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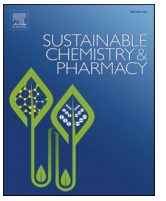

# Electrodialytic lithium extraction from secondary resources: Technical, environmental and economic assessment

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ABSTRACT

In transportation, large-scale electrification, particularly through lithium-ion batteries, is expected to drive significant emissions reductions while simultaneously increasing global lithium demand. However, the limited availability of lithium, compounded by geopolitical constraints, poses risks to the energy transition, namely in the European Union. Adopting circular economy models offers a sustainable approach to increase resource recovery. The present research aims to assess the potential of the electrodialytic process for lithium recovery from wastewater generated during lithium-ion battery recycling and aluminium-lithium alloy dust processing, as well as related environmental and economic impacts. Bench-scale experiments were conducted using two-compartment electrodialytic reactors, operated at 50 mA, 100 mA and 200 mA with a cation-exchange membrane interposed. Tests were performed over 24 h, 48 h and 72 h. Lithium recovery reached 91.54 % from the aluminium-lithium alloy dust and 97.23 % from the wastewater of lithium-ion battery recycling. The cradle-to-gate life cycle assessment resulted in 0.26 kg of $CO_2$ eq/g Li global warming impacts for wastewater of lithium-ion battery recycling, and 46.40 kg of $CO_2$ eq/g Li for aluminium-lithium alloy dust. Material flow cost accounting showed lower recovery costs for wastewater (0.36 €/g Li versus 129.26 €/g Li). Energy consumption in the reactor is the primary hotspot, where optimizing energy and time efficiency could reduce environmental and economic impacts.

| Abbreviation | Designation |
|---|---|
| 2C | Two Compartments |
| AL | Aluminium-Lithium Alloy Dust |

(*continued on next page*)

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(*continued*)

| Abbreviation | Designation |
|---|---|
| CAM | Cathode Active Material |
| CEM | Cation-Exchange Membrane |
| $CO_2$ | Carbon Dioxide |
| CRM | Critical Raw Material |
| DES | Deep Eutectic Solvents |
| ED | Electrodialytic |
| EU | European Union |
| EV | Electric Vehicle |
| EQ | Equivalent |
| GHG | Global Greenhouse Gas |
| GWP | Global Warming Potential |
| ICP-OES | Inductively Coupled Plasma Optical Emission Spectrometry |
| Li | Lithium |
| LCA | Life Cycle Assessment |
| LCI | Life Cycle Inventory |
| LCIA | Life Cycle Impact Assessment |
| LFP | Lithium-Iron-Phosphate |
| MFCA | Material Flow Cost Accounting |
| NMC | Nickel-Manganese-Cobalt |
| RPM | Rotations Per Minute |
| SC | Super Critical |
| SRM | Strategic Raw Material |
| Ti/MMO | Titanium/Metal Mixed Oxides |
| TRL | Technology Readiness Level |
| WWLB | Wastewater from Li-Ion Battery Recycling. |

## 1. Introduction

Global greenhouse gas (GHG) emissions increased by 1.3 % in 2023 compared to 2022, surpassing the average annual growth rate of 0.8 % observed between 2010 and 2019 (IEA et al., 2024). The power sector remains the largest contributor, emitting 15.1 $GtCO_2e$, followed by the transportation sector with 8.4 $GtCO_2e$ (United Nations Environment Programme, 2024). Since 1990, emissions from transportation have risen by nearly 80 %, now accounting for over 20 % of global $CO_2$ emissions from fossil fuel combustion (Filonchyk et al., 2024). Road transportation is the leading source, responsible for approximately 12 % of global GHG emissions in 2021, ranking second only to coal-fired power. Within this category, light-duty vehicles (LDV) are the primary contributors, with emissions growing by an average of 1 % annually since 2010, reaching over 3.5 $GtCO_2$ (Statista Research Department, 2024).

Achieving the global climate target of limiting temperature rise to 1.5 °C above pre-industrial levels requires a profound transformation of the transportation sector ((IPCC) Intergovernmental Panel on Climate Change, 2022). Achieving the global climate target of limiting temperature rise to 1.5 °C above pre-industrial levels requires a profound transformation of the transportation sector (IPCC, 2022). In this context, electric vehicles (EV) are central to decarbonisation strategies, given their significantly lower life cycle impacts compared to conventional internal combustion engine vehicles. Despite this potential, EV represented only 10 % of global new LDV sales in 2022. To align with the 1.5 °C scenario, this share must increase to at least 75 % by 2030 (Statista Research Department, 2024).

Battery production plays a crucial role in determining the environmental performance of EV. Effective decarbonisation policies, standardised life cycle assessment (LCA) methodologies, and enhanced supply chain transparency are essential. Currently, electricity-related emissions account for 20–25 % of the life cycle impacts of nickel-manganese-cobalt (NMC) and lithium-iron-phosphate (LFP) batteries. Addressing this requires further electrification of the battery supply chain beyond the existing 20–25 %, along with improvements in energy density and increased adoption of recycling practices (International Energy Agency, 2024).

Critical and strategic raw materials (CRM and SRM), such as lithium (Li), are indispensable for clean energy technologies and are included in the European Union's 2023b CRM list (European Commission, 2023a; Pires et al., 2023). Lithium is particularly important for the decarbonisation of the transport sector due to its use in Li-ion batteries. However, its limited availability and the geopolitical risks associated with its supply pose significant challenges, namely for the EU, which relies heavily on imported Li. In response, the EU's Critical Raw Materials Act(European Commission, 2024a) has set a target to recover 25 % of annual CRM consumption from recycled sources by 2030, aiming to foster circular supply chains and stimulate recycling industries(European Commission, 2024b). Achieving this goal requires diversifying pathways for CRM recovery.

Lithium is primarily extracted either from brines or hard rock sources, both of which are processed into stable compounds such as lithium carbonate or lithium hydroxide, suitable for storage and transport (Souza et al., 2025). Hard rock extraction is energy-intensive due to the processing and high-temperature treatment involved (Lappalainen et al., 2025; Mas-Fons et al., 2024), whereas brine extraction is comparatively lower-cost and uses solar evaporation. However, it is also associated with high water consumption, where between 39 and 70 $m^3$ of water are needed to produce 1 tonne of lithium carbonate (Lagos et al., 2024; Lappalainen et al., 2025; Mousavinezhad et al., 2024).

Driven by the European battery regulation (European Commission, 2023b), research into secondary lithium recovery has focused largely on recycling end-of-life batteries (Kang et al., 2023). More recently, attention has turned to alternative secondary sources across

various industries, which could play a critical role in expanding the availability of recycled Li. Nevertheless, research in this area remains limited.

A recent study demonstrated the feasibility of using an electro-based technology to recover lithium from mine tailings, presenting a promising approach to CRM recovery (Almeida et al., 2025). The ED process involves applying a low-intensity electric current across electrodes, generating an electric field that selectively migrates ions through ion-exchange membranes. This technique not only enables CRM recovery but also removes toxic elements and generates hydrogen gas, supporting broader sustainability goals (Almeida et al., 2020a,b). In this context, ED has been evaluated for its potential to recover Li from two distinct secondary sources: aluminium (Al)–Li alloy dust and wastewater from Li-ion battery recycling. The study investigated operational parameters — such as pH, electrolyte composition, current intensity, stirring rate, and processing time — to optimise lithium recovery, improve purity, and minimise environmental and economic burdens.

To support sustainable technology development, environmental and economic impacts were assessed using a cradle-to-gate LCA of the laboratory-scale ED process. Life cycle assessment (LCA) evaluates the environmental performance of products and systems across their life cycle—from raw material extraction to production, use, and end-of-life. The methodology consists of four phases: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation (ISO, 2006a, 2006b). In parallel, Material Flow Cost Accounting (MFCA) was applied to measure physical and cost flows of materials and energy, identify inefficiencies, and highlight opportunities for improvement (Pinto et al., 2022).

This study aims to: (i) explore the potential of closed-loop systems to enhance resource recovery; (ii) demonstrate the technical feasibility of recovering lithium from both liquid and solid secondary sources to reduce dependency on primary raw materials; and (iii) evaluate the environmental and economic performance of the ED process to identify key improvement areas. These elements are critical to advancing the technology readiness level (TRL) of emerging lithium recovery technologies and supporting the transition toward a more circular and sustainable battery value chain.

## 2. Materials and methods

### 2.1. Initial characterisation of samples

In the present study, both solid and liquid secondary Li resources were selected for investigation. The solid resource comprises Al–Li alloy dust (AL), a by-product extensively used in the aerospace industry. This waste material arises during the manufacturing of structural components from rolled, drawn, or forged semi-finished products and during the scalping of ingots for rolling or the skinning of billets in Al and titanium (Ti) processing facilities [7]. The liquid resource pertains to wastewater generated during the recycling processes of Li-ion batteries (WWLB).

The Li content, pH, and conductivity of the samples were evaluated for the initial characterisation. For the solid sample, these parameters were assessed in a water suspension with a liquid-to-solid ratio (L:S) of 40. Measurements of pH and conductivity were conducted using an EDGE electrode meters (HANNA Instruments, Rhode Island, USA).

Lithium concentrations were quantified using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) (HORIBA Jobin-Yvon Ultima, Japan) at the Chemical Analysis Laboratory of REQUIMTE/LAQV, NOVA School of Science and Technology, NOVA University Lisbon, Portugal. Lithium analysis by ICP-OES followed quality assurance (QA) and quality control (QC) protocols. The calibration curves were prepared with certified standards with $R^2 > 0.995$. Blanks were run to verify contamination. The samples were analysed in triplicate, with relative standard deviation (RSD) below 5 %. The recoveries for spikes ranged from 90 % to 110 %, corroborating accuracy. In each batch, certified reference materials were included, with results within ±10 % of certified values. The stability of instruments was regularly monitored, and interferences were evaluated and corrected as necessary. Detection and quantification limits were determined from blanks and low-level standards to guarantee method sensitivity.

### 2.2. Bench-scale electrodialytic experiments

The bench scale experiments were carried out in two (2C)-compartment ED acryl XT reactors (RIAS A/S, Roskilde, Denmark). The cylindrical reactors have an internal diameter of 8 cm, and 5 cm length in both electrolyte and sample compartments. The compartments were separated by a cation-exchange membrane (CEM), CR67, MKIII, Blank (Ionics, USA). The electrodes were made of Ti/MMO (titanium/mixed metal oxide) Permaskand wire with 3 mm in diameter, and 50 mm in length (Grønvold & Karnov A/S, Denmark). A power supply E3612A (Hewlett Packard, Palo Alto, USA) maintained a constant current of 50 mA, 100 mA, or 200 mA in the ED reactor. A volume of 250 mL of the sample was added to the anode compartment. All the experiments were conducted in a reactor not fully closed, in a fume hood at room temperature (approx. 24 °C). Appropriate ventilation was applied to mitigate any risk from potentially harmful gases. Additionally, the electrolyte composition and concentration were selected to reduce such side reactions.

For AL tests, a sample suspension at a L:S of 40 was prepared with deionised water (Water purification system from Thermo Fisher Scientific, Waltham, USA). A stirrer (Heidolph, Germany; VELP Scientifica Srl, Italy) was placed into the sample compartment, at 200 rpm, to prevent the sample from settling during the experiments. Fig. S1, in supplementary materials, depicts the ED configurations tested.

For the experiments, 250 mL of sodium nitrate ($NaNO_3$) or sodium sulfate ($Na_2SO_4$), at a concentration of 0.02 mol/L, was introduced as catholyte in the electrolytes' compartment. The solid sample suspensions were prepared with deionised water, AL, and different assistant agents, as listed in Table 1.

The experiments were conducted over durations of 24 h, 48 h, and 72 h. The ED reactors were placed within a fume hood (Industrial Laborum Ibérica, Portugal) and maintained at room temperature (~24 °C). Throughout the experimental period, the voltage of each reactor was recorded, and pH and conductivity of the electrolyte(s) and sample compartments were monitored daily. Samples were periodically extracted from the reactors, analysed using HANNA EDGE pH and conductivity meters, and subsequently reintroduced into the ED system.

### 2.3. Reagents and lithium determination

At the end of each experimental run, the ED reactor was disassembled. The electrodes and membranes were subjected to an immersion process in nitric acid ($HNO_3$) at concentrations of 5 mol/L and 1 mol/L, respectively, for a duration of 48 h to facilitate the complete removal of $Li^+$. Subsequently, the volumes of acid used for each electrode and membrane were quantified, and samples were collected for ICP-OES analysis.

To achieve the separation of solid and liquid phases from the AL tests, vacuum filtration was performed using a laboratory vacuum pump (KNF LABOPORT, Freiburg, Germany) connected to a 500 mL vacuum filtration flask (DURAN, West Germany). The solution was filtered through a Whatman filter paper (pore size: 11 μm; diameter: 150 mm, United Kingdom) positioned on a Büchner funnel. The collected liquid phase was stored in a clean, dry location at room temperature (~24 °C) until further analysis via ICP-OES.

The final solid phase was collected and allowed to dry under room conditions within a fume hood for one week. Subsequently, the dried sample underwent microwave-assisted digestion using a Milestone Ethos Easy Microwave Digestion Platform (Bergamo, Italy). The digestion process followed the EPA3051A (Environmental Protection Agency, 2007) microwave extraction protocol, wherein 0.5 g of the dried sample was placed in digestion vessels along with 9 mL of $HNO_3$ and 3 mL of hydrochloric acid (HCl), within a controlled fume hood environment. Following the completion of the microwave-assisted digestion program and subsequent cooling of the vessels, the resulting digests were subjected to vacuum filtration using MFV5 glass microfiber filters (pore size: 0.7 μm; diameter: 47 mm; Filter Lab, Barcelona, Spain). The filtrates were then diluted to a final volume of 25 mL with deionised water before undergoing ICP-OES analysis. For the bench scale tests, the reagents used are summarised in Table S1 (in supplementary materials section).

The ICP-OES measurements provided quantitative determination of Li content, expressed in mg/L. Hence, the percentage of $Li^+$ recovery ($Li_{Rec}$) for Al–Li alloy dust was obtained according to eq. (1):

$$Li_{Rec}(\%) = \frac{Li^+_{catholyte} + Li^+_{Liquid\ \ phase}}{Total\ \ Li^+} 100\% \quad (1)$$

and eq. (2) for wastewater from Li-ion battery:

$$Li_{Rec}(\%) = \frac{Li^+_{catholyte}}{Total\ \ Li^+} 100\% \quad (2)$$

### 2.4. Life cycle assessment

An LCA was conducted to quantify and analyse the potential environmental impacts of the ED process on a laboratory level. The inventory processes were collected from the life cycle database Ecoinvent v.3.10 (Wernet et al., 2016), using the Simapro software v9.6.0.1 (Database and Support Teams at PRé Sustainability, 2024) to connect to the background data in each flow.

The goal was to determine the main environmental hotspots of the ED Li recovery process, focusing on the maximum amount of Li recovered from the samples. A cradle-to-gate approach was followed with a functional unit of 1 g of Li recovered, serving as a reference for impact quantification. The LCI phase involved primary data collection to ensure the reliability and accuracy of the assessment,

**Table 1**
Conditions tested during electrodialytic experiments.

| Sample | Code | Current intensity (mA) | Time (h) | Assisting agent/ treatment | Stirring (rpm) | Electrolyte |
|---|---|---|---|---|---|---|
| Al–Li alloy dust (AL) | A1 | 100 | 72 | – | 200 | $NaNO_3$ 0.02 mol/ L |
| | A2 | | | ChCl:OA (1:1) | | |
| | A3 | | | ChCl:MA (1:1) | | |
| | A4 | | | Super Critical $CO_2$ | | |
| | A5 | | | OA 0.1 mol/L | | |
| | A6 | | | OA 0.5 mol/L | | |
| Wastewater from Li-ion battery recycling (WWLB) | W1 | 50 | 72 | – | – | $Na_2SO_4$ 0.02 mol/ L |
| | W2 | 100 | 48 | | | |
| | W3 | 200 | 24 | | | |
| | W4 | 50 | 72 | – | – | $NaNO_3$ 0.02 mol/ L |
| | W5 | 100 | 48 | | | |
| | W6 | 200 | 24 | | | |
| | W7 | 50 | 48 | | | |
| | W8 | 50 | 24 | | | |
| | W9 | 100 | 24 | | | |

considering all mass and energy flows at the laboratory scale, wastes and consumables. Energy consumption was calculated based on the maximum rated power of each equipment and the duration of its operation throughout the process.

The created LCI models were used to calculate the environmental impacts through the ReCiPe midpoint (H) quantification method (Huijbregts et al., 2017), focusing on global warming potential (GWP) and water consumption. These categories were selected based on the main impacts of the activities related to Li extraction and production (Halkes et al., 2024; Souza et al., 2025).

A sensitivity analysis was conducted to include the Li content variations between sample batches, to showcase how this affects the impacts of each sample. The obtained results aim to validate the environmental benefits of Li retrieval from waste streams and its contribution to reducing the overall impacts of EV Li-ion batteries.

An MFCA was performed to estimate the total cost per gram of Li recovered through ED separation for each sample. This analysis considered only the direct costs associated with material and energy consumption. Consistent with the LCA, the WWLB and AL samples were treated as "burden-free" in the MFCA, implying that they were not assigned any monetary value. Given that the analysis was based on a laboratory-scale process characterised by a low TRL, costs related to equipment, infrastructure, and waste management were excluded. These factors are not critical for decision-making in the context of technological upscaling.

The primary objective of this assessment was to identify the main cost drivers associated with Li recovery from secondary liquid and solid resources. Finally, based on the technical, environmental and cost assessment, a critical discussion on upscaling Li recovery from secondary resources was conducted, focusing on key drivers and barriers.

## 3. Results and discussion

### 3.1. Initial characterisation

To evaluate the impact of media conditions on the recovery processes, the initial samples were analysed for pH, conductivity, and the concentrations of Li, Al, potassium (K), phosphorus (P), and Na. The elemental composition was quantified using ICP-OES. Table 2 provides the initial characterisation data, wherein the pH and conductivity of AL were measured at a L:S ratio of 40 using deionised water.

The initial conductivity of the samples was measured at 20.97 mS/cm for AL and 43.29 mS/cm for WWLB. These values indicate that the samples can facilitate current passage during the ED process without requiring conductivity-enhancing agents. The pH values of the samples were 12.47 for AL and 13.06 for WWLB, indicating a strongly alkaline environment. The extraction efficiency of Li from secondary resources is significantly influenced by solution chemistry, particularly pH and conductivity. While each parameter independently affects Li recovery, their combined effect plays a crucial role in optimizing process performance.

Lithium concentration, as determined through ICP-OES, was found to be 2706.13 ± 196.07 mg/kg in AL and 1796.02 ± 60.35 mg/L in WWLB. During the ED process, interactions between Li and other elements present in the samples may lead to the formation of Li-containing compounds. Additionally, the use of assisting agents could promote the formation of Li complexes. As indicated in Table 2, there is a presence of K, Al, and Na in the samples. However, stable compounds with Li may not be formed due to the repulsive interactions among cations (Nie et al., 2023).

### 3.2. Electrodialytic experiments

The ED process was tested for Li recovery from AL and WWLB. For the solid sample, deep eutectic solvents (DES), supercritical (SC) $CO_2$ treatment, and an organic acid (oxalic acid at different concentrations) were considered (Table 1). The effect of the electrolyte composition in Li ED separation was addressed for the liquid sample, considering 0.02 mol/L of $NaNO_3$ and of $Na_2SO_4$. The ED process was performed according to Fig. S1 (supplementary materials), at room temperature (approximately 24 °C). The variations of pH, conductivity, and voltage in the ED reactor were measured before and after the experiments. Fig. 1 shows pH fluctuation in the reactor compartments.

In the ED process, the pH of the system is influenced by water electrolysis at the electrodes. At the cathode, hydroxide ions ($OH^-$) are generated, leading to an increase in pH, while at the anode, hydrogen ions ($H^+$) are produced, resulting in a decrease in pH (Almeida et al., 2025; Ribeiro and Rodríguez-Maroto, 2006). Consequently, during the ED process, the pH at the anode compartment (where the sample is placed) becomes acidic, and the pH at the cathode compartment becomes alkaline.

For the AL (Fig. 1a), a final pH from 1.00 (A6) to 3.7 (A3) was observed in the anolyte (sample) compartment, while the catholyte

**Table 2**
Initial characterisation of Al–Li alloy dust (AL) and wastewater from Li-ion battery recycling (WWLB).

| Parameter | AL | WWLB |
|---|---|---|
| pH | 12.47 ± 0.33 | 13.06 ± 0.07 |
| Conductivity (mS/cm) | 20.97 ± 1.60 | 43.29 ± 0.92 |
| Elements | mg/kg | mg/L |
| Li | 2706.13 ± 196.07 | 1796.02 ± 60.35 |
| Al | 82987.17 ± 4857.37 | 5.74 ± 0.16 |
| K | 52021.13 ± 4367.51 | 62.70 ± 2.48 |
| P | 135.11 ± 18.21 | 23.56 ± 1.32 |
| Na | 40164.69 ± 3485.52 | 229.65 ± 147.63 |

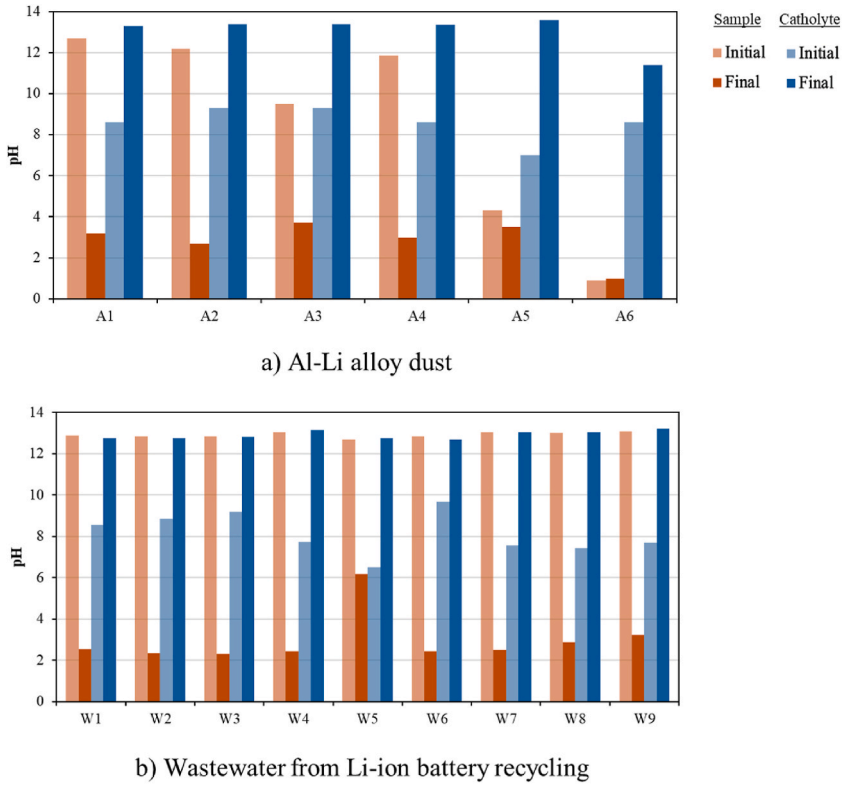



**Fig. 1.** pH variation during the electrodialytic experiments of (a) Al–Li alloy dust: A1 - no assisting agent; A2 - ChCl:OA; A3 - ChCl:MA; A4 - SC $CO_2$; A5 - OA 0.1 mol/L; A6 - OA 0.5 mol/L, and (b) wastewater from Li-ion battery recycling: $Na_2SO_4$ [W1 - 50 mA, 72 h; W2 - 100 mA, 48 h; W3 - 200 mA, 24 h], $NaNO_3$ [W4 - 50 mA, 72 h; W5 - 100 mA, 48 h; W6 - 200 mA, 24 h; W7 - 50 mA, 48 h; W8 - 50 mA, 24 h; W9 - 100 mA, 24 h].

exhibited pH values between 11.4 (A6) and 13.6 (A5). When an organic acid (OA) solution at a concentration of 0.5 mol/L was added to the sample compartment, a more acidic pH was observed both at the start and at the end of the ED process, with values ranging from 0.9 to 1.00. The acid dissociation constants (pKa) of oxalic acid (a diprotic acid) are $pKa_1 = 1.46$ and $pKa_2 = 4.40$, indicating higher acidity compared to other organic acids (PubChem, 2024). A lower pKa corresponds to a greater propensity for proton donation of the acid, favouring the formation of soluble metal complexes (Van Der Hagen and Järnberg, 2009), which may have accounted for the observed behaviours.

For WWLB (Fig. 1b), after the ED process, the pH in the anolyte was found to be below 6.2, while the catholyte exhibited a pH above 12.7. The electrolyte composition slightly affects pH. $Na_2SO_4$ generates a relatively neutral or slightly acidic environment due to its weak interaction with $H^+$ and $OH^-$. However, at the anode, sulfate ions ($SO_4^{2-}$) may lead to mild acidification due to sulfuric acid ($H_2SO_4$) formation (Karami et al., 2010). The final pH registered for the sample was W1 = 2.5, W2 = 2.4 and W3 = 2.3. On the other hand, $NaNO_3$ tends to shift pH in the cathodic region due to nitrate reduction, which can lead to the production of alkaline species as $NH_3$ (Bouzek et al., 2001). The final sample pH ranged from 2.4 (W4) and 6.2 (W5).

The pH controls the solubility and chemical speciation of Li and other coexisting metal ions. In AL, acidic conditions typically enhanced the dissolution of Li-bearing phases by promoting proton-driven breakdown of metal oxides or phosphates. Similarly, in WWLB, pH affected the stability and reactivity of $Li^+$, influencing both precipitation and separation pathways. The solubility of substances, such as hydroxides, oxides, and carbonates, is influenced by the pH of the solution (Król et al., 2020). In the sample compartment, the acidic pH may promote the dissolution of Li from the sample and its electromigration in the form of $Li^+$ toward the cathode compartment. On the other hand, conductivity reflects the ionic strength of the medium and directly impacts ion mobility. The conductivity behaviour during the ED experiments is presented in Fig. 2.

The initial conductivity of WWLB is higher than the solid sample (Table 2). Conductivity in the reactor is a crucial factor in facilitating current flow and promoting the electrochemically induced extraction of metals (J. Almeida et al., 2020a,b). As conductivity increases, resistance decreases, favouring ions' electromigration (Khedkar et al., 2016).

After all the experiments, the conductivity in the sample compartment decreased, while the conductivity in the catholyte increased. This change indicates the electromigration of ions from the sample compartment towards the electrolyte end. For AL, the higher initial conductivity was observed when OA at 0.5 mol/L (A6) was added to the sample compartment (59.6 mS/cm). Regarding W experiments, $Na_2SO_4$ dissociates into $Na^+$ and $SO_4^{2-}$ ions, where $SO_4^{2-}$ has a lower ionic mobility than nitrate $NO_3^-$, generated by the dissociation of $NaNO_3$ (Wu et al., 2015). This can lead to slightly lower conductivity compared to $NaNO_3$ at the same concentration. At the end, the final conductivity in the sample ranged from 0.01 (W8) to 1.4 (W7), and in the catholyte between 13.1 (W4) and 43.1

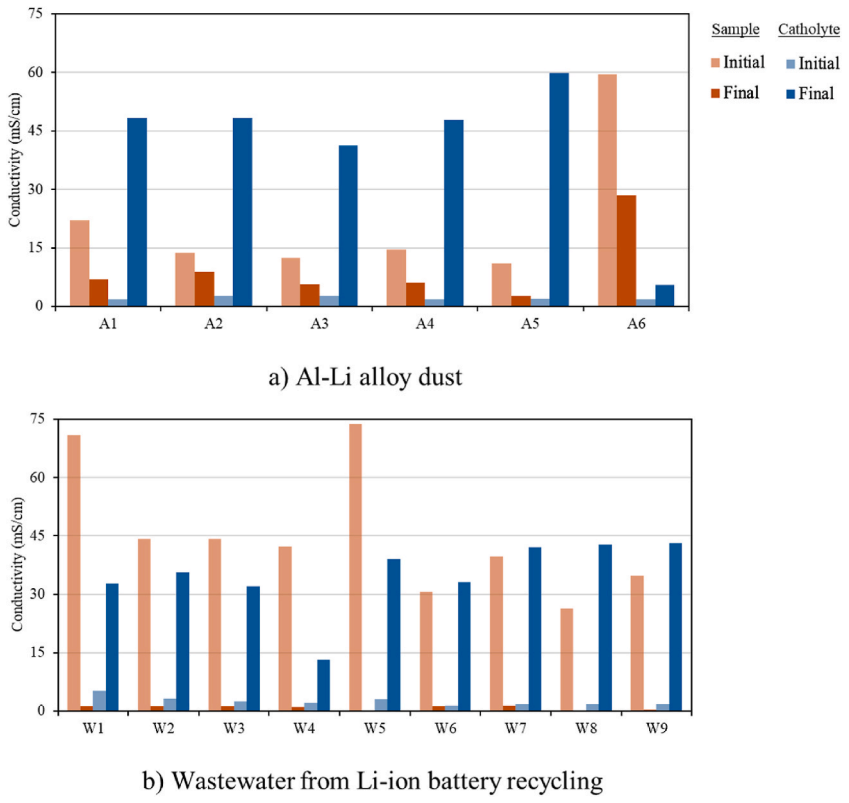



**Fig. 2.** Conductivity variation during the electrodialytic experiments of (a) Al–Li alloy dust: A1 - no assisting agent; A2 - ChCl:OA; A3 - ChCl:MA; A4 - SC $CO_2$; A5 - OA 0.1 mol/L; A6 - OA 0.5 mol/L, and (b) wastewater from Li-ion battery recycling: $Na_2SO_4$ [W1 - 50 mA, 72 h; W2 - 100 mA, 48 h; W3 - 200 mA, 24 h], $NaNO_3$ [W4 - 50 mA, 72 h; W5 - 100 mA, 48 h; W6 - 200 mA, 24 h; W7 - 50 mA, 48 h; W8 - 50 mA, 24 h; W9 - 100 mA, 24 h].

(W9). The relation between pH and conductivity determines the overall efficiency of ion transport, selectivity, and reaction kinetics. In the ED process, an optimal conductivity level is needed to facilitate efficient ion migration, while maintaining a pH range that prevents undesirable side reactions or precipitation. Moreover, improper pH control in highly conductive environments may lead to co-extraction of competing ions (such as $Na^+$, $Mg^{2+}$, or $Ca^{2+}$), thereby reducing Li purity. Voltage variation during the ED was also analysed, as shown in Fig. 3.

In general, the voltage increased after all the experiments, except for A6 and W8. For AL the highest voltage increase was observed for A5, from an initial value of 8.8 V–98.4 V. On W tests, the highest voltage increase was reported for W2 (11.7 V–53.9 V). According to Ohm's law, as ohmic resistance increases, so does the voltage increase. Ohmic resistance is caused by the ion migration and by friction of ions with the membranes and water during transfer between solutions (Strathmann, 1995). Therefore, the increase observed in the voltage is related to a conductivity increase in the electrodes' compartments (Almeida et al., 2021). The ED process involves complex compound formation and electrodes reactions, which requires high driving force to proceed efficiently. Although high voltages can lead to increased energy usage, based on LCA results, improvement actions aim to further adjust the process to improve efficiency and reduce energy input while maintaining product quality. The results obtained for Li recovery are presented in Table 3.

The maximum Li recovery from AL was achieved when OA was added to the sample compartment at 0.5 mol/L (A6). Approximately 92 % of the Li was solubilised in the liquid phase of the sample compartment. The properties of the bidentate oxalate ion may have contributed to form oxalates complexes with Li, since OA may react with other solubilised elements (Krishnamurty and Harris, 1961). Al–Li alloys are a class of low density, high strength and high stiffness monolithic metallic materials (Rao and Ritchie, 1992). The high reactivity of Li and the appearance of the surface oxide layer result in the continuous oxidation of the active metal during storage (Cao et al., 2020). This sample characteristics may have facilitated Li recovery.

The ED results obtained Li recovery efficiencies between 61.21 % (W8) and 97.23 % (W5) for WWLB. The solubility of Li is essential for its recovery (Almeida et al., 2025). Based on the highest recovery rates observed for each sample (Table 3), the final Li concentration in the electrolyte solution was 1746.3 mg/L for the WWLB test (W5) and 49.71 mg/L for the Al experiment (A6). The increased solubility of the targeted element in the liquid sample, turn it into a more mobile ionic species capable of electromigration towards the cathode compartment.

Additionally, a CEM positioned between the cathode compartment and the sample suspension functioned as a selective barrier, preventing the migration of negatively charged ions into the sample while permitting the passage of cations, such as $Li^+$, from the sample suspension into the cathode compartment. This selective ion transport enhances the specificity of the final solutions by

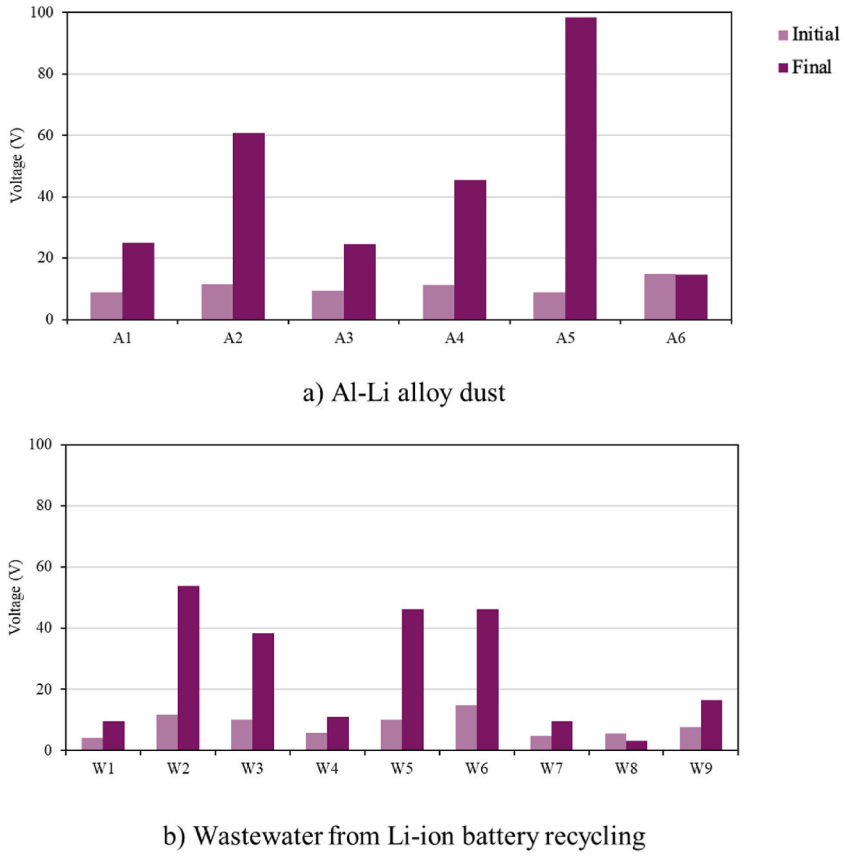



**Fig. 3.** Voltage variation during the electrodialytic experiments of (a) Al–Li alloy dust: A1 - no assisting agent; A2 - ChCl:OA; A3 - ChCl:MA; A4 - SC $CO_2$; A5 - OA 0.1 mol/L; A6 - OA 0.5 mol/L, and (b) wastewater from Li-ion battery recycling: $Na_2SO_4$ [W1 - 50 mA, 72 h; W2 - 100 mA, 48 h; W3 - 200 mA, 24 h], $NaNO_3$ [W4 - 50 mA, 72 h; W5 - 100 mA, 48 h; W6 - 200 mA, 24 h; W7 - 50 mA, 48 h; W8 - 50 mA, 24 h; W9 - 100 mA, 24 h].

facilitating targeted cation separation (Ribeiro and Rodríguez-Maroto, 2006).

The Li recovered in the catholyte can be subsequently separated and reused in various applications through methods such as precipitation, adsorption, ion-exchange, solvent extraction, or membrane electrolysis (Wu et al., 2023). Comparing to conventional methods, the ED process offers advantages in terms of chemical-free operation, lower secondary waste generation, energy efficiency for specific applications and potential for selective recovery. This could be beneficial for Li recovery technologies development. Some limitations of this technique include membrane fouling and higher initial costs.

### *3.3. Environmental and economic impact assessment*

To assess each sample's environmental impact, LCI data regarding mass and energy flows were compiled. A cradle-to-gate approach was followed, and data were collected for a functional unit of 1 g of Li. Table 4 presents the LCI for the ED process performed with WWLB and AL samples.

The total cradle-to-gate impacts of $Li_2CO_3$ production from spodumene range from 3.5 to 9 kg of $CO_2$ eq/kg $Li_2CO_3$, whereas the brine route ranges between 2 and 6.2 kg of $CO_2$ eq/kg $Li_2CO_3$ (Manjong et al., 2021). In addition, the impacts of solvent extraction, which is applied in conventional routes, could reach 52.7 kg $CO_2$eq/kg $Li_2CO_3$ (Nikfar et al., 2025). The cradle-to-gate LCA resulted in 0.26 kg of $CO_2$ eq/g Li global warming impacts for the WWLB sample, and 46.40 kg of $CO_2$ eq/g Li for the AL sample (Fig. 4). Other secondary routes of Li recovery also demonstrated potential to reduce overall impacts compared to conventional Li production, achieving 0.004–0.008 kg $CO_2$ eq/g Li at a pilot line and full-scale level (Hyder et al., 2025).

For water consumption, the total impacts were 0.0029 $m^3$/g Li for WWLB and 0.61 $m^3$/g Li for AL. A higher Li concentration results in a greater total yield, which, assuming equivalent processing efficiency for both samples, reduces the environmental impacts per gram of product. Two-stage electrodialysis Li recovery route demonstrated water consumption between $1.38 \times 10^{-5}$ $m^3$/g Li and $1.30 \times 10^{-4}$ $m^3$/g Li (Hyder et al., 2025). Inorganic acid leaching for spent LIB cathode scraps demonstrated water consumption (excludes background system contributions to the impact category) between $2.88 \times 10^{-5}$ $m^3$/g Li and $6.89 \times 10^{-5}$ $m^3$/g Li (Tang et al., 2023).

For both samples, most of the potential environmental impacts derive from electricity consumption. For WWLB impacts, electricity generates 98 % of global warming impacts and 97 % of water consumption impacts. In the AL results, electricity has the highest share

**Table 3**
Lithium recovery from Al–Li alloy dust (AL): Al - no assisting agent; A2 - ChCl: OA; A3 - ChCl:MA; A4 - SC $CO_2$; A5 - OA 0.1 mol/L; A6 - OA 0.5 mol/L, and wastewater from Li-ion battery recycling (WWLB): $Na_2SO_4$ [W1 - 50 mA, 72 h; W2 - 100 mA, 48 h; W3 - 200 mA, 24 h], $NaNO_3$ [W4 - 50 mA, 72 h; W5 - 100 mA, 48 h; W6 - 200 mA, 24 h; W7 - 50 mA, 48 h; W8 - 50 mA, 24 h; W9 - 100 mA, 24 h].

| AL | Li recovery (%) | WWLB | Li recovery (%) |
|---|---|---|---|
| A1 | 72.70 | W1 | 91.26 |
| A2 | 27.84 | W2 | 91.73 |
| A3 | 11.20 | W3 | 91.78 |
| A4 | 71.76 | W4 | 92.92 |
| A5 | 82.57 | W5 | 97.23 |
| A6 | 91.54 | W6 | 91.99 |
| - | - | W7 | 88.63 |
| - | - | W8 | 61.21 |
| - | - | W9 | 94.50 |

**Table 4**
Life cycle inventory of the electrodialytic process using the wastewater of lithium-ion battery recycling (WWLB) sample and Al–Li alloy dust (AL) to obtain 1 g of lithium.

| Sample | Flows | Mass and energy flows | Amount | Unit | Cost | Total Cost |
|---|---|---|---|---|---|---|
| WWLB | Inputs | Electrolyte (miliQ water) | 441.49 | mL | 2.25 €/$m^3$ | 0.001 € |
| | | Electrolyte ($NaNO_3$) | 0.75 | g | 0.11 €/g | 0.083 € |
| | | WWLB sample | 441.49 | mL | – | – |
| | | Electricity | 1.22 | kWh | 0.23 €/kWh | 0.28 € |
| | Outputs | Li in the electrolyte | 1.00 | g | – | – |
| | | WWLB sample | 441.49 | mL | – | – |
| | | Electrolyte | 441.49 | mL | – | – |
| AL | Inputs | Electrolyte (miliQ water) | 156862.7 | mL | 2.25 €/$m^3$ | 0.353 € |
| | | Electrolyte ($NaNO_3$) | 266.67 | g | 0.11 €/g | 29.33 € |
| | | AL sample | 78431.37 | mL | – | – |
| | | Electricity | 432.94 | kWh | 0.23 €/kWh | 99.58 € |
| | Outputs | Li in the electrolyte | 1.00 | g | – | – |
| | | AL sample | 78431.37 | mL | – | – |
| | | Electrolyte after treatment | 156862.7 | mL | – | – |

in global warming (96 %), whereas electricity is responsible for 83 % of total impacts in water consumption.

Electricity-related impacts are due to the energy mix from the 2020 Belgium grid (from the Ecoinvent database). In the global warming category, the potential impacts are generated mostly due to using natural gas in the energy mix (66 % of impacts) and the share of electricity imported from the Netherlands (19.4 %). In the water consumption category, most of the potential impacts are generated due to the nuclear energy share from the pressure water reactor (53 %), followed by the import share from the Netherlands (around 19 %) and the use of natural gas (12 %).

For the WWLB sample, the remaining flows do not generate significant impacts for global warming (around 2 %). In the water consumption category, water contributes to 17 % of total impacts derived from treatments needed before distribution. Waste treatment also has a significant contribution, with −15 % impacts, meaning water-related positive effects due to its purification treatment.

Following the electricity impacts in the AL sample, $NaNO_3$ and water are the main impact sources. $NaNO_3$ contributes 3 %, and water contributes less than 1 % for the global warming impact category. In the water consumption impact category, water usage accounts for 29 %, followed by $NaNO_3$ at 2 %. Waste treatment results in an impact mitigation of 13 %, attributed to the subsequent reuse of treated water. To significantly reduce the cradle-to-gate environmental impacts of the samples in both categories, it is crucial to effectively reduce the energy consumption, either through more efficient equipment or operation time reduction.

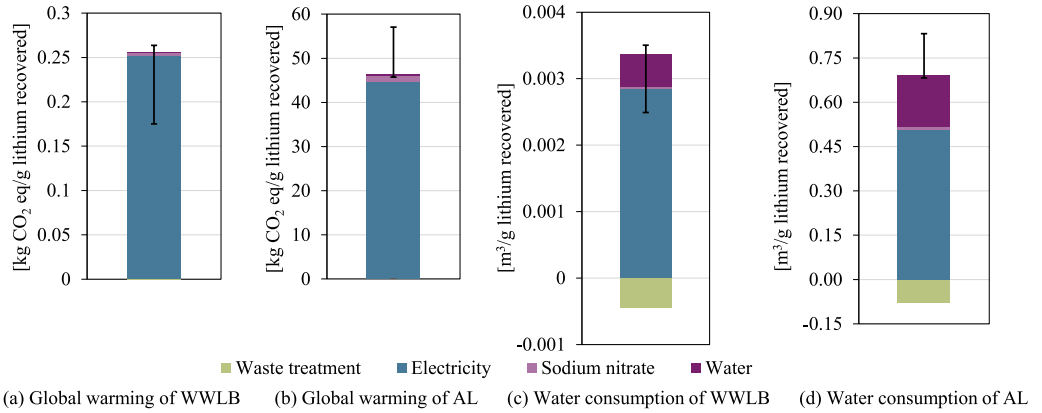
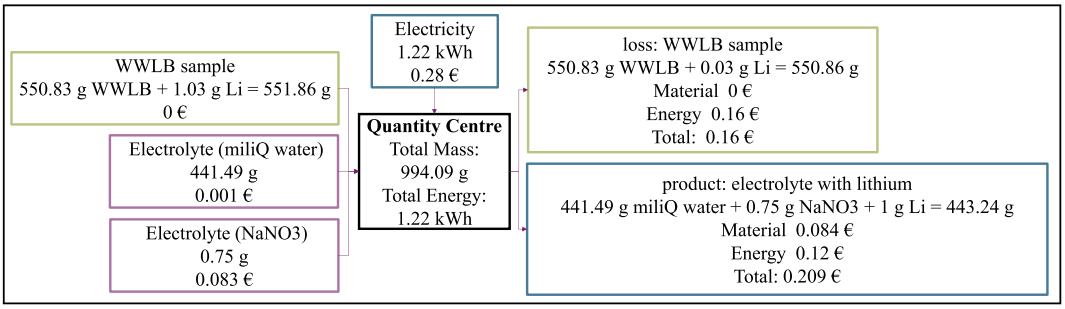



**Fig. 4.** Cradle-to-gate life cycle impacts for: (a) global warming of wastewater of lithium-ion battery recycling (WWLB), (b) global warming of Al–Li alloy dust (AL), (c) water consumption of WWLB and (d) water consumption of AL.

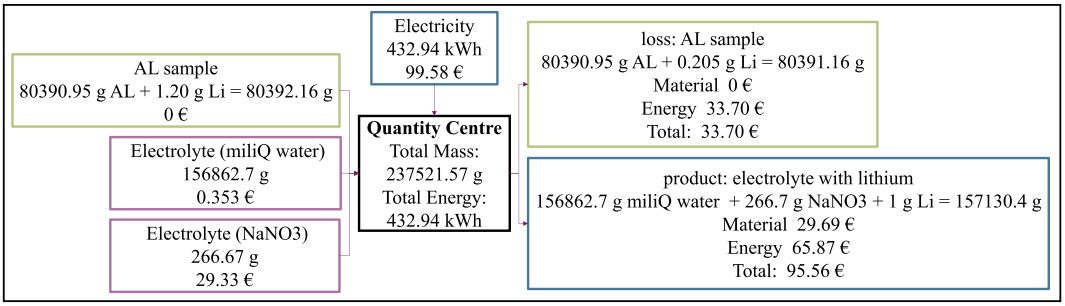
A sensitivity analysis assessed the environmental performance variation according to the obtained Li concentrations from several experiments. These results are depicted in Fig. 5. For the WWLB sample, the variation between average and maximum impact values is small (3.35 %) for global warming potential and water consumption. In contrast, the difference between minimum and average values is approximately 23.09 % for both impact categories. In total, a discrepancy of 25.67 % is observed between the minimum and the maximum values.

For the AL sample, the variation between average and minimum impact values is limited (1.90 %) for both GWP and water consumption. However, the maximum and average values are approximately 23.01 % for GWP and water consumption. In total, a discrepancy of 24.47 % exists between the minimum and the maximum values. When developing scale-up scenarios, the significant range between minimum and maximum impact values for the WWLB and AL samples needs careful consideration.

To analyse the material and energy costs of the ED process, a material balance was conducted for each sample to determine the costs allocated to the final product and material losses (Fig. 5). All materials were quantified in mass (g) and energy (kWh) and added to the




(a) Quantity centre assessment for WWLB.




(b) Quantity centre assessment for AL.

**Fig. 5.** Material balance and cost calculation in the electrodialytic process quantity centre to obtain 1 g of lithium from (a) wastewater of lithium-ion battery recycling (WWLB) and (b) Al–Li alloy dust (AL).

quantity centre. The WWLB amount was converted into mass through sample density (1.25 g/cm$^3$), and the AL amount was quantified assuming the liquid sample contained 6.25 g of AL powder in 250 g of water.

The monetary values for the $NaNO_3$ are the highest from the material flows, representing around 23 % of the total costs. However, the energy-related monetary values contribute most to the total costs (77 %). A mass allocation for the collected costs was conducted for each output (product and losses). The monetary quantification is summarised in Fig. 6, presenting the total process, energy, and material costs for each sample. The allocation of these monetary values to the final product and the waste generated (material losses) is also represented.

A significant amount of the total costs goes into material loss for both samples, derived from the energy consumption during the process. In WWLB, 55 % of energy-related costs are considered lost since the final sample is not further recovered, and most of the Li is transferred to the electrolyte, generating waste. This also occurs in the AL, but only 34 % of energy-related costs are lost to the original sample. When analysing the monetary flows that go into the product, 40 % of the product cost of the WWLB comes from the materials, and 60 % comes from the energy. In the AL, 31 % comes from materials and 69 % comes from energy consumption. In total, 57 % of all costs go into the final product, and 43 % go into waste for the WWLB. This indicates that any enhancements aimed at increasing the value derived from the waste samples have the potential to reduce overall costs significantly.

For AL, 74 % of the total costs go to the final product and 26 % to material loss. This suggests that the process is more efficient in generating monetary value; however, it could be further enhanced by implementing waste valorisation strategies. The costs associated with waste treatment fall outside the scope of this assessment and were therefore excluded from the MFCA analysis. Waste treatment was considered for the LCA, where local municipal facilities treated samples outside the laboratory.

Finally, considering the recovery of Li, the AL sample involves higher costs than the WWLB sample. The final cost of the ED Li recovery is 0.36 €/g Li for WWLB and 129.26 €/g Li for AL. Two-stage electrodialysis was applied to recover Li from WWLB, with costs between 0.20 €/g Li and 0.32 €/g Li for pilot-level TRL (Hyder et al., 2025). Additionally, an MFCA study demonstrated that the leaching of spent Li battery cathode ranged from 0.4 €/g Li to 0.6 €/g Li (Tang et al., 2023). These values are consistent with the cost estimated for WWLB (0.36 €/g Li), reinforcing this sample's economic advantage for process upscaling in alignment with environmental goals.

### 3.4. Critical discussion

Despite extensive research on electro-based technologies for environmental remediation over the past decades, their TRL remains low. While numerous systems demonstrate potential, many are still far from being commercially viable as efficient processes. Key challenges, including high energy consumption and operational costs, must be addressed to achieve a higher TRL and facilitate the upscaling of the technology (Magro et al., 2019).

The concurrent capture of self-generated $H_2$ within the ED reactor is a feasible consideration for mitigating the impacts related to energy consumption and costs observed for WWLB and AL, where electricity generates 98 % and 83 % of total global warming impacts, respectively (Fig. 4). Integrating electrokinetic processes with ED technology has been explored for $H_2$ production alongside substance removal/recovery (Almeida et al., 2021). Such improvement would support reducing the expected environmental impacts for both waste samples. The $H_2$ produced could be stored or used to self-feed the ED reactor. Based on experimental self-generated energy, savings on electroremediation of approximately 7 % could be achieved (Magro et al., 2019).

The selectivity of ion-exchange membranes is crucial to reduce contamination. While ED has demonstrated high Li recovery efficiencies in bench-scale experiments, scaling up introduces challenges. These challenges are related to fouling, energy consumption, and membrane durability, which exhibit degradation over time, reducing efficiency and increasing operational costs (Almeida et al., 2021).

The same set-up (2C reactor at 100 mA) demonstrated the highest efficiencies for Li recovery from AL (91.5 %) and WWLB (97.2 %), decreasing the need to adjust the system for different samples. However, Li extraction from solid samples may require initial leaching steps to dissolve Li into a recoverable liquid phase (Zhao et al., 2023). The efficiency of ED depends on the leachate composition and the ability to remove impurities. Pre-treatment and purification steps may have to be adjusted not to compromise the overall process viability at an industrial scale.

Following the previous work from Almeida et al. (2020a,b), the ED treatment of sewage sludge and mining residue suspensions

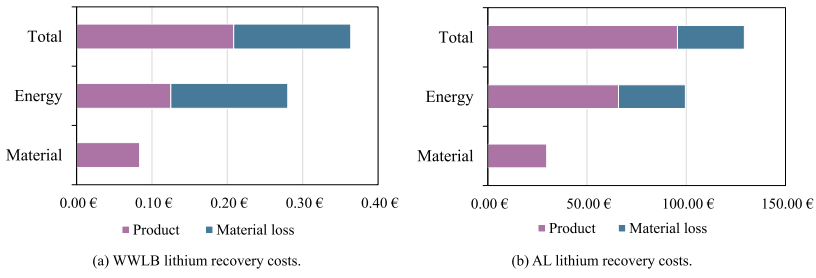



**Fig. 6.** Costs of the ED process to obtain 1 g of lithium from (a) wastewater of lithium-ion battery recycling (WWLB) and (b) Al–Li alloy dust (AL).

(combined with effluent or sewage sludge) was tested to recover CRM [21]. The extraction of P (71 %) and W (62 %) from sewage sludge and mining residues was more efficient when both matrices were merged (Almeida et al., 2020a,b). Bearing this in mind, combining both AL and WWLB into the same reactor to perform the ED treatment would decrease water consumption (by replacing the water content with WWLB), which revealed impacts of 0.0029 $m^3$/g Li (WWLB) and 0.61 $m^3$/g Li (AL) (Fig. 4). Also, if feasible, it would promote recovering Li in a high quantity in one step, decreasing energy consumption, operation time, and other consumable needs. In addition, using more efficient equipment for the reactor or implementing design improvements should be tested to reduce the reaction time and, consequently, its energy consumption.

The amount of waste streams generated in the industries is often not constant, leading to uncertainties in downstream systems' capacity and operation modes. Also, batch composition can vary greatly. Therefore, combining ED with other recovery techniques (precipitation, solvent extraction) is necessary to obtain Li at a high purity level. This next step will be crucial to produce Li battery precursors, such as Li carbonate or Li hydroxide, and allow a more effective integration into the battery supply chain. The recovered Li content must meet purity standards to be competitive with primary sources, impacting commercialisation prospects.

By 2030, LIB cost forecast increases by up to 67 % depending on cumulative production (Orangi et al., 2023), and LIB recycling foresees emissions of 3.7 Mt of $CO_2$ eq and an energy demand of 33.6 GWh (Bruno and Fiore, 2023). However, from an environmental perspective, using a waste stream as a secondary resource to obtain Li has benefits. In the LIB-supply chain, cathode active material (CAM) synthesis, cell manufacturing and hydrometallurgy recycling account for significant costs and emissions, with CAM alone responsible for 45 %. Recycling can decrease costs by 44 % and $CO_2$ emissions by 37 %, further improved by renewable energy (Gutsch and Leker, 2024). This approach encourages industries to reduce the volume of waste that requires treatment and disposal, thereby reducing associated environmental and health risks. Additionally, it highlights the repurposing of waste materials by giving them a second life through recycling, reuse, or recovery processes, ultimately promoting sustainability and resource efficiency.

The application of the ED process results in secondary waste streams that require proper management. In the assessment, incineration with energy recovery was assumed as the end-of-life treatment; however, this process was not included in the cost evaluation. Further treatment and in-house valorisation would significantly reduce monetary losses to waste flows. A significant portion of energy-related environmental impacts and costs is driving the main impacts of the ED process, meaning that energy-specific actions for improvement during upscaling, such as more energy-efficient machinery or the use of digital energy monitoring tools, would support cost and environmental impact reduction of the Li recovered by ED. In addition, the total costs and the carbon footprint of ED-based Li recovery must be assessed against conventional extraction methods or Li recovery from battery recycling processes to justify large-scale adoption. The heterogeneous composition of the samples is a limitation in performing LCA studies, resulting in significant variability.

To enhance the scalability of ED for Li recovery, advancements in membrane technology, process optimisation, and hybrid system integration are necessary. Therefore, future research should improve membrane selectivity, reduce energy requirements, and develop cost-effective pre-treatment methods. The economic and environmental optimisation of the ED technology for Li recovery will promote to leverage the TRL into a circular industrial scale activity.

## 4. Conclusions

Innovative and sustainable approaches for valorising secondary resources are gaining interest across various fields. Recovering Li from secondary solid and liquid resources offers a viable solution to mitigate the environmental impact of primary Li production, reduce EU dependence on raw material imports, and address challenges associated with waste accumulation. Implementing feasible circular economy strategies for Li recovery is crucial to support a sustainable and efficient energy transition.

The application of the ED process was tested for Li recovery from AL and WWLB. Coupling OA, a maximum of 91.5 % of Li was recovered from AL in 72 h. On the other hand, Li recovered from WWLB, in the electrolyte solution, reached 97.2 % in 48 h. Both maximum Li recoveries were achieved when experiments were performed at 100 mA, with a 2-compartment reactor and a CEM interposed.

The LCA results showed that energy consumption in the reactor is the main hotspot of the process, indicating that improvement measures related to optimisation of energy and time efficiency could significantly reduce impacts for both waste samples.

The cradle-to-gate LCA resulted in 0.26 kg of $CO_2$ eq/g Li global warming impacts for the WWLB sample, and 46.40 kg of $CO_2$ eq/g Li for the AL sample. Water consumption impacts were also higher for AL (0.61 $m^3$/g Li) than WWLB (0.0029 $m^3$/g Li).

This was also validated in the MFCA, where energy-related costs corresponded to most of the total process costs. Lower recovery costs were found for WWLB (0.36 €/g Li) compared to AL (129.26 €/g Li). In fact, cost-related losses were also driven by the waste generated from the process, indicating that further in-house valorisation of the waste samples after the ED process could significantly reduce waste-related costs.

The sensitivity analysis revealed a significant range between minimum and maximum impact values for the WWLB and AL samples. Therefore, prudence is required when defining scale-up scenarios. Nonetheless, these results highlight a promising alternative approach for recovering Li from secondary solid and liquid waste streams, offering a supplementary Li supply and a means to reduce environmental hazards. Future scale-up scenarios could vary key parameters and assumptions to assess technical, environmental, and cost performance, and benchmark against existing Li extraction technologies for competitiveness.

Summing up, the achievements of the present research support the circular use of secondary materials, tackling resource scarcity and progressing innovative recovery techniques, contributing to environmental and industrial goals. However, technical, economic, and environmental barriers must be addressed before large-scale implementation. Continued innovation and interdisciplinary efforts are essential to establish ED as a viable technology for sustainable Li recovery.

## CRediT authorship contribution statement

**Joana Almeida:** Writing – review & editing, Writing – original draft, Methodology, Data curation, Conceptualization. **Joana R. Gouveia:** Writing – review & editing, Writing – original draft, Methodology, Data curation, Conceptualization. **Inês S. Ribeiro:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Carolina Pires:** Writing – review & editing, Methodology, Data curation. **Eduardo P. Mateus:** Writing – review & editing, Validation, Resources, Formal analysis. **Alexandra B. Ribeiro:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Funding acquisition, Formal analysis.

## 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.scp.2025.102189.

## Data availability

All data was made available throughout the document and the supplementary information file.

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