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Addressing preliminary challenges in upscaling the recovery of lithium from spent lithium ion batteries by the electrochemical method: a review†

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The paramount importance of lithium (Li) nowadays and the mounting volume of untreated spent LIB have imposed pressure on innovators to tackle the near-term issue of Li resource depletion through recycling. The trajectory of research dedicated to recycling has skyrocketed in this decade, reflecting the global commitment to addressing the issues surrounding Li resources. Although metallurgical methods, such as pyro- and hydrometallurgy, are presently prevalent in Li recycling, they exhibit unsustainable operational characteristics including elevated temperatures, the utilization of substantial quantities of expensive chemicals, and the generation of emissions containing toxic gases such as Cl_2, SO_2, and NO_x. Therefore, the alternative electrochemical method has gained growing attention, as it involves a more straightforward operation leveraging ion-selective features and employing water as the main reagent, which is seen as more environmentally benign. Despite this, intensive efforts are still required to advance the electrochemical method toward commercialisation. This review highlights the key points in the electrochemical method that demand attention, including the feasibility of a large-scale setup, consideration of the substantial volume of electrolyte consumption, the design of membranes with the desired features, a suitable layout of the membrane, and the absence of techno-economic assessments for the electrochemical method. The perspectives presented herein provide a crucial understanding of the challenges of advancing the technological readiness level of the electrochemical method.

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Introduction

Mitigating the global environmental issues encompassing climate change and greenhouse gases through decarbonisation of the transport sector has influenced the current shift in the global vehicle market from the domination of the internal-combustion engine (ICE) to electric vehicles (EVs) in support of the net-zero carbon emissions agenda.[1,2] In the post-pandemic era, the EV sector has experienced a significant growth, with a projected revenue of USD 56.1 billion in the year 2023. Moreover, the market forecasts anticipate its expansion to a volume of USD 906.7 billion in 2028.[3] As EVs are powered using Li-ion battery (LIB) technology, the boom in the EV industry has contributed significantly to the increasing lithium (Li) demand. In addition, the multiplying number of spent LIB from EVs in the next few years is a cause for concern. The International Energy Agency (IEA) forecasted a significant surge in LIB waste, with the EVs manufactured in 2019 are expected to generate 500 000 tons of such waste, a figure that could escalate to 8 million tons by 2040.[4,5] These trajectories highlight the critical dependence of the global community on Li and the opportunity for spent LIB to become the future Li market.[6]

As the global dependence on Li is ever-growing, increasing the extraction and recovery efficiency of Li from spent LIB is pivotal to strike a balance between global Li reserves and consumption. The goal of recycling is threefold: (i) to decelerate

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resource depletion, (ii) to contribute to environmental conservation and (iii) to create a national domestic supply chain for the element.[4,6,7] In addition to the issue of sustainable Li supply, spent LIB present alarming concerns in terms of their threat to the environment. Their occupation of landfills, leaching of the hazardous elements from the spent sources into the receiving water, and potential harm to the ecosystem from the exposure to contaminated water sources requires an imminent solution.[7–10] Leachate from spent LIB may contain dissolved gases, heavy metals, additives, and electrolyte degradation products, leading to the potential release of heavy metals into the soil, nearby rivers, or lakes through rainwater runoff. The management of the spent LIB through recycling approaches not only benefits the environment, but also has potential in contributing to the establishment of a circular flow of the valuable elements in LIBs. The establishment of spent LIB recycling in the next few years would give local governments and the community a potential secondary supply, generating a secure supply chain as well as wealth, benefiting the local economy.

At present, the mature pathways for the recycling of Li and other precious metals from spent LIB are hydrometallurgy and pyrometallurgy. Pyrometallurgy employs the concept of heat treatment to induce chemical conversions in the mixture of metals in spent LIBs. Hydrometallurgy involves the separation or purification of the mixture of metals in spent LIB through leaching methods. Through hydrometallurgy, Li and other precious elements are leached into a leachate using either inorganic acid, organic acid, or bacteria. Despite the efficacy of metallurgical methods in extracting Li, the long-term setbacks are a matter requiring consideration. At the surface, the problems surrounding these techniques are excessive energy consumption, the use of toxic reagents, and the generation of wastewater, which highlights the need for an environmentally benign option for Li recovery.[11–13] Fig. 1 illustrates the circularity of LIB from the virgin products to their end-of-life, followed by the need for their recycling to sustain the mineral supply while handling the substantial number of LIB that will be disposed of in the coming years.

In the recent years, the electrochemical approach to recycle Li mediated by membrane selectivity, current-induced reaction, and safer reagent consumption has garnered significant attention in academia and industry.[14–17] This new approach has already been proposed as an alternative to pyro- and hydrometallurgy, and benefits from minimal-current operation and employment of water as the main reagent.[11,14] However, to the best of our knowledge, there are not yet any comprehensive reports or patents for the up-scaled electrochemical technology

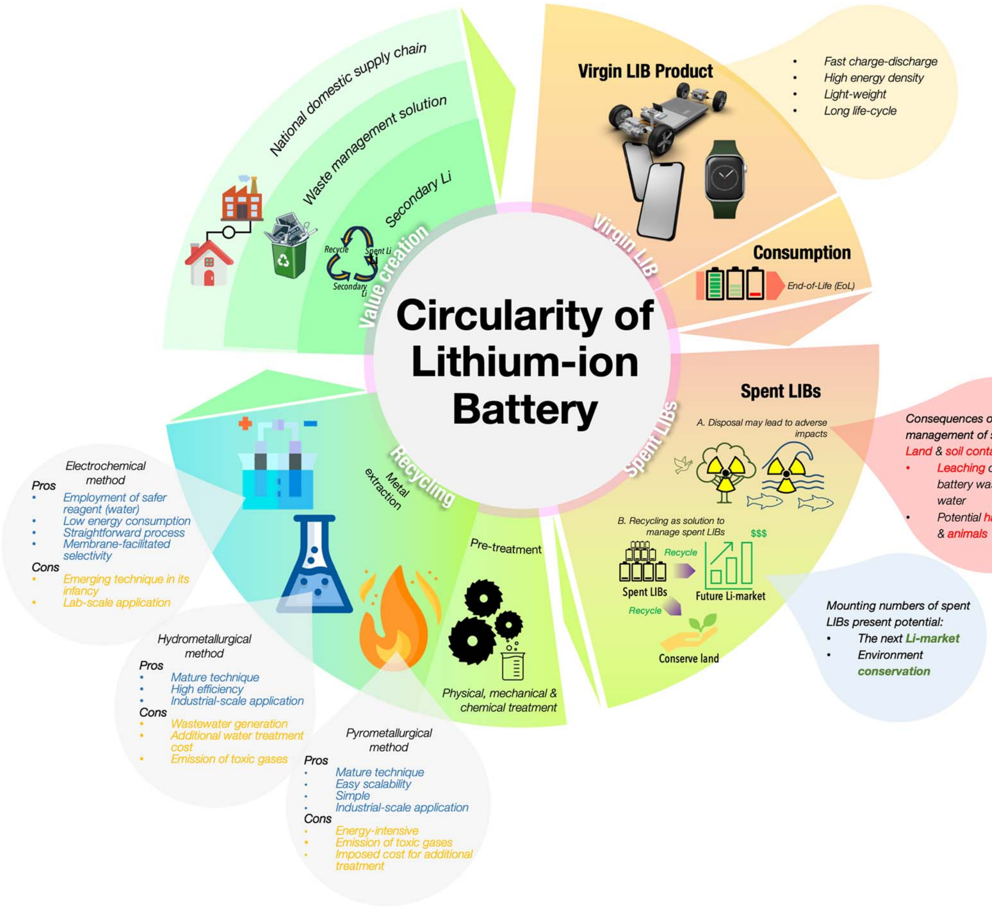

Fig. 1 Circularity of LIBs. Minerals like Li in LIB are precious elements, which are projected to face scarcity. Mounting numbers of disposed spent LIB must be recycled to address the waste management issues while solving the problem of mineral depletion.

in Li recycling. The method still requires solutions from both academia and industry before it can proceed to commercialisation. Despite the availability of metallurgical techniques like hydro- and pyrometallurgy at the industrial scale, their drawbacks in terms of adverse effects on the environment and high cost have ignited research into the emerging electrochemical recycling technique. This review aims to lay a foundation for addressing critical areas that remain to be tackled for the expansion of the electrochemical recycling technique and to provide insights into its path toward commercialisation.

Current trends in lithium consumption

LIB are the forebearers of energy storage technology and stand at the core of the clean energy and net-zero emission agenda. The first-ever LIB were commercialised by SONY Corporation in 1991, based on Asahi Kasei Corporation's first assembly of $Li_{1-x}CoO_2/C_6$.[18] The path of rechargeable batteries based on Li has advanced robustly since then. Their rechargeability, extended life cycle, broad operational temperature range, and lightweight nature position LIB for versatile applications, spanning from personal appliances like smartphones, smartwatches, tablets, and laptops to vehicles and grid energy storage.[19,20] As shown in Fig. 2A, from the year 2021 to 2022, Li production and consumption increased by 21% and 41%, respectively. Fig. 2B indicates that most of the Li consumed is directed toward battery manufacturing.[21]

As of January 2024, global Li reserves stand at 28 million tons (Mt). The ceiling for this figure could potentially increase in coming years with the availability and readiness of advanced technologies that enhance the efficiency of Li production.[22] However, despite this promising trend, the demand for Li is experiencing rapid growth. Electric vehicles (EVs) assume a pivotal role in achieving the pressing global agenda for net-zero emissions.[1,2] A research conducted by Goldman Sachs has forecasted that EVs will constitute 50% of new car sales worldwide by 2035 (Goldman Sachs, 2023). In 2025, it is estimated that the amount of spent LIB could reach 464 000 tonnes, and the forecast from International Energy Agency states that by 2040, the amount of spent LIB accumulated from EVs manufactured in 2019 could reach as high as 8 million tons (see Fig. 2C).[4,5,11] These unprecedented numbers present a detrimental threat to the environment and could hasten the shortage of global mineral reserves.[4]

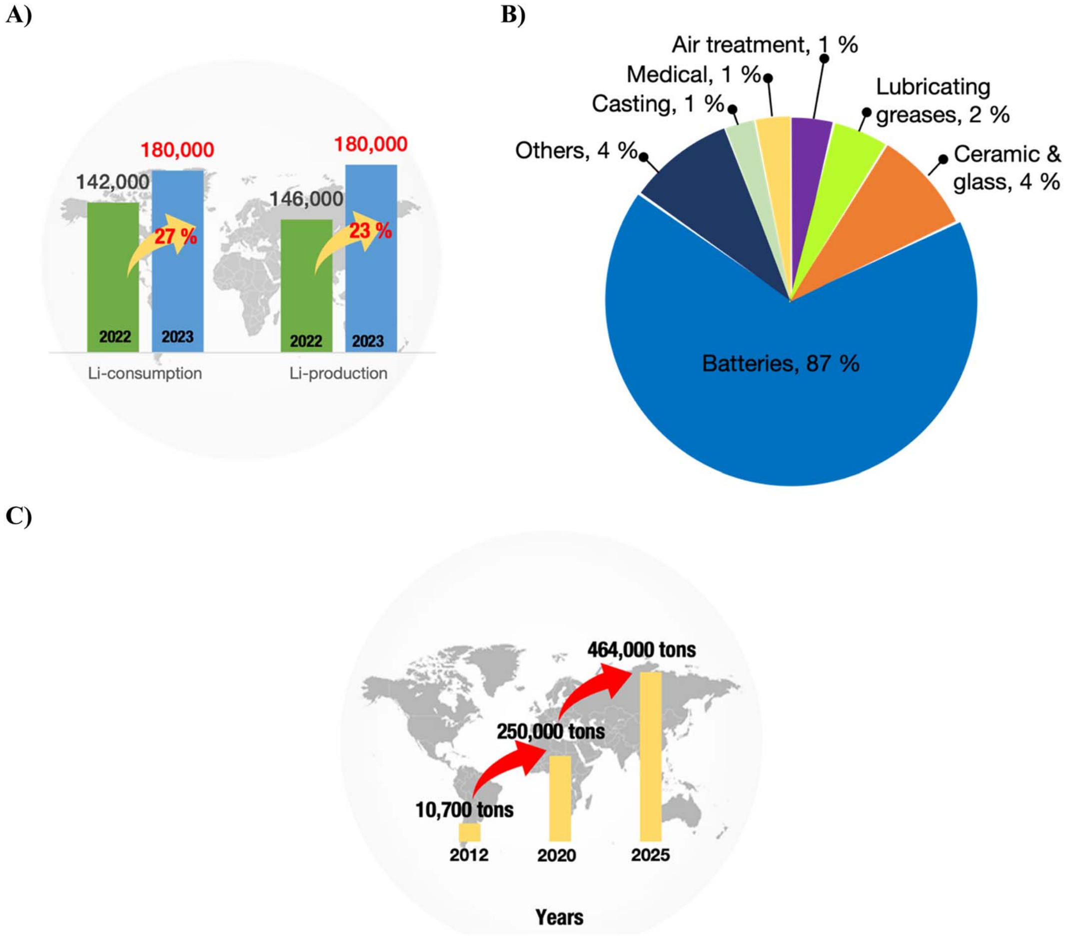

Fig. 2 (A) Li consumption and production in 2023, showing significant increases of about 27% and 23%, respectively. (B) Li usage distribution in 2023. (C) Estimation of the amount of spent LIB generated through the year 2025. It is imperative to note the Li production, consumption and generated spent LIB waste. The generated spent LIB could potentially be the next reserve for Li in LIB.

Confronted with these circumstances, research endeavours have been directed towards a sustainable or circular-path of Li consumption through the recycling of LIB waste, which contains precious metals including Li, cobalt (Co), copper (Cu), manganese (Mn), and nickel (Ni). Spent LIB recycling is a developing industry that is gaining interest worldwide, driven by the economic returns derived from recovering the precious metals such as Li, Co, Mn, and Ni. This approach also serves as a barrier to protect the supply chains of critical materials for energy storage and production.[12] Hydrometallurgy, pyrometallurgy, and electrochemical methods are being actively pursued for this purpose. However, the intricacies of these approaches have sparked a new concept of recycling spent LIBs. To relieve the raw material pressure for sustainable battery production, ongoing research on spent LIB recycling has focused on the concept of transforming or upcycling various types of degraded LIB cathodes, such as $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$, $LiMn_2O_4$, and $LiCoO_2$, into new regenerated forms for next-generation battery manufacturing.[1,23,24] The advantages of this approach lie in the simplicity of the process, its low cost and environmental friendliness, and the ability to directly reuse the regenerated products.[2] Recycling and upcycling uphold the greater goal of addressing the need for sustainable production of LIBs. It is notable that the diversity of approaches towards this common goal has ignited and contributed to the dynamic and rapid development of methods for the recycling of spent LIBs. Here, it is worth mentioning the work on spent LIB recycling has paid attention to the other integrated parts, such as the anode, which is composed of valuable materials such as copper, graphite and residual Li, and the liquid electrolyte, which is also of interest due to presence of the mineral Li.[2] This demonstrates that the prospective recycling industry for spent LIB could prosper in the coming years with a united vision of protecting the mineral reserves while preserving battery production.

Given that recycling stands as a crucial pillar of energy security, it is imperative for academic and industry stakeholders to collaborate in refining both existing and upcoming recycling technologies to achieve a technology that is cost-effective, sustainable, and efficient in serving its purpose.

Commercialised lithium recycling methods

Precious metals such as Li, Mn, Cu, Co, and Ni are present in abundance in spent LIBs. The recycling rates of most of these elements exceed 70%; however, disconcertingly, the recycling rate for Li is below 50%, and the total percentage of recovered Li is merely ∼9% (see Fig. 3).[25] The inconsistency in the recycling rates raises concerns about efficiency and stresses the urgency of Li recycling initiatives, highlighting the need for intensified efforts in the reality of escalating resource challenges. Fig. 3 details the recovery rates of minerals from spent LIBs, which encompasses the rates from the total generated spent LIB (X^{Total}) and the rates from the total spent LIB entering recycling plants (X^{Treat}).[25]

The prevalent approach to Li recycling involves metallurgical and electrochemical methods. As shown in Fig. 4, the processing route for the recovery of Li and other precious metals from spent LIB will include the following: (i) preliminary treatment/pretreatment and (ii) metal extraction. The pretreatment step involves discharging, dismantling, comminution, and separation of individual parts by means of physical methods like sieving, flotation, magnetic, or electromagnetic separation. This preliminary treatment plays a crucial role in achieving high purity for the recovered metals and contributes to the energy-saving aspect of the recycling process.[12,26–29] Considering the differences in battery assembly and cathode chemistry, and the wide range of metal compositions, the pretreatment step is

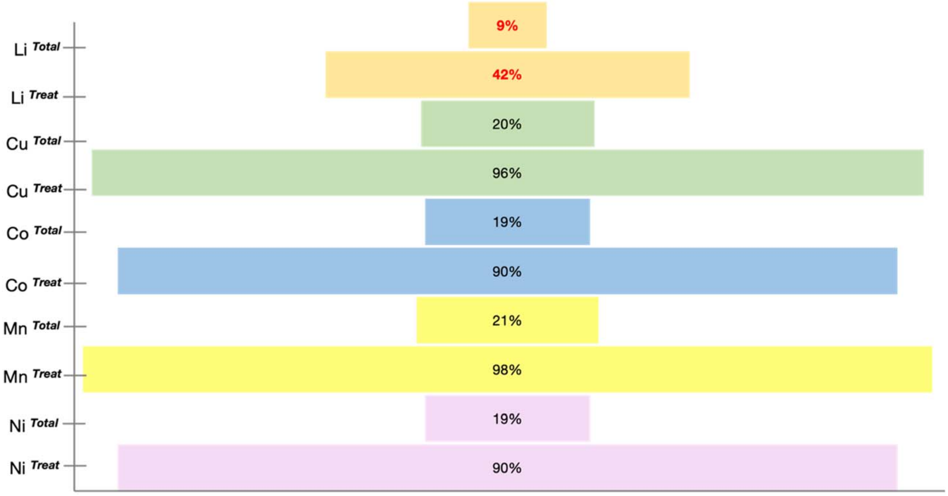

Fig. 3 Recycling rates of Li, Cu, Co, Mn and Ni from spent LIBs. The percentages of Li recycled based on both the total number of spent LIB generated (Li^{Tot}) and spent LIB subjected to recycling (Li^{Treat}) are both low, as highlighted from the chart. The difference in obvious in comparison with the other metals, which have recycling rates of more than 10% with respect to the total number of spent LIB generated (X^{Total}) and ∼90% accounting for the number of spent LIB entering a recycling plant (X^{Treat}).

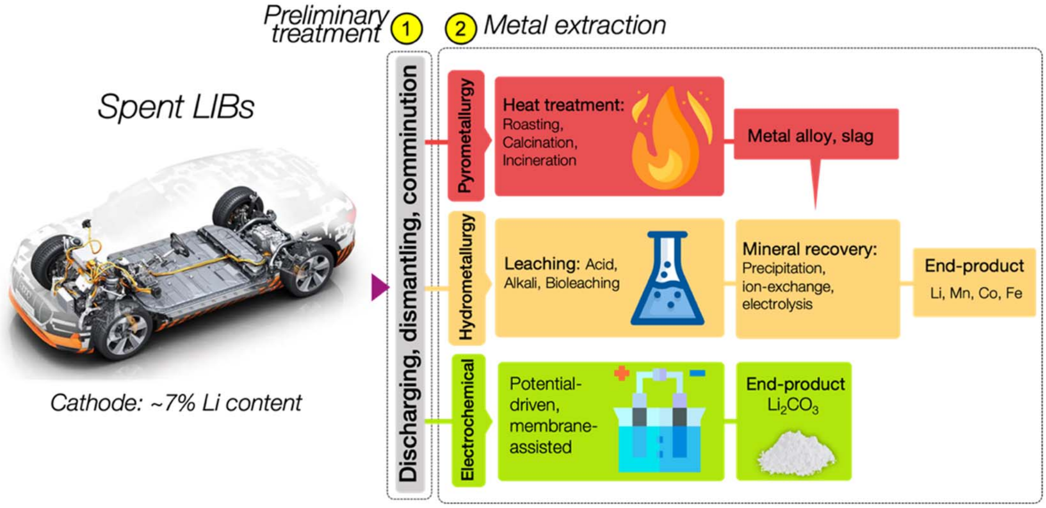

Fig. 4 Li recycling routes from spent LIBs. The process requires the preliminary treatment of the spent LIB prior the metal extraction from pre-treated waste or black mass. The mature processes in the recycling industry are pyro- and hydrometallurgy. The bottleneck in the commercialisation of the electrochemical method lies in addressing several metrics in the upscaling.

imperative to enable selective separation and recovery of metals from the waste.[12,30] In addition to the high purity of the product and energy savings, the pretreatment is important is to guarantee the safety in the subsequent recycling processes. The reactivity of Li in the LIB system is a feature that is beneficial to its performance, but at the same time presents a major concern in the recycling process, and thus the established process of recycling the spent LIB would require pretreatment steps. Thus, to guarantee the safety in the recycling procedure, a pretreatment would ideally aim to make the battery reach the 0% state of charge (SoC) through deactivation by subjecting the battery to a nitrogen cooling environment. There are also discharging methods using a conducting solution or salt-solution immersion discharge and non-electrochemical discharge in a conducting powder.[31–33]

Once the pretreatment phase is concluded, the pre-treated spent LIBs, commonly referred to as black mass, are subsequently subjected to a metal extraction process. At this phase, a specific recycling method or a combination of processes is typically employed to extract Li, particularly using the pyrometallurgy, hydrometallurgy, and electrochemical methods. In the realm of Li recycling, pyro- and hydrometallurgical technologies are already mature and commercialised. They have been widely adopted by recycling companies worldwide, and are at Technology Readiness Level 9 (TRL9). The TRL offers a reliable measure of the maturity of a specific technology, which is assessed by the parameters assigned to each level.[34,35] Fig. 5 briefly presents each TRL stage with its respective parameters. In comparison, the electrochemical method is currently at the lab-scale development and integration level which is TRL4, and hence, the critical phase now focuses on preparing it for an expanded scale of application.

These mature technologies have now been adopted in industry worldwide, but some regions would still require meticulous preparation before adopting these methods. Bahrudin *et al.*, in their writing on the Malaysian perspective towards spent LIBs, highlighted crucial points that are relevant to any country heading toward the adoption of Li recycling technology locally.[36] Emphasis is placed on the prerequisites for recycling interventions, infrastructure, and legislation to be researched and designed beforehand. This prior action should provide the government and local community with sufficient data, adequate infrastructure, and strong policy that sustains the local recycling industry.

Pyrometallurgy

The most studied method in Li and spent LIB recycling is metallurgy, a process that is utilised to extract or refine metals from their ore, or in this case, black mass.[37] Pyrometallurgy is understood as a metallurgical process involving thermal treatment to trigger physical and chemical transformations in a multi-element source or black mass, thereby segregating the elements into individual entities.[38]

The heat treatments used in the pyrometallurgy of black mass include roasting/calcination, incineration, pyrolysis, and smelting; these approaches are further classified into many processes based on their different extraction methods and operating atmospheres.[39–41] The employment of high temperatures ranging from >500 °C to 2000 °C gives pyrometallurgy the advantage of greatly enhancing chemical reactions, surpassing the effects achievable at lower temperatures, at which only phase transitions and structural changes in metals occur.[39,42,43] Companies worldwide including Umicore, Accurec, and Inmetco have employed pyrometallurgical treatment to give alloy metals and slag as the final products, which contain most of the Li remains.[12] However, given its substantial energy consumption, association with high amounts of atmospheric emission, and limited range of recovered products, the pyrometallurgical method for Li recycling is inefficient, environmentally destructive, and unsustainable in the long run.[12,13] It is important to mention the inability of the pyrometallurgical

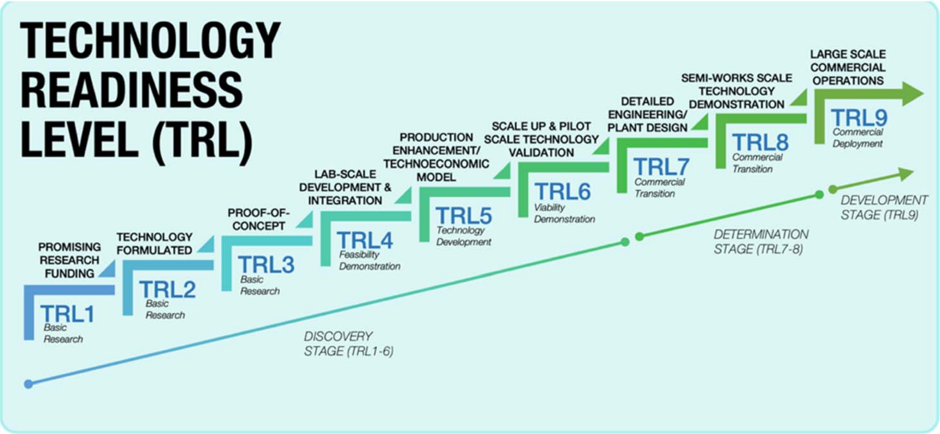

Fig. 5 Stages of the technology readiness level from level 1 until level 9 and their corresponding parameters. It is worth noting that the hydro- and pyrometallurgical methods have reached TRL9 with widespread industrial application. This has made them a viable choice for Li recycling, while the electrochemical method is still a developing technique in its infancy.

method to recycle Li directly. This is related to the greater oxygen affinity of Li, which contributes to Li slagging in the pyrometallurgical process.[44] Thus, the pyrometallurgical method necessitates subjecting the slag to a post-treatment process. This will involve a leaching or hydrometallurgy process; these metallurgical methods will be elaborated upon in a later section. Such post-treatment leads to a higher energy consumption.[45] Table 1 presents the diverse sets of parameters adopted by recycling companies worldwide for the recycling of spent LIB using the pyrometallurgy method. Here it is notable that in the approaches in which the products are not specifically subjected to post-treatment, Li is not recovered.

Fig. 6 displays the recycling route carried out by Umicore, one of the recycling companies with a pyrometallurgical recycling approach.[60] It is worth mentioning that the end-product of the Umicore process following the pyrometallurgy is in the form of alloy and slag. This product will usually necessitate further treatment to segregate the chemical entities of the desired minerals, such as Li.[4,27,61–64] In addition to the additional cost required for the post-treatment of the alloy and slag, the pyrometallurgy process is a high-energy-demand process that contributes to greenhouse gas emissions (GHG).[65,66] Moreover, environmental studies on pyrometallurgy have revealed possible risks including global warming, ozone layer depletion, photochemical ozone creation, carcinogenic and non-carcinogenic effects, and eutrophication due to the pyrometallurgical recycling process.[66–69] These dilemmas of the pyrometallurgical method have directed the interest of academia and industry toward optimising the pyrometallurgical method in recent years. One strategy is minimising the energy consumption to mitigate the GHG emissions through strategies such as synergistic pyrolysis and the utilisation of green reactants such as bio-fuels, hydrogen, and ammonia.[70,71]

Hydrometallurgy

The concept of hydrometallurgy is the transfer or leaching of valuable metals into a solution through a step referred to as 'leaching' employing a leaching agent from an alkali medium, acid medium or microorganisms like bacteria and fungi.[39,72,73] Subsequently, metal recovery from the leachate solution takes place using various processes such as electrolysis, solvent extraction, chemical precipitation, and ion-exchange.[72]

The primary advantage of utilising hydrometallurgy lies in its low energy consumption and higher recovery rates of metals in high purity compared to the pyrometallurgy technique. Despite that, the drawbacks associated with hydrometallurgy require careful consideration. In addition to the use of harsh chemicals as leaching agents in the metal leaching and recovery steps, research has progressed to the application of concentrated leachate to deliver a time-efficient and effective metal recovery. Table 2 lists the recoveries of Li and other precious minerals from spent LIB through hydrometallurgy techniques including alkali inorganic acid leaching, organic acid leaching, alkali leaching, and bioleaching. The hydrometallurgical method offers a solution to recover Li, which is not attainable using pyrometallurgy. As shown in the table, most studies successfully recovered Li. The flexibility of hydrometallurgy lies in the ability to choose among numerous reagents that can deliver the intended result. Compared with pyrometallurgy, hydrometallurgy operates at a lower temperature. Despite the ability to yield Li as the final product, the conventional use of inorganic acids as the reagent in hydrometallurgy generates toxic waste with adverse effects on the environment and human health, which has prompted a shift toward studies employing organic acids like citric acid and H_2O_2 (see Table 2). The efficiency of hydrometallurgical recycling is compromised by its substantial reagent consumption. When executed at the large industrial scale to process large quantities of spent LIBs, hydrometallurgy imposes significant chemical costs and additional expenses for wastewater treatment. Bioleaching hydrometallurgy for the recovery of Li has been recognized in recent years for its cost-effectiveness, environmental friendliness, low energy requirements, and high efficiency with minimal GHG emissions. However, challenges are encountered in the aspects

Table 1 Pyrometallurgy recycling of LIB adapted in industry as reported from Makuza *et al.*[39]

Company	Country	Recycling capacity (t per year)	Types of batteries processed	Pyrometallurgic process	Post-treatments	Products	Secondary products	Waste	Ref.
Valdi (Eramet)	France	20 000	LIB and other battery types	(Not reported)	—	FeNi/FeMn alloy	—	—	46
Umicore	Belgium	7000	LIB, Ni–Cd, Ni–MH	Pyrolysis, 700 °C; smelting 1200–1450 °C	Leaching, solvent extraction	$CoCl_2$, Co, Ni, Cu, Fe	Slag (Al, Si, Ca, Fe, Li, Mn)	Electrolyte, plastics, graphite	47–49
Xstrata (Glencore)	Switzerland	7000	All LIB chemistry	Conditioning (rotary kiln) and introduction into a Co–Ni winning process (EAF)	Hydrometallurgy	Alloy (Co–Ni–Cu)	Li slagged and ignorable metals	No recovery of Li, Al, electrolyte, graphite, plastics	47 and 50
Inmetco	USA	6000	LIB, Ni–Cd, Ni–MH	High-temperature melting recovery (HTMR), 1260 °C	Iron casting	Alloy (Co–Ni–Fe)	Li slag	The organic material used as a chemical reagent	47 and 51
Accurec	Germany	6000	All LIB chemistry	Pyrolysis, 250 °C; carbothermic reduction, 800 °C	Acid leaching	Li_2CO_3 Co-alloy	Metallic alloy	Electrolyte, polymers, graphite	52–54
JX Nippon mining and metals	Japan	5000	—	Smelting	Leaching, selective precipitation, electrowinning	Li_2CO_3, $MnCO_3$, Co	—	Electrolyte	46, 55 and 56
SNAM	France	300	—	(Not reported)	—	Black mass (Cu, Ni, Co)	—		47
Sumitomo	Japan	150	LIB ($LiCoO_2$)	Calcination, 1000 °C	Acid leaching, hydrometallurgy	CoO	Co–Ni–Fe alloy Cu, Al, Fe	Electrolyte, plastics, Li, Ni, graphite	41, 57, 58
G&P batteries	UK	145	LIB	(Not reported)	(Not reported)	—	—	—	59
LithoRec	Germany	2000	—	Drying, calcination	Undisclosed leaching agent	Li_2CO_3, metal oxides	Al–Cu, plastic fractions	Electrolyte	52
GRS batteries	Germany	—	$LiMnO_2$	Vacuum distillation	—	Co, Ni, Cu, FeNi and FeMn	—	—	59
Battery resources	Germany	—	LIB, Ni–MH	Sintering	Leaching by NaOH, H_2O_2, H_2SO_4, Na_2CO_3	Li_2CO_3, metal oxides	Al–Cu, plastic fractions	Electrolyte	52

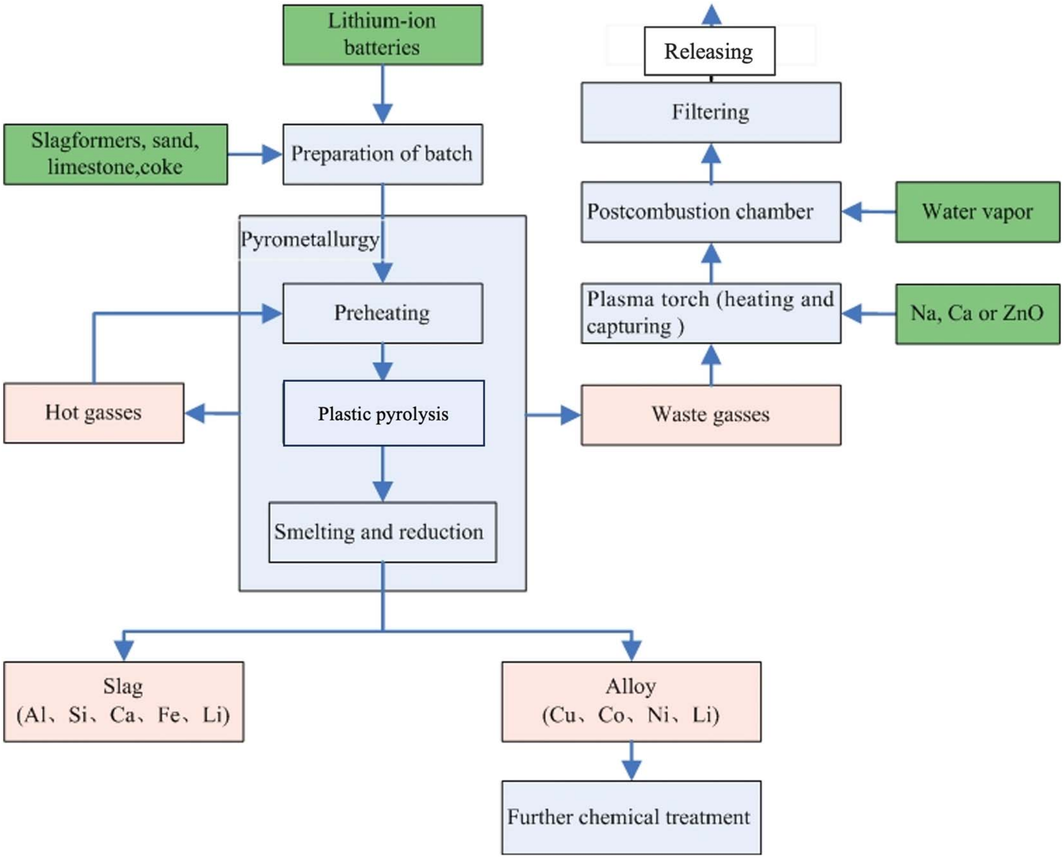

Fig. 6 Umicore pyrometallurgical recycling route.[60] The process involves initial preheating at a temperature <300 °C to evaporate the electrolyte, followed by pyrolysis of plastics at 700 °C, whereby the emitted hot gases are resupplied for the preheating. At the last stage, smelting/reduction takes place at 1200–1450 °C, resulting in slag (aluminium, silicon, calcium, iron) and alloy (copper, cobalt, nickel, Li, small quantities of iron). Beyond this stage, further leaching of the separated elements is required.

of scalability and commercialisation, notably when there is an excessive input time compared to the traditional acid and alkali leaching methods in hydrometallurgy.[74]

As described in the previous section, the implementation of pyrometallurgy mostly relies on post-treatment methods to leach or extract the metals from the alloy or slag formed, and thus includes hydrometallurgy as one of the necessary treatments, owing to its efficiency and selectivity. Worldwide, hydrometallurgy and combined pyro/hydrometallurgy techniques have been employed at the pilot and commercial scale. Provided in Table 3 are the companies that have committed to either the singular metallurgical method or combined metallurgical method. It is worth stating that the hydrometallurgical method is normally implemented to follow the recovery of metal through pyrometallurgy. The ability of hydrometallurgy to allow the recovery of most of the metals produced as alloy and slag in pyrometallurgy makes it a necessary post-treatment. On the other hand, the execution of the hydrometallurgy method alone can successfully recover Li and other precious metals from spent LIB without the need to perform pyrometallurgy. The use of thermal treatment to drive the reaction in pyrometallurgy leads to an excessive amount of energy being consumed compared to hydrometallurgy. It is unfair to say that hydrometallurgy is more cost-effective than the pyrometallurgy method by looking exclusively at the energy consumption, as the overall operation of hydrometallurgy imposes external costs including reagent costs and wastewater treatment expense. Nonetheless, when considering the energy consumption factor, it is simple to conclude the thermal treatment in pyrometallurgy is the most energy-consuming, followed by hydrometallurgy, as shown in Table 3 below.

While the recycling output holds promise, and such pyro/hydrometallurgy recycling strategies have been implemented on the pilot and commercial scale, the pursuit of this strategy may prove more harmful and costly considering the metrics of energy and chemical consumption. The trade-offs made to maximize recycling benefits lead to increased recycling costs, raising concerns. In addition to the GHG emissions from the energy-demanding process of pyrometallurgy, the hydrometallurgy process produces gases such as chlorine (Cl_2), sulfur dioxide (SO_2), and nitrogen oxides (NO_x), which are harmful to the environment and human health. A study also revealed that there is a risk of freshwater and terrestrial acidification from hydrometallurgy recycling, creating a requirement for wastewater treatment to mitigate the pollution threat in the receiving waters.[12,69,72,96] This scenario introduces complexities in

Table 2 Hydrometallurgical recycling approach from multiple studies as reported by Dobó *et al.*[72]

LIB type	Leaching agent	Leaching conditions	Recycling efficiency (%)	Ref.
Inorganic acid				
$LiCoO_2$	2 M H_2SO_4 + 5 vol% H_2O_2	80 °C, 1 h, 50 g L^{-1}	Li = 99, Co = 99	75
$LiCoO_2$	2 M H_2SO_4 + 50 g per L glucose	80 °C, 2 h, 35 g L^{-1}	Li = 92, Co = 88	76
NCM	1 M H_2SO_4 + 1 vol% H_2O_2	40 °C, 1 h, 40 g L^{-1}	Li/Co/Ni/Mn = 99.7	77
NCA	4 M HCl, 90 °C, 18 h with a 5% (*w*/*v*) solid to liquid ratio	90 °C, 18 h, 5% (*w*/*v*)	Li/Co/Ni/Al = 100	78
$LiFePO_4$	2.5 M H_2 SO_4	60 °C, 4 h, 100 g L^{-1}	Li = 97, Fe = 98	79
Mixture	1.34 M H_2SO_4 + 0.45 g per g $Na_2S_2O_5$	20 °C, 45 min, 10.9%	Mn = 94, Cd = 81, Zn = 99, Co = 96, Ni = 68	80
Mixture	1 M H_2SO_4 + 0.075 M $NaHSO_3$	95 °C, 4 h, 20 g L^{-1}	Li = 97, Co = 92, Ni = 96, Mn = 88	81
Organic acid				
$LiCoO_2$	1.25 M citric acid + 1 vol% H_2O_2	90 °C, 0.5 h, 20 g L^{-1}	Li = 99, Co = 91	82
$LiCoO_2$	0.1 M citric acid + 0.02 M ascorbic acid	80 °C, 6 h, 10 g L^{-1}	Li = 100, Co = 80	83
NCM	1 M citric acid + 12 vol% H_2O_2	60 °C, 40 min, 80 g L^{-1}	Total metals >98	84
NCM	3.5 M acetic acid + 4 vol% H_2O_2	60 °C, 1 h, 40 g L^{-1}	Li = 99.97, Co = 93.6, Ni = 92.7, Mn = 96.3	85
Mixture	1.5 M citric acid + 0.5 g per g glucose	80 °C, 2 h, 20 g L^{-1}	Li = 99, Co = 92, Ni = 91, Mn = 94	86
Mixture	2 M tartaric acid + 4 vol% H_2O_2	70 °C, 0.5 h, 17 g L^{-1}	Li = 97.7, Ni = 98.2, Co = 98.9, Mn = 98.4	87
Alkali leaching				
NCM	367.5 g per L $NH_3 \cdot H_2O$ + 140 g per L NH_4HCO_3 + 63.24 g per L H_2O_2	—	Li = 81.2, Co = 96.3, Ni = 96.4, Mn: hardly leached out	88
Mixture	15 wt% NaOH	50 °C, 1 h, 0.1 g mL^{-1}	Al = 58	89
Mixture	1.5 M NH_3 + 1 M $(NH_4)_2SO_3$ + 1 M NH_4HCO_3	60 °C, 3 h, 20 g L^{-1}	Li = 60.53, Co = 80.99, Ni = 96.32	90
Mixture	3 M $(NH_4)_2SO_4$ + 0.75 M $(NH_4)_2SO_3$	180 °C, 2 h, 83 g L^{-1}	Li = 98, Co = 81, Ni = 98, Mn = 92	91
Bioleaching				
$LiCoO_2$	Bacteria: *Acidithiobacillus ferrooxidans* + 0.75 g per L copper ions	35 °C,6 d	Co = 99.9	92
$LiCoO_2$	Bacteria: *Acidithiobacillus ferrooxidans* + 0.02 g per L Ag^+	35 °C, 7 d	Co = 98.4	93
Mixture	Bacteria: *Acidithiobacillus ferrooxidans* + elemental sulphur 1% + 3 g per L Fe(II)	30 °C, 20 d, 5 g L^{-1}	Li = 10, Co = 65	94

Table 3 Several industries worldwide implementing the combined metallurgical recycling techniques as reported by Jung *et al.*[a][95]

Company	Pyrometallurgy	Hydrometallurgy	Recovered mineral	Energy consumption	Status in the year 2020
America manganese	—	H_2SO_4, SO_2 reductive leach	LiOH, $Ni(OH)_2$, $Co(OH)_2$, $Mn(OH)_2$, Al	Low	Pilot plant
Brunpt recycling	—	H_2SO_4, H_2O_2 reductive leach, solvent extraction	Ni, Co, Mn, Al	Medium	Commercial plant
Duesenfeld	Calcination	Solvent leaching	Li_2CO_3 and metal oxide	High	Pilot plant
Li-cycle	—	H_2SO_4, H_2O_2, reductive leach, solvent extraction	$NiSO_4$, $CoSO_4$, $MnCO_3$, Li_2CO_3	Medium	Commercial plant
Lithion recycling	—	Leaching	Ni, Co, Mn, Al	Medium	Pilot plant
OnTo technology	Brief heating	Hydrothermal	Cathode material	Medium	Pilot plant
Sumitomo	Smelting	Electrowinning H_2SO_4 leaching	Ni, Co, Cu	High	Commercial plant
Umicore	Smelting	Leaching, solvent extraction, Co, Ni refining	$Ni(OH)_2$, $LiMeO_2$	High	Commercial plant

[a] High energy consumption reflects the employment of both the pyrometallurgy and hydrometallurgy methods together, while medium and low energy consumption reflect the employment of the hydrometallurgy method.

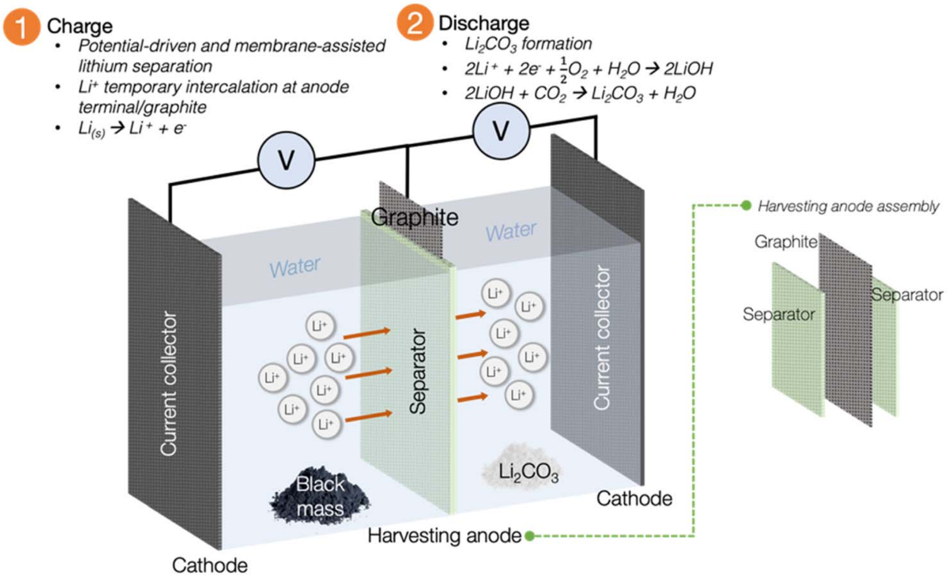

Fig. 7 Illustration of the electrochemical setup and separator-integrated electrode assembly (adapted from ref. 11 and 14). Employing water as the main reagent could considerably cut the chemical consumption cost. The operational mechanism, which relies on a minimal current supply can presumably be less energy-consuming than pyrometallurgy, while the selectivity of the process towards the element of interest is facilitated by the high ionic conductivity of the membrane.

addressing health risks and environmental impacts from the recycling process, requiring a nuanced approach to balance the potential benefits and the drawbacks.

The risks associated with the current recycling methods have pushed the boundaries of Li recycling beyond metallurgy. In addition, based on the pursuit of sustainability, the quest for greener recycling methods has prevailed. In the past years, the electrochemical technique has appeared as the better solution. This method is anticipated to succeed as a commercialised method, but the following key areas require attention and effective solution from experts.

Electrochemical method

The innovative approach of Li recycling *via* the electrochemical method has garnered a significant attention from researchers and industry due to its more sustainable operational mechanism. The initial exploration was first undertaken by the Asl group in 2012, and mainly concentrated on developing a Li-liquid battery using spent LIB as the Li for the energy source.[97] Building upon this pioneering study, the Bae group in 2016 leveraged the success achieved by Asl, directing their efforts towards a new emphasis on recycling Li from spent LIBs.[11,14] Their venture marked a transformative development in the landscape of the recycling industry's future. The quest for electrochemical recycling of spent LIB has been cultivated since then, with studies moving in the direction of emulating the electrochemical cell to harvest Li from spent sources.

Using the electrochemical concept, Li from spent LIB has been successfully harvested into one of the commercial precursor forms of Li_2CO_3 through reaction with water.[11,14] The advantage of accomplishing recycling through the electrochemical technique, offers flexibility of choosing a safer reagent to recycle Li and other precious minerals compared with traditional hydrometallurgical approach. The avoidance of toxic or harmful reagents, along with the environmentally friendly operating conditions and the simplicity of the charge–discharge process, make this method an economical and sustainable substitute to existing metallurgical recycling approaches. Fig. 7 is a representation of the first electrochemical recycling attempt by Bae's group. In brief, the black mass from the pretreatment procedure is placed in the ‘waste compartment’. Water is used as the main reagent, and the presence of a small amount of electrolyte in the system is crucial to facilitate the movement of Li ions across the system. The system's selectivity for Li^+ and its temporary intercalation are facilitated by the separator unit and graphite within the harvesting anode assembly, respectively. Following the completion of the charging process, the temporarily intercalated Li^+ undergoes a subsequent discharging procedure to be transformed into the common precursor Li_2CO_3 through reaction with water. This profound exploration by Bae was the inspiration that has propelled contemporary research towards the electrochemical method of Li recycling.

Table 4 compiles the studies conducted on the electrochemical recycling of Li, and Fig. 8A–G are schematic diagrams of the concept from the respective studies. Over the years, there have been diverse selections of electrode, electrolyte, and separator materials, which have contributed to increased efficiency in the recycling process. Water has consistently played a crucial role as the primary reagent for the electrolyte across various attempts. It is more cost-effective than other hazardous and toxic reagents employed in the hydrometallurgy technique. Importantly, all attempts employing the electrochemical

Table 4 Attempts at Li recycling using the electrochemical method as reported by Li *et al.*[98]

Spent Li source	Operating temperature	Electrode	Membrane	Electrolyte	Recycling efficiency (%)	Product	Commercialisation status	Ref.
$LiFePO_4$	Room temperature	Spent $LiFePO_4$ mixed with carbon black (cathode) stainless steel (anode)	Inorganic solid polymer electrolyte membrane	Water	—	Li metal	Laboratory scale research	97
$LiFePO_4$, $LiMn_2O_4$, $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	Room temperature	Copper (cathode) and graphite (anode)	Inorganic solid polymer electrolyte membrane	Water and $LiPF_6$ in EC-DMC	Li = 75%	Li_2CO_3		14
$LiFePO_4$	Room temperature	Ruthenium-plated titanium	Anion-exchange membrane	Water and NaCl	Li = 98.0%, Fe = 96.0%	NaOH, $FePO_4$, Li_2CO_3		16
$LiFePO_4$	Room temperature	Ruthenium-plated titanium	Cation-exchange membrane	Water, Li_2SO_4, and LiOH	Li = 96.0%	$FePO_4$, $LiOH\cdot H_2O$		15
$LiMn_2O_4$	90 °C	Graphite (cathode) and ruthenium-plated titanium (anode)	Filter cloth	Water, H_2SO_4 and $MnSO_4$	Li = 99.0%, Mn = 92.0%	Li_2CO_3, MnO_2		99
$LiCoO_2$	Room temperature	Spent Li_xCoO_2 and pristine $LiCoO_2$	Porous polymer membrane	1.5 M Li_2SO_4 solution	—	Regenerated $LiCoO_2$		100
$LiCoO_2$	60 °C	Platinum-plated titanium base electrode plates	Acrylic fabric	Solution of mixed NH_4HCO_3 and $(NH_4)_2SO_3$, and NaF	Li = 71.8%, Co = 55.4%	Regenerated $LiCoO_2$		101
$LiCoO_2$	100 °C	Pt cathode and Ni plate anode	—	LiOH solution	—	Regenerated $LiCoO_2$		102

technique resulted in minimal-to-no wastewater generation. This not only reduces the environmental impact associated with disposal into receiving water, but also lowers the costs related to wastewater treatment. The compilation of studies highlights that the electrochemical recycling approach has proven to be an efficient and environmentally conscious method for Li recycling. Employing electrical energy to drive the reaction is assumed to be a superior option to the thermal treatment of pyrometallurgy, which consumes more energy and emits harmful gasses.

The exploration of the electrochemical method in the field of spent LIB recycling has seen its application in the direct recovery of precious elements. Additionally, the unique concept of the electrochemical method allows its application in the regeneration or rejuvenation of spent LIB cathodes. Compared with other regeneration approaches, the electrochemical regeneration process is less energy-consuming and uses less reagent compared to solid phase regeneration and hydrometallurgical regeneration.[98] With lower cost and a facile process to produce a regenerated cathode, the electrochemical regeneration method has been deemed to be another attractive solution for sustainable battery production. Table 4 lists studies in which the electrochemical recycling method was attempted. The ability of the electrochemical method to make progress in achieving high efficiency of recovery is one of the excellent indicators of the performance of this method. With a facile operation and controlled environmental impact through usage of less detrimental reagents, this method is anticipated to be available in industrial scale in a few years. Apart from the recycling of minerals like Li, the electrochemical method is beneficial in the emerging technique of regenerating degraded cathodes from LIBs. In light of this electrochemical regeneration technique, which involves the relithiation of the Li-deficient spent cathode, it is important to understand that the reasons for the degradation of the electrochemical performance of LIB are not merely affected by the absence of Li. A wider perspective includes the influence of structural distortion and impurities on the spent cathodes. Therefore, an electrochemical regeneration process that includes the approach of the relithiation of degraded cathodes is undeniably needed to solve the identified issues. Hence, the electrochemical regeneration method, as well as other regeneration processes, must be able to handle the raw material impurities, the poor adaptability of the regenerated cathodes to the raw materials, and the inferior electrochemical performance of the regenerated products compared to virgin materials.[98]

Fig. 8 illustrates the electrochemical recycling concepts from several studies, which are referred to in Table 4. The initial discovery of Li recycling through the electrochemical technique was reported in the studies of Asl *et al.*, who originally envisaged the potential to make Li-liquid batteries powered by spent LIBs.[97] Their successful attempt to harvest Li powder from spent sources later inspired the studies of Li recycling using the electrochemical concept by Bae *et al.* in which more than 75% Li was recovered.[14]

Following these successful attempts, studies on the electrochemical recycling of Li prospered. Li *et al.* used a setup with

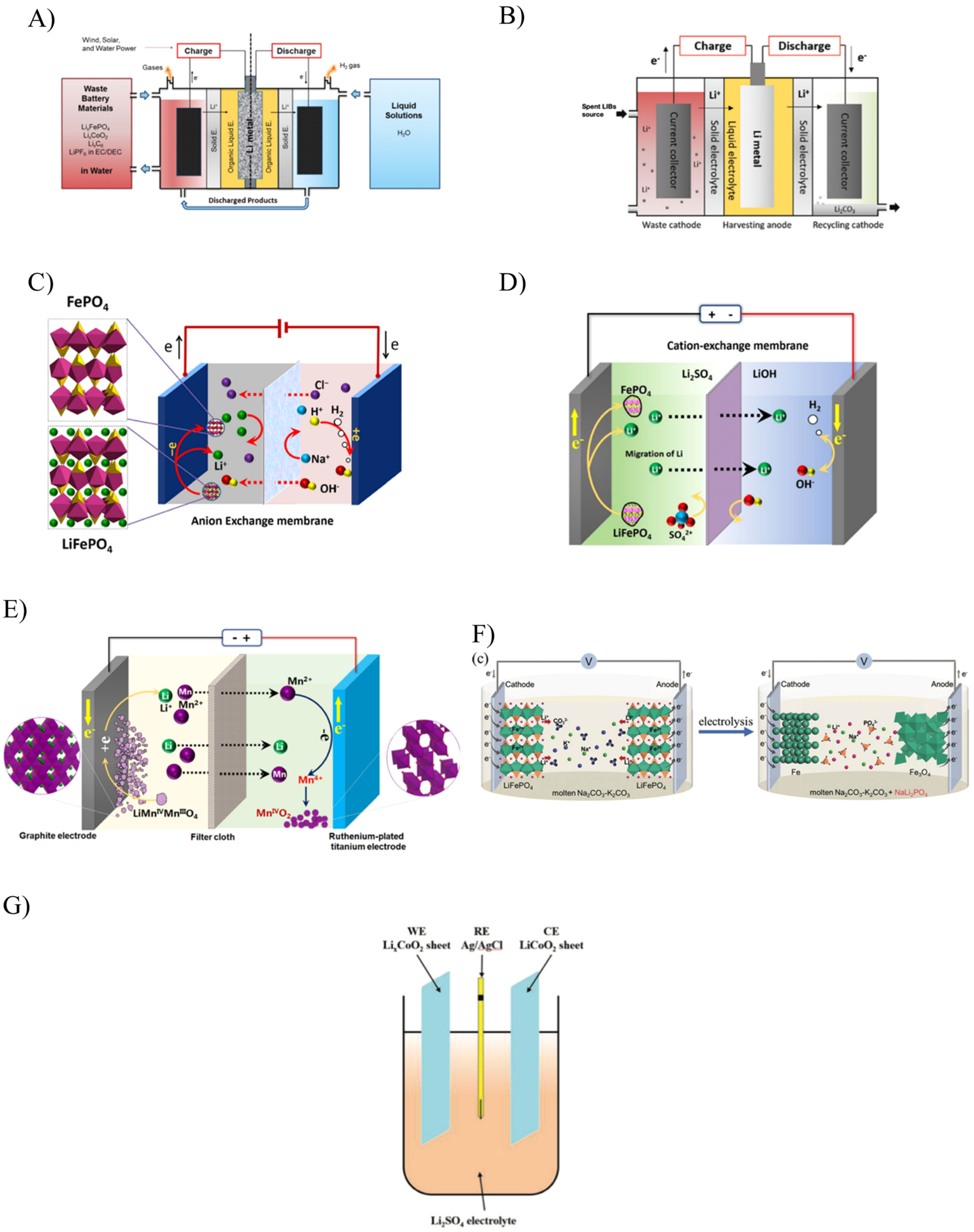

Fig. 8 A–G are schematic illustrations of Li recycling studies using the electrochemical method. The displayed figures are representations from ref. 14–16, 97, 99, 103, and 104. The use of a current-driven process in the electrochemical method greatly reduces the consumption of reagent while avoiding Li loss in the process of separation from its spent sources. Studies using the electrochemical method have successfully recycled other precious minerals in addition to Li from spent $LiFePO_4$, $LiCoO_2$, $LiMn_2O_4$ and $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$. A diverse range of setups have been explored in studies on the electrochemical recycling method, paving the way for its improvement, including improvement in the cost, which is dictated by the recovery efficiency.

a ruthenium-plated titanium electrode paired with the simple electrolyte of sodium chloride (NaCl), which delivered 98% Li recovery.[16] Using a different electrolyte, Li *et al.* were able to achieve 96% recovery for Li.[15] Using NaCl, the process was much more cost-saving, but it led to impurities, as reflected in the production of the by-product, sodium hydroxide (NaOH). Using a Li salt as the electrolyte avoids this scenario while also contributing to enhanced electrical conductivity.[98] Despite the issue of impurities, choosing NaOH as the electrolyte seems to be more reliable, delivering the desired product with less energy consumption and cost-savings. The recovery of Li in the form of Li_2CO_3 and manganese as Mn_2O_4 from spent $LiMn_2O_4$ was also realised by the electrochemical method, as reported by Li *et al.*[17] A 99% and 92% recovery efficiency for Li and Mn, respectively, were obtained in this trial, but the operating conditions rely on an elevated temperature, incurring an additional cost. The use of the electrochemical method to regenerate spent batteries, on the other hand, offers a very straightforward procedure, but heavily relies on the reagents used in the relithiation of the Li-deficient cathode. As discussed earlier, the issues with spent cathode of LIB is not exclusive only to Li-deficient. In a broader spectrum, the issues entail the structural distortion and impurities that require a much more holistic solution than the regeneration technique and this demands further extensive research.[103,104]

Despite the success of the electrochemical method to recover most metals with high efficiency, contemporary recycling research has underscored the deficient selectivity of the recycling methods in terms of the products. Although hydrometallurgical methods are an efficient process comparable to the electrochemical method, their selectivity remains problematic.[105,106] Research navigating around the specific selectivity of recycling methods has involved the electrochemical recycling technique.[105] A multifaceted strategy can be used to reach recycling output with high selectivity integrating the electrochemical method. Li *et al.* reported an approach to yield selective Li and Co recovery from $LiCoO_2$ using electrochemical leaching and electrodeposition.[105] The multiple electrochemical process strategy gave a recycling output with 97% of Li and 92% of Co recovered without contamination from impurities.

It is clear that the electrochemical recycling technique provides a solution. Its status as a safer and more economical method has positioned it as the keystone to sustaining the Li recycling industry. This contributes to the aforementioned threefold goal of recycling: to decelerate resource depletion, to contribute to environmental conservation, and to establish a national domestic supply chain for the elements. This vision, however, requires a collective effort from experts of multiple disciplines to ensure that this promising technique can be commercialised and mature as a technology to compete with and potentially replace the hydrometallurgy and pyrometallurgy techniques.

Challenges in upscaling the recovery of lithium from spent LIB using electrochemical methods

Despite the maturity of the pyro- and hydrometallurgical technology, there is a continuous pursuit of novel technologies that take into account the sustainability aspect. The intended

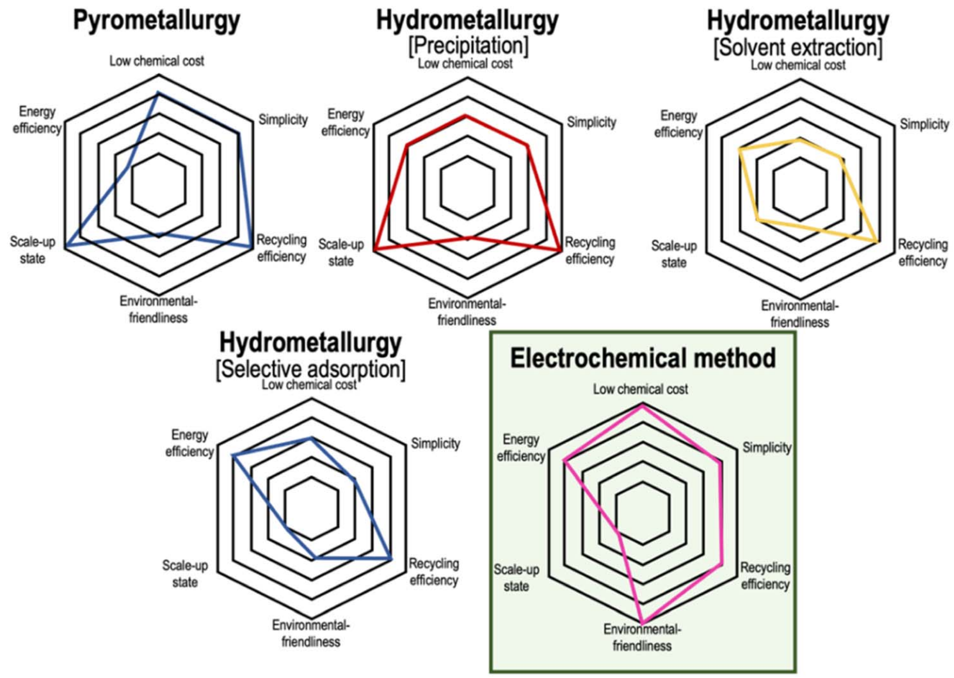

Fig. 9 Spider plot highlighting a comparison of several metrics between recycling techniques. The electrochemical method is a much safer choice, but it is still some time away from commercialization (ref. 10).

refinements will encapsulate guarantees of environmental safety and cost-effectiveness. Although the electrochemical method appears to be the suitable pick, it is still a considerable time away from commercialisation. The spider plot in Fig. 9 delineates various metrics that serve as prominent aspects of comparison and portray the electrochemical method as a green alternative to metallurgy. However, the transformative potential of this method hinges on overcoming the challenge of upscaling, which is a crucial hurdle that demands immediate attention for the successful commercialisation of this groundbreaking approach.

The subsequent section briefly explores the essential elements pivotal to the electrochemical technique, emphasizing a fresh perspective focused primarily on each integrated part or unit involved in its setup. This will serve as a basis for achieving the extensive and enduring application of the electrochemical method on a large scale.

Feasibility of a large-scale setup

For the recycling industry to embrace the electrochemical technology, the pivotal factor is feasibility assessment of this technology on a substantial scale. This stresses the critical need for larger dimensions and expansive setups enabling higher operational capacities.[98] The anticipation is that these larger-scale operations will process substantial quantities of spent waste. This aspect of scalability gives rise to a vital concern regarding the feasibility of electrochemical setups capable of processing or recycling >1000 tonne per year of spent LIBs, which is equivalent to the industrial scale capacity. Table 5 below enumerates the global recycling companies that have effectively implemented recycling techniques on an industrial scale. The prevailing trend among these companies is the utilization of metallurgical methods, with none presently employing electrochemical techniques.

Most of these recycling companies employ pyro- and hydrometallurgy at the industrial scale, while the electrochemical method has yet to reach this scale. The full realisation of the potential and benefits offered by the electrochemical method as an alternative to metallurgical techniques hinges on intensive efforts to transition from laboratory-scale applications to industrial-scale operations. The inability to bridge this gap would mean that the true potential of this method would remain untapped, prolonging the dominance of traditional metallurgical approaches in the recycling industry. Early findings from Halizan *et al.* in 2023 revealed that a few companies had attempted to commercialise this method, including OnTo Technology, Farasis Energy, Green Li-ion and Evonik.[108–110] This signals a future trend toward a potential transition of recycling technology to the electrochemical technique.

Despite uncertainties surrounding the feasibility of establishing an industry-suitable setup for electrochemical techniques, a potential solution lies in drawing inspiration from other successful electrochemical-related industries, including electrowinning and electroplating, which have successfully operated at industrial-scale capacities. Emulating their scale-up strategies could provide a pathway to leverage the advantages of electrochemical methods and facilitate the scalability of this

Table 5 Recycling companies worldwide and their scale of operation as reported in the literature; adapted from ref. 107[a]

Scale	Company (country)	Recycling method
Industrial scale (>1000 tonnes per year)	Umicore (Belgium)	Pyro + hydro
	Accurec (Germany)	Thermal + mech + pyro + hydro
	Nickelhütte Aue (Germany)	Thermal + pyro + hydro
	SungEel (Korea)	Mech + (thermal +) hydro
	Kyoei Seiko (Japan)	Pyro
	Dowa (Japan)	Thermal + pyro + hydro
	Brunp (China)	Thermal + mech + hydro
	GEM (China)	Mech + hydro
	Huayou Cobalt (China)	Mech + hydro
	Ganzhou highpower (China)	Mech + pyro + hydro
Pilot scale (<1000 tonnes per year)	SNAM (France)	
	EDI (France)	Mech (aqueous shred + unknown)
	TES-AMM (formerly "Recupyl") (France)	Mech (inert gas)
	AkkuSer (Finland)	Mech + unknown
	Duesenfeld (Germany)	Mech + hydro
	Promesa (Germany)	Mech (aqueous shred) + unknown
	Redux (Germany)	Thermal + Mech + unknown
	Retriev (US)	Mech (aqueous) + hydro
	Kobar (Korea)	Mech + hydro
	JX Nippon (Japan)	Thermal + mech + hydro
	Telerecycle (China)	Mech + hydro
	Guanghua (China)	Mech + hydro
Lab scale	Erlos (Germany)	Mech + reconditioning (direct recycling)

[a] Thermal: thermal pretreatment. Mech: mechanical treatment. Pyro: pyrometallurgy. Hydro: hydrometallurgy. Industrial scale: >1000 t/a. Pilot scale: <1000 t/a (*e.g.*, a few hundred tons per year). Lab-scale: small-scale trials (a few grams or kilograms), or sometimes pilot-scale trials (a few hundred kilograms or more), but not yet commercialized.

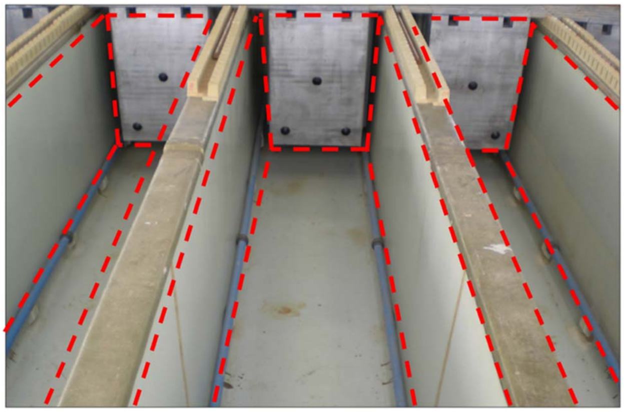

Fig. 10 Industrial scale setup of copper electrowinning. The implementation of such a setup could be a 'blueprint' for feasible large-scale setup of the electrochemical recycling process.

infant technology. Fig. 10 depicts the setup for the electrowinning of copper in industry.[111] This type of layout permits multiple electrowinning procedures to be performed simultaneously, directly impacting the efficiency and cost of the process. Apart from the compartment design, many nuances and intricacies were successfully addressed in transforming electrowinning technology from the laboratory-scale to the industrial-scale; most of the addressed concerns could shed light on addressing the upscaling dilemma in the electrochemical recycling technique.

Substantial electrolyte consumption

In the broader perspective, it becomes evident that while laboratory-scale operation of the electrochemical recycling method only requires a manageable volume of electrolyte, the transition to an industrial scale, with its expansive setup, demands a comparable increase in electrolyte volume to accommodate a higher quantity of spent LIBs. The consumption of electrolyte emerges as a central concern in expanding the electrochemical method, not only impacting costs but also affecting the sustainability of this technique.

Considering that a huge amount of electrolyte will be consumed and transformed into wastewater from the implementation of electrochemical technique, one approach to sustainable electrolyte consumption is to 'reuse' the used electrolyte with a prior treatment that is deemed necessary. In addition to this, while reusing the electrolyte is a sustainable step for minimising electrolyte consumption in the recycling process, the approach should 'reduce' any possible health effects and environmental threats from the disposal of the used electrolyte. This has been practiced in the electroplating industry to tackle the issue arising from the generation of considerable amounts of effluent with heavy metal ions such as iron (Fe), cobalt (Co), chromium (Cr), cadmium (Cd), nickel (Ni), lead (Pb), zinc (Zn), and mercury (Hg), which could bring about some detrimental health effects including kidney failure, thyroid dysfunction, sleeplessness, tiredness, rheumatoid arthritis, negative effects in the circulatory system and neural system, irritation of the gastrointestinal mucosa, and lung cancer.[112] In the electrochemical recycling approach, similar concerns are probable for, the recycling of spent LIBs, as most of their composition is made up of heavy metal ions like Ni, Co, Fe, and manganese (Mn).

In the context of Malaysia, within the scheme of environmental regulations, the authority overseeing industrial wastewater is the Department of Environment (DOE). According to DOE, the wastewater generated from any processes in industrial premises is termed as "industrial effluent". In line with the Environmental Quality Act of 1974, the DOE has enacted the "Environmental Quality (Industrial Effluent) Regulations 2009." Within the scheme of these regulations, there exists a clear obligation for the owners or occupiers of any industrial premises to design and construct a system in strict compliance with the specifications issued by the DOE for treating industrial effluent on-site.[113] Table 6 presents some of the regulation imposed by countries and regions including Malaysia (MY), China (CN), Singapore (SG), Korea (KR), and the European Union (EU).

Through the approach of reuse of the effluent and reduction of the adverse effects from the effluent, it is anticipated that these approaches will contribute to 'Sustainable Blue

Table 6 Imposed regulations regarding wastewater processing (reuse or treatment)

Imposed regulation	Region	Mechanism	Ref.
Regulation (EU) 2020/741 of the European Parliament and of the council of 25 May 2020 on minimum requirements for water reuse	EU	Wastewater reuse	114
Environmental quality (prescribed activities) (environmental impact assessment) order 2015	MY	Wastewater treatment	115
Action plan for prevention and control of water pollution 4	CN	Wastewater treatment	116
Environmental protection and management act 1999, part 5 water pollution Control	SG	Wastewater treatment	117
Wastes control act, article 18	KR	Wastewater treatment	118

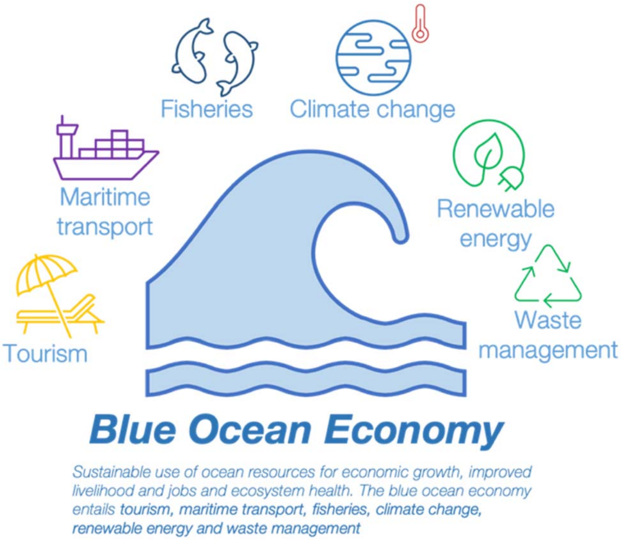

Fig. 11 Illustration of the key aspects contributing to the blue economy agenda. One way for the electrolyte to contribute to this economy is through the implementation of the 'reduction' and 'reuse' of the wastewater or used electrolyte.

Economies'. In detail, a blue economy is the sustainable use of ocean resources for economic growth, improved livelihoods and jobs, and ocean ecosystem health. One of the segments that is highlighted as contributing to the blue economy is waste disposal management, alongside with fisheries, tourism, maritime transport, climate change and renewable energy (see Fig. 11).[119,120]

Membrane separator features and layout

The membrane is a vital component in the mechanics of the electrochemical recycling setup, as underscored before. In attempts to upscale an electrochemical setup, the focal point is on the manufacturing or fabrication of a membrane with the features of interest and the design of the assembly of membranes in a large setup.

The fabrication of the membrane in an electrochemical recycling technique, like the membrane separator in a battery cell, must take into account its ionic conductivity and mechanical stability. These are issues that require prominent focus. The ionic conductivity of the membrane guarantees Li^+ ion mobility, which is conducive to high recycling efficiency. Fortunately, numerous approaches to fine-tune these properties have been explored in studies to ensure high Li^+ mobility throughout the membrane, including the strategies of alternative electrolyte hosts, integration of fillers, blending materials, interpenetrating networks, and electrochemical and heat treatment of the membrane.[64,121–126] Table 7 presents several approaches to enhance the ionic conductivity in the membrane separator of LIB systems. These approaches focus on enhancing the ionic conductivity as the main feature of the membrane. Assimilating these formulations and the strategies used in LIB systems in the development of membranes for electrochemical recycling technique is the most viable approach. This takes into account the similar purpose of the membrane in both LIB and the electrochemical technique, which is to facilitate in the mobility of Li ions across the system. The strategies of using either singular oxide-based or polymer-based materials or the use of composites is an interesting topic being explored in the development of membranes in LIBs. With successful outcomes being achieved using a wide range of materials, future membrane formulation and manufacturing must further probe the effectiveness of the membrane in its applications, the economic returns reflected from its manufacturing and outputs, and the feasibility, stability and cyclability of the

Table 7 Studies with various approaches to address ionic conductivity in the membrane separator in LIB systems

Membrane materials	Li source	Approach	Conductivity achieved, S cm^{-1}	Ref.
Polyethylene oxide/$Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$	Li perchlorate ($LiClO_4$)	Inorganic fillers	1.70×10^{-4} at 30 °C	127
Polyethylene oxide/poly(propylene carbonate)	Li bis(trifluoromethanesulfonyl)imide (LiTFSI)	Materials compositing	2.82×10^{-4} at 60 °C	128
Glycerol propoxylate triacrylate/poly(ethylene glycol) diacrylate/polymethylpyrrole	Li bis(trifluoromethanesulfonyl) imide (LiTFSI)	Electrochemical treatment	7.0×10^{-4} at 70 °C	126
Aluminium-doped $Li_{6.75}La_3Zr_2O_{12}$	Li metal	Thermal pulse treatment	3.2×10^{-4}	129
Polyacrylonitrile	Li triflate ($LiCF_3SO_3$)	Alternative polymer host	3.04×10^{-4}	130
Octakis(3-glycidyloxypropyl dimethylsiloxy) octasilsesquioxane and amine-terminated polyethylene glycol	Li bis(trifluoromethane sulfonyl)imide (LiTFSI)	Interpenetrating network	1.5×10^{-5} at 40 °C	131
Star-shaped siloxane cross-linker with the oligo(ethylene oxide) acrylate	Li perchlorate ($LiClO_4$)	Interpenetrating network	7.8×10^{-4} at 30 °C	132

membrane in large-scale setups of electrochemical recycling technique.

The absence of research investigating the aspects of feasibility, stability and cyclability of the membrane in electrochemical recycling techniques represents a notable void, obscuring insight into the sustainability of the technique. This gap stresses the need for intensive analyses to gain a full understanding and ensure the long-term viability of the process. The opportunity to tailor a membrane with significant cyclability, allowing one membrane to be subjected to multiple recycling cycles, is imperative to give the electrochemical recycling method superiority over the hydrometallurgical method in terms of cost-savings. This would potentially position the electrochemical method as a more efficient and cost-effective solution. Affecting the cyclability and stability is a factor called membrane aging, which is a common issue for electrochemical cells. The aging effect is unavoidable, but research towards its mitigation and strategies to develop a high-performance separator is intensely sought after, derived from the understanding of the aging fundamentals and mechanism.[133,134] Establishing such important fundamentals and mechanism could advance membrane formulation and development for both LIB and electrochemical techniques to achieve much more stable and long-cycle-life membranes.

When further discussing the design of membranes for large electrochemical cells, the focus is often on a scheme that effectively addresses the concerns around the mechanical strength of the membrane while simultaneously mitigating pressure exertion throughout the entire cycle, especially when subjected to a substantial volume of electrolyte in longer-term operation. Failure to accomplish this delicate balance may lead to membrane separator leakage, in turn leading to unwanted side reactions and incidences that are detrimental to the overall operation. In the electrochemical cells of LIBs, every facet is precisely considered and embodied in the internal design to deliver a high performance battery, which has resulted in winding, stacking and folding designs for the electrode–separator composites (ESCs), as shown in Fig. 12.[135–137] Emulating this design in a large-scale electrochemical recycling technique setup could mitigate the issues of stress or pressure on the separator to contribute to stable and safe electrochemical operation.[137] Fig. 13 shows envisioned layouts of the ESC for electrochemical recycling methods. Inspired by the designs implemented in LIBs, real-world scale and larger-

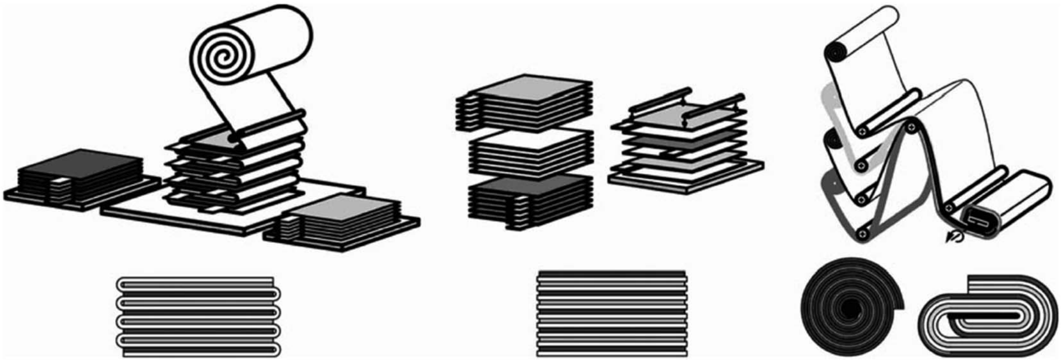

Fig. 12 ESC designs in LIBs.[136] Included are the folded (left), stacked (middle) and winding (right) design, primarily to confront the stress and bending issues in the separator.

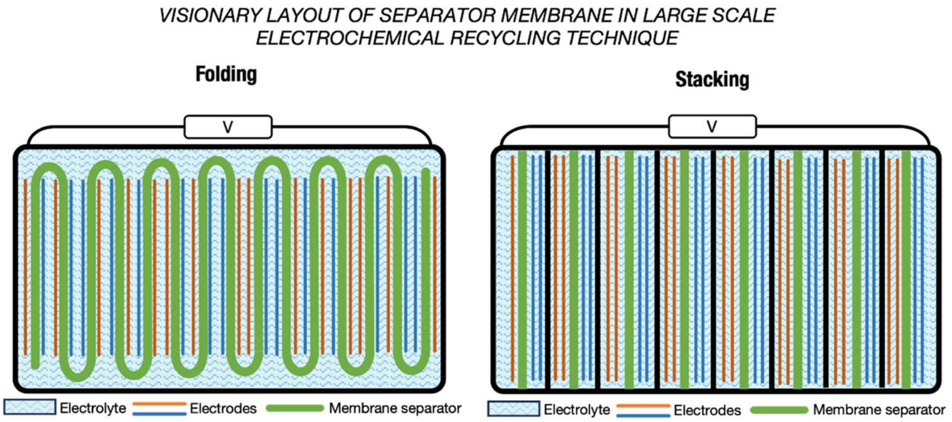

Fig. 13 Envisioned layout designs for the separator and electrode arrangement using a folding and stacking concept in a large-scale electrochemical recycling setup, emulating the ESC arrangements in LIBs. Consultation with experts from the engineering field should be considered to assess the process efficiency.

dimension electrochemical recycling setups integrating such a separator require further studies to determine the efficiency of these layouts. Practically, the processing of a large membrane would require consideration of the raw material cost and availability. Additionally, the properties of the membrane formulated in bulk size may require several compensations in order for it to serve its practical purpose, which will demand further fine-tuning and validation of the formulation to fit the intended application. In the folding design, the benefits are the facile assembly of the membrane and whole setup compared with the stacking design. However, the folding design would face the issues of manufacturing one large-dimension singular unit membrane. This is different from the stacking concept, which offers the simplicity of manufacturing smaller dimension membranes. The stacking design may offer flexibility to perform the recycling of multiple spent LIB sources or different types of spent LIB concurrently. The target output may need to be considered before choosing and implementing any envisioned design in a real-scale setup.

Techno-economic assessment (TEA) and life-cycle analysis (LCA) of recycling techniques

Techno-economic analysis (TEA) and life-cycle analysis (LCA) are two vital components requiring that require equal attention to the technical parts of the electrochemical recycling technique. TEA is a metric to evaluate the economic viability of a process through profit and cost analyses, while the LCA assesses the environmental performance using suitable analyses that include parameters like GHG emissions and global warming potential (GWP).[138–141] Within the thriving industry of spent Li-ion battery (LIB) recycling, both components play crucial role in assisting industry stakeholders making informed decisions regarding the adoption of recycling techniques. The studies outlined in Table 8 focus exclusively on TEA and LCA studies performed for pyrometallurgical and hydrometallurgical technologies; these assessments serve as pivotal factors facilitating the global industrial-scale adoption of these techniques.

Checking the scalability of the recycling technology necessitates TEA to assess the aspect of economic viability. Table 8 below provides the preliminary economic analysis conducted from studies using the electrochemical recycling method for 1 ton of spent $LiMn_2O_4$ (LMO) and spent $LiFePO_4$ (LFP) cathodes. Pyrometallurgy and hydrometallurgy are also included in the table. The emphasis here is on indicating that the electrochemical recycling method is a more economical option than pyro- and hydrometallurgy. The table presents the preliminary cost analysis involving the raw material costs: reagents, operation cost, energy, revenue, and net profit of the recycling technologies. In this defined scope of techno-economic assessment, one could come to conclude the economic viability of the electrochemical method from the revenue and profit gained. The reagent cost and energy cost for the electrochemical method are both the lowest among the recycling methods. The energy cost for hydrometallurgy may be interpreted as lower than that of the electrochemical method considering the much lower temperature required during operation (<150 °C) to facilitate the leaching procedure compared to the electrochemical method. Nonetheless, it has the great drawback of enormous reagent cost. Despite encompassing the metallurgical and electrochemical method in the perspective, the inaccessibility of economic analysis extending to other cathode chemistries, including ternary cathodes such as Li/Ni/Mn/Co (NMC) and Li/Ni/Co/Al (NCA), is another failing in the economic viability forecast of the recycling methods. The study of Lander *et al.* highlighted the economic feasibility of NMC and NCA recycling *via* hydrometallurgy in the United States (U.S.), United Kingdom (U.K.), and China.[142] This is not the case for LFP and LMO recycling, which have diminished net benefits due to emissions and constrained recycling profit.[142] Despite contributing to a simple judgement on the profitability of recycling different types of cathode chemistries, the findings lack inclusion of the electrochemical recycling technique. Hence, any conclusions made may not be translated into transparent decision-making aimed at choosing most economically viable recycling method. The presence of this gap in the current research is a call for action to researchers to supply insights on the economic analysis of each recycling study carried out as early as at the laboratory scale. This valuable perspective could dictate the commercialisation potential of conventional and developing recycling techniques.

Table 8 Preliminary economic analysis of Li recovery from spent sources

Recycling technique	Reagent cost (USD)	Energy cost (USD)	Revenue (USD)	Profit (USD)	Ref.
Recovery of 1 ton of spent $LiMn_2O_4$					
Pyrometallurgy	—	559.05	1687.14	1128.09	143
Hydrometallurgy	—	387.53	1237.78	850.25	144
Hydrometallurgy	659.28	415.5	1303.10	228.32	81
Electrochemical	442.43	392.00	2260.85	1426.42	17
Recovery of 1 ton of spent $LiFePO_4$					
Hydrometallurgy	275.70	250.1	1272.2	510.8	145
Hydrometallurgy	274.22	106.50	1329.90	949.18	15
Hydrometallurgy	858.00	77.88	1901.13	965.25	15
Electrochemical	—	416.26	1566.65	1150.39	15

Table 9 TEA and LCA studies for recycling technology of spent LIBs

Recycling techniques	Key focus	Ref.
Hydrometallurgy	Key enhancement for hydrometallurgical recycling through modified pretreatment methods	146
Pyrometallurgy	Evaluate profitability of pyrometallurgical recycling for various cathode chemistries in spent LIBs	147
Hydrometallurgy	Assess cost and environmental impacts of producing cathode active material for LIB through hydrometallurgical recycling of spent LIBs	148
Pyro- and hydrometallurgy	Economic value and environmental benefit of the metallurgical recycling of spent LIBs	149
Hydrometallurgy	Assess environmental impacts of hydrometallurgical recycling for two Li-ion traction batteries *via* life-cycle analysis	150
Pre-treatment, pyro- and hydrometallurgy	Evaluating the environmental and economic impacts of prevalent recycling methods through detailed analysis of energy and material flows at the process unit level, considering both physical and chemical aspects	151
Pyro- and hydrometallurgy	Economic, environmental, and geospatial analysis of future LIB recycling in United Kingdom (UK)	152
Pre-treatment, pyro- and hydrometallurgy	Examining the economic aspects of Li-ion battery (LIB) recycling involving evaluation of the pros and cons of recycling, as well as investigation of the factors that impact the cost and economic viability of battery disposal	153
Pyro- and hydrometallurgy	Techno-economic cost model for electric vehicle battery recycling	142

Even if it is only at a preliminary stage, the availability of economic analysis provides stakeholders with information on the risks and potential of the commercialisation of this technology, along with the tangible and intangible returns. Addition to this preliminary analysis is needed for an advanced and comprehensive TEA and LCA analysis that covers a broader scope involving the electrochemical recycling technique. Displayed in Table 9 are the sets of studies performed with a focus on the various extensive aspects of TEA and LCA. Accessing this information could take time, but once completed, the data would provide a bigger picture of the economic and environmental performance of existing and developing technologies. For Table 8, the detailed calculations of the numerical data can be accessed in the ESI, Tables S1–S4.†

Table 10 Costs of hydrometallurgical recycling of black mass with a plant capacity of 16 560 tons per year[148]

Description	Canadian dollars per year
Reagents	50 260 153
Shipping and disposal	9 325 384
Labour	12 041 607
Other	5 872 877
Contingency	7 749 734
Total	85 249 755

A direct approach for establishing TEA and LCA analysis is to perform them locally. This should minimise the intricacies in obtaining data and samples, which is complex when considering international or cross-continent data and samples. Furthermore, the findings could precisely reflect local returns and costs. Tables 10 and 11 present data from a study that briefly covers the TEA and LCA aspects in the sustainability analysis of battery material supply chains specifically in Canada.[148] The study conducted in Canada considers the circularity of the battery materials, including Li, *via* a hydrometallurgy recycling approach. It should be noted that the study's methodology involved translating bench-scale hydrometallurgy process parameters into a conventional hydrometallurgy plant with an annual processing capacity of 16 560 tons of black mass. This simulation emerges as a pivotal element, providing the study with concrete findings regarding the advantages of sustaining the circularity of battery materials through the recycling approach.

The study's key revelation underscores that the production of Li-ion battery (LIB) cathode active materials *via* recycling incurs costs that are 48% and 54% lower in overall expenses and environmental impacts, respectively. These findings derived from the TEA and LCA analyses serve as valuable guidelines and benchmarks for the development of the electrochemical technique, shedding light on the potential benefits of adopting

Table 11 Analysis of the environmental impact of producing the cathode active material of LIB through recycling compared with the typical mining, beneficiation, extraction, and refining processes.[148] The high values obtained of 35% and 90% emission reduction strongly indicate the necessity to perform recycling as an alternative to the traditional processes of producing LIB cathode materials

Battery precursor cathode active materials (CAM)	Recycling Processing (kg CO_2e per kg CAM)	Mining, beneficiation, extraction, and refining	Emission reduction
Li_2CO_3—brine	0.8810	1.3500	−35%
Li_2CO_3—ore		8.7300	−90%

similar recycling approaches in the realm of battery material production.

Scrutiny using TEA and LCA analyses are the gears bridging the transformation and rational adoption of technology that includes the emerging electrochemical Li recycling technique. The purpose of TEA implementation is wide-ranging, encompassing the evaluation of economic factors, payback period, internal rate of return, return on investment, discounted cash flow rate of return, capital cost, general costs, profit or revenue, economic potential, overall economic feasibility, process factors, efficiency of operation and environmental factors.[138,154–163] LCA covers a broad spectrum of environmental performance parameters, including greenhouse gas emissions, global warming potential, human toxicity potential, terrestrial ecotoxicity potential, ozone layer depletion potential, water consumption, freshwater ecotoxicity, and land use.[141,164,165] In addition to informed decision-making, the variety of scopes covered in the assessments put all stakeholders in the know about the key issues to be resolved for successful scale-up of a particular technology.

A thorough technological and economic evaluation driven by meticulous data analyses underscores the imperative of accumulating a substantial dataset for the electrochemical method. Addressing challenges, including the availability of large electrochemical setups, strategic planning for significant electrolyte consumption, and customization of the membrane and layout, is crucial for gathering precise data and facilitating a comprehensive TEA. This detailed dataset could contribute to valid LCA and TEA assessments. The primary focus should be directed towards deriving definite insights into the technological profitability and environmental performance. Obtaining clear results in these areas is crucial for determining the feasibility of implementing the emerging electrochemical technique on an industrial scale. Fig. 14 serves as a visual representation, briefly highlighting the key metrics essential for addressing challenges in electrochemical recycling techniques and thereby facilitating a transparent pathway towards successful commercialization.

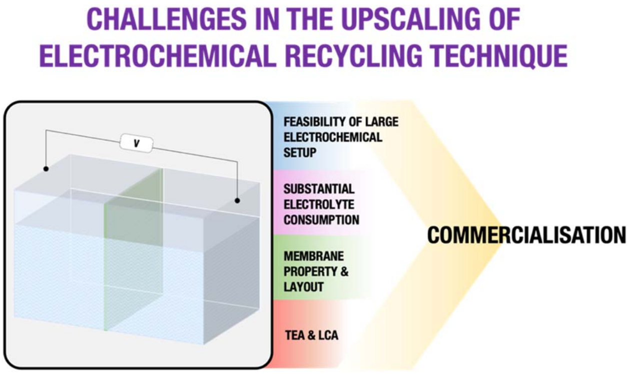

Fig. 14 Major challenges in upscaling the electrochemical recycling technique. Intense efforts should be directed toward addressing these prominent issues, as further exploration to contribute to the progress of the electrochemical method toward a commercialisation path is imperative.

Future directions: lithium circularity

The essence of this review is to underscore the paramount importance of synergy among experts and professionals from diverse fields in preparation for the upscaling of the electrochemical recycling method. Envisioning collaborative efforts from professionals such as process engineers, chemical engineers, techno-economists, and experts from recycling and electrochemical-related industries, the objective is to successfully implement the strategies outlined in this review and effectively address the prominent issues at hand. In the review, it has been identified that the endeavour to expand electrochemical recycling encompasses several challenges. Establishing a feasible setup for broad application is imperative for this emerging technique. The likely compensation of membrane features in a large-scale application poses a challenge that demands an appropriate counter-strategy from both academia and industry. Additionally, a solution for efficient membrane layout design to allow it to retain its properties while considering process efficiency is required. It is crucial to emphasize the significance of techno-economic analysis (TEA) in analysing the economic feasibility of the electrochemical technique.

In addition to the technical hurdles inherent in developing efficient electrochemical recycling methods, the battery recycling landscape in general faces significant wider challenges that include the battery materials, battery design, recycling setup and economic feasibility. It is well known that the extraction and sourcing of critical materials indicate the substantial importance of recycling. If not coupled with management of hazardous components and sufficient resources, the battery recycling effort will fail.

The aspects of battery performance and architectures must evolve to meet the demands of emerging technologies. Optimisation of the composition of new batteries and utilisation of novel materials contributing to facile recycling efforts are areas that require exploration. At present, research on post-LIB is a burgeoning field. The development of metal-based batteries such as magnesium metal batteries, which are safer when being recycled, is ongoing. Being relatively stable in the atmosphere with a high melting point and boiling point, the handling of spent magnesium metal batteries would guarantee safer operation. There is also the option of zinc-based batteries, which are being investigated as a better alternative to LIB with greater safety compared to Li containing a metal that is easier to recycle and less toxic than Li. By reviewing and choosing possible candidates to replace the current LIB technology, a facile and safer recycling method could be established. The wider compromises in each choice of material are well understood, but the vision of positioning recycling as one of the priorities should not be neglected. In addition to the choice of materials for battery production, another interesting topic is research into choosing materials or reagents that could translate into successful or efficient recycling outputs. With recycling technologies like the electrochemical method, the choice of membrane materials is broad, which offers many options and at the same time presents the puzzle of choosing a particular option that is profitable.

Additionally, eliminating obstacles to easy recycling represents an aspect of battery design. Academic and industry should instil the idea of designing batteries with a sustainable production–recycling concept. This concept could be considered at the early stages of designing the battery packaging. With the current design of LIB opting for physical fastenings like clips and welds, the strategy of opting for an easy-to-remove adhesive in LIB packaging could speed the disassembly and avoid the need to subject the battery to shredding, preserving the purity of the battery. A standardised design battery pack design could immensely improve the rate of recycling. The current situation of varying cell packs of LIB from brand to brand and from device to device truly complicates the recycling process. This would require imposing strong regulations and policy, giving a clear indication of our commitment to the sustainability of the existing technology. The emphasis on developing batteries with recyclability in mind from the outset would mitigate end-of-life challenges effectively. In addition to battery design considerations, tailoring the recycling technology could also be an additional initiative that could be executed. As discussed earlier, the layout of the ESC in the electrochemical recycling technique could imitate the concept used in LIB systems. The efforts made to further validate the feasibility and efficiency of this layout would have a noteworthy influence on the recycling industry.

Moreover, economic feasibility has emerged as a central factor determining the scalability of battery recycling initiatives. The cost-effectiveness of recycling methods, particularly electrochemical techniques, is paramount for their widespread adoption and long-term sustainability. In general, it is believed that recycling is much more expensive than mining the materials. This paramount issue could cause existing and potential recycling practitioners and investors to deviate from the goal of sustainability. Economic instruments like rebates and subsidiaries from local and international parties could alleviate the costs that may be the deciding factor influencing the efficiency and rapid development of the recycling industry.

It is overwhelming to look at each of the multifaceted challenges. There should be a pressing call for a joint effort among diverse stakeholders, including the industry players, consumers, and government. There is an obligation to consult and engage with professionals from various disciplines, including process engineering, chemical engineering, economics, and the recycling industry, to cultivate the development of an all-inclusive solution. By aligning these efforts with the ultimate Sustainable Development Goals (SDG) and of Net-Zero Emission policies, the battery industry can spearhead innovation in recycling technologies and facilitate the transition to a circular economy.

Conflicts of interest

The authors declare that they possess no conflict of interest.

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