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# Advancing hydrometallurgical recycling of spent lithium-ion batteries: an AI-based readiness and sustainability assessment

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Recycling of spent Li-ion batteries presents technical and environmental challenges that must be addressed. The hydrometallurgical route offers greater advantages over pyrometallurgy in obtaining pure products, recovering lithium, and handling feedstocks composed of diverse chemical compositions. However, current technologies still face efficiency, economic feasibility, and environmental issues. New advances are motivated by new battery technologies in the market, efficiency, productivity, and low environmental impact. This literature review aims to comprehensively evaluate current and emerging technologies and rank them as per their technology readiness. We compared ongoing approaches with emerging technologies (*e.g.*, membranes, new adsorbents, deep eutectic solvents, ionic liquids, supercritical fluids, nano-hydrometallurgy, and direct regeneration) reported in the literature focusing on complexity, energy usage, emissions, economic potential, and adaptability. The qualitative analysis was used to rank technologies based on a scoring system. The scoring was obtained using a novel approach utilizing artificial intelligence (AI) models eliminating personal preferences in qualitative data evaluation. The results indicated the technology readiness of the processes: direct regeneration > adsorbent materials > supercritical fluids > deep eutectic solvents > membrane technologies > ionic liquids > nano-hydrometallurgy. Future directions highlight the necessity of testing the identified technologies in emerging battery chemistries (*e.g.*, sodium-ion batteries) to ensure efficient and sustainable recycling processes.

### Sustainability spotlight

The rising demand for lithium-ion batteries (LIBs) presents significant environmental and resource challenges, underscoring the need for efficient recycling strategies to recover critical raw materials and minimize waste. This review highlights recent advances in sustainable LiB recycling, focusing on innovative recovery processes that maximize material reuse while reducing environmental impact. These approaches support the transition to a circular economy and align with key United Nations Sustainable Development Goals (SDGs): SDG 9 (Industry, Innovation, and Infrastructure) by driving technological advancements in recycling, SDG 11 (Sustainable Cities and Communities) by improving battery waste management, and SDG 12 (Responsible Consumption and Production) by enhancing resource efficiency. By reducing reliance on virgin materials and mitigating environmental risks, these innovations contribute to a more sustainable and resilient battery supply chain.

## 1. Introduction

Li-ion batteries (LIBs) are important for different markets such as energy storage, electric vehicles, and electronic equipment. The transport sector is a major emitter of greenhouse gases; while electric vehicles reduce local emissions, efficient battery recycling is essential to support material circularity.[1,2] In portable electronics industries, the demand for the use of batteries is increasing.[3] The battery market increased twice between 2014 and 2019, which represents 94.8% of Li-ion batteries and might increase exponentially during the next 5 years.[4]

These batteries are composed of a case (Al alloy or stainless steel), polymeric separator (polypropylene or polyethylene), electrolytes (salt solutions such as lithium hexafluorophosphate, $LiPF_6$) mixed in organic solvents such as propylene carbonate, ethylene carbonate, or dimethyl sulfoxide, anode (graphite or graphene) with Cu foil and cathode with Al

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foil.[5,6] Safety, energy density, power energy, and lifetime are related to the cathode material which can be $LiCoO_2$ (LCO), $LiFePO_4$ (LFP), $LiMn_2O_4$ (LMO), $LiNiCoAlO_2$ (NCA) or $LiNi_xMn_yCo_zO_2$ ($x + y + z = 1$, NMC). In addition, there are three physical structures of battery cells: cylindrical, prismatic, and pouch.[7] Many different charge accumulators can be found in the market and new configurations/chemistries are coming.

Li-ion batteries are composed of critical minerals (*e.g.*, graphite, Li, Ni, Co, Al, Cu) that can be subjected to supply cutoffs in a short and medium term as they are produced by a limited number of countries and replacement possibilities are limited.[2] For instance, of the global cobalt production, about 70% is produced by the Democratic Republic of Congo, while 77% of global lithium production is centered in Chile, Argentina, and Australia.[8,9] As a result, battery recycling will play a crucial role in raw material supply, decrease the dependence on other countries and extractive mining, and contribute to the circular economy.[10]

The wide variety of battery configurations complicates the recycling process. After physical processing (*e.g.*, discharging, milling, and separation), the material generated contains different metals in high concentrations (mostly from the cathode) also known as black mass due to its color and powder characteristics. For instance, cobalt content is 28.2% and that of lithium is 3.5% in LCO processing,[11,12] while cobalt content is 4.5%, that of nickel is 16.2%, and that of lithium is 3.7% in NMC processing.[12] As current recovery techniques were designed based on extractive mining techniques, development in optimization and technologies is necessary for higher concentrations,[13,14] even in complex systems.[15–18]

This study aimed to analyze hydrometallurgical recycling technologies and new advancements in metal recovery from the spent cathode of LIBs and promote sustainable mining.[19] The focus on aqueous hydrometallurgical processing was made based on the lowest environmental impact,[20–22] flexibility to recycle most batteries,[23,24] recovery of most of the materials present in the batteries (*e.g.*, graphite, plastic and electrolyte), and production of high-quality products (>98%).[25,26] Proper analysis of new technologies is important for the future development of industrial facilities. Special attention was given to cathode materials due to their high content of critical metals (Ni, Co, and Li) and their complex chemistry, which poses challenges for efficient recovery. Graphite is not the goal of this study although its importance and classified as critical in Europe, USA and UK.[14,27,28]

We analyzed innovative and regenerative technologies in light of the current Li-ion battery market. Furthermore, this study aims to provide a qualitative comparison of emerging hydrometallurgical technologies, carefully selected based on their compatibility with new battery technologies, operational efficiency, productivity, and minimal environmental impact. Moreover, the study aims to rank the identified technologies based on their technology readiness level under specific criteria established (complexity, energy usage, emissions, economic potential, and adaptability) within the study. The qualitative analysis was converted into a quantitative score utilizing a novel approach where Artificial Intelligence (AI) tools were involved.

## 2. Review methodology and analysis by AI

The literature identification was performed using the SCOPUS database with the keywords: “Li-ion batteries” and “cathode” and “recycling” combination. Research articles in English were chosen for the period of 2016–2024. Initially, 221 documents related to the recycling of cathode active materials in LIBs were identified. After excluding those not focused on hydrometallurgy, 74 were retained. A manual review was conducted to identify emerging technologies with high potential for metal recovery. These were then classified into two categories: emerging hydrometallurgical technologies and direct recycling technologies. The term “emerging technologies” used in this study stands for “the novel or trending technologies that can be used to extract critical minerals available in the cathode (or black mass)” as per the experience of the authors. An overview of these technologies is provided as extensive technology explanation is not the aim of the manuscript.

In the third phase, a qualitative comparison was carried out between the emerging and the direct regeneration technologies. A quantitative comparison was not possible at this point due to limitations on data availability. For instance, one technique can be applied in the leaching of the metals while another will be adopted for separation/purification. Hence, extracting the data for quantitative analysis poses difficulties. Moreover, the applications and products were also different in emerging and direct regeneration technologies: direct regeneration makes a new cathode while recycling methods give raw materials to produce new cathode materials or other industrial applications (*e.g.*, Co and Ni for steel, manganese for alkali batteries and lithium carbonate for pharmacy). Accordingly, a qualitative data extraction was carried out focusing on five major sectors: complexity, energy usage, economic viability, and adaptability. Assessing the technologies in these sectors will help us to decide later which technologies have greater potential to be applied in larger scale sooner (depending on their technology readiness). To minimize human error during data extraction, three authors independently extracted qualitative data for each criterion. A consensus description was then developed based on the three extractions. AI models were not employed at this stage due to the sensitivity of qualitative content and current limitations in AI's ability to accurately interpret such context-specific data. In the final phase, the qualitative analysis was converted to numerical values using a non-biased AI model – Open AI's gpt-4-turbo engine – to remove any personal preferences from the authors. The language model uses the vocabulary to assign the score, giving more robust and unbiased results. The AI model was fed a summary result table and was commanded to provide scores based on the word combinations/patterns and vocabulary. The command used in the study is as follows; “Assign scores for each technology based on qualitative descriptions in the table, into numerical values on a scale (*e.g.*, 1 to 5) to reflect their respective impacts on complexity, energy usage, emissions, economic potential, and adaptability, ensuring that lower values indicate better performance in most cases”.

## 3. Analysis of current and emerging technologies for recycling of Li-ion batteries

### 3.1. Current technologies for recycling of Li-ion batteries

Here we introduce the state-of-the-art Li-ion battery recycling and hydrometallurgical approach. The dominant recycling process for battery recycling is pyro-based and hydrometallurgical routes. The pyrometallurgical process involves high temperature thermal techniques including polymer degradation (carbon source), smelting, pyrolysis, and carbothermic reduction. On the other hand, hydrometallurgical processes occur in aqueous media by leaching (acid or alkali) followed by separation and purification techniques such as ion exchange resins, precipitation, solvent extraction, and cementation.[3,7,29,30]

Before processing, the batteries are discharged to avoid explosions. Salt solutions have been explored for short-circuits discharging to remove residual electricity within 24 h. Fig. 1 presents a typical optimization of discharging of spent LIBs using salt solutions. However, iron from the casing oxidizes and partially dissolves into the solution, posing a risk to hydrometallurgical processes by causing co-precipitation and contaminating products.

This corrosion primarily affects the terminals, releasing the cathode material and potentially hindering the complete discharge of residual electricity in the battery.[32–34] The literature reports patented and industrial routes for Li-ion battery recycling. Detailed flowcharts are depicted in the literature.[2,29] For instance, the Umicore process is a pyrometallurgical-based process with a capacity of 7000 tons/year for Li-ion and NiMH batteries.[35,36] Here, in a vertically mounted, preheated furnace (heated up to 30 °C), the material is further heated to 300 °C in the presence of coke and slag formers to facilitate gas removal and subsequent treatment. Then, plastic pyrolysis occurs at 700 °C followed by smelting by injection of oxygen-rich air preheated to obtain an alloy of nickel–copper–cobalt and lithium in the slag along with aluminum, silicon, and calcium. The AkkuSer Oy plant has a recycling capacity of 1000–4000 tons per year through physical processing as described for LCO batteries;[37] after milling and particle size separation, the material is smelted to obtain a copper–cobalt rich material. Retriev Technologies operates in Canada and the USA processing 4500 tons per year of spent batteries as a closed-loop process. The process includes crushing them in a hammer mill in an inert atmosphere and water, followed by physical separation to isolate the black mass from graphite. In this case, plastic is recovered before the thermal treatment at 400–700 °C which is used for binder removal, and then, carbon is separated by flotation. Lithium is then obtained at 400–850 °C.[38,39]

The French company Recupyl primarily operates in Europe, while new commercial recycling lines have also emerged in Asia and North America, with a processing capacity of 110 tons per year. The process involves automated separation and chemical treatment of pretreated battery materials. Physical processing is conducted under controlled conditions using $CO_2$ or argon, followed by magnetic separation to remove iron, and a densiometric table to recover aluminum and plastics. Soluble Li is obtained from particles lower than 500 μm, while Co is obtained by leaching in sulfate media followed by electrolysis.[40,41] LAREX-Tupy process is the first in South and Latin America to recycle Li-ion batteries through a hydrometallurgical process where all cathode materials (*e.g.*, LCO, NMC, NCA, and LMO) can be fed. After physical separation involving knife milling and particle size separation, plastic, case, and electrolytes are recovered before acid leaching; then, aluminum, cobalt, manganese, nickel and lithium can be recovered, while graphite and aluminum/copper foil are further recovered from the leach residue.[12,20,42–48] Examining laboratory-scale recycling approaches is also necessary. Guimarães *et al.* (2022) proposed hydrometallurgical processing by discharging the batteries using copper wires until 0 V (∼24 h) without material losses.[12] This approach proved successful for cylindrical, pouch, and prismatic batteries.[11,45,47] Comparing the discharging time, both techniques require about 24 h, but the use of a mechanical

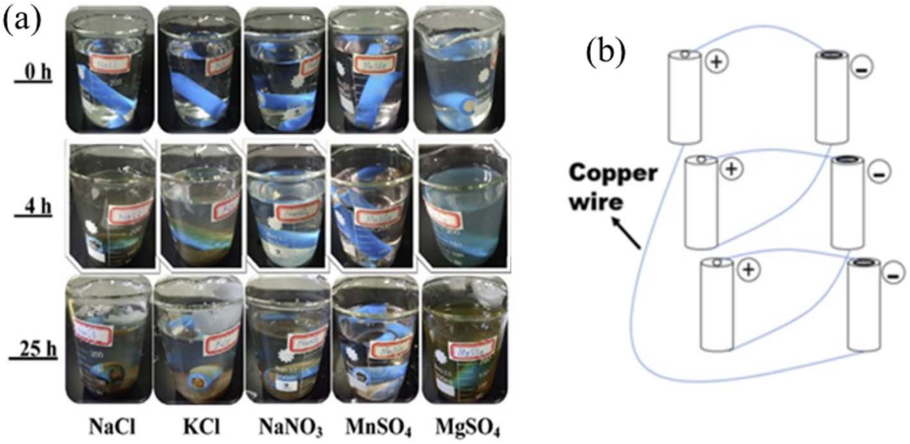

Fig. 1 (a) Discharging process using different salts to promote short-circuiting by removing remaining electricity.[31] (b) Discharging process using copper wire.[12]

approach instead of a chemical reaction avoids material losses and recovers electricity for future use.[49]

Pyrometallurgical processes for battery recycling can be used as a pretreatment, organic binder (around 350 °C, to release cathode and anode materials from respective current collectors) conductive carbon or acetylene black (over 600-700 °C, under air atmosphere or absence to burn carbon, binder, electrolyte, and plastic) decomposition, and metallurgy separations (roasting/calcination as chlorination, sulfation, and nitration and smelting as $CaO–SiO_2–Al_2O_3$ slag from 800 to 1500 °C).[50–52] Redwood Materials' process involves a combination of reductive calcination followed by hydrometallurgical treatment for recycling and production of valuable products from Li-ion batteries.[25]

The main drawbacks are energy consumption, low material recovery rate (losses of graphite and plastic), high $CO_2$ footprint, and, mostly, loss of Li to the slag phase. In this case, the hydrometallurgical route has great advantages including recovery of most materials, low energy consumption and $CO_2$ footprint, and high-purity products; on the other hand, low kinetic rates are achieved, and a large volume of wastewater is generated.[53] After pretreatment, including discharging and mechanical steps, the leaching reaction is carried out to convert the cathode material into ions. The main reactions occur in acid media using inorganic (mineral) acids such as $H_2SO_4$ (most used for economic and technical feasibility), $HNO_3$, or HCl. Organic acids (*e.g.*, citric acid ($C_6H_8O_7$)) have been explored to reduce corrosion and increase overall sustainability.[54–57] During the reaction, Co(III) is converted into Co(II) from the cathode material, and usually, a reducing agent such as $H_2O_2$ is added during the process; however, literature reports that aluminum foil can also act as a reducing agent if it is present in black mass ($Al(s) \rightarrow Al^{+3}$).[11,12] Eqn (1)–(6) present the leaching of the LCO cathode as an example using inorganic (eqn (1) and (2)) and organic (eqn (3)–(6)) acids, which are described in detail elsewhere.[58] Alkali leaching is also reported for aluminum selective leaching.[2]

$$4LiCoO_{2(s)} + 6H_2SO_{4(aq)} \rightarrow 4CoSO_{4(aq)} + 2Li_2SO_{4(aq)} + 6H_2O_{(l)} + O_{2(g)} \quad (1)$$

$$4LiCoO_{2(s)} + 6H_2SO_{4(aq)} + 2H_2O_2 \rightarrow 4CoSO_{4(aq)} + 2Li_2SO_{4(aq)} + 8H_2O_{(l)} + 2O_{2(g)} \quad (2)$$

$$6LiCoO_{2(s)} + 6C_6H_8O_{7(aq)} \rightarrow 2Co_3(C_6H_5O_7)_{2(aq)} + 2Li_3C_6H_5O_{7(aq)} + 9H_2O + 3/2O_{2(g)} \quad (3)$$

$$6LiCoO_{2(s)} + 6C_6H_8O_{7(aq)} + 6H_2O_2 \rightarrow 2Co_3(C_6H_5O_7)_{2(aq)} + 2Li_3C_6H_5O_{7(aq)} + 15H_2O + 9/2O_{2(g)} \quad (4)$$

$$4LiCoO_{2(s)} + 3C_4H_6O_{5(aq)} + H_2O_2 \rightarrow 2CoC_4H_4O_{5(aq)} + Li_2C_4H_4O_{5(aq)} + 4H_2O + O_{2(g)} \quad (5)$$

$$4LiCoO_{2(s)} + 7C_2H_2O_{4(aq)} \rightarrow 2Co(C_4H_4O_4)_{2(aq)} + LiC_2HO_{4(aq)} + 4H_2O_{(l)} + 2CO_{2(g)} \quad (6)$$

After leaching, the metals in the ionic form are separated by separation/purification methods to obtain the products. Solvent extraction is the main technique and involves liquid–liquid separation using an organic extractant (organic phase) in contact with the leach solution (aqueous phase). Literature reports the use of bis(2,4,4-trimethyl pentyl)phosphonic acid (cCyanex 272),[59] 5-nonylsalicylaldoxime organic extractant (Acorga M5640),[60] bis (2-ethyl hexyl) phosphoric acid (D2EHPA),[61] 2-hydroxyl 5-nonyl acetophenone (LIX 84-IC),[62] trioctyl tertiary amine (N235), and tributyl phosphate (TBP),[63] and high separation rates are observed. Ion exchange resins differ from solvent extraction as the technology uses a solid–liquid separation rather than flammable diluents. Chelating resins such as iminodiacetate (Lewatit TP 207, Purolite S930, and Amberlite IRC 748),[64–67] bis-polyamine (Lewatit TP 220 and Dowex M4195),[68,69] amino phosphonate acid (Purolite S950, Amberlite IRC747, and Lewatit TP 260)[70,71] have been reported for battery recycling in scientific publications, but no industrial applications were shown. Precipitation is also used for both separation and obtaining the product, as for lithium carbonate production by sodium carbonate reaction,[72] but for separation, it is not ideal and causes coprecipitation when applied in similar pH levels (*e.g.*, around 6.7–8.0);[73] for this case, sulfide and carbonate precipitation seems to be an alternative.[73–75] Ozone precipitation has been reported as selective for manganese over cobalt.[76,77]

These technologies are mature and well-known to scientists and industries.[78] However, technical advances are considering the environmental impacts of recycling activities.[79] Current technologies were developed with a focus on extractive recovery, often overlooking their $CO_2$ footprint and alternative, more environmentally sustainable approaches.[80] When it comes to battery recycling, the variety of Li-ion batteries available in the market—differing in both composition and physical structure—poses significant challenges for existing recycling methods.[29] For instance, there are iron-rich (also containing phosphate in the cathode material) and nickel-rich batteries, which might not be mixed for recycling as these elements make separation problematic causing product losses and contamination.[81]

### 3.2. Emerging technologies for recovery of critical minerals from spent Li-ion batteries

Approximately 38% of the selected articles focused on emerging technologies for LIB recycling. These studies were categorized by technology, and additional sources were reviewed using specific technological keywords to provide a comprehensive evaluation. This section discusses the identified emerging technologies.

**3.2.1. Membrane-based separation technology.** The membrane technique has been largely used for water recovery and desalination.[82] Among emerging technologies for hydrometallurgical processes, it has received attention for the separation of metals, including microfiltration (particle distribution 0.1 and 10 μm and 2 bar pressure), nanofiltration (particle distribution 0.001–0.02 μm and 1–10 bar pressure), ultrafiltration and reverse osmosis (particle distribution 0.001–0.02 μm and 5–150 bar pressure).[83] In addition, electrodialysis has been explored to separate metals from battery recycling using electricity as the driving force and cationic and anionic membranes for ion-selective separation. However, after leaching, all ions are cationic, and Li as a monovalent ion can be separated selectively using a monovalent ion-selective membrane. Cobalt, manganese, and nickel can be further purified using cationic membranes by EDTA complexation as negative compounds (anions) in different pH ranges, resulting in the selective separation of metals in each step.[84] Electrodialysis is a mature technology for water desalination, but several developments are necessary for battery recycling to improve membrane resistance, selectivity, and economic feasibility.[85]

Another membrane approach involves the combination of solvent extraction and supported liquid membranes and polymer inclusion membranes (Fig. 2).[86] For supported liquid membranes, the organic extractant comes in contact with the diluted extractant (for instance, Cyanex 272 in kerosene[87]), and this carrier promotes the selective migration of ions as nickel/cobalt separation occurs in the traditional solvent extraction process.[88] The feed chamber (Fig. 2a) contains the ions that react selectively with the organic extractant filled in the porous membrane, and protons from the receiving chamber (on the right) react with the organic phase releasing the metals. It occurs continuously without direct contact between organic fractions and the aqueous phase operating in different reactors as extraction and stripping steps. However, loss of organic phase from the membrane into the aqueous phase and kinetic separation are the spotlight now.[86] To improve kinetic separation, electrodialysis has been explored to change the driven force from the concentration gradient to electricity and make the process faster.[88,89] Sadyrbaeva *et al.* (2020) evaluated the ionic liquid [P66614][Cl] as a carrier in a polyvinylidene fluoride (PVDF) membrane and achieved 96% cobalt separation from nickel; moreover, Sadyrbaeva (2013) increased the kinetic separation by applying electricity as the driving force using supported liquid membranes based on tri-*n*-octylamine and trialkyl benzyl ammonium chloride ionic liquids.[90] Polymer inclusion membranes consist of polymers (such as poly(vinyl chloride) or cellulose triacetate), as a carrier (commercial organic extractant or ionic liquids) immobilized usually by a plasticizer (Fig. 2b), and the ions migrate from feed to receive chambers through the membrane.[91] Emulsion and bulk liquid membranes are also reported.[86]

3.2.2. **Adsorbent-based materials.** In the hydrometallurgical process, chelating ion exchange resins can be used for selective separation and purification. In battery recycling, most of commercial chelating resins focus on nickel, cobalt, and iron, such as iminodiacetate (Lewatit TP 207 and Amberlite IRC748), amino phosphonic (Purolite S950, Amberlite IRC-747, and Lewatit TP 260), bis-polyamine group (Dowex M4195 and Lewatit TP 220).[58,67,93] In the case of Strauss *et al.* (2021), they could recover 99% of nickel and 98.5% of cobalt from the initial cathode content (synthetic) introduced in the form of hydroxides. The authors used 1 M $H_2SO_4$ with 10 mM $FeSO_4$ to leach the metal oxides into the solution using an electrochemical leaching technique, and then used "Dowex M4195" to adsorb nickel and cobalt from the leachate with a minimum addition of chemicals. The method used seems to be green, however, the resulting materials need further purification to reach the final product.[93] In another case, adsorbent materials (ceramic and polymers) have been developed focusing on lithium separation and have the potential for application in battery recycling. Chitrakar *et al.* (2014) reported the synthesis of $H_2TiO_3$ for lithium recovery from brine.[94] The material is prepared using $Li_2TiO_3$ as an ion-sieve. In this case, lithium is adsorbed by the material due to the lower ionic radii: 0.074 nm for lithium, 0.102 nm for sodium, 0.138 nm for potassium, and 0.100 nm for calcium.[94,95] It can be applied for lithium separation from larger ions such as nickel, cobalt, and manganese. Lithium separation can be performed using magnetic graphene mixed with potassium with selectivity achieving around 14 grafted with methacrylic acid which, unlike resins, the separation is facilitated by

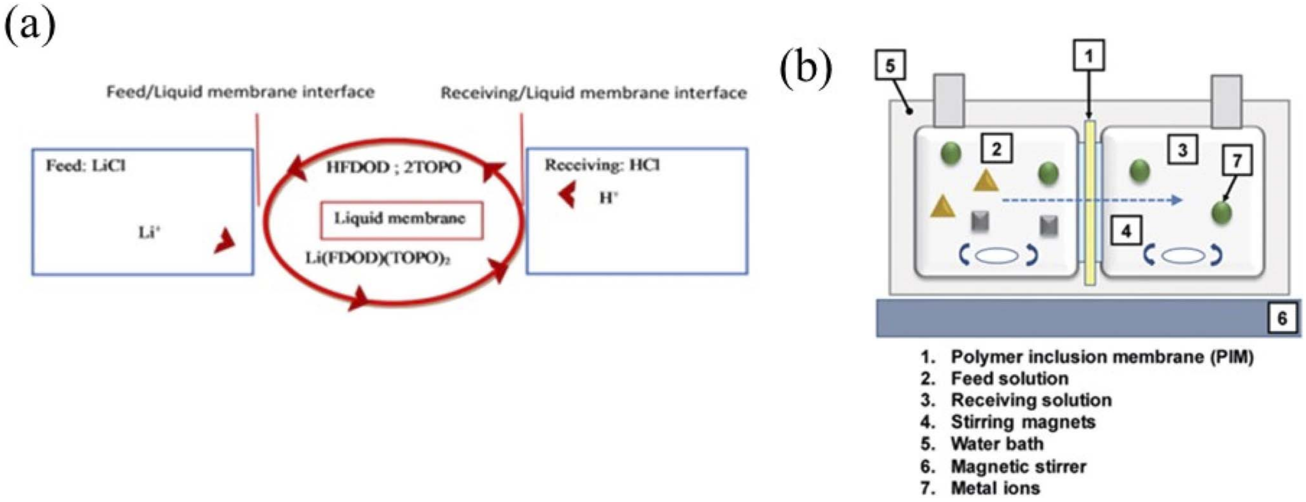

Fig. 2 (a) Scheme of the mechanism of transport using a supported liquid membrane (modified based on ref 92); (b) schematic diagram of the polymer inclusion membrane transport cell.[91]

magnet.[96] Functionalized materials improve lithium separation, as observed by Zhou *et al.* (2022) wherein amino ethyl benzo-12-crown ether (B12C4–NH2) covalently immobilized lithium bonding onto polymers, achieving a selectivity separation rate of almost 10 over sodium and potassium.[97] Another example is functionalized cellulose whose synthesis was reported by Chen *et al.* (2019). According to the authors, the mechanism might involve complexation achieving the following selectivity order: $Li^+ >> Na^+ > K^+ > Cs^+$.[98] Although the literature on lithium selectivity over nickel, cobalt, manganese, and iron is lacking, the potential application for battery recycling is focused on the recovery of sodium or potassium, which might be found due to its possible presence in electrolytes and use of alkalis (such as NaOH and KOH) in the separation and purification steps.[29,56,99]

**3.2.3. Deep eutectic solvents (DES).** Eutectic mixtures are specific compositions of mixtures of two or more solid components that change to the liquid phase at a certain temperature.[100] However, DES has a recent history that starts when Abbott *et al.* first experimented with mixing choline chloride (ChCl) with urea. The resulting solvent had an extra low eutectic point at 12 °C which is much lower than the corresponding freezing points of its two ingredients separately.[100] Since then, DES has been extensively studied by different researchers for various applications, including hydrometallurgy.[101,102]

For the dissolution of metal oxides, DES which is made from hydrogen bond acceptors and donors was used (HBA–HBD type). This is because such solutions provide the required properties for dissolution, such as low pH range, reducibility, and the ability to coordinate.[101] For instance, different metal oxides (various sources with above 98% metal oxide availability) were dissolved in three different DES solutions (ChCl with urea, ChCl with ethylene glycol, and ChCl with malonic acid) to analyze the metal oxide dissolvability in the DES used. Results showed that the metal oxides used dissolve better in DES that contains malonic acid. Importantly, a better correlation was also observed between malonic acid-based DES and aqueous HCl solution in metal oxide solubility.[101] Further studies undertaken for metal oxide leaching from LIBs revealed that DES that contains organic acids as hydrogen bond donors improves the solubility of the metal oxide significantly due to two reasons: the low pH of the solution and the presence of hydrogen as the oxygen atom acceptor.[103] Another attempt to examine the solubility of metallic oxides in thiol (sulfur substitution instead of oxygen in alcohol groups) substituted alcohol groups in DES reported improved physical and chemical properties. The researchers achieved lower viscosity, wider liquid range, and elevated solubilities for late transition metals (copper and zinc). Furthermore, they showed that it would be possible to use thiol-DESs for selective dissolution of metals incompatible with aqueous processing (*e.g.*, cadmium and lead).[104] These studies have shed light on using DES in LIB processing and possible material recoveries, especially using HBA–HBD.

In the LIB recycling application, Wang *et al.* (2019) used DES for the pretreatment of LIB cathode materials. ChCl was used with glycerol (2.3 : 1 molar ratio) to produce the DES to dissolve the aluminum foil from the cathode at 190 °C for 15 minutes. They were able to recover 99.86% of the cathode material successfully through the dissolution of aluminum by deactivating the PVDF binder. The method was identified to be greener and cost-effective than most of the conventional methods as it requires low energy, does not release HF, and non-availability of corrosive acid or alkalis for processing.[105] In another study, the authors came up with a novel method to identify the possible DES to recycle LIB cathode materials. The method was based on electrochemical principles and determined the reduced power of the DES quickly. The study demonstrated that using DES with more negative reduction potential reduces reaction temperature and time. Two DES solutions, ChCl–urea and ChCl–ethylene glycol were used for comparison studies, where ChCl-urea has more negative potential and showed higher extraction of lithium and cobalt (95%) at 170–180 °C within 12 hours.[106] Chang *et al.* (2022) developed a DES-based selective recovery strategy to recover transition metals from NMC-type LIBs (Fig. 3). The approach aims to establish an optimally constructed dual leaching and separation mechanism that is founded on the regulation of the coordination circumstances. An average of 95% recovery was obtained for each element: nickel, cobalt, and manganese using ChCl and oxalic acid dihydrate as the DES ingredients. Optimal conditions were recorded as 20 : 1 solid–liquid ratio at 120 °C.[107]

Zante *et al.* (2020) used a hybrid method using an ionic liquid and DES to extract metals from a simulated LIB cathode material leaching liquor. Two different ionic solutions were used to separate manganese first from the liquor followed by

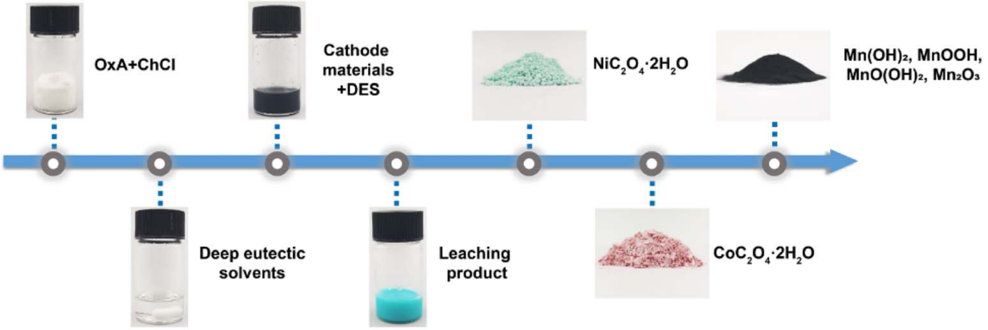

Fig. 3 Flow chart for DES-based separation of Ni, Co, and Mn from exhausted LIBs.[107]

cobalt. The following subchapter (Section 3.2.4) describes the application of ionic liquids in the respective study. However, nickel and lithium were separated from the remaining liquor using a DES made using lidocaine and carboxylic acid. This separation left a higher concentration of lithium in the liquid phase which can then be deposited during the next steps. However, the method proposed needs further optimization using more compact systems to apply it in practical applications.[108] Another study was carried out to extract metals from LCO-type LIBs using DES. The authors used DES made of ChCl and ethylene glycol with a 1 : 2 molar ratio. Researchers recorded over 99% leaching efficiency and 90% recovery of lithium and cobalt. More importantly, the researchers assessed the recyclability of the recovered DES after electrodeposition of the metal ions onto a substrate. It was reported that the recovered DES can be used for another round of leaching without lowering its efficiency.[109]

**3.2.4. Ionic liquids (ILs).** Generally, ILs are organic molten salts that contain dissolved ions with freezing points close to zero or below the ambient temperature,[110,111] and possess several physicochemical properties that allow them to be used in multiple applications. Acting as perfect organic solvents or extractants is one among many applications that are justified by their properties such as high selectivity, stability, and very low volatility[112] This makes ILs one of the candidates to separate metals in black mass-leaching liquors.

In the experiments done by Zante *et al.* (2020), ILs were used to separate manganese and cobalt from the leaching liquor. Researchers used an IL of 95% pure N, N, N′, N′-tetra(*n*-octyl) diglycolamide (TODGA) diluted using 99.5% pure 1-butyl-3-methylimidazolium bis(tri-fluoro methylsulfonyl)imide ([C4mim][NTf2]) to separate manganese from metal (Li, Mn, Co, and Ni) sulfate leaching liquor in 1 : 1 ratio. 99% of manganese separation was observed when the molarity of TODGA increased to 0.1 mol.$L^{-1}$ and at a pH of 3.3. Cobalt extraction was then done by using 97% pure tri-hexyl tetradecylphosphonium chloride ([P66614][Cl]) with a separation factor of Mn/Co 40–60. However, using a higher volume of IL makes the process economically inefficient and requires further development.[113] Another attempt was made to extract lithium from a sulfate-based simulated black mass leaching solution containing cobalt, nickel, and lithium using ILs. Generally, it requires a high amount of extractants to achieve higher separation of lithium from the solution with lower pH values. However, a satisfactory level of lithium extraction was achieved using a lower amount of IL in higher pH values (pH > 3). As the organic liquid, they used a mixture of 99% pure tri-*n*-butyl phosphate (TBP) with 97.5% pure sodium salt of tetrakis(trifluoromethyl)phenylboron (NaTFPB) ILs. However, since lithium extraction occurs through ion exchange, loss of ions from the ILs was observed, which was hard to rectify fully.[108] Zheng *et al.* (2021) also attempted to extract lithium from real LIB active material-leaching liquor made using sulfuric acid. About 80% TBP and 20% carboxymethyl trimethylammonium bis(trifluoromethyl)sulfonimide ionic liquids were used to prepare the organic phase and 95% lithium was recovered from the leachate with 2 : 1 organic/aqueous phase ratio. Importantly, the recyclability of the organic phase used in the study increases the potential for the real application of carboxyl-functionalized ILs soon.[114] Morina *et al.* (2023) used a novel technique to isolate and extract metals (lithium and cobalt) from an LCO-type electrode-leaching liquor made from organic acids. The IL used in the study was 0.1 mol.$L^{-1}$ 3-methyl-1-octylimidazolium thenoyltrifluoroacetone (Omim-TTA) under optimum conditions of organic to aqueous phase ratio of 1 : 3 and 120 minutes reaction time at room temperature. Over 70% of LiCl deposition was achieved using HCl after separating lithium from the aqueous solution. Interestingly, it was shown that the IL was not altered after the experiment and can be reused for the same purpose.[115]

While for separation and extraction IL has been used in recent years with little success, successful regeneration of NMC-type electrodes was also reported by Wang *et al.* (2020). In the study, mixture of three ILs, namely, ethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)-imide ([C2mim][NTf2]), [C4mim][NTf2], and 1-ethanol-3-methylimidazolium bis(trifluoromethanesulfonyl)-imide ([C2OHmim][NTf2]) were used as the flux solvent in an ionothermal lithiation at 150 °C. As per the lithium source, lithium chloride and lithium bromide were used. The process enables the ionothermal re-lithiation of the spent cathode (NMC111) material, and it was identified that the IL can be recycled readily for another round without decreasing the efficiency. The study shows that the re-lithiated cathode performed similarly to the original cathode material.[116]

**3.2.5. Supercritical fluids (SCFs).** Studies show the possibility of using SCFs to recover materials from LIBs due to their extremely high solubility and high stability when the materials reach their temperature and pressure at a critical point.[117] A study has been conducted to recover electrolyte materials from LIBs using the SCF technique. Initially, researchers placed the battery material in a supercritical fluid reactor. Then, the $CO_2$ supplied to the reactor was converted to the SCF state by adjusting the temperature or the pressure. Super Critical $CO_2$ (SC–$CO_2$) would dissolve the electrolyte which can be later separated using vacuum. Afterward, the electrolyte will be further treated to separate valuable contents while the $CO_2$ can be recycled and forwarded to the reaction chamber to extract more electrolyte materials.[118] A different study conducted by Grutzke *et al.* (2014) evaluated the use of SC-$CO_2$ for electrolyte and lithium salt extraction. Though they succeeded in recovering the electrolyte material, they found that the recovery rate highly depends on the composition of the solvents, temperature, and the solid–liquid interface production. Also, they could recover only a trace amount of lithium salt.[119] In a second attempt, the same group of researchers tried to improve the recovery rate of electrolytes by adding a mixture of acetonitrile/propylene carbonate and liquid $CO_2$ (25 °C, 60 bar) in a 3 : 1 ratio. Authors reported 89% recovery of electrolytes along with lithium salts in their original composition within 20 minutes, a greater achievement compared to their previous study.[120]

Mu *et al.* (2017) also used SC-$CO_2$ to recover materials from LIBs. Authors reported over 90% total recovery of the materials, including organic solvents and electrolyte materials, along with

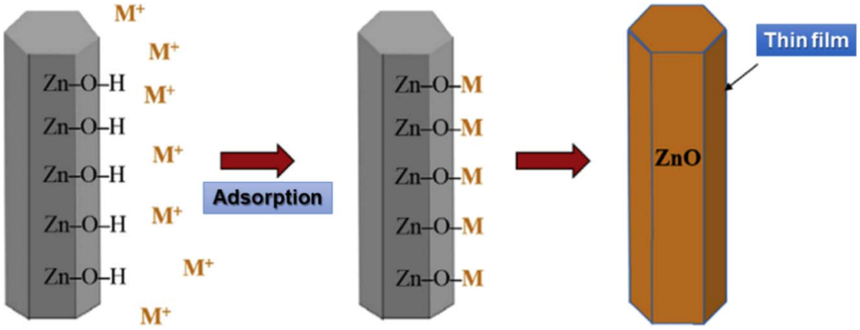

Fig. 4 Scheme of the adsorption process using ZnO nanoparticles (based on ref. 136).

lithium salts and additives under optimized conditions. The authors also showed the reusability of the electrolyte materials recovered when mixed with organic solvents and lithium salts in the right amounts. The pros of the method were the high purity of materials, non-degradation of organic solvents or lithium salts, and very high recovery rates. However, some disadvantages were also reported, such as high CAPEX and OPEX for the instrument and energy, and weak intercalation of the electrolyte due to the weak polarity of $CO_2$.[121] In another attempt to recover electrolytes from used LIBs, experimental conditions were determined through a predictive model. It was determined that the extraction pressure is important in the process. Researchers used the model's predicted value of 23 MPa and 40 °C for 45 minutes to achieve 85.7% electrolyte recovery using SC-$CO_2$.[122,123]

Other than organic solvent recovery, Bertuol *et al.* (2016) showed that using SC-$CO_2$ in cathode metal recovery from LIBs is also possible and beneficial. The study was intended to determine the efficiency of cobalt recovery with the presence of SC-$CO_2$ (at 60 bar pressure) and with atmospheric gas (atmospheric pressure). The study concluded that incorporating SC-$CO_2$ as a co-solvent with $H_2O_2$ and $H_2SO_4$ can increase the efficiency of recovery significantly in terms of time (reduction to 5 minutes from 60 minutes), resource use (50% reduction in $H_2O_2$ usage) and purity (99.5 wt%) of the recovered materials.[124] The same team of researchers also used SC-$CO_2$ to recover copper from printed circuit boards using the same mechanism and reported that the efficiency of copper recovery can be enhanced by nine times when SC-$CO_2$ is used instead of atmospheric extraction.[125] Furthermore, Han *et al.* (2023) discussed the possibilities of utilizing SC-$CO_2$ in new cathode material synthesis and modifications.[126]

**3.2.6. Nanohydrometallurgy.** A recent technology named nanohydrometallurgy has been helping in waste management mostly with the separation of elements with great efficiency tested on a laboratory scale.[127] The aim of this technology is similar to that of hydrometallurgy, focusing on the recovery of valuable metals from aqueous solutions, in which the substrate can be waste from electrical and electronic equipment (WEEE). However, the difference remains in the application of nanotechnology.[128–130] In a nutshell, the addition of nanoparticles (NPs) or nanomaterials in the solution is responsible for the adsorption of the metals.

The application of this technology in the recycling processes consists of the adsorption of critical and strategic metals by nanomaterials like nanofibers, graphene, and graphene oxide.[131,132] Nevertheless, many nanomaterials have been used for the application of the nanohydrometallurgy technology, such as polymer-derived nano adsorbents, polymer-modified and metal-based nano adsorbents, biogenic, carbon-based, magnetic, and superparamagnetic nanoparticles, graphene, graphene oxide, nanofibers, and nanocellulose.[129,132,133] Considering the use of this technology in the battery recycling field and compared with the literature, it presents great efficiency in the recovery of nickel, cobalt, manganese, and copper meeting the principles established by green chemistry.[129,130,134]

A study made by Melo *et al.* (2019) reported the synthesis of superparamagnetic nanoparticles that function with diethylenetriaminepentaacetic acid (DTPA) using a crystalline $Fe_3O_4$ core with a $SiO_2$ protective coating.[135] The recovery of metals from LIBs, such as copper, nickel, manganese, and cobalt by commercial sorbents were found in the literature.[131,132] Another study found in the literature regarding the use of nano hydrometallurgy in the recycling process of LIBs was developed by Le *et al.* (2019), who studied the adsorption by ZnO nanoparticles (Fig. 4) in the separation and recovery of metallic ions, such as copper, nickel, and manganese. Their results stated that all copper was adsorbed in 15 min under ultraviolet (UV) or visible light as the photocatalyst. Meanwhile, nickel had an adsorption of less than 10%.[136]

### 3.3. Direct regeneration technologies

The technologies used for direct regeneration were recently developed and are responsible for regenerating the LIBs' cathode without compromising its original structure.[137–139] Direct regeneration methods (Fig. 5) are seen as a long-term approach for recycling LIBs, with substantial economic advantages and a low carbon impact.[137,140] When compared to the most popular methods, such as hydro- and pyrometallurgy, it can achieve electrochemical characteristics extremely comparable to those seen in novel cathode materials. Furthermore, these technologies consume little energy and emit no hazardous gases throughout the processes.[141,142] These advantages have made direct regeneration a method of current research interest.

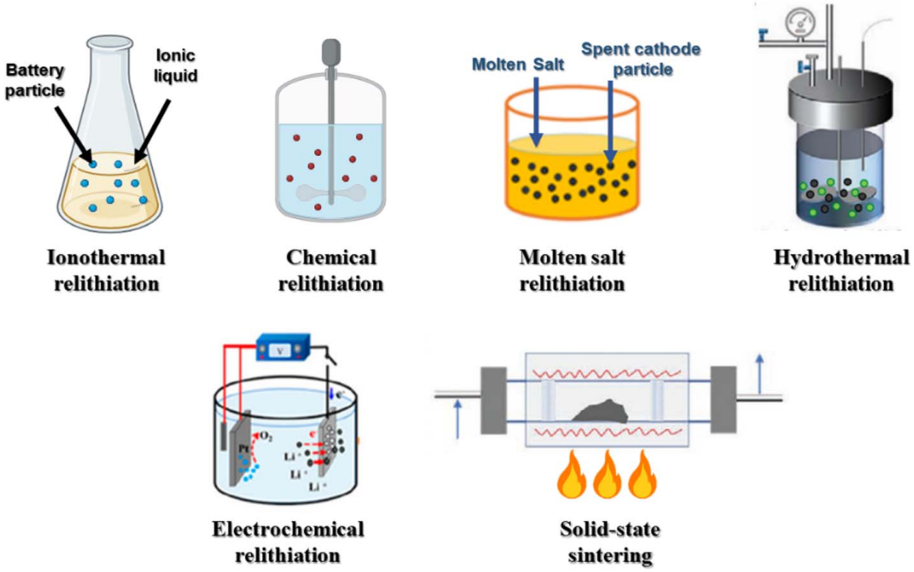

Fig. 5 The different technologies used in the direct regeneration processes (modified based on ref 80,137,140).

In general, spent cathodes are deficient in $Li^+$ and contain a significant number of high-valence transition metal ions, which are important for charge conservation. As a result, the emphasis of the direct regeneration operations is the incorporation of $Li^+$ ions into the degraded cathode active material, hence encouraging complete crystal structure recovery. This regeneration is caused by redox reactions between cathode-active materials and reductive lithiation agents.[137,138] Solid-state sintering and relithiation methods (chemical, hydrothermal, electrochemical, molten salt, and ionothermal) are popular direct regeneration strategies for discarded cathode materials which will be discussed further.[138,143–146]

3.3.1. **Solid-state sintering.** This approach resembles the cathode material generation process in which $Li^+$ flows back into the crystal structure of the waste cathode materials. This occurs at high temperatures and results in a regenerated cathode with high crystallinity and excellent stoichiometry.[137,138] Solid-state sintering offers advantages such as minimal waste generation and process simplicity, but it is highly energy intensive. This is due to its one-step regeneration process, which eliminates the need for additional purification or separation and produces no liquid waste.[147] The scalability of this technology, however, is a matter that needs more studies and applications.

To summarize, in order to obtain a regenerated second-life battery, a certain quantity of lithium source may be supplied to the cathode material in order to replace the loss of lithium ions in the crystalline structure *via* this method. Meng *et al.* (2019), for example, employed $Li_2CO_3$ as a lithium source to regenerate waste NMC111 using a combination of solid-state sintering and mechano-chemical activation processes.[148] The solid-state sintering technique uses homogeneous mixing of spent LFP battery components with a particular quantity of lithium salt in a reductive environment.[137,138]

3.3.2. **Relithiation processes: chemical, hydrothermal, electrochemical, molten salt, and ionothermal relithiation mechanisms.** Several relithiation mechanisms have been examined using the direct regeneration approach. To begin with, chemical relithiation technology is utilized to recover discarded cathode materials at lower temperatures (≤120 °C) resulting in decreased energy consumption and atmospheric pressure.[143] The type of reductant agent utilized for the relithiation process will be determined by the composition of the cathode in the LIB. For example, to achieve chemical relithiation on a spent LMO cathode, 1,2-dimethoxyethane (pyrene-Li) can be utilized under ambient conditions.[149,150] A chemical relithiation developed on a spent LCO type battery can be carried out using lithium ethylene glycolate, betaine, and urea (BEU) under ambient pressure and 80 °C.[151] Furthermore, the chemical relithiation procedure proved the ability to regenerate many spent cathode materials with only minor changes.[137,138,142] Wu *et al.* (2021) demonstrated that the use of chemical relithiation technology completely renews LFP battery cathodes in 10 minutes.[138]

Ouaneche *et al.* (2023) demonstrated the creation of a chemical relithiation technique at ambient temperature to renew waste LFP cathode materials while keeping the aluminum foil (current collector) attached where ethanol was used as a solvent and LiI as a precursor of lithium. Iodine reduced $Fe^{3+}$ in $LiFePO_4$ to $Fe^{2+}$ during the relithiation process, and iodine was oxidized to $I_2$.[152]

The hydrothermal relithiation process, like chemical relithiation, uses a reductant and is carried out under adequate temperature and pressure conditions. When high temperature and pressure are utilized, relithiation processes can reestablish the crystalline structure of a cathode and be beneficial in renewing the materials. Gao *et al.* (2020) used hydrothermal relithiation to renew used LMO batteries. Various amounts of LiOH were tested as a lithium source. The regenerated LMO material had a lithium/manganese molar ratio of 1 under the circumstances of 0.1mol $L^{-1}$ LiOH solution, 12 h, and 180 °C.[150]

Another type of relithiation process is electrochemical relithiation. This technology is based on the flow of $Li^+$ back into the cathode materials electrochemically, in order to perform the regeneration of the waste cathode.[137,153] This is a promising technique for directly regenerating spent LFP and LCO batteries for instance. Liu *et al.* (2022) demonstrated the restoration of depleted LFP electrodes through an electrochemical process that utilizes the electrochemical driving force to effectively reinsert lithium ions into the degraded LFP crystal structure. This method reduces the need for chemical reagents and minimizes waste generation. Additionally, the electrolyte used for relithiation contains lithium recovered from the spent LFP electrodes themselves.[153]

Zhang *et al.*'s study (2020) utilized a spent $LiCoO_2$ cathode (LCO) directly obtained from spent LIBs, and a platinum plate served as the anode electrode, while Ag/AgCl acted as the reference electrode. To enable electrochemical regeneration, $Li_2SO_4$ solution was employed as both the electrolyte and the source of lithium. By applying a constant cathodic current, they effectively facilitated the insertion of $Li^+$ ions into the crystal structure of the lithium-deficient spent $LiCoO_2$. Simultaneously, this process led to the liberation of $O_2$ at the anode. Notably, the $LiCoO_2$ regenerated from this relithiation process demonstrated electrochemical properties comparable to commercially available $LiCoO_2$.[154]

The method of molten salt relithiation involves employing a eutectic salt mixture with a lower melting point than any individual salt component under standard atmospheric pressure for the regeneration process.[146] When heated to a suitable temperature, the molten salt enables improved dispersion of lithium ions, thereby aiding in the regeneration of cathode materials. Additionally, the lithium remaining as a source post-regeneration reaction in the cathode is easily soluble and extractable.[155]

Under mild conditions, ionic liquids—which were discussed in Section 3.2.4 of this work—can also be used as fluxing agents or even in novel processes for the regeneration of spent cathode materials. Ionic liquid media are thus used in the ionothermal regeneration process to accomplish that in waste battery materials.[156] Wang *et al.* (2020) reported the full recovery of the structure of NMC batteries using ionothermal relithiation of a spent cathode of this type of battery. This success was attained by employing inexpensive lithium halides as the lithium source, and recyclable ILs as the solvent.[116]

## 4. Evaluation of technologies based on the scoring by AI

The information presented in Sections 3.2 and 3.3 is manually summarized using two-way tables, with columns representing evaluation sectors for recycling technologies and rows representing the individual technologies. The summary tables are useful in feeding data to the AI-language model. The AI model was instructed to extract data only from the summary tables for the evaluation. Accordingly, Tables 1 and 2 summarize emerging recycling and direct regeneration technologies across five sectors, including complexity, energy usage, emissions, economic potential, and adaptability. An AI model was used to score each technology based on linguistic patterns—such as comparative adjectives and descriptors—from the summary tables. This approach offers clear advantages over expert judgment by minimizing bias and ensuring objectivity and reproducibility. Unlike subjective assessments, which may vary due to personal preferences or differing familiarity with emerging technologies, the AI model draws conclusions solely from the literature, enabling consistent evaluation across all technologies. In contrast, the AI approach systematically interprets text data, providing a robust, unbiased evaluation directly aligned with the presented evidence. Although the methodology benefits from consistency and neutrality, potential limitations, such as dependence on input data quality, should also be acknowledged to contextualize the results comprehensively.

As discussed in Sections 3.2 and 3.3, regeneration technologies are generally more efficient than conventional recycling methods. However, their effectiveness is limited to cases where the same cathode chemistry is reused, meaning the introduction of new chemistries may require significant adaptation. Similarly, recycling technologies must also evolve to accommodate changing feed materials. Fig. 6 presents the AI-derived scores for various emerging technologies across different evaluation sectors.

Complexity is the highest in DES, membrane, and nanohydrometallurgy technologies. The lowest complexity is reported for adsorbent materials with a score of 2 under the considered criteria. Considering energy usage, the best results were shown by nanohydrometallurgy and adsorbent materials. DES seems to be the worst in this sector. Membrane, IL, and SCF showed medium-range performance. Emissions were reported to be the highest in nanohydrometallurgy. However, this is due to the non-availability of emission data for the technology in question. All assessed technologies show low emissions, aligning with future sustainability targets and potentially accelerating their adoption over current high-emission recycling methods.

Considering the economic potential of the technologies the lowest scores were acquired by the DES-based technology. Adsorbent materials and SCF follow this with lower economic viability compared to DES. The worst score was reported for nanohydrometallurgy. Adaptability is very high again in DES-based leaching and membrane-based purification technologies. IL and SCF also show higher adaptability compared to the other technologies. The lowest adaptability was reported for nanohydrometallurgy. The total score reported for each technology was taken into consideration to decide the best among the emerging technologies. Since lower scores indicate better potential, the technology with the lowest total score is considered most applicable for future use. Accordingly, adsorbent material-based technologies rank the highest, followed by supercritical fluid (SCF) technologies with the second-lowest score. Deep eutectic solvents (DES) and membrane technologies share the same application potential, each with a score of

**Table 1** Summary of the qualitative evaluation of the upcoming technologies discussed

| Technology | Complexity | Energy usage | Emissions | Economic potential | Adaptability |
|---|---|---|---|---|---|
| Membrane technology | Can be highly complex when increasing the kinetics of the reaction as different solutions need to be introduced | Relatively lower due to the low pressure applied and lower electricity requirement | Lower emissions, as the process is more likely to do a purification | Low economic potential due to frequent membrane changes and material consumption | High adaptability can be seen as the changes in membrane and the carrier materials can be effectively adjusted according to the feed solution or metal ions |
| Adsorbent materials | Lower complexity due to the lower chemical usage and interactions. However, could be complex when separating Li from the feed | Lower energy usage as the electricity consumption is only used for the feeding pumps with lower flow rate for effective adsorption | Lower emissions, as the process mainly involves separation of metal ions | Economic potential can be lower owing to the frequent change of ion-exchange resins | Low adaptability for new feed flows. Might need to optimize all the parameters. Also, frequent changes in resins can be problematic if the feed concentration is higher |
| Deep eutectic solvents | Highly complex due to the optimization of various parameters before application | Higher energy usage due to prolonged leaching and high temperature. Low energy usage for pretreatment applications | Lower emissions were reported by many authors due to the coordination ability and selectivity towards metals | Highly economical industrially due to the lower prices of organic acids, high recovery rates, and high recyclability | Versatile applications (pretreatment and leaching) make it adaptable. However complex optimization processes can be highly sensitive for the feed materials |
| Ionic liquids | Complex setting due to the optimization of parameters | Lower energy use is reported due to the room temperature leaching. However, use in cathode regeneration can be highly energy intensive | Lower emissions due to the high boiling points of ILs | Can be costly as the IL is required in larger quantities for the treatment | Medium adaptability due to costly material requirements. However, high recycling rates reported can reduce the overall cost |
| Supercritical fluids | Less complex, as the method relies on a highly pressurized chamber with elevated heat | Comparably lower, researchers have shown that the reaction time is lower than most of the existing methods | Lower emissions, in an industrial line, $CO_2$ can be recycled | The economic potential of the technology can be lower due to the lower recovery rates reported, the cost of instruments, and safety | Adaptable technology due to its versatility, however not industrially ready for leaching |
| Nano hydrometallurgy | Highly complex, no studies have yet reported the applicability in LiBs directly | Lower energy usage is reported as additional temperature is not required | No data | Controversial; due to the usage of nanomaterials the cost can be higher as well | Less adaptability for LiB recycling, and the technology needs further studies |

Table 2 Summary of the qualitative evaluation of the regenerative technologies discussed

| Technology | Complexity | Energy usage | Emissions | Economic potential | Adaptability |
|---|---|---|---|---|---|
| Solid-state sintering | Simple mechanisms as it uses a single step sintering process | Higher energy use is reported for the sintering process | Lower emissions as no extra purifications and separations are involved | Moderately economical method given the simplicity of the process and high energy use | With less adaptability, further studies are needed to shape up the technology |
| Relithiation | Average complexity, some relithiation technologies can be complex due to optimizations | Lower energy use compared to other methods. Lower temperatures will be utilized for a limited period | Lower emissions as minimum purifications are required | Highly economical due to less chemical usage and fast reactions | Can adapt after brief modifications |

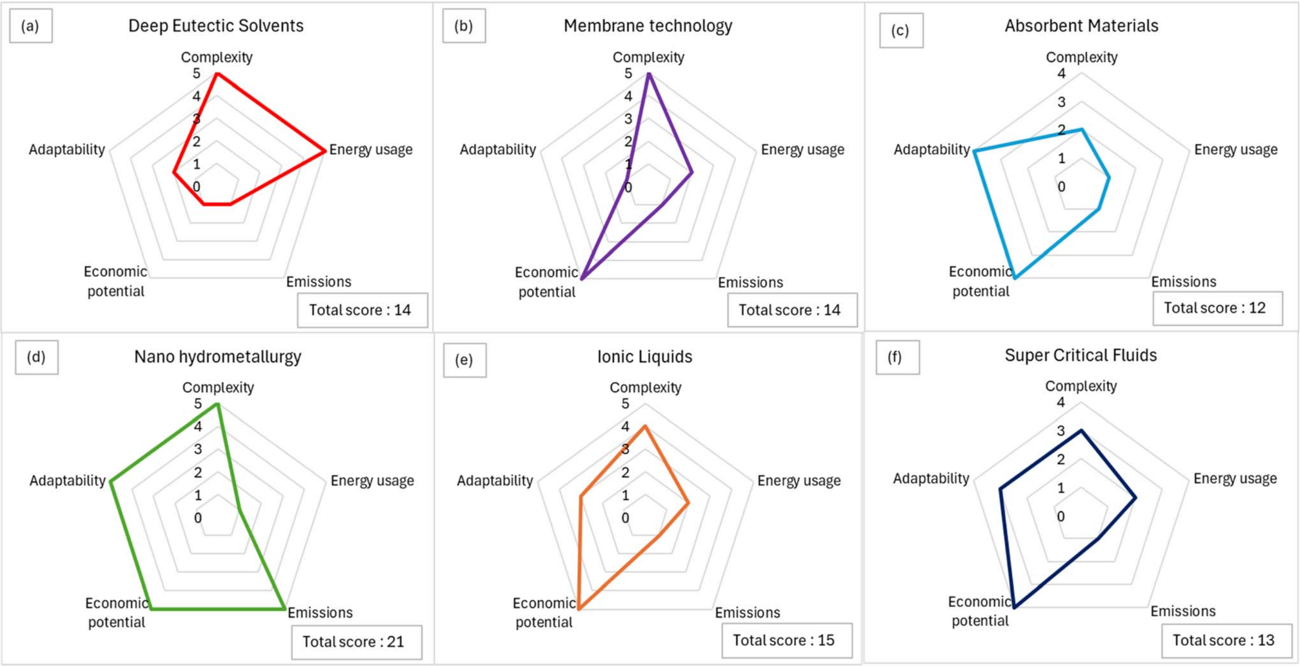

**Fig. 6** Score generated for each emerging technology, (a) deep eutectic solvents, (b) membrane technology, (c) adsorbent materials, (d) nano hydrometallurgy, (e) ionic liquids, and (f) supercritical fluids in different sectors (0 = best and 5 = worst).

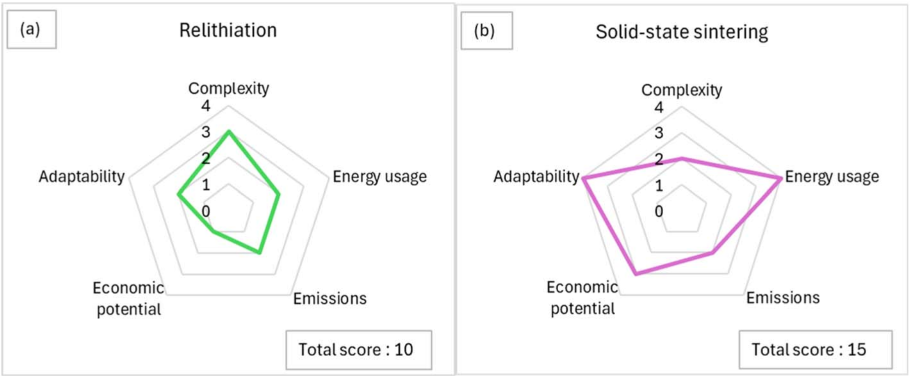

**Fig. 7** Score generated for each regenerative technology (a) relithiation technologies and (b) solid state sintering technologies.

14. Nanohydrometallurgy has the least possibility with a significantly high score of 21.

Considering the regenerative technologies assessed, scoring was done using the AI language model to evaluate the two technologies. Accordingly, Fig. 7 was developed to demonstrate the results.

Solid-state sintering has a lower adaptability and economic potential with higher energy usage. However, relithiation is only worse off than solid state sintering under the complexity criteria. Moreover, the overall score of the two technologies further convinces us that relithiation has a higher possibility to be applied sooner in larger scales than solid-state sintering.

## 5. Conclusions

This literature review aimed at the analysis of current recycling and upcoming technologies in light of the current Li-ion battery market. Established hydrometallurgical techniques were discussed in detail alongside emerging approaches such as membrane technologies, adsorption, deep eutectic solvents, ionic liquids, supercritical fluids, and nano-hydrometallurgy. A novel approach using AI language models was applied to convert qualitative results into a quantitative scoring system for evaluating technology readiness levels.

The study and the analysis demonstrated that the knowledge gained from pilot-scale studies might be sufficient for the first industrial facilities, given the urgency of recycling the current stock of spent batteries. However, adsorbent materials and supercritical fluid-based recycling show a stronger tendency to emerge as next-generation technologies for upscaling with the lowest overall scores of 12 and 13 respectively. In addition, deep eutectic solvents (score: 14), membrane technologies (score: 14), and ionic liquids (score: 15) also present significant potential to replace existing recycling facilities due to their efficiency and lower emission possibilities. Nanohydrometallurgy, currently at the lab scale with the highest score of 21, is not necessarily less important or efficient, rather, the literature emphasizes the need for more focused research on this technology for Li-ion batteries.

Direct regeneration appears to be a trending approach that could significantly lower treatment costs and emissions compared to emerging technologies. Among these, relithiation (score: 10) seems to offer better potential compared to solid-state sintering (score: 15) which can lead to its application in industrial lines sooner. Future directions should focus on applying these technologies to the recycling of new battery chemistries, ensuring the development of more sustainable and efficient recycling processes.

## Disclaimer/publisher's note

## Author contributions

Conceptualization, methodology, formal analysis, investigation, data curation, writing – original draft preparation, D. S. P., L. F. G., A. B. B. J., J. A. S. T., D. C. R. E., MV; writing – review and editing, D. S. P., L. F. G., A. B. B. J. All authors have read and agreed to the published version of the manuscript.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The authors declare that the data used will be available under proper request.

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