Garbage in, metal out: A perspective on recycling battery metals using organic molecules

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Global demand for batteries is increasing at a rapid pace, precipitating the equally rapid generation of hazardous battery waste. Recycling, which holds high potential for both mitigating this waste and recovering raw materials for subsequent battery manufacture, is often recognized as a necessary component of the battery life cycle. A critical step in many battery recycling schemes is the use of solvent to recover valuable metals such as lithium, cobalt, manganese, nickel, and others. This recovery typically involves the use of harsh mineral acids and peroxides, which pose their own environmental and safety hazards. The use of more benign organic acids and other organic compounds has emerged as a promising means to mitigate the hazards posed by purely inorganic solvents. In this article, we review recent research on organics-based metal recovery for battery recycling and provide our perspective on the extant challenges and opportunities in the field.

Introduction

The ever-increasing demand for rechargeable batteries in contexts ranging from transportation[1] to grid-scale energy storage[2] is well recognized. Lithium consumption is estimated to increase by 18% year over year through 2050;[3] such a dramatic increase in lithium utilization presents the undesired concomitant increase in related waste (**Figure** 1) due both to increased mining activity (to obtain Li and cathode metals such as Co) and end-of-life battery disposal via landfilling and other means of non-reuse. Unfortunately, strategies to safely manage battery waste are lagging behind, leading to substantial environmental and safety hazards. For example, both lithium and cobalt accumulation in soil can stunt plant growth.[4]

Recycling has long been recognized as an important part of the battery life cycle, well before the emergence of Li-ion batteries as the dominant form of portable energy storage for electronic devices.[6] For example, lead-acid batteries enjoy a high rate of recycling—up to 99% according to industry groups.[7,8] Given the central role of Li-ion batteries in modern technology, and the expectation demand will continue to grow, Li-ion battery recycling is of critical importance. However, the diversity of Li-ion battery construction and chemistries makes their recycling challenging. The case of these batteries (steel and plastic) accounts for ca. 25% (wt%) of the battery, current collectors account for 13%, and electrolyte, separator, and binder together account for 18%, leaving 44% of battery weight comprised of the cathode and anode.[9] As the value proposition of some of these components (e.g., plastic casing) is not necessarily attractive, many recycling processes focus strictly on recovering valuable cathode metals, and to a lesser extent, lithium. Recycling thus typically[10] begins with discharging and dismantling the battery and/or pyrolysis. The resulting product is then processed mechanically to obtain a particulate mixture known as "black mass."[11,12] Black mass contains both valuable metals and undesirable waste such as separator and binder materials. This can pose a challenge, as heat treatment to remove the undesirable fraction of black mass can yield extremely toxic hydrofluoric acid (e.g., as a byproduct of poly(vinylidene fluoride) binder pyrolysis).

Following these preprocessing steps, pyrometallurgy, hydrometallurgy, or a combination thereof is used to collect and isolate the valuable components of the battery. Current commercial-scale operations for these processes leave room for improvement: they involve high-energy inputs (in pyrometallurgy) and/or the use of strong mineral acids (in hydrometallurgy). As such,

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doi:10.1557/s43577-024-00745-2

substantial research has been devoted to the discovery and development of more benign approaches to the collection and purification of Li, Co, Ni, Mn, and other battery metals that have high potential for economic, environmental, and geopolitical value for their recycling. In this perspective, we first summarize the most popular approaches of collection and purification, noting that the interested reader may find many general and comprehensive reviews on these approaches elsewhere.[13–21] We then focus on recent advances, specifically in the use of organic molecules to extract these metals and provide our perspective on the remaining challenges in the field. We recognize that ca. 25–50% of a typical LIB is not composed of precious metals (*vide supra*); recycling techniques dedicated to these components are beyond the scope of this manuscript and have been reviewed elsewhere (**Figure 2**).[22–24]

Pyrometallurgical metal extraction

While pyrolysis at temperatures below 500°C can be used to remove organic residue as a pretreatment step during the battery recycling process, pyrometallurgy at temperatures as high as 1500°C can also be used to recover transition metals, including Ni, Co, and Cu.[15,25,26] This approach has already been implemented at industrial scale, with hundreds of thousands of tonnes of spent batteries processed in this manner annually.[12] At lower temperatures, where battery waste is heated to 500–800°C, liquids (e.g., electrolytes) and carbonaceous materials such as separators can be removed through volatilization. Temperatures below 500°C can produce a mixture of metal oxides and reduced species.[25,27] After pyrolysis, metals are separated through a variety of approaches, including hydrometallurgy, electrorefining, and precipitation at high temperature[15,25,27,28] (noting that lithium is typically lost in waste slag[29]). Pyrometallurgy has been quickly adopted in industrial battery recycling[15] due to its well-established prevalence, relative ease of adoption, and deep historical roots.[30,31] However, the required high temperatures necessitate large energy inputs; other challenges include potentially toxic emissions (and thus the need to manage them), necessity for post-treatment separation of recovered metals, and the loss of Li at higher temperatures (noting that Li recovery is an active area of research in this context[26,32–34]).

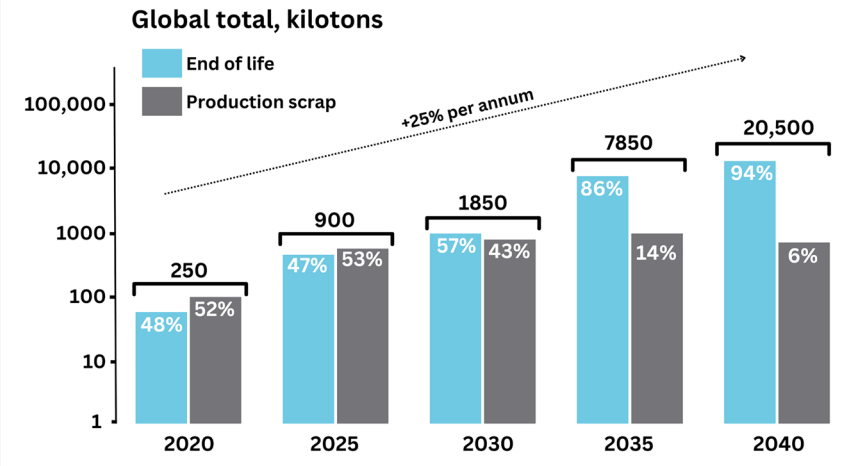

Figure 1. Estimated increase of global end-of-life Li-ion battery waste and production scrap by mass through 2040.[5]

An overview of methods to leach battery metals from battery waste

Battery preprocessing yields a complex mixture containing valuable cathode transition metals as well as other elements, including Li, C, and/or Al. To recover the transition metals (and in some recycling schemes, Li), metal leaching with aqueous solutions (hydrometallurgy) is commonly employed. Leaching is a process in which valuable metals are rendered into a solution from which they can subsequently be isolated (*vide infra*). The standard approach to leaching, and that which has enjoyed the majority of industrial implementation, is by mineral acids such as HCl, HNO_3, and H_2SO_4.[13,29,35–37] In addition, alkaline solutions, organic molecules (typically acids), and bacteria (i.e., bioleaching), have also been examined as potential candidates for isolating the valuable components of spent batteries. We briefly summarize each of these approaches before focusing on recent work in the area of organic leaching for the remainder of this perspective.

Leaching with aqueous inorganic acids

Inorganic acids are inexpensive, are effective leachants, and have a strong track record in industry for battery recycling.[12,14,38] Once a metal-containing precursor mixture is prepared, the mixture is immersed in an acid-containing solution, most typically aqueous HCl, HNO_3, or H_2SO_4. Hydrochloric acid has shown excellent leaching efficiency (e.g., >99% for Co and Li[39–41] and nearly that amount for Mn and Ni[42]). Similar efficiencies are achieved for Co and Li with HNO_3[37,43] and H_2SO_4.[44–46] The use of H_2O_2 as a reducing agent,[47] although not necessary for HCl leaching, is featured in the studies exhibiting the highest leaching efficiencies for HNO_3 and H_2SO_4. Reducing agents such as H_2O_2, which is the most commonly studied, reduce the oxidation state of metals that, in their higher oxidation states, exhibit poor solubility.[48] The reduction of insoluble Co^{3+} to soluble Co^{2+} is a common example.[49] Reducing agents have also been shown to reduce the acid concentrations needed for effective leaching.[44]

Leaching with mineral acids such as those described above necessitates careful management of health and environmental hazards. Mineral acids

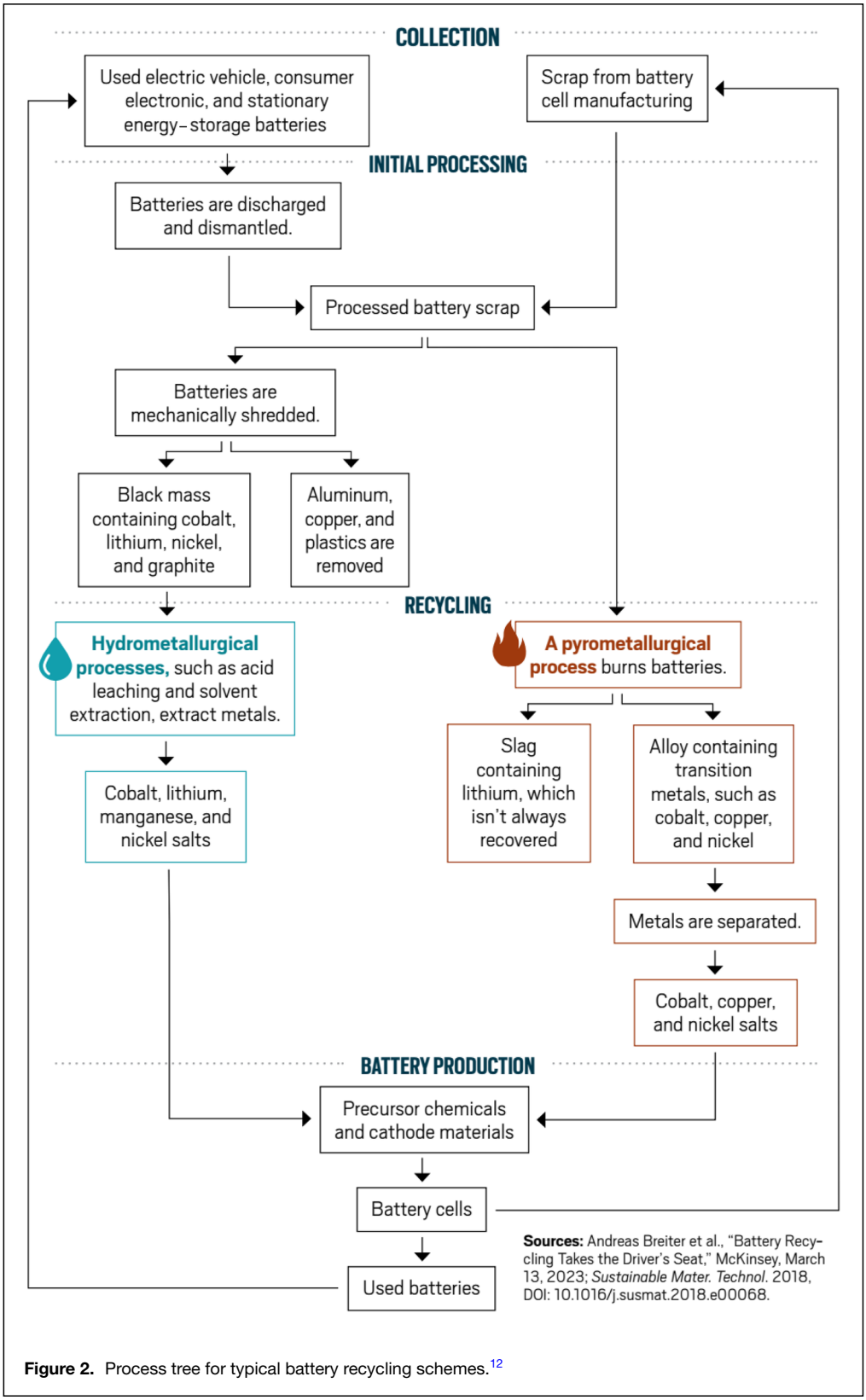

Figure 2. Process tree for typical battery recycling schemes.[12]

are typically used at concentrations of 1 M and higher; these concentrations pose acute safety risks and can lead to corrosion of infrastructure (e.g., metal pipes) if not appropriately managed. The leaching process can produce hazardous gases, including Cl_2, NO_x, and SO_x when using HCl, HNO_3, and H_2SO_4 for leaching, respectively.[50] The acid waste itself must

be treated and disposed of properly; both safety measures and necessary waste handling escalate the overall cost of the recycling process and reduce the overall environmental benefits.[13]

Leaching with aqueous organic acids

Given the challenges associated with using strong mineral acids for a process meant in part to reduce the detrimental environmental impact of widespread battery use, organic acids present potentially attractive alternatives. The first reports using organic acids (citric,[51] malic,[52] oxalic,[53] and ascorbic[54] acids) to recycle the valuable metals in spent batteries were published at the outset of the previous decade. Since then, the topic has received substantial attention, with more than 300 studies having been published to date.[55] These acids present attractive alternatives to mineral acids due to their competitive leaching efficiencies, potential for selectivity,[56–59] smaller environmental footprint, mild conditions (i.e., less corrosive to recycling hardware compared to inorganic strong acids), and recyclability.

Citric acid is perhaps the most-studied example of this class of leachants. At high concentrations (up to 3 M), it can leach greater than 90% of Li, Co, Ni, and Mn from preprocessed battery waste.[56,60] Other well-studied organic acids include malic,[52] tartaric,[61] and oxalic acids,[62] each of which has shown high extents (>90%) of metal recovery. The temperature dependence of leaching has been thoroughly examined; optimal leaching temperatures, where kinetics are rapid but decomposition of the hydrogen peroxide reducing agent is avoided, tend to fall within the range of 70–90°C.[55]

Despite the advantages offered by organic acid leaching agents, they do present several drawbacks. The kinetics of leaching with organic acids tend to be slower[48,63] than those achievable with mineral acids. Furthermore, organic acids typically exhibit incomplete acid dissociation, necessitating large excesses relative to the battery metals being leached. Finally, although inexpensive, organic acids are not currently cost-competitive with mineral acids on a raw materials basis.[55]

Leaching with microorganisms (bioleaching)

More recently,[64] bioleaching has been examined for battery recycling.[65] In bioleaching, microorganisms most commonly utilize biochemical pathways to generate acids (biogenic H_2SO_4 or organic acids) to drive the dissolution of battery metals in battery waste.[66] In the former case, H_2SO_4 is produced by the microorganism aerobically through the reaction of elemental sulfur with dioxygen and water.[66] Organic acids can be produced through bacterial metabolism of sugars; for example, the aerobic oxidation of glucose to form citric acid.[67] Although bioleaching can be used to quite effectively recover Li, efficiencies for recovering other battery metals are lower than those achievable with mineral and organic acids.[66] Furthermore, the kinetics of bioleaching must be improved for this process to enjoy commercial adoption.

Recent advances in leaching battery metals with organic molecules

Organic acids

There have been many publications concerning the most popular organic acids in the past few years.[68] We highlight several recent findings that further the potential of organic acids for battery recycling.

First, the use of a sacrificial reducing agent, considered critical in achieving high extents of metal leaching, was shown recently to be potentially unnecessary in the presence of Al, which itself can act as a reducing agent under battery metal leaching conditions.[69] First shown for sulfuric acid-containing leaching solvents,[69,70] the use of Al, which is commonly the material used for battery current collectors, was recently used in a citric acid-based leaching scheme.[68] However, citric acid can dissolve Co(III) as well as Co(II) and thus does not require the use of a reducing agent;[71] as such, it would be beneficial to examine the use of Al as a reducing agent more deeply. Doing so might obviate the need for external sacrificial reducing agents and thereby improve the recyclability of the leaching solution. We note that other organic acids are capable of metal capture without reducing agents. For example, a recent report showed that EDTA effectively captures Ni, Co, and Mn directly from lithium-nickel-cobalt-manganese oxide without the deliberate introduction of a reducing agent.[72]

While we focus on leaching in this article, it bears mentioning that organic compounds can play important roles in preprocessing steps. In a recent example, glycerol triacetate was used to delaminate the cathode from its Al current collector by dissolving the PVDF binder, allowing the direct collection of the spent cathode and clean Al sheets.[73] Spent cathode material that is recovered directly can then be reprocessed and used directly in new batteries, obviating the need to separate its individual chemical constituents.[10,74]

It has also been shown that the use of external stimuli such as ultrasound or exposure to microwaves can improve the kinetics of leaching with organic acids.[75–79] While these approaches have promise in the laboratory scale, it remains an open question as to how these processes might be used at commercial scales and at costs low enough to be practically adopted.

Deep eutectic solvents

Deep eutectic solvents can be broadly defined as mixtures of organic compounds whose melting points are dramatically reduced compared to those of their individual pure components (**Figure 3**).[80] A common example is the mixture of choline chloride (ChCl; Figure 3) and urea. Individually, these compounds have melting points of 302 and 133°C, respectively. However, their combination in a 1:2 ChCl:urea mole ratio results in a liquid with a melting point of 12°C, well below room temperature.[81] Upon their discovery, it was recognized that these solvents were capable of dissolving metal salts

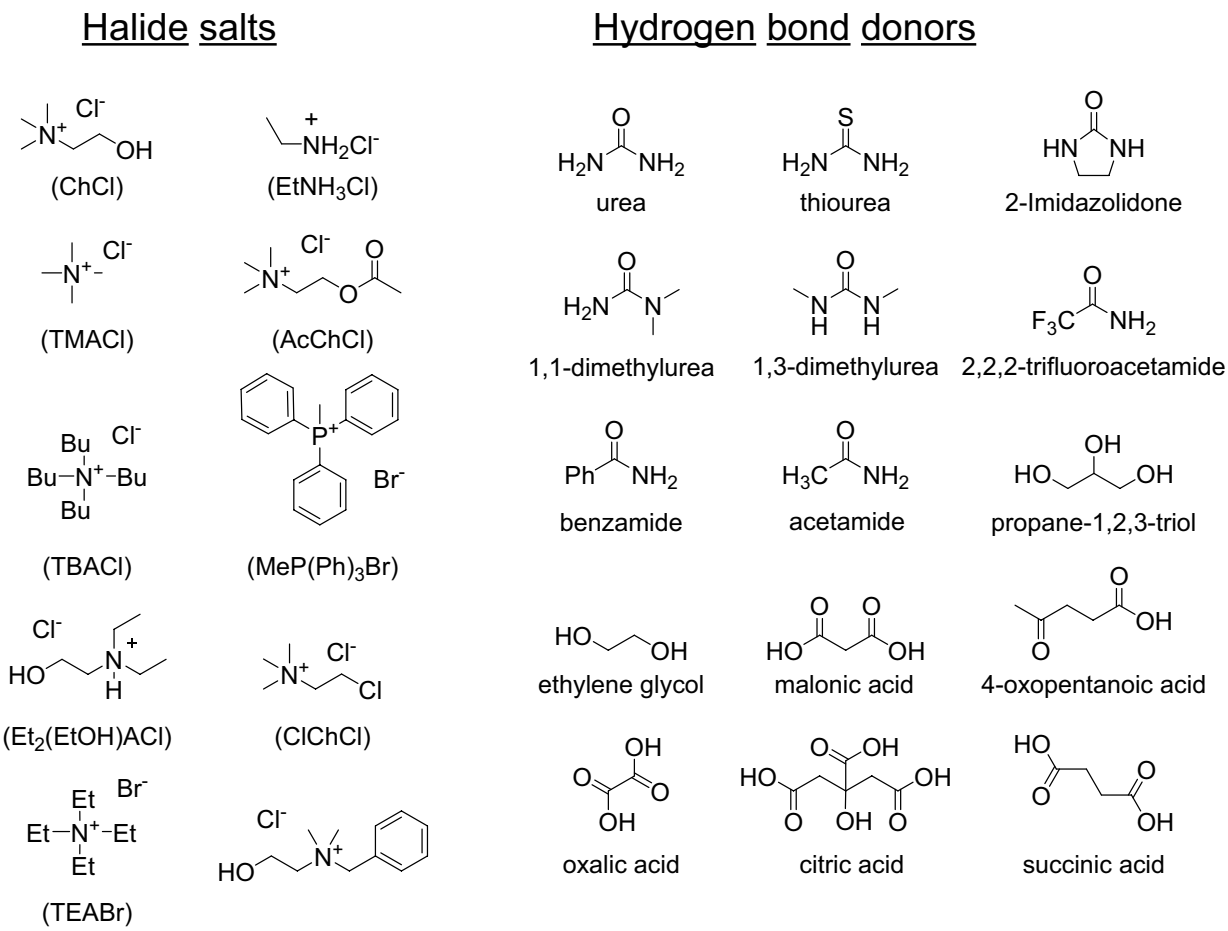

Figure 3. Examples of compounds used in combination to form deep eutectic solvents.[80]

(e.g., LiCl)[81] and some metal oxides (e.g., CuO).[81,82] These characteristics led naturally to the demonstration that deep eutectic solvents could be used to recover metals from waste.[83]

A seminal report using a deep eutectic solvent for collecting recycling metals from spent lithium-ion batteries was published in 2019.[84] In this work, the authors used a ChCl:ethylene glycol deep eutectic solvent to leach cobalt and lithium from $LiCoO_2$ directly from the battery electrode without further preprocessing (e.g., heat treatment). After incubating at 180°C for 24 h in the solvent and stirring, leaching efficiencies of up to 90% for Li and 50% for Co were achieved. Recovery of cobalt by electrodeposition and subsequent solvent reuse was also demonstrated.

The notable leaching characteristics and recyclability of ChCl:ethylene glycol drove substantial interest in examining deep eutectic solvents for Li-ion battery recycling.[85,86] Of these solvent systems, leaching efficiencies approaching 100% at elevated temperatures (above 90°C) have been achieved.[87] Recent studies have explored deep eutectic solvents for selective leaching[88,89] and post-leaching metal separation,[90,91] and the mechanisms by which these solvents operate have begun to receive some attention as well.[92–94]

Although they have many attractive characteristics for battery recycling, deep eutectic solvents also face challenges before being practical for industrial-scale implementation. Their viscosities can range from approximately 50[95] to 10,000[82] × that of water.[96] Other potential challenges include thermal and chemical stability, recyclability, and cost,[97] among others—these are described in detail in a recent report by Neguse et al.[98]

The next battle: Separation

Once metals are leached from battery waste into solution, they are typically separated and isolated as solids capable of reuse (noting that direct cathode recycling is also an important and active area of research, reviewed elsewhere[74,99–101]). Such separation schemes vary widely—they range from collection of mixed products useful for applications with less stringent materials requirements (e.g., catalysis,[102,103]) to recovery of purified metals[104,105] or metal salts.[106] Where

organic molecules are used to leach metals, the most common methods used to subsequently separate those metals are salt precipitation[55] and liquid–liquid extraction.[107]

In precipitation, solubility differences in metal salts are used to precipitate and separate individual cationic components of the leachate solution. Typical examples are using oxalic acid to preferentially induce the precipitation of Co (as the oxalate salt),[62] using sodium carbonate to induce the precipitation of Li (as the carbonate salt),[108] and the use of potassium permanganate to induce the precipitation of Mn (as the tetravalent oxide).[42]

In liquid–liquid extraction, coordinating organic molecules are dissolved in a solvent immiscible with the (typically aqueous) leachate phase (e.g., kerosene). The two immiscible solutions are placed in contact with one another; differences in metal-ion partitioning between the phases can be large enough to selectively capture ions of one identity (e.g., migration of Co^{2+} into the organic phase with Li^{+} remaining in the aqueous phase[46]).

An underexplored area for post-leaching separation of battery metals is the use of sorbents. Lithium ions have been separated from mixtures containing other transition metals with lithium-selective chemisorbents[109] (e.g., lithium-ion sieves[110]). A recent study has shown that a bismuth metal–organic framework is capable of selectively adsorbing Ni^{2+} from a Ni^{2+}- and Co^{2+}-containing solution.[111] Computation suggested that ion differentiation was achieved through differences in the solvation sphere of the adsorbed species within the MOF.

We briefly mention that electrodeposition has received some attention as a means to collect valuable metals after leaching. This technique has primarily been used after leaching by traditional means (i.e., mineral acids).[104,112–114] We note that electrodeposition from multicomponent mixtures can lead to codeposition. In this respect, mixtures of cobalt and nickel present a particularly challenging case.[112,115] Using strongly coordinating organic acids to leach metals from battery waste may also provide a means by which to differentiate their electrochemical deposition windows[116]—to our knowledge, this concept has not been exploited for post-leaching separation.

Outstanding questions

Although the recycling of lithium-ion batteries already enjoys industrial-scale implementation,[12,21,117,118] these industrial processes leave much room for improvement. The use of organic molecules for metal leaching might obviate the need for mineral acid processing and therefore substantially improve the sustainability of the battery recycling process (and ultimately, the battery life cycle itself). However, while many creative approaches to organic-derived leaching solutions have been disclosed, and many more are anticipated, we wish to present here our own perspective of the most pressing needs and where academic laboratories might best contribute.

First and foremost, there is no agreed-upon standard for leaching conditions. This is a natural occurrence stemming from the wide variety of potential conditions that might be optimal for particular battery waste compositions and form factors. However, this presents a challenge for the academic: is it possible to rationally compare the efficacies of different compounds used in leaching based on reports using such disparate inputs (ranging from pure cathode materials to commercial battery waste) and varied leaching agents (organic acids, neutral organic chelating agents, etc.)? And even for a given set of conditions (chemical identity of the leaching agent, concentration, temperature, time, etc.), when battery waste is so varied, are data such as extent of leaching meaningful? Given the uncertainty associated with transitioning recycling processes from laboratory scale to factory scale, it is our view that the academician may contribute most effectively by focusing on mechanistic insight and leachant discovery, rather than on the optimization of a process that might be carried out under quite different conditions at larger scales.

Questions that we find surprisingly understudied, despite a rather copious literature, are:

1. Do the constituents of battery waste interfere with or enhance dissolution of battery metals into leaching solutions, and if so, how? This question can be investigated by examining dissolution behavior in the presence or absence of the expected components of battery waste (e.g., studying the dissolution of Co in Co oxides versus that of $LiCoO_2$ to determine how lithium itself might or might not play a role in enhancing or reducing Co dissolution). Given the large number of possible chemistries that may make up battery waste, we believe this area is ripe for exploration.
2. To what extent, if any, do redox processes play during dissolution? For example, it has been often shown that Co(III) is challenging to dissolve, and that a reducing agent (typically hydrogen peroxide) enhances its dissolution. Several organic reducing agents have also been examined to this end.[119–122] However, while dissolution rates and extents improve with the presence of a reducing agent, the fate of the reductant is rarely monitored.
3. What is the role, if any, of dioxygen (O_2) on metal recovery? Industrial processes can often be carried out under anaerobic conditions;[25] given the potential reactivity (and redox reactivity) of dioxygen and thus potential influence on metal recycling outcomes, this is an important and underexplored question.
4. The speciation of metals in leaching solutions is not well understood. Although there are notable studies attempting to address this question,[123] the multitudes of possible solution compositions beg for deeper scrutiny. Knowledge of metal speciation in solution, for example, might aid in extracting and purifying those metals via precipitation, electrochemistry, etc.
5. Can the leaching solution be recycled? Many studies focus on metal recycling alone, but a truly sustainable process would involve a recyclable leaching solution. This is of special importance given the often-unstudied fate of

reductants used during the leaching process. The reuse of leaching solutions should be investigated as a key component in studies focused on the discovery of new leaching and separation processes.

6. Studies focused on leaching with organic molecules often provide less attention to post-leaching separation of valuable metals. This is a particularly important and difficult problem to solve, especially where mixed battery wastes are concerned (Li-ion batteries can possess cathodes of substantially different cathode chemistries[124]). The appropriate metal separation process necessarily depends on the leachant solution composition, but leaching and separation are often studied independently. Hence, there is much to be learned about the relationship between metal speciation in the leachate and chemistries needed to collect these metals in pure or pure salt forms.
7. Finally, while we focus here on battery waste, this is by no means the only waste that contains valuable and recoverable metals. By focusing on optimization of a particular combination of battery waste composition, leaching agent(s), and other experimental conditions, the scope of the work narrows. Going "back to basics" and studying the solution and interfacial chemistry that drives metal dissolution may provide substantial benefits not only to the recycling of lithium-ion batteries, but to broader recycling goals and even industries such as mineral extraction, where harsh acid leachants[125] could potentially be replaced with more benign alternatives.

Summary

Approximately 250 tons of lithium ore are needed to make 1 ton of new lithium-ion battery, whereas it only takes 28 tons of recycled lithium-ion battery to generate 1 ton of new battery.[126] Yet, industrial battery production is reliant on using fresh ore or brine for sources of lithium as well as the other precious metal components of the LIBs. As we have reviewed here, extensive work has been undertaken toward turning the recycling of LIBs into an environmentally and economically sustainable alternative to the mining of ore. However, there are remaining questions that must be addressed. As laid out in this perspective, the charge lies with the academician to discover the fundamental principles that govern the challenges of LIB recycling (solubility, partitioning, etc.), while exploring alternatives to traditional approaches (e.g., mineral acids and high-temperature pyrolysis). Investment in basic science and fundamental research can then lead to innovation from both academic and industrial laboratories alike. Global reliance on LIBs is not diminishing. As such, establishing the sustainability of LIBs is a necessity. Thankfully, with the appropriate investment of resources, it is within reach.

Author contributions

All authors contributed equally to the conceptualization, writing, and editing of the manuscript.

Funding

This work received generous support from the National Science Foundation Grant Nos. DMR-2337183 (P.A. and J.I.F.) and DMR-2337184 (A.E.S. and D.T.G.).

Data availability

Not applicable.

Declarations

Conflicts of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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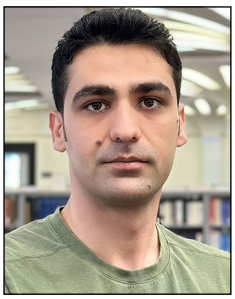

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