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Techno-economic and environmental assessments for hydrometallurgical treatment of waste lithium-ion batteries (LIBs): The ELiMINATE project

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Abstract: The increasing demand for consumer electronics and electric vehicles (EVs), in particular, results in a rapid increase in the production of end-of-life (EoL) lithium-ion battery (LIB) waste. Compared to pyrometallurgical treatment, hydrometallurgical treatment of LIBs offers several advantages for recovering strategic and critical metals. The ELiMINATE project, which is a multidisciplinary project, involved mapping of LIB recyclers located in the EU as well as life cycle assessment of hydrometallurgical processes using mineral acids (HCl, H_2SO_4) and certain organic acids for the recovery of metals from cathode active materials present in EoL LIBs. Material mapping and logistic network modelling studies for the recycling of EoL LIBs were carried out for different contexts (European and South African). These assessments were used to determine optimal locations for collection centres and processing plants. In addition, experimental tests were carried out to develop novel hydrometallurgical recycling processes using inorganic and organic acid solutions to leach metals in the presence of different reducing agents (H_2O_2 and $Na_2S_2O_5$). Further tests were performed to remove impurities and selectively recover metals from pregnant leach solutions in different salts/compounds. The developed processes at TRL 3-4 were then tested at a pilot scale (TRL 6) with the cooperation of EXITCOM Recycling, the SME partner of the project. The findings have demonstrated that high metal recoveries can be achieved at the leaching stage under optimum conditions, and that critical/strategic metals can be selectively separated/recovered using suitable methods.

Keywords: lithium-ion battery (LIB), life cycle assessment (LCA), material flow analysis (MFA), hydrometallurgy, recycling

1. Introduction

There is an increasing effort to realise the green transition through supporting research and investments to improve renewable energy and oil-free technologies and make their use more widespread (European Commission, 2019). The development and improvement in the technology of electric vehicles (EVs) also play a significant role in aligning with global climate targets since they contribute to reducing the carbon emissions produced by cars with internal combustion engines (ICE) (IEA, 2021). Lithium-ion batteries (LIBs) are the key component of EVs as they store energy through electrochemical reactions between cathode and anode. LIBs have already been used in portable electronic devices such as cell phones and

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laptops. However, the increasing demand for EVs is driving a significant expansion in their production, which also brings challenges related to the treatment (disposal and recycling) of end-of-life (EoL) LIBs.

Certain raw materials, which are defined as "Critical/strategic raw materials (CRM/SRM)" (Table 1) are crucial for the production of green transition technologies, including LIBs in EVs (European Commission, 2023; IRENA, 2023). The EU encourages recycling of these raw materials from secondary sources such that 15% of strategic raw materials consumed by EU countries should now be produced from recycling (European Commission, 2023). In addition, the European Union (EU) announced a new battery regulation, which mandates the incorporation of recycled raw materials from end-of-life batteries in the composition of newly manufactured batteries (European Union, 2023). In this regard, recycling of waste LIBs is of critical importance for the sustainable utilisation of essential raw materials for LIB production, i.e., lithium, nickel, cobalt, manganese, aluminium, copper, graphite, etc. Recycling these raw materials from waste LIBs is of paramount importance in establishing a more reliable and sustainable supply chain by reducing external dependence on raw materials and the negative impacts on the public and the environment (Baum et al., 2022; Circular Solutions, 2022). Recycling of CRM/SRM would also contribute to decreasing the demand for primary raw materials (Martinez et al., 2019; Pražanová et al., 2022).

Table 1. Critical/strategic raw material list of European Union (EU) (bold ones are also strategic raw material) (European Commission, 2023)

Aluminium/bauxite	Coaking coal	**Lithium**	Phosphorus
Antimony	Feldspar	**LREE (light rare earth elements)**	Scandium
Arsenic	Fluorspar	**Magnesium**	**Silicon metal**
Baryte	**Galium**	**Manganese**	Strontium
Berilium	**Germanium**	**Natural graphite**	Tantalum
Bismuth	Hafnium	Niobium	**Titanium metal**
Boron/Borate	Helium	PGM (platinum group metals)	**Tungsten**
Cobalt	**HREE (heavy rare earth elements)**	Phosphate rock	Vanadium
		Copper *	**Nickel ***

** Copper and nickel do not meet the CRM criteria but included in the list as SRM*

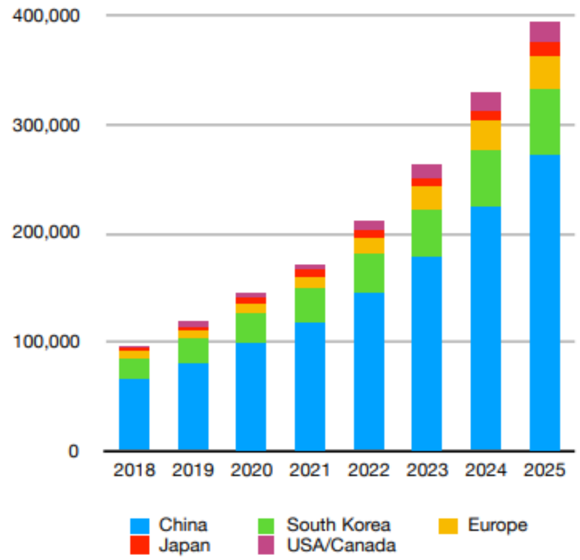


Fig. 1. Amount of lithium-ion batteries recycled (tonnes) (2018-2025) (Melin, 2020)

The increased utilisation of electric vehicles (EVs) worldwide has led to a concomitant rise in the amount of waste lithium-ion batteries (LIBs), with the recycling rate of batteries also showing an upward trend (Figure 1). China is the leading country in LIB recycling, followed by South Korea, the

European Union (EU), Japan, Canada and the United States of America (USA) (Fig. 1). However, current recycling rates remain low in comparison to the amount of waste LIBs generated worldwide. This is partly because there is a lack of recycling processes that satisfy technical, environmental and economic aspects (Martinez et al., 2019).

Four primary methods are employed to recover raw materials from EoL LIBs. These include direct recycling, physical separation, hydrometallurgy and pyrometallurgy-based methods (Martinez et al., 2019; Jena et al., 2021; Celep et al., 2023). Direct recycling aims to recover cathode/anode active materials (CAM/AAM) by preserving their chemical structure. Other methods enable the separation of the components of CAM/AAM by physical/chemical processes. Physical separation methods provide pre-separation before extractive metallurgical methods (hydrometallurgical and/or pyrometallurgical). Hydrometallurgical processes offer several benefits over other extractive metallurgical techniques, such as considerable operational flexibility with comparatively low to medium costs. Although many hydrometallurgical methods have been proposed for the recovery of metals from EoL LIBs, their techno-economic feasibility, possible environmental effects, and scale-up challenges continue to be major obstacles to their adoption to industrial scale (Celep et al., 2023; Jung et al., 2021; Musariri et al., 2019; Neumann et al., 2022; Pinegar and Smith, 2019; Rouquette et al., 2023). Even though inorganic acids such as H_2SO_4 and HCl have been commonly preferred, in recent years, organic acids such as oxalic acid ($H_2C_2O_4$), citric acid ($C_6H_8O_7$) and formic acid (CH_2O_2), ascorbic acid ($C_6H_8O_6$) and malic acid ($C_4H_5O_6$) have also been tested in the leaching of metals from LIB (Jena et al., 2021; Celep et al., 2023). Considering the fact that inorganic acids are generally cheaper than organic acids (Celep et al., 2023; Zhang et al., 2018; Zhao et al., 2019), they (particularly H_2SO_4) are predominantly utilised in industry for the extraction of metals from LIB through hydrometallurgical processes. Hydrogen peroxide (H_2O_2) exhibits strong oxidising or reducing properties and is frequently employed with inorganic acids. However, in addition to its high cost, peroxide consumption is relatively high due to its susceptibility to decomposition, particularly through the catalytic effect of metal ions at high temperatures (Yazici, 2005 and 2012). Innovative approaches with reduced environmental footprint are needed to tackle these challenges to improve the LIB recycling industry and build a circular economy. In this regard, research activities should be supported to develop technically, environmentally and economically sound recycling processes.

In this context, the aim of the ELiMINATE project (End-of-life Li-ion battery management integration and technology evaluation) which was supported under the ERA-MIN 2 Call, is to accomplish an implementation framework for establishing facilities for the recycling of metals from EoL LIBs (van Schalkwyk et al., 2023; Wu, 2022; Yazici et al., 2023). The project consortium is coordinated by Stellenbosch University (SUN; South Africa) with partners from Sweden (IVL Swedish Environmental Institute and Chalmers University of Technology (CUT)) and Türkiye (Karadeniz Technical University (KTU) and EXITCOM Recycling as the SME partner).

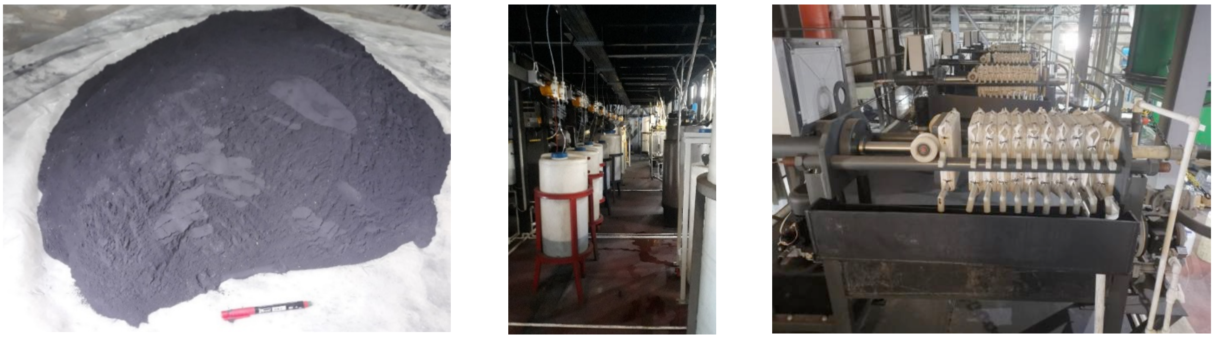
The ELiMINATE project covers the following areas of research (i) Comparative technical and environmental impact analysis of alternative hydrometallurgical processes for the recovery of critical/strategic metals from LIBs, (ii) Market analyses and business development to understand value chain integration for the selected processes within European and South African contexts, (iii) Material flow analyses and reverse logistics optimisation to improve resource efficiency of the recycling industry in Europe and South Africa and (iv) Development of novel hydrometallurgical processes at laboratory scale followed by their demonstration at pilot scale. This paper presents the key findings obtained during the studies carried out during the project.

2. Materials and methods

The project was executed through six work packages. IVL & CUT, and SUN conducted comprehensive studies to analyse the market structure and its estimated evolution within both European and South African contexts. The business case development followed a structured approach covering the needs-approach-benefit-competition framework, focusing on three key processes: (1) a mineral acid-based process, (2) an organic acid-based process, and (3) a thermal pre-treatment process, all aimed at maximizing value creation. Based on literature data, nine flowsheets were designed and modelled, utilising three selected two inorganic lixiviants (HCl, H_2SO_4) and one organic (citric acid), each paired with three downstream processing routes: sequential precipitation, direct production of mixed NMC, and sequential precipitation combined with solvent extraction (SX). Additionally, a tenth flowsheet was assessed, involving H_2SO_4-based leaching of black mass (NMC) following thermal pre-treatment. The black mass feedstock was assumed to be a composite of various LIB types, including NMC, LCO, and LFP. NMC, LCO and LFP refer to lithium nickel manganese cobalt oxide, lithium cobalt oxide, and lithium iron phosphate, respectively.

Life cycle assessment (LCA) studies were performed for the chosen flowsheets, integrating multivariate uncertainty and weak point analyses to identify optimal processes and potential alternative technologies. Screening LCAs were utilised to assess and compare different flowsheet options to select the most suitable processing option. A comprehensive LCA was performed for the selected processes within European and South African contexts to determine key process units requiring further development and evaluate their alignment with environmental regulations.

Novel hydrometallurgical processes were tested by KTU in the confines of the laboratory using a black mass sample (~120 kg) sourced from various cell phone brands at EXITCOM Recycling facilities (Fig. 2). The black mass was generated through a pre-treatment process that included size reduction, impurity removal via screening, and electrolyte removal. Chemical analysis of the sample indicated the presence of 3.25% Li, 23.6% Co, 2.06% Ni, 8.36% Mn, 1.78% Al, 0.92% Cu, and 0.28% Fe. In the project, hydrogen peroxide (H_2O_2) (as the common reductant used in LIB recycling) or sodium metabisulphite ($Na_2S_2O_5$) (as an alternative to H_2O_2) were employed in sulphuric acid (H_2SO_4) or organic acid (methanesulphonic acid; MSA) leaching tests. The impact of several factors on the leaching of metals (Co, Li, Ni, Mn, Cu, Al, and Fe) was thoroughly investigated. Leaching tests were followed by metal recovery from PLSs, for which the solvent displacement crystallisation (SDC) method was suitably adopted. The tests at a high technology readiness level (TRL 6) were also carried out for the novel processes developed at the pilot plant of EXITCOM Recycling.



(a) (b) (c)

Fig. 2. The black mass material produced from EoL LIBs at EXITCOM Recycling facilities and sent to KTU for hydrometallurgical tests (a); pilot plant facilities of EXITCOM (b, c)

3. Results and discussion

3.1. Value chain integration

In order to develop a business case plan to determine the best position of a LIB recycling process, market analysis was conducted to assess the recycling sectors in the European Union (EU) and South Africa (SA). Strategies for incorporating recycling businesses into the current value chain were also developed. According to the market analysis for South Africa (2022–2030), laptops and, to a lesser extent, mobile phones will be the main sources of waste LIB, with EVs making up a very small portion of the total. This suggests that the most common battery type will be LCO-type batteries, in which the dominant CRM is cobalt. It is relevant to note that there is no LIB recycling facility in South Africa. Future projections suggest that, for a single recycling plant to be economically viable, at least 37% of waste LIB must be recovered.

Recent assessments of the battery recycling market in Europe indicate an upward trend in terms of EoL LIBs generation, pre-processing, and material processing capacity. This growth is supported by European policies to ensure the secure supply of critical raw materials within the continent. The increasing number of EoL EVs is also expected to contribute to this growth. The adoption of nickel-based cathode chemistries in EVs is projected to rise, and the optimal locations for establishing recycling centres include Nordic countries and Germany. The optimal number of facilities required to establish these centres ranges from one to four, depending on the selected technology and the associated transportation costs. A comparison of battery markets in Europe and South Africa is presented in Table 2. In the South African context, nine hydrometallurgical processes were considered, and H_2SO_4-H_2O_2 leaching followed by the precipitation of a nickel-cobalt-manganese-hydroxide (NMC) mixture was identified as the optimal process option in terms of economic viability and environmental impact. The NMC and lithium carbonate produced in this process can be utilised to produce precursor cathode active material (*p*CAM).

The process was determined to have the least environmental impact (as measured by LCA) and the easiest implementation in terms of plant operability (Kuhn, 2023; Ozturk, 2024). The economic feasibility and business development studies of two novel hydrometallurgical LIB recycling processes for cathodes were evaluated. These processes were developed and tested at a pilot scale as part of the ELiMINATE project by KTU and EXITCOM Recycling, and three different business model scenarios were created in the EU context. A techno-economic assessment was also performed for each of the novel process technologies. The results showed that the different business model scenarios had a significant impact on the profitability of the two novel recycling processes. The most profitable scenario for both technologies was identified as one in which battery manufacturers possessed an in-house recycling facility and offered a buy-back option for their batteries.

Table 2. Comparison of the South African and European battery markets

	Europe	**South Africa**
Battery recycling market	Pre-processors and material recovery recyclers	No recyclers on the market
Future applications (+10 years)	Electric vehicles	Portable devices
Dominant chemistries	Nickel-based	LCO (i.e., cobalt)-based
Battery regulation	EU Battery Regulation: to set targets for material recovery and min. recycled content in new batteries	Extended producer responsibility (since May 2021), ban on battery waste landfilling (from August 2021)

3.2. Process modelling and life cycle assessment (LCA) of hydrometallurgical processes

Simplified process flowsheets were modelled using modelling/simulation software (Fig. 3). It is important to note that, during hydrometallurgical processing of LIBs, Ni, Mn, and Co remain in the leach solution after the impurity removal stage. In some of the process flowsheets (i.e., HCl-NMC, H_2SO_4-NMC, and citric acid-NMC), those metals are precipitated as an NMC hydroxide precursor material (*p*CAM) with adjusting the metal ratios (e.g., if the target chemistry is NMC 811 then proper additions of Ni/Mn/Co compounds should be made to maintain the 8:1:1 ratio), enabling the direct resynthesis of NMC cathode active materials (CAM) (Maritz, 2024; Maritz et al., 2025). LCAs were evaluated for the midpoint impact categories (indicators) of abiotic depletion (ADP elements), acidification potential (AP), eutrophication potential (EP), global warming potential excluding biogenic carbon (GWP), and ozone layer depletion potential (ODP) covered by Centrum voor Milieuwetenschappen (CML) 2001 methodology. Considering these midpoint indicators, endpoint values were calculated to quantify the overall environmental impacts of hydrometallurgical processes. The simplified representation of environmental impacts through endpoint values helps to interpret the key insights more easily. Standardised values (%) were calculated using endpoint values, considering the hydrochloric acid sequential precipitation process as the base process, having a value of 100%. Processes with standardised values greater than 100% therefore performed better on average in terms of its environmental impact compared to the HCl-sequential process. The results demonstrated that processes incorporating an SX stage generally exhibited the worst environmental impact, as indicated by the lowest standardised) values (Maritz et al., 2022; Maritz, 2024) (Table 3). Europe (i.e., EU) case demonstrated lower environmental impact in comparison to South African case, primarily due to the fact that electricity is the primary source of adverse impact and that the South African electricity supply mix is much more reliant on traditional coal-based electricity generation capacity compared to the EU where renewable energy contributes significantly to the electricity supply mix (Maritz, 2024).

Furthermore, thermal pre-treatment was found to adversely affect the environmental impact of the process. Processes with low standardised values were eliminated based on the results of screening LCAs and the selected four flowsheets (i.e., H_2SO_4 – mixed NMC precipitation, HCl – mixed NMC precipitation, citric acid – mixed NMC precipitation, and H_2SO_4 sequential precipitation) and novel hydrometallurgical processes developed by KTU were used in full LCA studies.

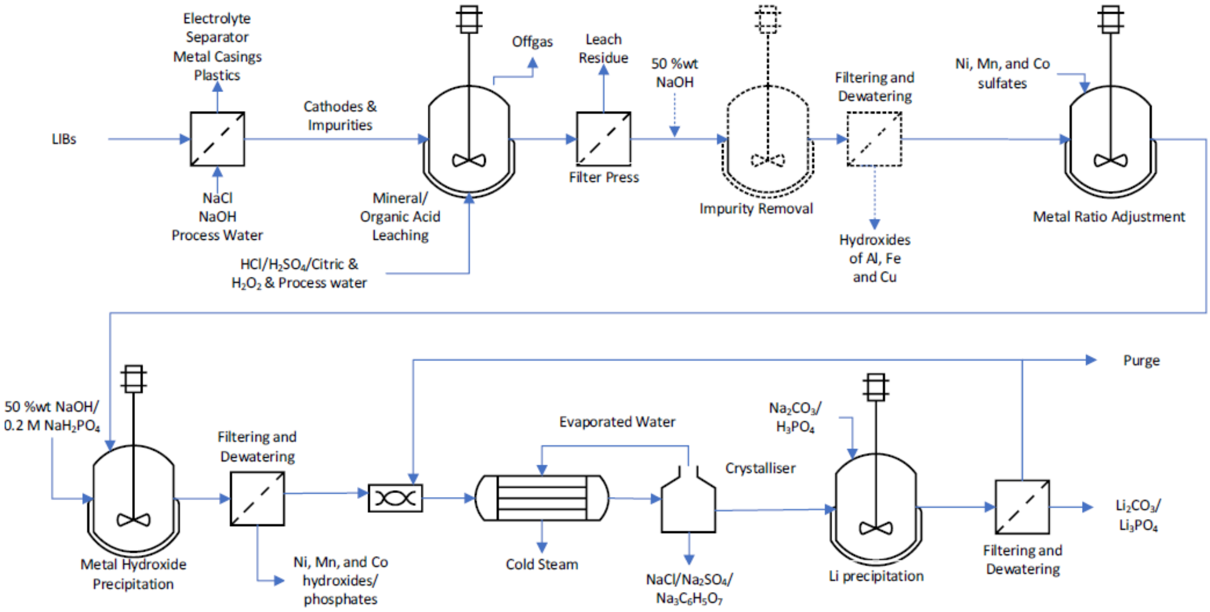


Fig. 3. Simplified process flowsheet for the metals recovery from EoL LIBs using mixed NMC precipitation (Maritz, 2024)

Table 3. Endpoint values for the nine hydrometallurgical process options (Maritz, 2024)

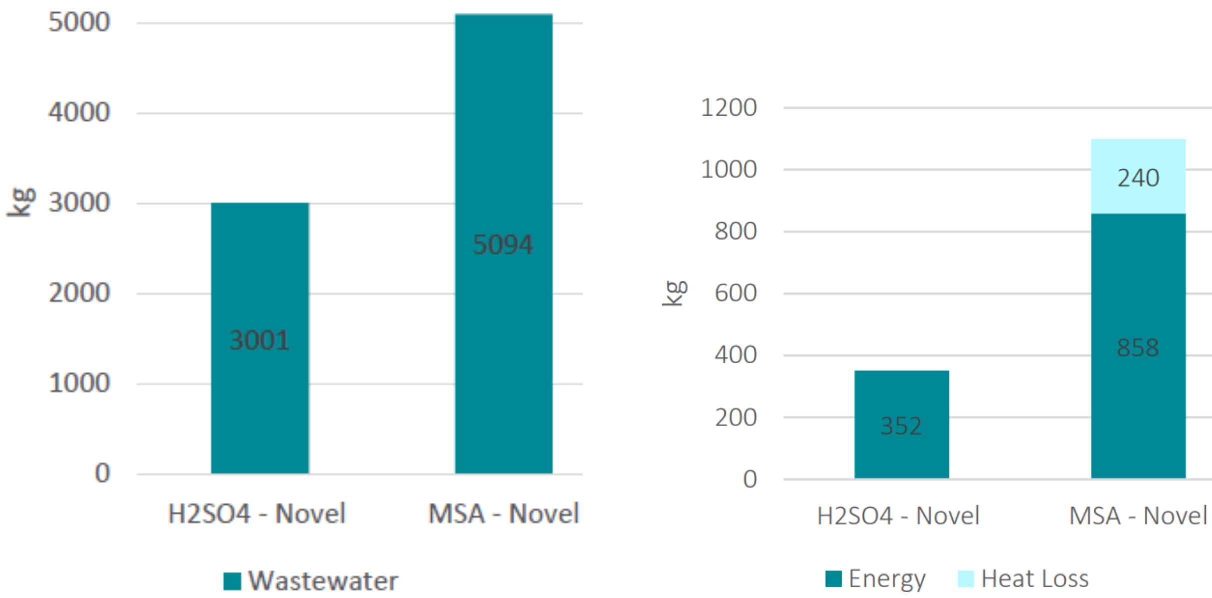
Process Type	**Endpoint Value (Pt)**	**Standardised Value**
Hydrochloric acid - NMC	-5.44E-09	169%
Hydrochloric acid - Sequential	-3.22E-09	100%
Hydrochloric acid - SX	-3.97E-09	123%
Sulphuric acid - NMC	-5.83E-09	181%
Sulphuric acid - Sequential	-5.01E-09	155%
Sulphuric acid - SX	-2.08E-09	64%
Citric acid - NMC	-3.88E-09	120%
Citric acid - Sequential	-3.35E-09	104%
Citric acid - SX	-1.47E-09	46%

The full LCA studies demonstrated that the H_2SO_4-mixed NMC process is more environmentally friendly than the novel technology (i.e., H_2SO_4 based novel process) developed in the project within the respective European and South African context (Table 4) (Ozturk, 2023; Elginöz Kanat et al., 2025a). Table 4 shows the normalised and standardised results relative to the value of novel technology (in EU context) with more negative values confirming a higher environmental benefit. The results also demonstrated that EU context is superior to South African in terms of environmental benefit (Table 4).

The comparison of the two novel processes indicated that the H_2SO_4-based novel process was superior to the organic acid-based novel process in terms of consumptions of lixiviant (leaching agent), raw materials (other reagents/materials used in the process), water and energy, as well as wastewater generation (Fig. 4) (Ozturk, 2023; Elginöz Kanat et al., 2025a).

Table 4. Normalised total LCIA results for current (H_2SO_4-mixed NMC) and novel technology (H_2SO_4 based novel process) based on the impact categories

Impact Categories	Novel Technology (EU)	Novel Technology (SA)	Current Technology (EU)	Current Technology (SA)
Abiotic Depletion Potential (ADP) of Elements	-100%	-100%	-140%	-139%
Acidification Potential (AP)	-100%	-99%	-119%	-118%
Eutrophication Potential (EP)	-100%	-97%	-126%	-126%
Climate Change (GWP)	-100%	-52%	-415%	-308%
Ozone Layer Depletion (ODP)	-100%	-100%	-126%	-126%



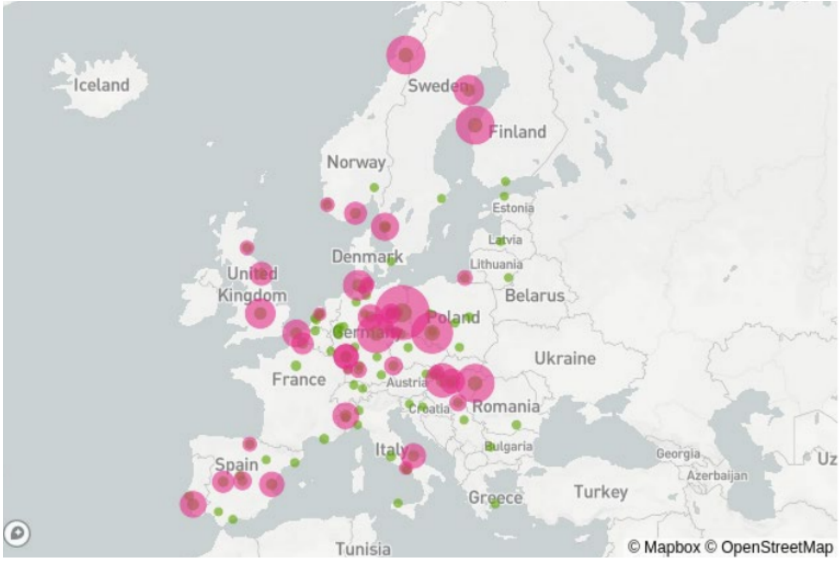
(a) (b)

Figure 4. Comparison of novel processes (H_2SO_4- and organic acid-based) in terms of leaching (Lixiviant) wastewater generation (a) and energy consumption (b)

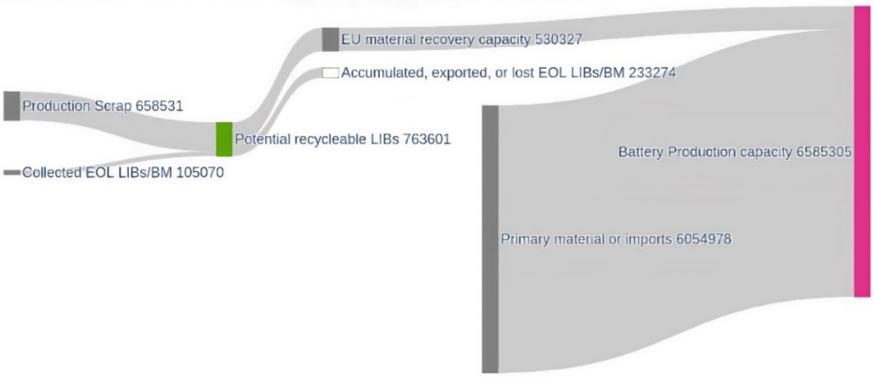
3.3. Material flow analysis and reverse logistics network modelling & optimisation

Material flow analysis and reverse logistics network modelling and their optimisation for EoL LIB recycling were performed by IVL & CUT and SUN for the European and South African contexts (Figs. 5-7). In Figs. 5-6 nodes serve as points on the map that signify different stakeholders. Each node possesses specific coordinates and manages the flow of EOL LIBs or materials associated with them.

The objective of the optimisation studies for a single- or multi-facility network was to determine the best reverse logistics network setup in order to minimise the economic burden. This was achieved by considering pre-treatment and developing one of the novel hydrometallurgical processes. The results showed that optimisation the number of processing facilities led to potential savings that ranged from 1-41% to over 100% of the minimum value. The developed reverse logistics network model shows the locations and capacities of collection points, dismantling centres, and pre-treatment and processing options (Figs. 6 and 7). Fig. 6a,b indicates that the current situation in EU LIB recycling activities mainly relies on pre-processing, however, it is projected that in 2030, the capacity will expand for material recovery. Fig. 6 also implies that Germany stands out as a key player in the recycling sector, followed by other EU countries. The transition from 2022 to 2030 reflects a growth in recycling infrastructure in Europe, likely driven by increasing demand for CRMs and regulatory obligations.

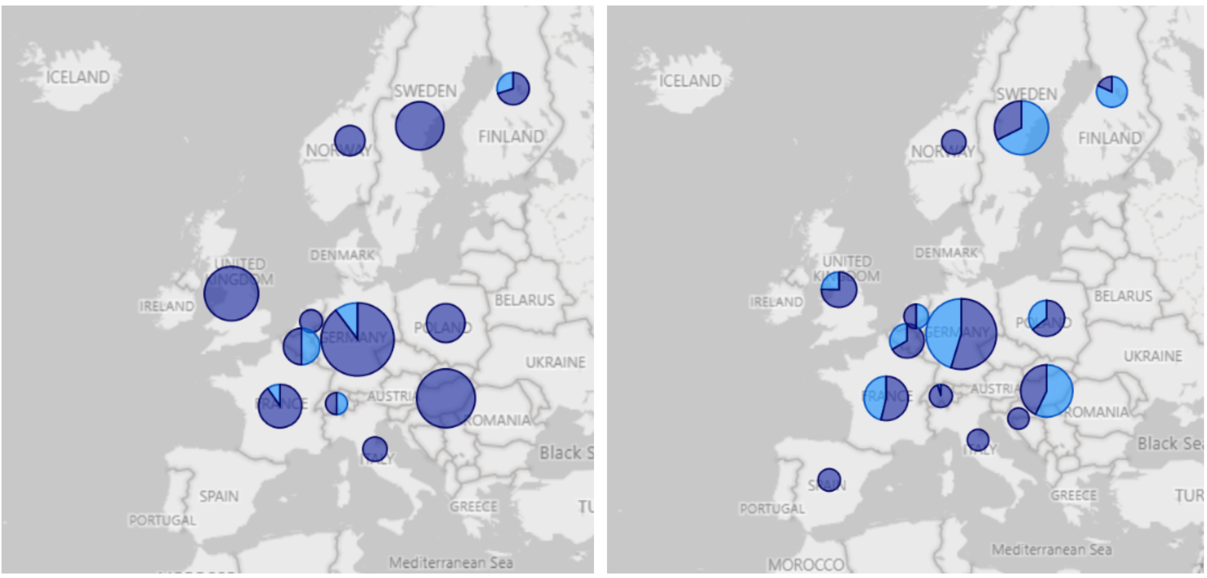


(a)



(b)

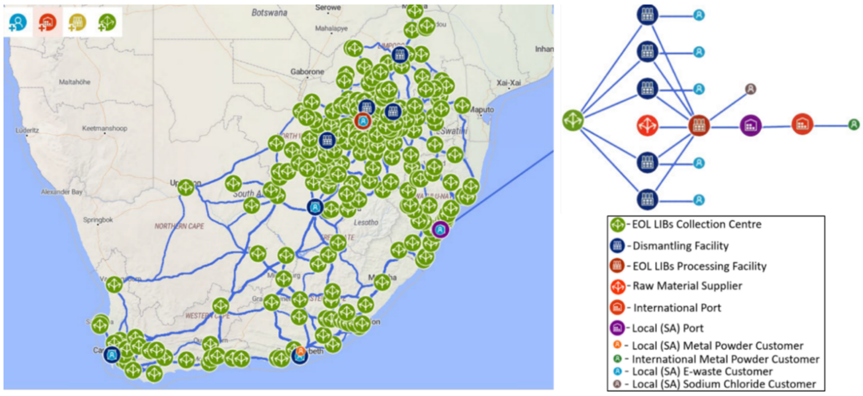
Fig. 5. Locations and relative volumes from supply (green-coloured) and demand nodes (pink-coloured) for 2030 (a) and Sankey chart for total recycling flows in comparison to battery production capacity across Europe, projected for 2030 (b) (Emillson and Ozturk, 2023)



(a) (b)

Fig. 6. Mapping of current (2022) (a) and forecasted (2030) (b) EoL LIB flows for material processing (in light blue) and pre-processing (in dark blue) within EU

Fig. 7 shows a reverse logistic network simulations (RLN) design for South Africa consisting of 212 EoL LIB collection centres, 9 dismantling facilities, and a single LIB processing facility, particularly for the HCl-NMC scenario. These centres and facilities were strategically placed to effectively balance CO_2 emissions and economic performance to establish a model for sustainable reverse logistics in battery recycling. The findings showed that the most convenient strategy to establish an efficient recycling network is to set up collection and dismantling centres for WEEE throughout the country, with a single recycling facility in Johannesburg (South Africa). The pre-treatment stages of LIBs should be integrated with the dismantling facilities to make the process more economic by increasing profit. The sale of e-waste from the dismantling facilities has the potential to significantly improve profitability. The discounted payback period and the projected internal rate of return (IRR) were estimated to be 11 years and 16.73%, respectively, depending heavily on the feed composition and LIB collection rates.



(a) (b)

Fig. 7. (a) The reverse logistics network diagram and (b) echelon structure showing preferred locations of EoL LIB collection centres, dismantling facilities and processing facilities for the HCl-NMC process scenario in the South African context.

3.4. Development and scale-up of novel hydrometallurgical processes

Leaching and solution purification and metal recovery tests were carried out to develop novel hydrometallurgical processes for the recovery of metals from EoL LIBs at a laboratory scale (Fig. 8). LIBs contain cobalt/nickel/manganese in CAM with varying ratios depending on the LIB type, in the oxidised forms. Therefore, applying the leaching process under reducing conditions is essential to achieve the dissolution of these metals at a high rate. For LCO ($LiCoO_2$) type LIBs, metals (i.e., Li and Co) dissolve in H_2SO_4 solutions (in the absence or presence of H_2O_2 as the reducing agent) according to Eqs. 1 and 2. In LIB production, metals such as Ni and/or Mn are also added in specific proportions to enhance the technical properties of the CAM to produce. The dissolution reactions of manganese-containing CAM in H_2SO_4/H_2O_2 are presented in Eqs. 3 and 4 (Jung et al., 2021):

$$2LiCoO_2 + 3H_2SO_4 \rightarrow Li_2SO_4 + 2CoSO_4 + 3H_2O + \frac{1}{2}O_2 \quad (1)$$

$$2LiCoO_2 + 3H_2SO_4 + H_2O_2 \rightarrow Li_2SO_4 + 2CoSO_4 + 4H_2O + O_2 \quad (2)$$

$$4LiMnO_2 + 6H_2SO_4 \rightarrow 2Li_2SO_4 + 4MnSO_4 + 6H_2O + O_2 \quad (3)$$

$$2LiMnO_2 + 3H_2SO_4 + H_2O_2 \rightarrow Li_2SO_4 + 2MnSO_4 + 4H_2O + O_2 \quad (4)$$

$$4LiMO_2 + Na_2S_2O_5 + 6H_2SO_4 \rightarrow 2Li_2SO_4 + 4MSO_4 + 2NaHSO_4 + 5H_2O \quad \text{(M: Co,Ni,Mn)}. \quad (5)$$

In the leaching tests, it was demonstrated that high recoveries (i.e., >90 %) can be achieved for metals, independent of the acid (H_2SO_4 or organic acid) or reducing agent (H_2O_2 or $Na_2S_2O_5$) under suitable conditions. Sodium metabisulphite ($Na_2S_2O_5$) was tested as an alternative reductant based on the fact that it decomposes to source strong reducing species (SO_3^{2-}, SO_2) (Vieceli et al., 2018). The dissolution of Co, Ni and Mn from cathode materials such as $LiCoO_2$ in the presence of $Na_2S_2O_5$ in sulphuric acid solutions proceeds according to Eq. 5. There have been relatively limited studies on the use of $Na_2S_2O_5$ in the leaching of LIBs. In accordance with the findings of the current study, numerous researchers (Sun and Qiu, 2011; Shin et al., 2005; Chen et al., 2011; Zhu et al., 2012; Jha et al., 2013; Meshram et al., 2015) reported that the recovery of metals is improved with the increase in reagent (H_2SO_4 and H_2O_2)

concentrations and temperature. However, H_2O_2 was superior to $Na_2S_2O_5$ due to the fact that it provides higher selectivity in the recovery of metals from pregnant leach solutions (PLS).

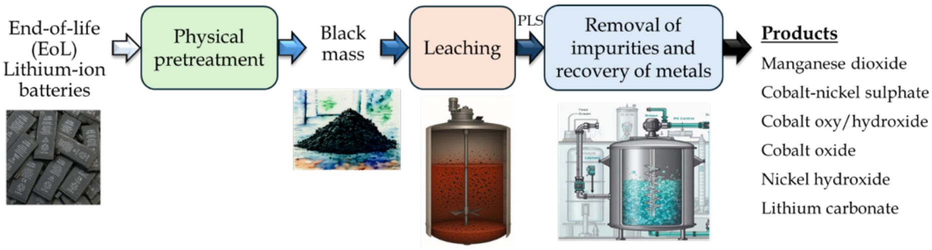


Fig. 8. General flowsheet of the developed H_2SO_4-based novel hydrometallurgical process

Following the leaching stage, suitable techniques involving different precipitation methods were utilised to separate/recover metals and obtain solid products, i.e., manganese dioxide, mixed Co-Ni-sulphate, cobalt (III) oxy/hydroxide, cobalt oxide (Co_3O_4), nickel hydroxide and lithium carbonate (Li_2CO_3). Developed hydrometallurgical processes were also successfully demonstrated at a high technology readiness level (TRL) of 6 with the collaboration of KTU and EXITCOM Recycling at the company's facilities located in Türkiye (Fig. 9). The innovative aspects behind the novel processes are the implementation of sodium metabisulphite ($Na_2S_2O_5$) as an alternative reducing agent to commonly investigated reagents and adoption of solvent displacement crystallisation (SDC) method in a hydrometallurgical process flowsheet. Patent applications for the developed novel hydrometallurgical processes are in progress.

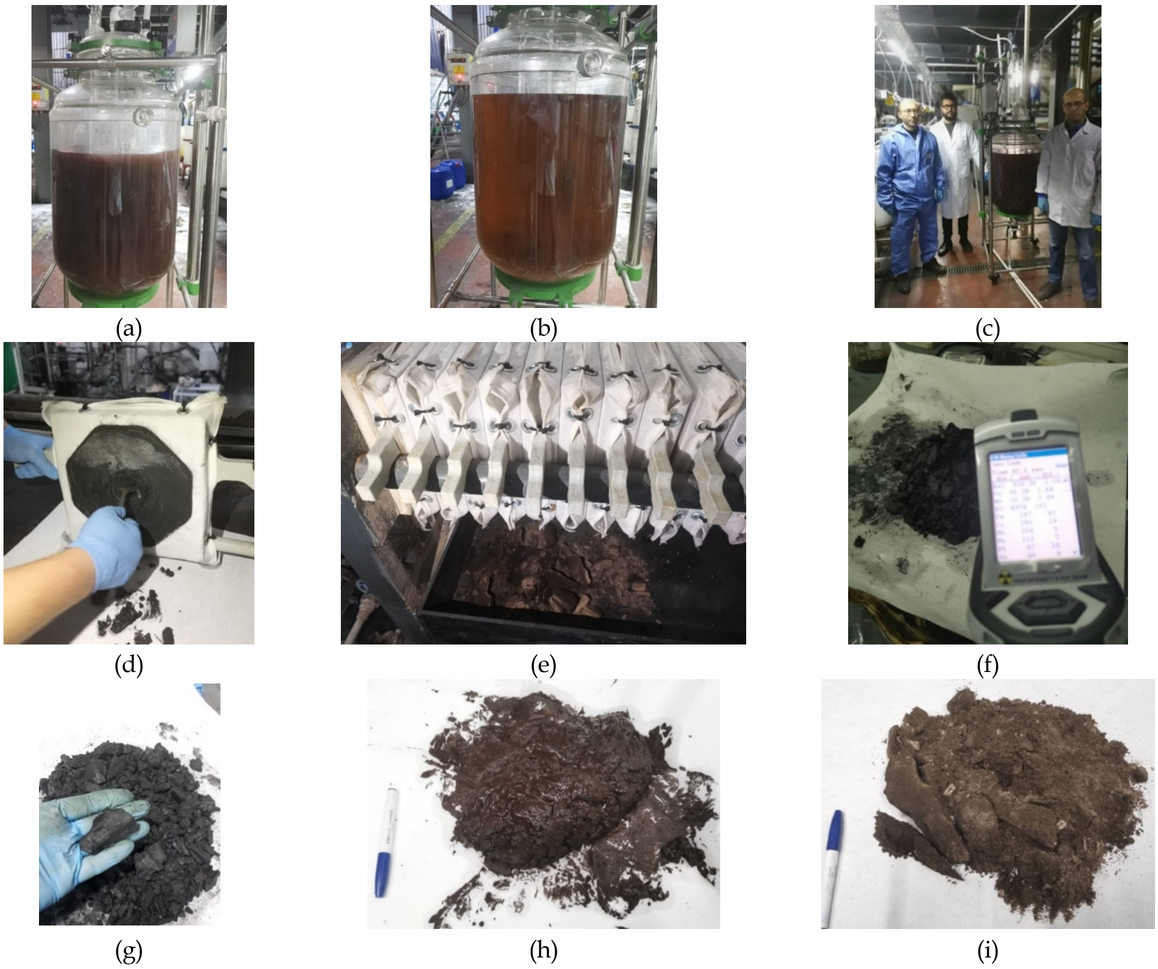


Fig. 9. Photos from the tests carried out at EXITCOM Recycling pilot plant (Kocaeli/Türkiye) (a, b, c, d, e, f) and some solid products produced from novel hydrometallurgical processes including graphite (g), manganese dioxide (h), mixed Co-Ni sulphate (i)

4. Conclusions

The multidisciplinary ELiMINATE project assessed a number of hydrometallurgical methods for the recovery of critical/strategic metal from black mass, as well as their effects on the environment, reverse logistics networks (RLNs), and value chain integration. A framework with recommendations for the development of LIB recycling industries in South Africa and Europe was brought. Market research clearly indicates that the demand for recycling is expected to rise. According to life cycle assessment (LCA) results, hydrometallurgical processes with a solvent extraction (SX) step often have a greater environmental impact than those without SX. The best methods in terms of economics and environmental effects were acid leaching followed by mixed NMC precipitation, according to techno-economic and LCA studies. The findings also indicated that the novel processes (H_2SO_4 leaching followed by different process stages including the use of solvent displacement crystallisation (SDC) for precipitation of metals) developed had a slightly higher environmental impact than the H_2SO_4-based leaching followed by the NMC precipitation technique. A high TRL of 6 was achieved by successfully demonstrating novel hydrometallurgical processes at the pilot plant.

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References

BAUM, Z.J., BIRD, R.E., YU, X., MA, J., 2022. *Lithium-ion battery recycling─overview of techniques and trends.* ACS Energy Lett 7, 712–719.

CELEP, O., YAZICI, E.Y., DEVECI, H., DORFLING, C., 2023. *Recovery of lithium, cobalt and other metals from lithium-ion batteries*. Pamukkale Univ Muh Bilim Derg 29 (4), 384-400.

CHEN, L., TANG, X., ZHANG, Y., LI, L., ZENG, Z., ZHANG, Y., 2011. *Process for the recovery of cobalt oxalate from spent lithium-ion batteries.* Hydrometallurgy 108, 80–86.

CIRCULAR SOLUTIONS (2022. https://www.cirbasolutions.com/news/lithium-ion-battery-recycling-benefits-and-risks-analyzed/.

EMILSSON, E., OZTURK, A. N., 2023. O*ptimizing Resource Recovery of Lithium-Ion Battery Recycling*, IVL Swedish Environmental Research Institute, Report no: C801

ELGINOZ KANAT, N., OZTURK, A. N., MARITZ, R., VAN SCHALKWYK, F., DORFLING, C., 2025a. *Full LCA report of selected process technologies for the South African and European contexts*, IVL Swedish Environmental Research Institute Ltd., Report no: C896

ELGINOZ KANAT, N., OZTURK, A. N., MARITZ, R., VAN SCHALKWYK, F., DORFLING, C., 2025b. *Techno-environmental screening of hydrometallurgical techniques for Li-ion battery recycling,* IVL Swedish Environmental Research Institute Ltd., Report no: C897

EUROPEAN COMMISSION, 2019. *Communication from the Commission to the European Parliament, The European Council, The Council, The European Economic And Social Committee And The Committee Of The Regions* – The European Green Deal, European Commission, Brussels, 11.12.2019, 24, https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1588580774040&uri=CELEX%3A52019DC0640

EUROPEAN COMMISSION, 2023. *European Commission, Study on the critical raw materials for the EU 2023* – Final Report. Luxembourg.

EUROPEAN UNION, 2023. *Regulation (EU) 2023/1542 of the European Parliament and of the Council of 12 July 2023 concerning batteries and waste batteries*, amending Directive 2008/98/EC and Regulation (EU) 2019/1020 and repealing Directive 2006/66/EC, Document no: Document 02023R1542-20240718, http://data.europa.eu/eli/reg/2023/1542/2024-07-18

IEA (2021. *The role of critical minerals in clean energy transitions*, International Energy Agency (IEA), Paris.

IRENA, 2023. *Geopolitics of the energy transition: Critical materials, International Renewable Energy Agency*, Abu Dhabi.

JENA, K. K., AFANTAZI, A., MAYYAS, A. T., 2021. *Comprehensive Review on Concept and Recycling Evolution of Lithium-Ion Batteries (LIBs)*, Energy&Fuels, 35, 18257-18284.

JHA, M.K., KUMARI, A., JHA, A.K., KUMAR, V., HAIT, J., PANDEY, B.D., 2013. *Recovery of lithium and cobalt from waste lithium ion batteries of mobile phone*. Waste Manag. 33, 1890–1897.

JUNG, J.C.Y., SUI, P.C., ZHANG, J., 2021. *A review of recycling spent lithium-ion battery cathode materials using hydrometallurgical treatments*. J Energy Storage 35, 102217.

KUHN, M. W. J., 2023. *Towards a business model for lithium-ion battery recycling in South Africa*, MSc thesis, Department of Industrial Engineering, Stellenbosch University

MARITZ, R.F., 2024. *Comparing the environmental impact of different hydrometallurgical processes for the recycling of lithium-ion batteries using a life cycle assessment approach*, PhD thesis, Department of Chemical Engineering, Stellenbosch University

MESHRAM, P., PANDEY, B., MANKHAND, T., 2015. *Recovery of valuable metals from cathodic active material of spent lithium ion batteries: Leaching and kinetic aspects*. Waste management, 45, 306-13.

MARITZ, R.F., VANSCHALKWYK, R.F., ELGINÖZ, N., AKDOGAN, G., DORFLING, C., 2022. *Investigating options for reducing the environmental impact of lithium-ion battery recycling plants using life cycle assessment*, Circular Materials Conference, 14-15 September, Skellefteå, Sweden.

MARITZ, R.F., VANSCHALKWYK, R.F., ELGINÖZ, N., AKDOGAN, G., DORFLING, C., 2025. *Using life cycle assessment to aid process development for hydrometallurgical recycling of end-of-life lithium ion batteries*, Waste Management 200, 114763

MARTINEZ, V. O., VALIO, J., SANTASALO-AARNIO, A., REUTER, M., SERNA-GUERRERO, R., 2019. *A critical review of Lithium-Ion battery recycling processes from a circular economy perspective*. Batteries, 5(4), 68.

MUSARIRI, B., AKDOGAN, G., DORFLING, C., BRADSHAW, S., 2019. *Evaluating organic acids as alternative leaching reagents for metal recovery from lithium ion batteries*. Miner Eng 137, 108–117.

NEUMANN, J., PETRANIKOVA, M., MEEUS, M., GAMARRA, J.D., YOUNESI, R., WINTER, M., NOWAK, S., 2022. *Recycling of lithium-ion batteries – current state of the art, circular economy, and next generation recycling*. Adv Energy Mater, 2102917.

OZTURK, A. N., 2023. *Techno-economic assessment and business development of two novel Li-ion battery recycling technologies*, MSc thesis, Uppsala University

PRAŽANOVÁ A., KNAP V., STROE. D. I., 2022. *Recycling of Lithium-Ion Batteries from Electric Vehicles, Part II: Environmental and Economic Perspective. Energies*. Energies 2022, 15(19), 7356

PINEGAR, H., SMITH, Y.R., 2019. *Recycling of end-of-life lithium ion batteries, part I: commercial processes*. Journal of Sustainable Metallurgy 5, 402–416.

ROUQUETTE, L.M.J., PETRANIKOVA, M., VIECELI, N., 2023. *Complete and selective recovery of lithium from EV lithium-ion batteries: Modeling and optimization using oxalic acid as a leaching agent*. Sep Purif Technol 320, 124143.

SHIN, S.M., KIM, N.H., SOHN, J.S., YANG, D.H., KIM, Y.H., 2005. *Development of a metal recovery process from Li-ion battery wastes*. Hydrometallurgy 79, 172–181.

SUN, L., QIU, K., 2011. *Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries*. J. Hazard. Mater. 194, 378–384.

VAN SCHALKWYK, R., YAZICI, E. Y., OTS, J., EMILSSON, E., PETRANIKOVA, M., EKEN TORUNOGLU, E., WU, A., DEVECI, H., AKDOGAN, G., LOUW, L., ELGINÖZ KANAT, N., MARITZ, R., VOOGHT, D., KUHN, T., OZTURK, A. N., LASSESSON, H., DORFLING, C., 2023. *End-of-life li-ion battery management ıntegration and technology evaluation*. 2nd International Conf. on Raw Materials and Circular Economy (RawMat 2023), 28 August- 2 September, Athens, Greece

VIECELI, N., NOGUEIRA, C.A., GUIMARÃES, C., PEREIRA, M.F.C., DURÃO, F.O., MARGARİDO, F., 2018. *Hydrometallurgical recycling of lithium-ion batteries by reductive leaching with sodium metabisulphite*. Waste Manag. 71, 350–361.

WU, A., 2022. *The ELiMINATE project: How to integrate battery recycling innovations in future society?*, Circular Materials Conference, 14-15 September, Skellefteå, Sweden.

YAZICI, E. Y., DEVECI, H., CELEP, O., DORFLING, C., VAN SCHALKWYK, R. F., EMILSSON, E., WU, A., PETRANIKOVA, M., EKEN TORUNOGLU, E., CUHADAR, E., EYUPOGLU, C., 2023. *Preliminary findings of the ERA-MIN 2 project (ELiMINATE) on the management and treatment of end-of-life lithium ion batteries*, The 5th International Conference of Materials and Engineering Technology (TICMET'23), 13-16 November, Trabzon, Türkiye

YAZICI, E.Y., 2005. *Removal of cyanide from waters using hydrogen peroxide, activated carbon adsorption and ultrasonic waves,* MSc Dissertation, Graduate School Natural and Applied Sciences, Karadeniz Technical University, 141p (In Turkish.

YAZICI, E.Y., 2012. *Recovery of metals from electronic wastes using physical separation and hydrometallurgical methods*. PhD Thesis, Graduate School Natural and Applied Sciences, Karadeniz Technical University, 192p., In Turkish)

ZHANG, W., XU, C., HE, W., LI, G., HUANG J., 2018. *A review on management of spent lithium ion batteries and strategy for resource recycling of all components from them.* Waste Management &Research, (36)2, 99-112.

ZHAO, J., QUA, X., QUA, J., ZHANGA, B., NINGA, Z., XIEA, H., ZHOUA, X., SONGA, Q., XINGA, P., YINA, H., 2019. *Extraction of Co and* Li_2CO_3 *from cathode materials of spent lithium-ion batteries through a combined acid-leaching and electro-deoxidation*. Journal of Hazardous Materials, 379, 120817.

ZHU, S.G., HE, W.Z., LI, G.M., ZHOU, X., ZHANG, X.J., HUANG, J.W., 2012. *Recovery of Co and Li from spent lithium-ion batteries by combination method of acid leaching and chemical precipitation*. Trans. Nonferrous Met. Soc. China 22, 2274–2281.