which facilitates cation exchange during the extraction process [1]. When the organic extractant is in contact with the aqueous solution, the pH of the aqueous solution reduces and additions of 10 M NaOH can be used to obtain the desired equilibrium condition. After a single contact, the extraction efficiency of Ni was found to be ~ 99.1%, resulting in 0.036 g/L in the aqueous raffinate. Interestingly, the distribution ratio between the Li and Ni increased with the concentration of organic extractant, with the co-extraction of Li found to be negligible for all the concentrations of Versatic Acid 10 tested [1,28]. The mechanistic explanation for selective Ni^{2+} extraction over Li^{+} is basically coordination chemistry and nature of complex they form with the Versatic Acid 10. The carboxylic acid group (-COOH) of the Versatic Acid 10 deprotonates during the mixing process and the resulting carboxylate anion binds with Ni^{2+} ion, while H+ ion release into the aqueous phases. Ni^{2+} forms stable organometallic complex with carboxylate ligands of Versatic Acid 10 and transported to the organic phase [28]. The formed complex is a chelate structure, which provides strong coordination and stability in the non-polar organic solvent [23].

However, Li is an alkali metal having smaller ionic radius and lower charge density compared to Ni^{2+} doesn't form relatively strong and stable complex with carboxylate ligands [40]. Li^{+} has a much weaker affinity and strong hydration shell in aqueous phase, resulting in it preferring to stay in the polar aqueous environment. Based on these preliminary experiments and the obtained results, the optimal concentration for Ni extraction was determined to be 0.9 M Versatic Acid 10 diluted in Isopar L at an equilibrium pH of 7.0 ± 0.1. Shuya et al. reported 99.18% Ni extraction using 50 vol% Versatic Acid 10, with 60% saponification at pH 6, $\theta = 1$, 25 °C for 7 min, followed by a secondary extraction under the same pH, θ, time and temperature conditions with 40 vol% of the same saponified extractant [28]. Nonetheless, additional steps such as saponification and secondary extraction using varying concentrations of organic extractant may pose operational challenges at the pilot scale.

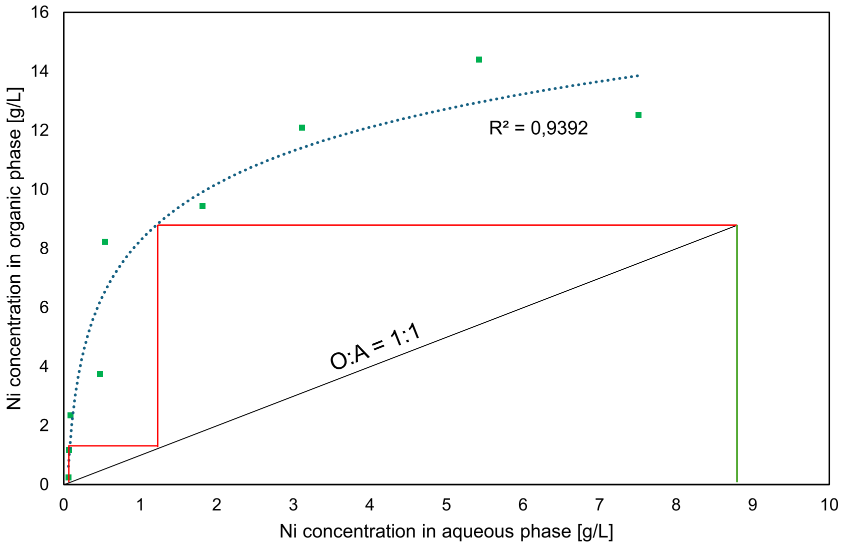


Fig. 3. M-C-T diagram for Ni extraction at 0.9 M Versatic Acid 10, equilibrium pH = 6.8 ± 0.1, T = 25 ± 1 °C, t_{eq} = 15 min, Operating line calculated for Ni extraction of 99.9% and $\theta = 1$.

Table 3
Elemental compositions after extraction with 0.9 M Versatic Acid 10.

Element	Co (PPM)	Li (PPM)	Na (PPM)	Ni (PPM)	Ni purity in loaded organic
Aqueous raffinate	ND	3715.07	54,903.59	36.08	
Loaded organic	43.48	6.82	32.90	8272.75	99.11%

3.1.2. *Kinetic study*

Extraction rate and volumetric mass transfer coefficient for nickel extraction were studied using 0.9 M Versatic Acid 10 at a phase ratio (θ) of 1. Fig. 2a and b illustrate the influence of mixing time on nickel extraction efficiency and the volumetric mass transfer coefficient, respectively. Initially, the pH was slightly higher (6.8 ± 0.1) after phase contact but stabilized at approximately 6.5 ± 0.1 within 15 min. The kinetics of nickel extraction were rapid, reaching equilibrium within approximately 3 min, which indicates a fast mass transfer process.

The initial increase in nickel extraction with mixing time reflects the rapid complexation reaction between Ni^{2+} ions and Versatic Acid 10, to form the nickel carboxylate complex that facilitates the transfer of nickel ions from the aqueous to the organic phase. The volumetric mass transfer coefficient was initially high, suggesting a strong driving force and efficient interfacial mass transfer, however, it declined sharply as the system approached equilibrium as the concentration gradient of Ni^{2+} ions between the two phases decreased, reducing the mass transfer driving force.

Approximately 90% of nickel was extracted within the first minute, demonstrating the high extraction rate and efficient kinetics of the process. This rapid extraction is likely controlled by chemical reaction rates at the interface, combined with convective mass transfer in the mixer-settler system. As nickel ions are quickly depleted from the aqueous phase, the mass transfer rate slows down, causing the observed decrease in the volumetric mass transfer coefficient over time.

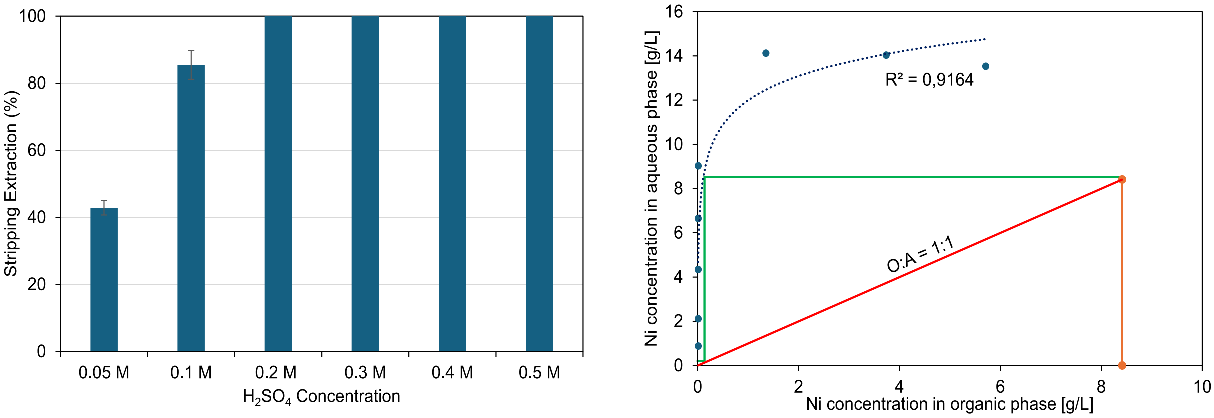
3.1.3. *Counter current extraction*

Graphical analysis of the McCabe-Thiele (M-C-T) diagram for the countercurrent Ni extraction is shown in Fig. 3. It was observed that two extraction stages are required for the counter-current Ni extraction when the organic to aqueous phase ratio is 1. The process was scaled up using a mixer-settler, utilizing the optimized parameters obtained from the batch-scale experiments. Based on these optimized extraction parameters, a two-stage mixer-settler system was constructed to facilitate the counter-current extraction of Ni. The extraction process using mixer-settler was performed with an organic-to-aqueous phase ratio of 1:1 and an equilibrium pH maintained at 6.8 ± 0.1 in both stages. Kinetic studies for the extraction of Ni revealed that only a short time (3 min) was required to reach the equilibrium. Therefore, the flow rate of both the organic and aqueous phases in the mixer-settler system was maintained at 15 mL/min resulting in a calculated residence time in the mixing chamber of 4 min (described in the supplementary information S1), which ensured sufficient contact time for phase interaction and equilibrium establishment.

Elemental composition of both the aqueous raffinate, and the loaded organic solution obtained after the mixer-settler extraction process are displayed in Table 3. Results indicate that 99.6% of nickel (Ni) was successfully extracted and transferred from the aqueous phase to the organic phase, demonstrating the high efficiency and selectivity maintaining 6.8 ± 0.1 pH and θ = 1. Moreover, under the same extraction conditions, the co-extraction of Li was negligible, highlighting that the system has selectivity for Ni over Li ions, which leads to minimal contamination and enhanced recovered Ni product purity. The presence of other metal impurities in the loaded organic phase was found to be negligible, confirming the high selectivity of the extraction system for Ni. Consequently, nickel purity in the organic phase increased to 99.11% (Table 3), demonstrating the efficiency of the extraction conditions in reducing the co-extraction of undesired metals.

3.1.4. *Stripping of the loaded organic*

As the purity of the nickel-loaded organic phase exceeded 99%, with lithium remaining unextracted and thus absent as an impurity, the scrubbing step was deemed unnecessary. Therefore, the process proceeded directly to the stripping stage to recover nickel from the organic phase. To determine the optimal stripping conditions, batch-scale experiments were conducted using sulfuric acid solutions of varying molar concentrations, as shown in Fig. 4a. It was observed that 0.2 M Sulfuric acid was sufficient to completely extract all the Ni ions from the loaded organic phase without any pH adjustment. As approximately 0.14–0.15 mL/L Ni was transported in the loaded organic phase during the extraction step, 0.1 M sulfuric acid was found to be insufficient to transport all the Ni from loaded organic due to a lack of H^+ ions as the stripping process involves the back-extraction of nickel ions from the organic phase into the aqueous phase by proton displacement. In this mechanism, hydrogen ions from the sulfuric acid react with the nickel carboxylate complex, breaking the metal-ligand bond releasing Ni^{2+} ions into the aqueous phase. The M-C-T diagram was constructed with 0.2 M sulfuric acid to determine the number of stripping stages required for the counter-current stripping process. According to Fig. 4b, it is observed that at least 2 stripping stages at θ = 1 are required to complete removal of Ni from the loaded organic phase. Consequently, a two-stage



(a) (b)

Fig. 4. (a) Various diluted concentrations of H_2SO_4 and (b) M-C-T diagram for stripping of loaded organic, Operating line calculated for Ni stripping of 99.9% and θ = 1.

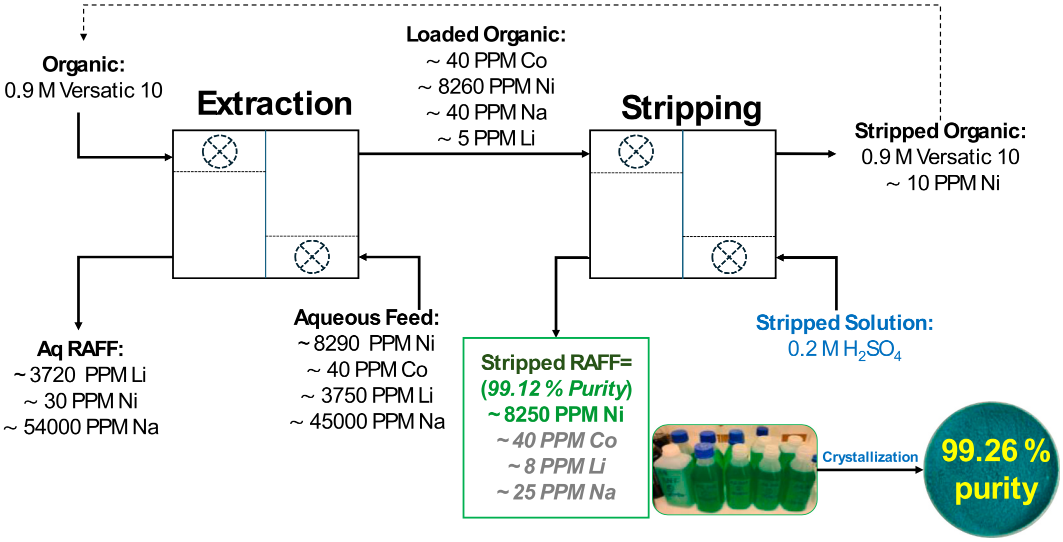


Fig. 5. Mass Balance for the Ni extraction using solvent extraction from PLS of the discarded NMC 111 batteries.

mixer-settler was constructed for the counter current stripping process, and this was attached to the extraction stage set-up.

3.2. Mass balance

A pilot run using the newly assembled mixer-settler set-up was undertaken to evaluate the parameters optimized during batch-scale laboratory extraction experiments. Aqueous PLS from the treatment of spent NMC 111 batteries, with a volume of 15 L, was processed through the mixer-settler under the conditions previously determined. In each extraction and stripping stage, the aqueous solution was initially introduced to approximately 50% of the stage volume, followed by the addition of the organic extractant into the extraction stages. pH regulation during the extraction process was achieved through the controlled addition of 10 M NaOH near the mixing zone to maintain favorable extraction conditions. In contrast, pH control was not implemented during the stripping process of the loaded organic phase. Samples were collected hourly from both the extraction and stripping chambers to monitor the process performance and analyzed by ICP-OES to determine the elemental concentrations.

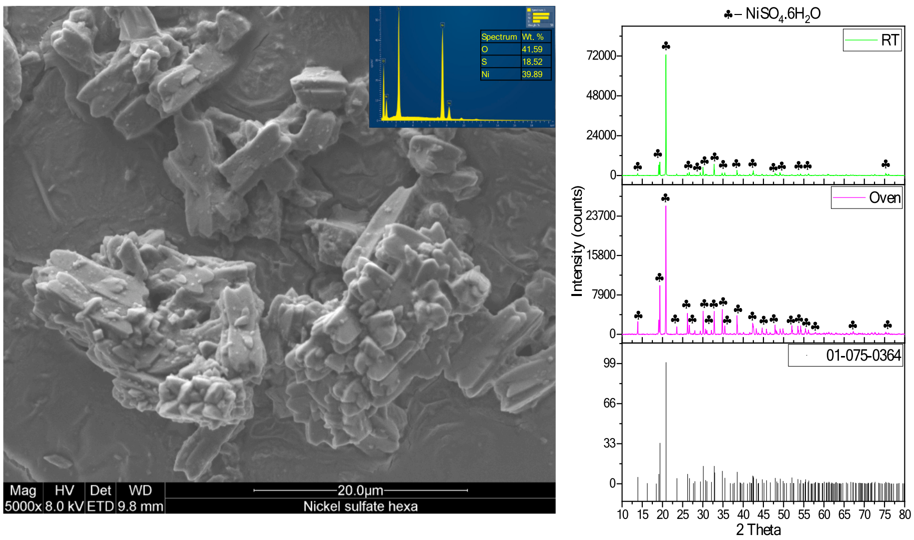
The extraction process resulted in the transfer of 99.63% Ni, 100% Co, 0.8% Li, and 0.08% Na from the aqueous phase to the organic phase and the mass balance of the solvent extraction of Ni using mixer-settler is displayed in Fig. 5. It is observed from Fig. 5 that behavior of Sodium (Na), which is also not transported to the organic solution with optimized condition. Na from feed as well as NaOH solution during the extraction was not transferred to the organic and it is reported to the aqueous raffinate. The increment of Na from feed solution (1.9 g/L) to aqueous raffinate solution (5.4 g/L) is because of addition of 10 M NaOH for maintaining the equilibrium pH during the extraction. The successful removal of Na also provides smooth extraction during mixer settler operation and subsequently helps to eliminate the scrubbing process. Concentration of cobalt in the feed solution was negligible (~ 40 ppm), as most of the Co had been previously removed in an earlier step, consequently, the loaded organic phase primarily contained Ni (~ 8250 ppm), with only minor amounts of Co and Na (~ 40 ppm each), and Li (~ 5 ppm). Given the low concentrations of co-extracted impurities, a scrubbing step was deemed unnecessary, therefore, the loaded organic phase was directly subjected to the stripping stage. Stripping was conducted using a 0.2 M H_2SO_4 solution, resulting in the efficient recovery of 99.87% Ni from the organic phase. A total of 99.12% pure Ni (~ 8250 ppm Ni) was successfully stripped from the organic phase, with only minimal levels of impurities, including Co, Na, and Li.

3.3. Crystallization

Stripped raffinate obtained from the mixer-settler operation was used for the recovery of high-purity Ni-sulfate salt. An evaporative crystallizer was employed to recover the salt at 45 °C at 35 RPM. The aspirator and reflux vacuum pressure were maintained constant at 0.1 MPa. A total of 6 L of stripped raffinate solution was introduced into the rotary evaporation flask and properly sealed before starting the process. As time progressed, the concentration of Ni in the solution increased with evaporation and Ni salt crystallization commenced when the degree of supersaturation of the solution exceeded a critical point. Increasing Ni concentration raises the supersaturation ratio, which helps generate a driving force for the necessary prerequisites for nucleation and growth, leading to crystallization [38,41]. After the process was complete, the crystallized salt, along with a small amount of residual supersaturated liquid, were removed from the rotary evaporative flask. This residual liquid may have contained sulfuric acid and Isopar L, as the boiling points of both liquids are higher than that of water. The product was thoroughly washed with ethanol to remove any contamination remnants and then subjected to vacuum filtration. Initially, this filter residue was kept in a desiccator at room temperature for 2 days and analyzed with XRD to allow phase determination. Following XRD, a sample of the recovered material was also placed in a drying oven at 60 °C for 5 h prior to product purity analysis. 214 g NiSO4.6H2O salt

Table 4
ICP-OES analysis of recovered products dried in room temperature (RT) and oven.

Condition	Al	P	Zn	Co	Ni	Li	Purity
	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(ppm)	(%)
RT	0.45	1.18	0.38	4.88	943.18	0.08	99.26
Error	±0.02	±0.06	±0.007	±0.04	±3.76	±0.002	±0.008
Oven	0.43	0.81	0.39	5.53	961.56	0.004	99.26
Error	±0.006	±0.04	±0.02	±0.09	±3.98	±0.009	±0.013



(a) (b)

Fig. 6. (a) SEM images with EDS and (b) XRD spectra of the recovered crystallized salt.

with 99.26% purity was recovered after washing and drying. The yield of the recovered NiSO$_4$6H$_2$O salt was calculated using Eq. 6 after the crystallization process. 95.8%. The detailed calculation for yield was determined in the supplementary information.

$$Yield\ (\%) = \frac{Actual\ amount\ obatined}{Theoretical\ amount\ possible} \times 100 \tag{6}$$

3.3.1. Characterization of the recovered crystallized salt

A solution of 1 g/L of crystallized nickel sulfate hexahydrate was prepared in the ultrapure water, and the composition was analyzed with ICP-OES. The elemental analysis and purity of the salt is shown in Table 4. From the results, it can be seen that nickel sulfate hexahydrate ($NiSO_4.6H_2O$) with a purity of 99.26 ± 0.01% was recovered through the sequential solvent extraction-crystallization process.

The recovered crystallized salt was characterized using SEM-EDS and XRD as shown in Fig. 6a and b respectively. The SEM micrograph reveals crystalline particles of nickel sulfate hexahydrate exhibiting a rod-like or prismatic morphology, densely clustered due to agglomeration which suggests significant interparticle interactions during the crystallization process. The accompanying EDS analysis confirms the elemental composition with 41.59% oxygen, 18.52% sulfur, and 39.89% nickel by weight, which corresponds to the expected composition of $NiSO_4·6H_2O$. XRD spectra for room temperature and oven dried product was characterized and only $NiSO_4.6H_2O$ peaks were observed in both the samples. Notably, the salt dried at room temperature resulted in high intensity diffraction peaks, however, lower intensity diffraction peaks were observed in the oven dried sample possibly as a consequence of product crystallinity deterioration due to heating.

The high purity (99.26%) of the crystallized nickel sulfate hexahydrate ($NiSO_4·6H_2O$) recovered has a strong potential for direct application as a precursor cathode active material (pCAM) for the fabrication of nickel-based lithium-ion batteries [41–43]. Furthermore, the efficient and selective recovery of high-purity nickel sulfate salt from real spent battery leach solutions presents a viable and sustainable pathway for the battery industry, aligning with the principles of a circular economy.

4. Conclusions

The current study focuses on investigating efficient recovery of Ni from the real spent NMC 111 batteries leach solution. PLS obtained after the separation of impurities, Mn and Co was used as the feed for the extraction of Ni utilizing Versatic Acid 10 diluted in the Isopar L. A kinetics study and volumetric mass transfer of the Ni with the time was undertaken using previously optimized extraction parameters and the findings demonstrated that a majority of the Ni was rapidly transferred to the organic solution within a short contact time (3 min). The counter-current process for the selective extraction of nickel was also conducted on a pilot scale using a mixer-settler, applying the optimized process parameters and M-C-T diagram data obtained from batch-scale experiments. An extraction efficiency of 99.63% Ni was achieved using 0.9 M Versatic Acid 10 with organic-aqueous phase ratio of 1:1 at 6.8 ± 0.1 pH in two stages. The scrubbing steps were omitted due to the absence of significant impurities in the loaded organic phase. Complete stripping of Ni from the loaded organic was achived at 0.2 M H_2SO_4 with $\theta = 1$ in two stages. The evaporative crystallization process was further used for the recovery of Ni salt from the aqueous raffinate obtained from mixer-settler operation. As a result, Ni-sulfate salt ($NiSO_4·6H_2O$) with a high purity of 99.26 ± 0.01% was recovered from the aqueous raffinate solution at 45 °C under a vacuum pressure of 0.1 MPa. The resultant high-purity crystallized nickel sulfate hexahydrate has significant potential for direct use in the production process of pCAM needed for the NMC batteries, thereby providing a pathway aligned with circular economy principles.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

org/10.1016/j.seppur.2026.137059.

Data availability

Data will be made available on request.

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