

# Recycling of Lithium-Ion Batteries—Current State of the Art, Circular Economy, and Next Generation Recycling

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Being successfully introduced into the market only 30 years ago, lithium-ion batteries have become state-of-the-art power sources for portable electronic devices and the most promising candidate for energy storage in stationary or electric vehicle applications. This widespread use in a multitude of industrial and private applications leads to the need for recycling and reutilization of their constituent components. Improving the “recycling technology” of lithium ion batteries is a continuous effort and recycling is far from maturity today. The complexity of lithium ion batteries with varying active and inactive material chemistries interferes with the desire to establish one robust recycling procedure for all kinds of lithium ion batteries. Therefore, the current state of the art needs to be analyzed, improved, and adapted for the coming cell chemistries and components. This paper provides an overview of regulations and new battery directive demands. It covers current practices in material collection, sorting, transportation, handling, and recycling. Future generations of batteries will further increase the diversity of cell chemistry and components. Therefore, this paper presents predictions related to the challenges of future battery recycling with regard to battery materials and chemical composition, and discusses future approaches to battery recycling.

the lithium-ion battery (LIB), as an established technology, is a promising candidate for the energy-storage of the future.<sup>[1]</sup> Consequently, LIBs are already the first choice for energy storage in modern day portable consumer electronics like laptops, smartphones, or tablets. Furthermore, as the most attractive battery technology for pure and hybrid electric vehicles, as well as a strong candidate for stationary storage solutions, there is a widespread use of LIBs in private and industrial applications.<sup>[2–4]</sup> This broad distribution goes hand in hand with the need for recycling. Partially, this necessity is economically driven by the value of the applied metals, which is significant given the high Ni, Cu, or Co contents. Spent LIBs usually contain 5–20% cobalt (Co), 5–10% nickel (Ni), 5–7% lithium (Li), 5–10% other metals (copper (Cu), aluminum (Al), iron (Fe), etc.), 15% organic compounds, and 7% plastics.<sup>[5,6]</sup> Currently, the market is dominated by LiPF<sub>6</sub>-based organic solvent electrolytes due to the excellent properties of LiPF<sub>6</sub>

## 1. Introduction

Increasing energy demands, due to the world population growth, as well as the changing lifestyle and the depleting fossil-fuel resources are creating a dependence on renewable energy sources. One problem of renewable energy is the unsteady electricity generation. Accordingly, surplus energy must be stored in order to compensate for fluctuations in the power supply. Due to its high energy density, high specific energy and good recharge capability,

regarding ion conductivity, supporting solid electrolyte interphase (SEI) formation at the anode, and protection of the Al current collector at cathode.<sup>[7–12]</sup> Additionally, the state-of-the-art LIB cell consists of a graphite anode, and in most cases a layered lithium metal oxide (LiMO<sub>2</sub>, M =, e.g., Co, Ni, Mn, Al) with varying metal contents or a lithium transition metal phosphate as a cathode.<sup>[13–19]</sup>

There are also a variety of non-Li chemistries currently investigated, including batteries based on naturally highly abundant elements such as sodium, zinc, magnesium, calcium,

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etc. Among these non-Li batteries, the sodium ion technology as the most similar to the commercial LIBs is considered as a drop-in solution.<sup>[20]</sup> However, from a recycling perspective, such battery chemistries with low-cost elements are of little economic interest. Therefore, the recycling of such batteries must be supported by legislation.

Battery recycling is encouraged by the legislation through different directives, mainly because of risks to human health or the environment deriving from hazardous battery constituents.<sup>[21–26]</sup> Recycling processes are the only option to re-introduce end-of-life (EOL) batteries and their components into the economic cycle, reducing the need for primary raw materials<sup>[27,28]</sup> and promoting an improved acceptance of pure and hybrid electric vehicles. However, the complexity of an LIB with its diverse components, cell chemistries, and aging mechanisms offers certain challenges for recycling.<sup>[29,30]</sup> In addition the varying lifetimes for batteries in different applications (cell phones: about 2 years, other consumer electronics 3–4 years and electric vehicles >10 years) produce a mixed upcoming stream of spent batteries.<sup>[31]</sup> Beyond this, the development of next generation batteries leads to even more complex mixtures of battery scrap, increasing the need for universal and flexible recycling processes.<sup>[32–35]</sup> Furthermore, in contrast to the lead acid battery, only high amounts of metals such as nickel or cobalt provide financial viability for LIB recycling.<sup>[36]</sup> And last, so far no standardizations for cell designs of LIBs exists.<sup>[37]</sup>

Nevertheless, recycling processes are used to handle the upcoming stream of spent batteries. Today, mainly pyro- and hydrometallurgical processes, or a combination of both, are applied to process current cell chemistries.<sup>[32,38,39]</sup> Yet, a closed loop in view of a holistic circular economy is not achieved, since several components like anode, electrolyte or binder are still not, or only recently subject to recycling. However, activities in this area are increasing, so that potential methods and processes for the recycling of these components are emerging. In contrast to this, the recycling of next generation batteries is neither focused by industry nor by research activities. Only less than a handful studies have been published recently so far.<sup>[40,41]</sup>

In this review, we provide an overview about the current state of the art in LIB recycling, addressing topics like regulations (EU, China, and USA), handling, transport, and current technologies which are applied during recycling in research and industry. In addition, a critical assessment on the different types of next generation cell chemistries (batteries of the future) and emerging recycling approaches such as design for recycling and direct recycling are given.

## 2. Regulatory Framework

### 2.1. Recycling Regulations

Legislation plays an important role in controlling the recycling of any waste material. By setting targets for collection rates and recycling efficiencies, and regulating disposal responsibilities and safety requirements, government authorities can contribute to the establishment of an effective circular economy. An important concept in this regard is the extended producer responsibility (EPR), which assigns the responsibility for

the treatment of EOL products to the producer. In general, a distinction is made between physical and financial responsibilities.<sup>[42,43]</sup> Physical responsibility refers to ensuring the treatment of waste products, including collection, transport, sorting, reuse, recycling, and disposal.<sup>[42]</sup> These tasks can usually be delegated to third parties.<sup>[42–44]</sup> The financial responsibility relates to the financing of the aforementioned activities and allows producers to internalize the costs of waste treatment and incorporate them into their prices.<sup>[42,43]</sup> Regulations regarding the treatment of EOL batteries vary from country to country. In the following, the legislation in three of the largest battery markets, EU, USA, and China, is presented (Table 1).

#### 2.1.1. EU

In the EU, present regulations include the Battery Directive (Directive 2006/66/EC) and the Waste Electrical and Electronic Equipment (WEEE) Directive (Directive 2012/19/EU). These policies include a physical and financial EPR. Member countries are required to set up collection schemes for end-of-life portable batteries in the form of collection points located in the vicinity of end-users. The costs for collection, treatment, recycling, and disposal must be financed by the battery producers. Producers and distributors are further required to take back portable, automotive, and industrial batteries (including electric vehicle (EV) batteries) free of charge. Industrial, automotive, and collected portable waste batteries must undergo treatment and recycling using the best available techniques to protect health and the environment before residual compounds can be landfilled or incinerated. In order to maximize the separate collection of spent batteries from mixed municipal waste, the directives set minimum collection targets and recycling efficiencies for member states. The collection rate is calculated by dividing the mass of portable waste batteries collected in one year by the average annual mass of portable batteries placed on the market in the previous three years. The minimum collection rates were set at 25% by 2012 and 45% by 2016. For Pb–acid, Ni–Cd, and other battery types, the directive sets recycling efficiency targets of 65%, 75%, and 50% by average weight, respectively.<sup>[44,45]</sup>

In a revision report from 2019, the European Commission evaluated the effectiveness of the 2006 Battery Directive. According to the report, most countries achieved the collection target of 25% by 2012. However, only 14 member states have reached the subsequent target of 45% by 2016. In total, 56.7% of all waste portable batteries are not collected annually and about 35 000 tons end up in municipal waste streams. The report concludes that the current collection targets are not sufficient and further targets should be defined for the future. Another concern is that collection targets have so far only been defined for waste portable batteries and not for automotive and industrial batteries (including EV batteries).<sup>[45]</sup> Another statement from the European Association of National Collection Systems for Batteries (Eucobat) describes the collection rate calculation as inappropriate. Accordingly, the calculation does not consider the varying lifetimes for different battery types, as well as possible battery exports, and is therefore not realistic. As a solution, the calculation of collection rates based on the battery cells available for collection is proposed.<sup>[46]</sup>

**Table 1.** Selection of the most important federal policies regarding the recycling and treatment of EOL batteries in the EU, USA, and China.

Year	EU	USA	China
1995		Universal Waste Rule as part of the Resource Conservation and Recovery Act (RCRA)	Law of the People's Republic of China on the Prevention and Control of Solid Waste Pollution
1996		Mercury-Containing and Rechargeable Battery Management Act (Battery Act)	
2006	Battery Directive (Directive 2006/66/EC)		
2012	Waste Electrical and Electronic Equipment (WEEE) Directive (Directive 2012/19/EU)		Notice of the State Council on Issuing the Planning for the Development of the Energy-Saving and New Energy Automobile Industry
2014			Guiding Opinions of the General Office of the State Council on Accelerating Promoting and Application of New-Energy Automobiles
2016			Policy on Pollution Prevention Techniques of Waste Batteries Implementation Plan of the Extended Producer Responsibility System
2018			Interim Measures for the Management of Power Battery Recovery and Utilization of New Energy Vehicles
2020	Proposal for a regulation of the European Parliament and of the Council concerning batteries and waste batteries, repealing Directive 2006/66/EC and amending Regulation (EU) No 2019/1020		

As part of the European Green Deal, in 2020 a legislative proposal was submitted by the European Commission to replace the 2006 Battery Directive. The proposed regulations considerably exceed previous legislation in many respects and are intended to support the development of the EU toward a modern, resource-efficient, and competitive economy. Accordingly, new collection targets for waste portable batteries (excluding batteries for light means of transport, e.g., e-bikes) are 45% by 2023, 65% by 2025, and 70% by 2030. The proposal does not include collection targets for industrial, automotive, and EV batteries, but sets a legal framework for the establishment of appropriate collection schemes for these battery types. In addition, the proposal is called for a revision of the collection targets in 2030, including the consideration of adjusting the calculation method for collection rates to be based on the waste batteries available for collection. New targets for recycling efficiencies are 65% for LIBs and 75% for Pb-acid batteries by 2025. Moreover, target material recovery rates of 95 % for cobalt, 95% for copper, 95% for lead, 95% for nickel, and 70% for lithium by 2030 have been defined. Further requirements include extended battery labelling, a battery passport for batteries with capacities above 2 kWh, minimum contents of recycled materials in new industrial and automotive batteries, minimum performance and durability requirements, and more.<sup>[44]</sup>

### 2.1.2. USA

The only federal policy in the U.S. regarding battery recycling is the Battery Act of 1996, which primarily focuses on facilitating the recycling of nickel–cadmium (Ni–Cd) and small sealed lead-acid (SLA) rechargeable batteries, as well as phasing out the use of mercury in batteries. The directive includes a national standardization of labelling requirements, the prohibition of selling certain

mercury-containing battery types, and requires the Environmental Protection Agency (EPA) to establish a public education program on battery recycling, proper handling, and disposal of used batteries. Moreover, the Universal Waste Rule, as part of the Resource Conservation and Recovery Act (RCRA) from 1995, is made effective for all 50 states.<sup>[47]</sup> The Universal Waste Rule prohibits the disposal of certain hazardous wastes and sets standards for the collection, storage, and transportation of these wastes.<sup>[48]</sup>

In addition to the Battery Act, some U.S. states have enacted further legislation on battery recycling. In 25 states, regulations that are more specific apply to the recycling of lead acid batteries. In most of these states, landfilling or incineration of lead acid batteries is prohibited, and consumers are required to dispose such batteries separately from mixed municipal wastes. Furthermore, retailers are required to take back lead acid batteries in certain quantities and deliver them to manufacturers or permitted secondary treatment facilities. Only four states, namely California, Minnesota, New York and Puerto Rico, have also introduced regulations for the collection and recycling of LIBs.<sup>[49–52]</sup> For example, the Rechargeable Battery Recycling Act of 2006 introduced the EPR in California. Thus, producers are required to internalize the costs for handling, recycling and safe, environmentally sound disposal of used rechargeable batteries. In addition, retailers are required to take back used batteries free of charge, the content of hazardous substances in rechargeable batteries should be reduced, batteries should be designed for longer life and reusability, and consumers must be provided with comprehensive information on battery recycling.

### 2.1.3. China

In China, first legislation regarding battery products were introduced in 1995. Initially, the regulations mainly focused

on phasing out batteries containing mercury and cadmium. Later, increasing attention was given to the recycling and proper disposal of lead-acid batteries.<sup>[53]</sup> However, until the 2010s, there was a significant lack of regulation governing the collection and treatment of waste LIBs.<sup>[42,53]</sup> With the Notice of the State Council on Issuing the Planning for the Development of the Energy-Saving and New Energy Automobile Industry from 2012 and the Guiding Opinions of the General Office of the State Council on Accelerating Promoting and Application of New-Energy Automobiles from 2014, the State Council set the foundation for establishing a system for collection and treatment of waste EV batteries, including the development of technical standards and management rules as well as their enforcement. Subsequently, in 2016, the Policy on Pollution Prevention Techniques of Waste Batteries by the Ministry of Ecological and Environment (MEE) and the Implementation Plan of the Extended Producer Responsibility System by the General Office of the State Commission (GOSC) were issued. These policies specify the development of standards related to pollution prevention and the collection, transportation, storage, utilization, and disposal of waste LIBs. Furthermore, the development of a monitoring system for waste batteries is encouraged, an EPR is introduced for EV and battery manufacturers and specific recycling targets of 40% by 2020 and 50% by 2025 for major waste products, including LIBs are set. With the Interim Measures for the Management of Power Battery Recovery and Utilization of New Energy Vehicles issued in 2018, the Ministry of Industry and Information Technology (MIIT) and six other ministries and commissions consolidated existing regulations. Along with several subsequent guidelines, the Interim Measures provide an overall policy framework for today's battery recycling industry in China. The key elements of this policy framework are: a) encouragement of manufacturers to design batteries for easy disassembly; b) obligation of manufacturers to provide the technical information necessary for EOL battery treatment; c) promotion of cascaded application and second life of EOL batteries; d) responsibility of EV and battery producers for battery waste treatment, based on the EPR concept; e) responsibility of cascaded application companies, EV makers and battery producers for establishing waste battery collection outlets; f) material recovery targets of 98% for nickel, cobalt, and manganese, 85% for lithium, and 97% for rare earth and other metals. Further specification of the financing mechanisms for waste battery treatment is not included in the policies.<sup>[42,53]</sup>

## 2.2. Material Collection and Sorting

The establishment of an efficient collection system for EOL batteries is a key element in a successful recycling strategy. The collection rate determines the number of spent batteries that enter the recycling stream and is therefore decisive for the extent of economic and ecological output of the overall recycling system.

One of the major challenges for setting up a performant collection infrastructure lies in the heterogeneity of battery types available on the market. LIBs are used for a wide range of applications, resulting in a large variety of battery designs that differ with regard to their capacity, shape, size, and chemical

composition.<sup>[54–57]</sup> Three of the main markets for LIBs are consumer electronics, stationary battery energy storage (SBES), and EVs.<sup>[55,58,59]</sup> While the consumer electronics market (cell phones, portable computers, medical devices, power tools, etc.) is mature, the EV market in particular is expected to be the main driver for an increasing LIB demand.<sup>[58–60]</sup> Since these markets show significant differences, it is necessary to establish different types of collection systems. The smaller household batteries from electronic devices can be collected in containers at retail partner and manufacturer locations, whereas the collection of larger modules from EVs and SBES devices requires disassembly and must be performed by trained personnel.<sup>[61]</sup>

The availability of EOL batteries is especially important in this context. Consumer electronics currently account for a large share of the LIB market. Batteries from this segment have lifetimes of approx. 3–10 years, depending on the type of device.<sup>[62]</sup> The service life is usually much shorter, but the widespread storage of devices after their use, often referred to as hibernation, leads to an extension of the overall lifetime. The lifetime of EV batteries is often warranted by car manufacturers for a minimum of 8 years or 100 000 miles/160 000 km (e.g., Tesla Model 3 Standard Range, VW eGolf, Nissan Leaf, BMW i3). The accurate prediction of the EV battery life is difficult because many factors, such as the battery type, the number of cycles, the charging conditions and the annual driving distance must be considered.<sup>[63,64]</sup> For example, using predictive models, Yang et al. have estimated EV battery lifetimes of 5–13 years under average driving conditions in different U.S. states. Considering a second-life application of retired EV batteries in SBES systems, the total battery lifetime could be increased to about 15–25 years depending on the application. Due to this long battery life and the immaturity of the EV sector, waste streams from consumer electronics are an important near-term source for battery recycling.<sup>[60,65]</sup> Currently, only a small portion of the electronic waste generated each year is collected and properly recycled.<sup>[66]</sup> According to the Global E-Waste Monitor 2020, the rate of collected and recycled e-waste is 42.5% in Europe, 11.4% in Asia, 9.4% in the Americas, 8.8% in Oceania, and 0.9% in Africa. Overall, 17.4% of the e-waste generated worldwide is documented to be collected and recycled.<sup>[66]</sup> Among the largest generators of e-waste, Asia leads with 24.9 Mt, followed by Europe with 12.0 Mt and North America with 7.7 Mt. To ensure effective battery recycling, it is therefore necessary to maximize the collection rate for spent batteries and e-waste and thus optimally use available resources. Furthermore, suitable structures for the collection and recycling of larger battery modules should be installed at an early stage in order to prepare for the rapidly growing EV and SBES markets.

In addition to the heterogeneity regarding different module sizes and fundamentally different cell chemistries, such as Pb-acid, Ni–Cd, and Ni–metal hydride, LIBs also significantly vary for similar applications within the same market segments.<sup>[54,57]</sup> Due to continuous development, changing performance requirements, increasing raw material prices and innovations, the composition and design of LIBs is constantly changing. While for many years LiCoO<sub>2</sub> (LCO) was the commercially dominant cathode material for LIBs, increasing costs for cobalt, as well as the limited thermal stability and rapid capacity fading of LCO-LIBs, have led to the commercialization of alternative



cathode materials. The most widely used cathode types today also include  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA),  $\text{LiNi}_{0.5}\text{Co}_{0.3}\text{Mn}_{0.2}\text{O}_2$  (NMC),  $\text{LiFePO}_4$  (LFP), and  $\text{Li}_2\text{Mn}_2\text{O}_4$  (LMO, Spinel type). Beyond that, several other materials, including  $\text{LiMnO}_2$  (LMO, Layered type),  $\text{LiMnPO}_4$  (LMP) and conversion-type cathodes, such as sulfides, are under development.<sup>[2]</sup> New developments are also progressing in the fields of anode materials, electrolytes, and separators. Although graphite and other carbons are considered the state-of-the-art LIB anode materials, alternative LIB anode types such as silicon and silicon composites, as well as lithium metal anodes, and zero-excess anodes for lithium metal batteries (LMBs) are being researched. At the same time, new electrolyte systems continue to find application. Especially solid-state electrolytes, which are usually based on polymers, ceramics, or hybrid systems, are attracting more and more attention.<sup>[2,67–70]</sup>

The different LIB cell chemistries pose a major challenge to current recycling systems. Not all processes are economically viable for the recycling of any battery type. Due to the high investment costs, pyrometallurgical treatment, for example, is primarily suitable for the recycling of batteries with high cobalt and nickel content, such as LCO, NMC, and NCA batteries. Lithium and aluminum end up in a slag and can only be recovered with considerable additional effort. Hydrometallurgy, on the other hand, also enables the recovery of lithium and is applicable for a mixture of different cathode types. However, due to the low intrinsic material value of LFP batteries both processes fail to recover valuable products from this battery type. Another approach, direct recycling, aims at the recovery of cathode materials with a still usable morphology. Since the number of further processing steps for the re-synthesis of cathode materials is reduced, this method has a comparatively low environmental impact and currently represents the only process by which significant value can be generated from used LFP and LMO cathodes. A major disadvantage of direct recycling is that the process requires single cathode types as input in order to recover high quality materials.<sup>[60,71–74]</sup>

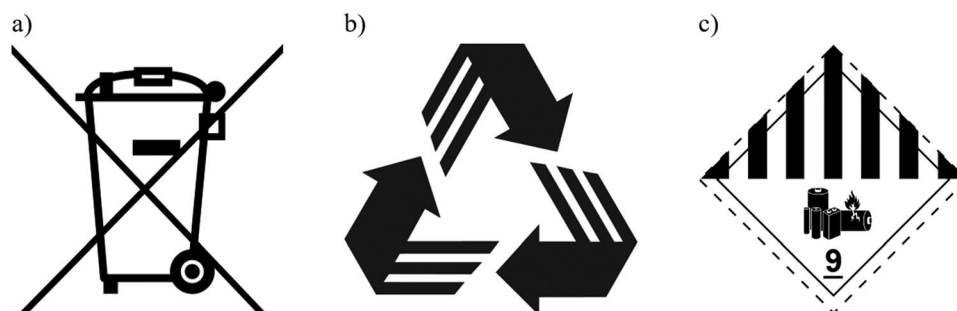
To overcome the challenge posed by the material mix, an effective separation of different battery types based on their chemical composition is required.<sup>[31,75]</sup> The sorting could be implemented through a more differentiated collection of batteries by the end-users. However, this would lead to a considerable additional effort for consumers and in order to achieve high collection rates, more convenient collection schemes could be advantageous. Another option is to sort batteries

directly in recycling facilities. Since not all recycling facilities are capable of processing every type of battery, this could result in additional transport routes. A possible solution would be the setup of fully integrated recycling plants capable of specifically treating various types of cells. Building such plants, however may require large capital investments and should be difficult to achieve in a private economy. A third option, the establishment of a decentralized pre-treatment infrastructure, could therefore be beneficial. Sorting of battery types, as well as other processes such as battery deactivation, removal of casings, and other physical separation methods, could already be applied as a pre-treatment, enabling an early separation of material streams, minimizing transportation costs, and improving the overall efficiency of battery recycling.<sup>[31,60]</sup>

Since the chemical composition of a battery is not visible from the outside, appropriate labelling is an important prerequisite for battery sorting. In addition to the essential information on the battery type and chemical composition, labels could contain data regarding the manufacturer, the date of manufacture and the use of the battery. This information would enable effective sorting but could also significantly improve safety during the handling of used battery cells.<sup>[60]</sup>

In the EU, current legislation mainly aims at providing end users with necessary information for the proper handling of battery cells. This includes the symbol shown in **Figure 1a**, which indicates that batteries must be collected separately from household waste. In addition, the battery capacity and the presence of the heavy metals Hg, Cd, and Pb must be indicated.<sup>[76]</sup>

In 2020, the European Commission has proposed a new directive with more comprehensive battery labelling requirements. The proposed legislation requires additional labelling of cells with information about the manufacturer, date of manufacture, date of market introduction, battery type, battery model, chemistry, hazardous substances, carbon footprint, recovered materials, and critical raw materials contained. Through a so-called battery passport, the information should be made available with a QR code printed or engraved on the housing and an electronic battery information exchange system. Independent operators shall also be given access to the battery management system of rechargeable industrial batteries and EV batteries with internal storage with a capacity above 2 kWh, for the purpose of assessing and determining the state of health and remaining lifetime of batteries in order to enable second-life applications. Parameters for determining the



**Figure 1.** Battery labels required by different federal policies in the EU and USA. a) Label indicating separate collection from mixed municipal waste; required by EU Battery Directive 2006/66/EC. b) Möbius loop indicating recyclability of batteries; required by US Mercury-Containing and Rechargeable Battery Management Act. c) LIB transport label indicating classification as Class 9 Dangerous Goods; required by US law 49 CFR § 172.447.

state of health of batteries include: remaining capacity, overall capacity fade, remaining power capability and power fade, remaining round trip efficiency, actual cooling demand, evolution of self-discharging rates, and Ohmic resistance and/or electrochemical impedance.<sup>[44]</sup>

A comparable regulation was already issued in China in 2018 with the Interim Provisions on the Traceability Management of Power Battery Recovery and Utilization of New Energy Vehicles. This policy requires the establishment of a national management platform for the traceability of batteries throughout their entire life cycle, including production, sale, use, disposal, and recycling.<sup>[42,53]</sup> In the U.S., there is no comprehensive federal regulation for the labelling of LIBs. Current laws only include the labelling of Ni–Cd and Pb–acid batteries with a reference to the respective battery type, as well as the requirement for battery recycling. Additionally, the batteries must be labelled with the recycling symbol shown in Figure 1b.<sup>[47–49]</sup> Further labelling requirements for LIBs in the U.S. are in place in the context of transportation. These regulations are targeted at transportation safety and include, for instance, certificates for safety tests, packaging guidelines, an additional LIB label (Figure 1c) and transport volume limitations.<sup>[47]</sup>

As a central part of battery recycling, the development of a suitable collection infrastructure for EOL batteries is essential. In particular, the better use of currently available resources and the expansion of collection capacities for the fast-growing EV market are crucial. In addition, the establishment of a suitable pre-treatment system for sorting as well as further pre-treatment steps, such as cell deactivation and physical separation of cell components, is promising. Policy plays a particularly important role in this regard, as the introduction of standardized battery labels, mandatory collection rates for spent batteries and an EPR could contribute to a more effective battery collection.

### 2.3. Transportation and Handling

The high energy density and the presence of toxic and flammable substances lead to significant safety risks when handling and transporting LIBs. One of the most serious hazards is the thermal runaway, which can be described as a cascade of uncontrolled exothermic reactions that is caused by an initial overheating of a battery cell,<sup>[77–79]</sup> and depends on the age of the used cells.<sup>[80–84]</sup> Potential reasons for the initial overheating are external heat sources, overcharging or external and internal short circuits. The risk for internal short circuits of LIBs is increased by lithium metal plating, due to inaccurate battery assembly, or inappropriate operation, such as overcharging, over discharging or low temperatures and high current densities during charging. Several strategies, such as the addition of electrolyte additives that support the formation of effective SEIs,<sup>[85]</sup> or protective coatings with artificial SEIs can reduce the dendrite formation in state-of-the-art LIBs.<sup>[78,86]</sup> However, the problem still remains and represents one of the most important challenges for next-generation anodes, such as lithium metal. Short circuits can also occur during handling, due to external mechanical stress or incorrect storage. A single short-circuited cell can thereby cause the initiation of the thermal runaway of many other cells. The consequences are the combustion of

flammable electrolyte components and the decomposition of cathode materials, which can lead to fires, explosions and the release of toxic substances.<sup>[25,87–91]</sup> There have been numerous incidents in the past where LIBs have led to fires at recycling and waste treatment facilities. In the UK, for example, it is estimated that approximately 48% of all waste fires occurring each year are caused by LIBs, costing the UK economy approximately 158 million pounds annually.<sup>[92]</sup> Unfortunately, official statistics on LIBs as cause of fires in waste transportation and waste treatment facilities are rare, as they are often included in the category of self-ignition. Still, the fire hazard associated with spent LIBs is generally considered to be high.<sup>[90,93]</sup>

As a result, lawmakers have set specific safety requirements for the transportation and handling of newly produced and EOL LIBs. Different national and international laws define restrictions on transport quantities, specifications for safe packaging and package sizes, labelling requirements, and prescriptions for safety tests. One of the most important international standards for the transport of LIBs is specified in the UN Manual of Tests and Criteria, Part III, Subsection 38.3. This provision is applied in most regions of the world, including the EU, US, and China, and requires that battery cells (excluding small production runs and prototypes) must pass certain safety tests in order to be transported. The safety tests include altitude simulations as well as heat, vibration, shock, external short circuit, impact, overcharge and forced discharge tests. In the EU, LIB transport by road, rail and sea freight is regulated by the Agreement concerning the International Carriage of Dangerous Goods by Road (ADR), the Regulations concerning the International Carriage of Dangerous Goods by Rail (RDI), and the International Maritime Dangerous Goods Code (IMDG Code).<sup>[94–97]</sup> Comparable provisions in the U.S. are provided by Part 49 of the Code of Federal Regulations. In China, there are few national transportation regulations, but responsible authorities follow international provisions such as the ADR, RDI, and IMDG Code.<sup>[77]</sup>

According to these regulations, LIBs with energies >100 Wh are classified as Class 9 Dangerous Goods. Transport quantities for these batteries in road transport are limited to 333 kg per transport unit and strong outer packaging that prevents short circuits and unintentional activation is required. For smaller batteries with energies ≤100 Wh no transport quantity limits apply, but similar package requirements must be met, and package sizes may not exceed 30 kg. Further, batteries must be labelled adequately and pass the UN safety tests. Defective batteries must be classified as critical or non-critical. Batteries that are liable to decompose rapidly, react dangerously, produce a flame or a dangerous evolution of heat, or emit toxic, corrosive, or flammable gases or vapors are classified as critical. The transport of critical battery cells must be approved by a competent authority. For non-critical batteries, as well as batteries designated for disposal and recycling, there are no restrictions on transport quantities, but special packaging regulations apply. The batteries must be protected against short circuits, dangerous heat generation and leakage. This requires, for example, individual securing of the battery terminals, inner packaging to prevent contact between batteries, specially designed battery terminals, use of a suitable cushioning and absorbing material, leak proof inner packaging and venting devices.<sup>[94–97]</sup>

Further transport regulations include the International Civil Aviation Organization Technical Instructions (ICAO TI) and the International Air Transport Association Dangerous Goods Regulations (IATA DGR), which specify the international transport of dangerous goods by air. According to the directives, LIBs can generally be transported by air if the previously mentioned UN safety tests have been passed and ventilation, short circuit protection, reverse current flow prevention and additional packaging conditions are met. The transport of LIBs intended for disposal and recycling is prohibited, unless approved by national authorities. Defective or damaged LIBs may not be transported by air and standalone LIBs (UN number: UN3480) must have a state of charge (SOC)  $\leq 30\%$  to be transported. National regulations regarding air transport of LIBs in the EU, US and China are in accordance with the IATA DGR and the ICAO TI.<sup>[94–97]</sup>

The security risks and the resulting safety regulations have a significant impact on the costs for transport and handling of LIBs. Along with the transport distance, the transport quantities, capacity utilization, and additional safety precautions are important cost factors. Taking into account emissions trading and CO<sub>2</sub> prices, additional transport routes can have a great impact on the future profitability of battery production and recycling. Several studies have estimated the transportation costs as a percentage of total recycling costs. In a review of these studies, Slattey et al. found an average contribution of 41%. The transport cost estimates vary significantly from \$0.24/kg to \$5.51/kg for a standard distance assumption with an average value of \$1.54/kg.<sup>[98]</sup> Reasons for these deviations include regional differences in fuel and labor costs, as well as different calculation methods. The high transportation costs directly affect the profitability of recycling. Especially batteries with low material value (e.g., LFP or LMO) could become a financial liability to owners and recyclers, so that there are fewer incentives for battery recycling.

One possible solution to this problem would be the introduction of an EPR through legislation, including a physical, as well as financial responsibility. This way, costs for collection, transport, recycling, and disposal would be covered by battery producers and the profitability of the recycling step would increase. As an additional incentive for end-users to return batteries to collection points, a deposit system could also be useful. Beyond that, the costs for collection transport and recycling should generally be reduced. An option to minimize transportation costs is the strategic siting of collection points and recycling facilities.<sup>[98,99]</sup> The resulting reduction of transport distances has an impact on the total transport costs, but also reduces potential safety risks during transport. Another option is the establishment of a decentralized pre-treatment system for EOL batteries. This would enable the sorting, deactivation, dismantling and physical separation of batteries into different waste streams prior to recycling. After pre-treatment, the different materials could be transported to centrally located recycling facilities. A major advantage of this type of system is that pre-treated battery components (e.g., black mass or battery casings) may not be classified as hazardous goods, which could significantly reduce transportation costs and safety risks.<sup>[98]</sup>

## 2.4. Battery 2030+ Initiative in Europe

The Battery 2030+ roadmap suggests research actions to radically transform the way to discover, develop, and design ultra-high-performance, durable, safe, sustainable, and affordable batteries for use in real applications. The purpose is to make a collective European research effort to support the urgent need to establish European battery cell manufacturing. Additionally, recyclability is a key cross cutting topic, which is considered from the very beginning of the Battery 2030+ research program.<sup>[100]</sup> New battery materials engineered interfaces and smart battery cell architectures will be developed bearing in mind the manufacturability, scalability, recyclability, and life-cycle environmental footprint of the novel technologies. The future recycling process, in which materials escape from the technosphere into the biosphere as non-reusable materials, will be dramatically improved by novel recycling approaches. This approach that will lead to a drastic reduction of the proportion of batteries to be treated in classical recycling processes. Challenges to overcome regarding the reconditioning of the active materials are numerous and require further fundamental studies. As an example, for the cathode material, being the most expensive battery materials cost factor, new processes need to be further developed to restore its original chemical composition, crystallography, and coatings (Figure 2).

Would the material/components not be suitable to be reconditioned to battery grade because of, for example, structural or purity constraints, a fallback alternative in the last stage of the new process could consist of converting them to precursors and eventually changing the composition ratios, anticipating future chemistry changes and new generation materials as shown in Figure 3.

## 3. State of the Art Recycling Technologies

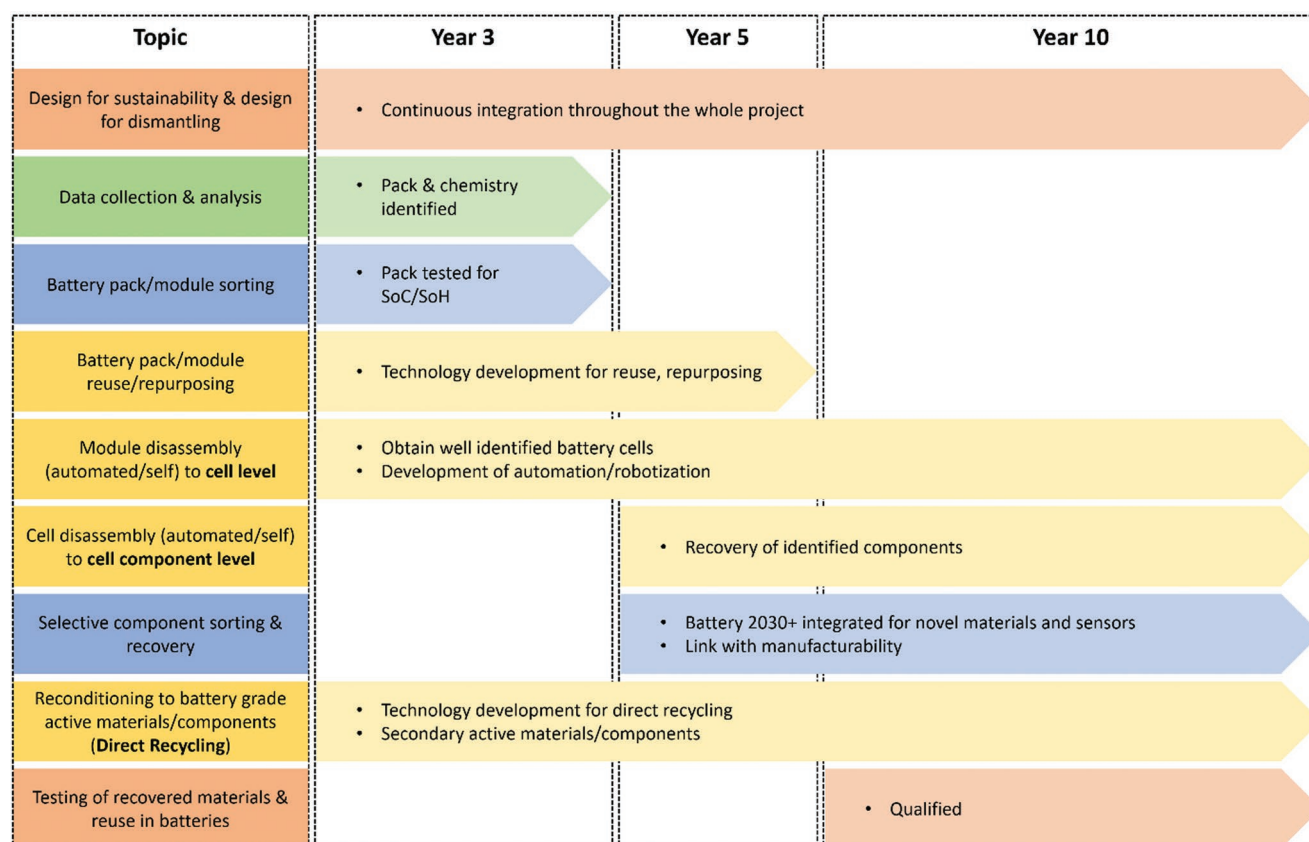
### 3.1. Pre-Treatment

Handling of Li-ion modules consists of two main processes: discharging, and dismantling of battery modules, including disconnecting of major components (cables, battery cells, frames, electronics, etc.). After cell removal, those go under the mechanical processing and separation. Mechanical separation techniques separate cell components based on their different physical properties such as particle size, density, conductivity, magnetic properties, etc.

#### 3.1.1. Discharging

Batteries are discharged before manual dismantling primarily to secure the safety of the personnel and to eliminate the risk of electric shocks. Additionally, sparks might cause the ignition of volatile organic compounds during the crushing process<sup>[21,101]</sup> and thus cause fires.

Discharging can be performed by different methods. Several companies use thermal pre-treatment (e.g., Accurec or REDUX in Germany),<sup>[96]</sup> salt-water based baths (NaCl or Na<sub>2</sub>SO<sub>4</sub>),<sup>[102–105]</sup> or controlled discharging via external circuits. NaCl and



**Figure 2.** Roadmap for the Battery 2030+ initiative.

alternative salts ( $\text{Na}_2\text{S}$  and  $\text{MgSO}_4$ ) were tested to discharge portable batteries. It was concluded that  $\text{NaCl}$  provided the best discharge profile. Also,  $\text{Na}_2\text{S}$  was a sufficient discharge media, while solutions containing  $\text{MgSO}_4$  were not capable to fully discharge the batteries.<sup>[106]</sup>

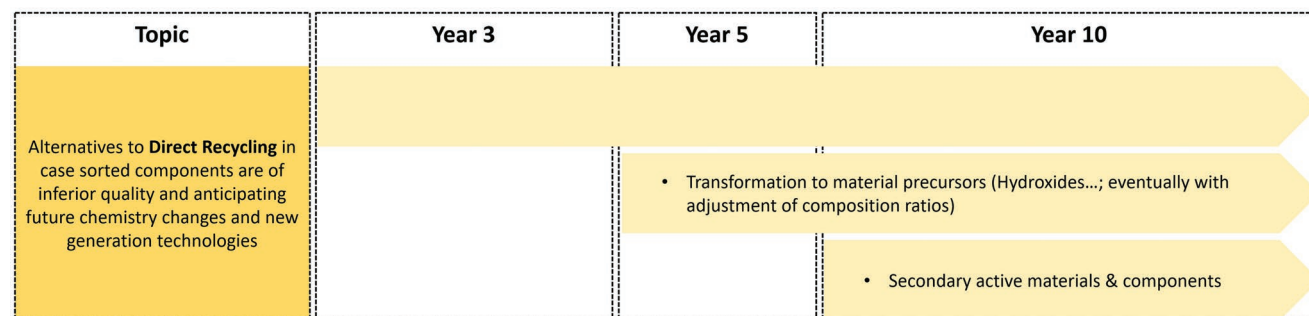
In some cases, batteries are discharged via cryogenic methods using liquid nitrogen or in vacuum atmospheres. Nevertheless, these methods are connected to higher OPEX and CAPEX.<sup>[106,107]</sup>

However, salt-water based discharge is still the most preferable method in the industrial environment. It was reported that it is a flexible and comparably safe way to stabilize different types of high-energy cells. The rate of discharge varies

depending on several factors, but predominantly on the solution resistance—itsself depending on the conductivity of the solution and the distance between the electrodes.<sup>[105]</sup>

### 3.1.2. Mechanical Pre-Treatment

Mechanical pre-treatment is used to disintegrate batteries and separate particular components into several streams. The main scope is to separate metallic particles (casing, copper and aluminum foils) and to concentrate the black mass, which is the mixture of cathode and anode active materials. Black mass is the most valuable battery cell component and mechanical



**Figure 3.** Battery 2030+: Anticipation of future chemistry changes and new generation materials.



pre-treatment is mostly designed to achieve its maximum recovery and separation. Mechanical processes involve crushing, sieving, magnetic separation, fine crushing, and classification, which are performed in sequence.<sup>[101]</sup> Multistage crushing and sieving result in quite effective separation. Magnetic separation is used to remove pieces of steel casing or joints. Traditional mechanical pre-treatment is performed by companies such as Akkuser (Finland), Batrec (Switzerland), or Duesenfeld (Germany). At Akkuser Oy, the batteries are crushed using a two-phase crushing process. Disintegrated material is separated using air separation and magnetic separation. An inert atmosphere is used during the crushing process. At Batrec the batteries are also crushed in a controlled atmosphere of CO<sub>2</sub> gas. The individual components, such as aluminum and steel casing, black mass, nonferrous metals, and plastic are separated in a multistage separating plant and sold to other producers. Duesenfeld is a start-up company, which also applies mechanical treatment of the batteries coupled with electrolyte recovery. The LIBs are discharged, shredded under nitrogen, and the electrolyte is evaporated and condensed. The dry materials are then separated using their physical properties via air separation and sieving.<sup>[108]</sup>

A flowchart for black mass recovery was proposed by Widi-jatmoko.<sup>[109,110]</sup> Cells were shredded and sieving was applied to separate the components into different size fractions. An attrition scrubbing technology was used to liberate fine black mass from coarse foils. In the milling step, the 850 µm size fraction gives the best composition of the black mass recovery with minimum presence of copper and aluminum. However, most of the black mass was held together by the PVDF binder.<sup>[110]</sup>

Beside the mechanical pre-treatment, mineralogical technologies such as flotation have been applied to recover the black mass.<sup>[111]</sup> Flotation is using the difference in hydrophilicity and hydrophobicity of electrode materials and other battery components. Yet, there are still challenges in using this technique since the efficiency of cathode material recovery is not sufficient.

Due to the low evaporation temperature of most electrolyte components (below 280°C), their recovery remains a key challenge in mechanical pre-treatment. Several approaches have been tested. The most common is the thermal pre-treatment below 300°C to evaporate the electrolyte. This method has been applied in the industry, but still results in insufficient electrolyte recovery.<sup>[101]</sup>

Some novel methods such as high-voltage treatment or the use of supercritical CO<sub>2</sub> for more effective separation have been reported.<sup>[112,113]</sup> By using high-voltage treatment 94 % of cathode particles were separated from the Al foil by sieving and 99 % of the particles maintained the chemical structure of original NMC.<sup>[112]</sup> Use of supercritical CO<sub>2</sub> lead to the separation of cathode material from the current collector when almost 99 wt% of polyvinylidene fluoride (PVDF) dissolved in a supercritical CO<sub>2</sub>-dimethyl sulfoxide system at 70 °C and 80 bar pressure after 13 min.<sup>[113]</sup> Despite promising results more investigation is required before the application of these new methods in industrial processing.

### 3.1.3. Thermal Pre-Treatment

Thermal pre-treatment methods are applied to safely deactivate the battery with regards to the combustible components of the electrolyte. Discharging of the battery can be achieved during

those processes.<sup>[114,115]</sup> Since the remaining carbon from the different components would interfere with the lithium recovery, the removal is mandatory. The used conductive carbon and acetylene black decompose above 600 °C.<sup>[116]</sup> Also, the binder can be removed by applying the respective decomposition temperatures which are typically lower compared to the carbon species.<sup>[117]</sup>

Mostly pyrolysis and incineration steps are applied for the pre-treatment. While similar processes, they can be distinguished by the presence or absence of oxygen or air.<sup>[118]</sup> By adding oxygen or air, incineration can be used to remove the binder or carbon species. However, all plastic parts and electrolytes are lost during the process.<sup>[38,119]</sup> Nevertheless, by choosing an appropriate temperature range, the subsequent recovery of lithium and other metals can be supported.<sup>[114,117,119–122]</sup>

In comparison, in an oxygen-free environment pyrolysis allows the transformation of the organic compounds into lower molecular compounds or their recovery by recondensation.<sup>[123]</sup> Further literature reports the separation of current collector foils and active material by pyrolysis.<sup>[124–126]</sup>

## 3.2. Hydrometallurgy

There are more than fifty companies worldwide, which process spent LIBs. The majority of the companies operate in China and South Korea, followed by the European Union, Japan, Canada, and the USA. It is predicted that over 700 000 tons of batteries will become obsolete by 2025. The worldwide recycling capacity is estimated to be around 400 000 tons at that time.<sup>[127]</sup> Recycling companies in China have different outcomes and achieve different grades of final purities, however almost all companies use hydrometallurgical approaches. The main recovery products are salts such as NiSO<sub>4</sub>, CoSO<sub>4</sub>, and Li<sub>2</sub>CO<sub>3</sub> or products in form of mixtures for precursor production based on NMC and NCA.<sup>[127]</sup> In the European Union, the most common recovery methods are pyrometallurgy, hydrometallurgy, and combinations of both. Due to the requirements of the new EU Battery Directive, the high demands on the precursor materials for battery production, and the goal of creating a circular economy, hydrometallurgy will be the most preferable process. The main reason is the ability to recover larger amounts of battery components and reach very high purities of metal salts. One of the downsides of hydrometallurgy is the need for mechanical pre-treatment consisting of two main processes: disintegration of the batteries and separation of the particular components.

Hydrometallurgical recycling of LIBs is based on the dissolution of metallic components mostly coming from the active material (mixture of cathodes and anodes), preferably with mineral acids, followed by metal separation by solvent extraction, ion exchange, and precipitation. A thermal pre-treatment (pyrolysis or calcination) can be applied as well.<sup>[41,28]</sup>

Leaching is applied as the first step in the hydrometallurgy process after mechanical or also thermal pre-treatment. The leaching media are usually mineral acids (H<sub>2</sub>SO<sub>4</sub>, HCl, and HNO<sub>3</sub>), in some cases also alkali and organic acids.<sup>[129,130]</sup> Studies show that inorganic acids are highly effective for the recovery of metals (>99%) in the leaching procedure when applied under optimal conditions. The most common acid is

sulfuric acid due to its effectiveness, costs, and compatibility with metal separation techniques. Generally, under comparable conditions, leaching efficiencies of Li and Co from LCO cathodes are  $\text{H}_2\text{SO}_4 \approx \text{Cl} > \text{HNO}_3 > \text{H}_2\text{SO}_3$ .<sup>[131]</sup> In the leaching process, graphite stays in a solid residue after filtration. A significant number of leaching studies have been published and concluded that the temperature (40–60 °C), solid to liquid ratio S/L (1/10–1/20) and the presence of the reduction agent (e.g.,  $\text{H}_2\text{O}_2$ ) are the most important factors affecting the complete recovery of metal ions.<sup>[132–134]</sup> A different approach has been published by,<sup>[135]</sup> where foils from current collectors were used as a reduction agent. It was reported that more than 99% of Li and Co can be extracted by addition of Al/Cu containing fractions.<sup>[135]</sup> Iron scrap has also been applied as a reduction agent utilizing the reduction potential of ferrous ions.<sup>[23]</sup>

The use of organic acids such as ascorbic, citric, malic, tartaric, and succinic acid has been reported as well.<sup>[136–139]</sup> To eliminate the use of inorganic reductants, organic reduction agents were also tested, including ascorbic acid and carbohydrates (e.g., cellulose, etc.).<sup>[138]</sup> Still  $\text{H}_2\text{O}_2$  was the most common reduction agent applied. The organic reagents might represent a lower risk for the operations and equipment used. Such reagents also decompose under aerobic and anaerobic conditions with limited gas generation. From the tested acids lactic acid showed a high recovery of Li (98%), Co (99%), Ni (98%), and Mn (98%) using 1.5 M lactic acid, S/L of 20 g L<sup>-1</sup>, 70 °C, and 0.5 vol%  $\text{H}_2\text{O}_2$ , with very short leaching time of 20 min.<sup>[140,141]</sup> However, very high leaching efficiencies were reported for the majority of tested organic acids when a reduction agent ( $\text{H}_2\text{O}_2$ ) was applied.

A novel approach is based on applying ultrasound when leaching with mineral acids<sup>[142]</sup> or organic acids.<sup>[143]</sup> In all cases a higher efficiency has been reported.

Bioleaching and biotechnologies are using microorganisms to recover metals from different resources.<sup>[144]</sup> The group of microorganisms involved in the bioleaching of metals from LIBs were usually chemolithotrophic prokaryotes, heterotrophic bacteria, and fungi.<sup>[145]</sup> Relatively high recoveries of Ni (90%), Mn (92%), Co (82%), and Li (89%) from spent LIBs in 72 h with a solid content of 100 g L<sup>-1</sup> were achieved using bacteria—*Acidithiobacillus ferrooxidans*.<sup>[146]</sup> However, despite extensive research, biotechnologies are not commonly applied in battery recycling due to comparably slow process kinetics, leading to long processing times. Some battery components such as typically used organic solvent based electrolytes and selected, mostly fluorinated binders are also toxic to microorganisms. Another disadvantage is that at high pulp density, the efficiency of the process is much lower. Using bioleaching, there is no potential for changing the metal valence state, which is commonly needed for a better cobalt recovery.<sup>[145]</sup>

After the leaching step an effective separation of metal ions is necessary. The most common separation methods are solvent extraction (SX), chemical precipitation, ion exchange (IX), electrochemical deposition, and more novel eutectic freeze crystallization.<sup>[147–149]</sup> The separation procedure is usually a combination of several separation techniques. Precipitation is most common for the removal of the impurities Fe, Al, and Cu. Solvent extraction is usually applied to separate NMC metals such as Mn/Co/Ni. In case that the Cu concentration in the leachate

is high, solvent extraction is applied for its recovery instead of precipitation. Lithium is usually recovered via precipitation with sodium carbonate, carbon dioxide, or via ion exchange.

The first step in leachate processing is the impurity (Fe/Cu/Al) removal via pH adjustment and precipitation.<sup>[101]</sup> However, these methods are related to a significant loss of valuable metals (sometimes Co loss can be around 10%). Recently a novel approach via ion exchange has been reported.<sup>[150]</sup> The suggested process produced a >99.6% pure Li/Co/Ni solution (battery grade) with a Co loss of around 1% only.

The most common method for Mn/Co/Ni separation is solvent extraction.<sup>[134,151–153]</sup> The acidic extractant di-(2-ethyl-hexyl) phosphoric acid (D2EHPA) is very often applied for Mn recovery.<sup>[152,154,155]</sup> The extractant concentration reported is usually around 0.4–0.5 M D2EHPA diluted in kerosene. Operation temperature is very often ambient, and the process is performing sufficiently at pH 2.2–5. The contacting time is reported to vary from 5–15 min.<sup>[156]</sup>

Co and Ni are separated using organophosphorus acids, chelating oximes, carboxylic acids, and high molecular weight amines (HMA). The organophosphorus acids (e.g., Cyanex 272) and chelating oximes are most widely used and commercially acceptable.<sup>[157]</sup> When using acidic extractants cobalt is extracted at lower pH before Ni. Co is tetrahedrally coordinated in Cyanex 272 as  $\text{Co}(\text{R}_2\text{H})_2$  and nickel forms the hydrated octahedral complex  $\text{Ni}(\text{R}_2\text{H}_2)_2(\text{RH})_2$ . Co is extracted at pH around 5 while Ni is efficiently recovered around pH 6.5.<sup>[158,159]</sup>

After the separation, Co and Ni salts are recovered using controlled crystallization.<sup>[101]</sup> Recently a new approach was tested using eutectic freeze crystallization to separate and recover metal ions after hydrometallurgical treatment of LIBs.<sup>[147]</sup> The eutectic freeze crystallization (EFC) is a method to separate aqueous inorganic solutions into ice and solid solutes via freezing at the eutectic point. The advantages of EFC are lower energy requirement and the possibility of complete conversion into water and solidified solutes.<sup>[149]</sup> It was reported that with suitable control of supersaturation, ice and cobalt and nickel salt crystals can be recovered as separate phases below eutectic temperatures.<sup>[148]</sup>

Li is usually the last metal recovered from the stream. The most common way is the precipitation with sodium carbonate or sulfate. The concentration of lithium ions in the solution directly affects the recovery efficiency and the purity of the compound precipitated. Larger concentrations involve higher recovery efficiencies with the drawback of a maximum concentration value after which the purity starts to decrease.<sup>[160]</sup> According to ref. [161], when the solid  $\text{Na}_2\text{CO}_3$  is used directly as the precipitant, the lithium recovery rate is higher than using a saturated  $\text{Na}_2\text{CO}_3$  solution while the purity obtained would not be affected. The concentration of  $\text{Na}_2\text{CO}_3$  or  $\text{Na}_3\text{PO}_4$  added in the solution is also significant and affects the recovery efficiency, and the purity of the compound precipitated. Several temperature values (from 25 to 95 °C) have been studied for the precipitation of lithium carbonate or lithium sulphate and studies show that the closer the temperature is to 100 °C, the more effective is the precipitation.<sup>[162]</sup> Also, the pH of the solution is a determinant factor in the precipitation of lithium carbonate/phosphate. When using  $\text{Na}_2\text{CO}_3$ , optimal pH of operation is between 10 and 11.5, while for  $\text{Na}_3\text{PO}_4$  this range is between 11

**Table 2.** Examples of hydrometallurgical recovery of valuable metals from LIBs in a form of different products and achieved purities.

Reference	Hydrometallurgical procedures	Products	Reported purity
[164]	1) Precipitation in pH 5.5	Li <sub>2</sub> CO <sub>3</sub>	>98%
	2) Carbonization	CoCO <sub>3</sub>	36–37 %
[164]	1) Precipitation at pH 5.5	Li <sub>2</sub> CO <sub>3</sub>	>98%
	2) Solvent extraction	CoCO <sub>3</sub>	47%
	3) Carbonization		
[165]	1) Precipitation of Ni using (CH <sub>3</sub> C(NOH)C(NOH)CH <sub>3</sub> )	CoC <sub>2</sub> O <sub>4</sub> ·H <sub>2</sub> O	98%
	2) Solvent extraction of Co and Mn using D2EHPA	Li <sub>2</sub> CO <sub>3</sub>	99%
	3) Co-precipitation (NH <sub>4</sub> OH) and Li-precipitation (Na <sub>2</sub> CO <sub>3</sub> )	MnSO <sub>4</sub> Ni <sup>+</sup>	98%
[166]	1) Solvent extraction of Mn using D2EHPA	MnO <sub>2</sub>	>99.5%
	2) Scrubbing the impurities (MnSO <sub>4</sub> )		
	3) Stripping with acid		
	4) Oxidative precipitation using KMnO <sub>4</sub> and MnO <sub>2</sub>		
[167]	1) Extraction with (I)P204 (II)P507	Co-metallic	99%
	2) Electrowinning		
[168]	Using different lithium solubilities	Li <sub>2</sub> CO <sub>3</sub>	99.5%
[169]	1) Precipitation of Fe	Co <sub>3</sub> O <sub>4</sub>	83%
	2) Solvent extraction	CoC <sub>2</sub> O <sub>4</sub>	96%
	3) Li precipitation using Na <sub>2</sub> CO <sub>3</sub>	NiO	89%
		Li <sub>2</sub> CO <sub>3</sub>	99%
[170]	1) Reductive hydrogen roasting	LiOH·H <sub>2</sub> O	99.9%
	2) Water leaching of lithium		
	3) Crystallization of lithium hydroxide		

and 13.<sup>[162]</sup> The time of agitation usually has a minimum value of 35 min and 60 min is a common standard to get recovery efficiencies over 80%. A novel approach to recover Li before the extraction of heavy metals was reported by ref. [163] using tributylphosphate (TBP) and FeCl<sub>3</sub> with a reported purity of lithium of almost 99%. Also, Solvay recently developed a phosphorus-based extractant with the trade name CYANEX 936P for selective recovery of Li. There are different approaches of hydrometallurgical processing listed in the **Table 2**.

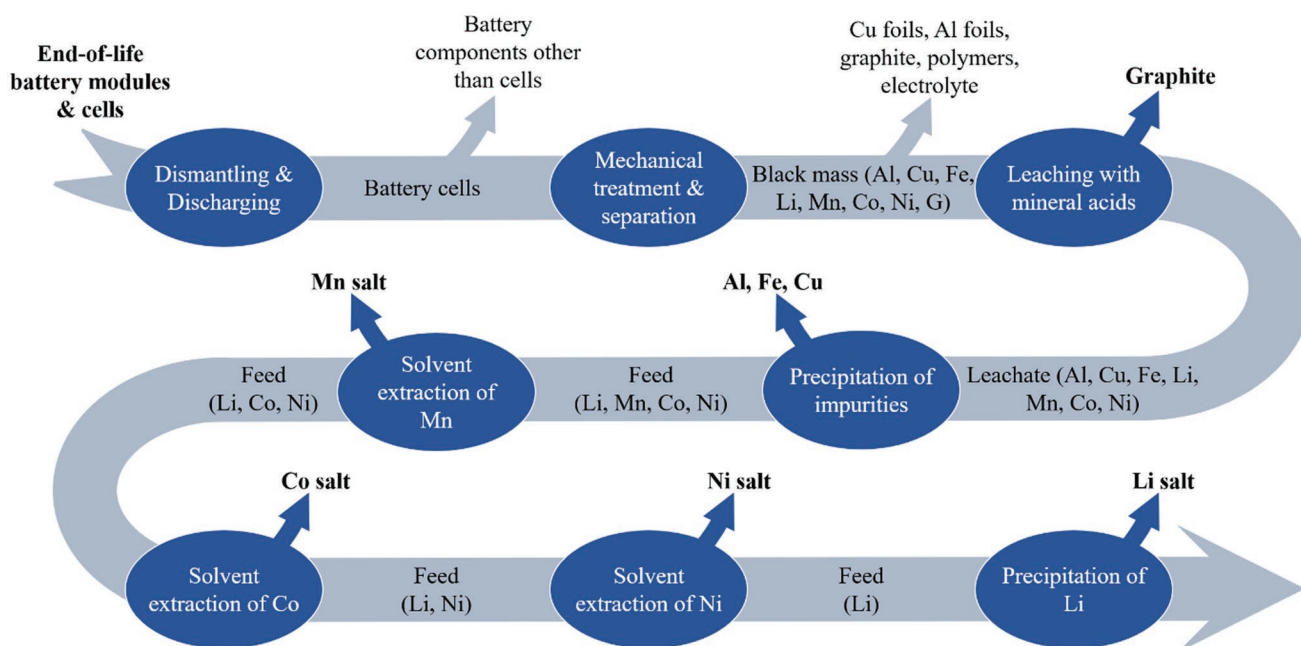
Industrially hydrometallurgy is mostly used in China and South Korea (e.g., Brunp, Soundon New Energy, GEM, Huayou Cobalt, Ganpower, etc.). Since the majority of batteries are produced in these countries, the infrastructure is very well organized to gather sufficient volumes of production scrap and spent batteries. In Europe, hydrometallurgical processes are used for instance in Sweden, where the battery producer Northvolt AB integrates a hydrometallurgical recycling process in the production cycle in order to secure the raw material supply and to decrease the environmental impact of battery production. Hydrometallurgical processing is also used in companies such as Eramet (France) and Fortum (Finland), etc. At Northvolt, the initial collection and handling of batteries is followed by the processing of batteries to recover aluminum, copper, steel, plastics, electronics, and electrolyte. The pilot plant has been running for a few years and the full-scale recycling plant will process 8500 t of black mass per year. At Fortum, LIBs are first disassembled and treated during a mechanical process at a plant in Ikaalinen, Finland. The black mass is collected and then taken to hydrometallurgical processing at another plant in Harjavalta, Finland. The recycling process achieves recovery rates of up to 95% and involves a chemical precipitation method. There

are several companies that apply a combined approach. Nickelhütte Aue GmbH (Germany) or Umicore (Belgium) use a hydrometallurgical treatment after smelting of the batteries to recover metals from the alloy (matte). Nickelhütte Aue GmbH has a hydrometallurgical plant and produces approximately 3900 t of Ni per annum. Matte processing starts with comminution followed by pressure oxidation leaching at 6 to 8 bar. Afterward, impurities are removed prior to solvent extraction of Cu, Co, and Ni. Before extraction, Fe is precipitated as goethite (FeOOH) by using H<sub>2</sub>O<sub>2</sub> as an oxidizing agent and basic nickel carbonate for pH adjustment. Depending on the process setup, copper sulfate, cobalt sulfate, and nickel sulfate, or nickel carbonate and chloride are produced.<sup>[171]</sup> In general, the processing and steps are depicted in **Figure 4**.

The main advantage of hydrometallurgy is the possibility to produce new battery precursors from waste with the sufficient purity. Despite the large demand for chemical reagents, hydrometallurgy allows the re-utilization of many solvents and by-products for several years, minimizing the overall secondary waste generation.<sup>[14]</sup> With future battery legislation and demands for higher material recovery rates, hydrometallurgy is one of the most promising approaches to meet the requirements but also to create a path to circular economy in the battery market.

### 3.2.1. Deep Eutectic Solvents

Within the realm of hydrometallurgy lies solvometallurgy in which organic phases are exclusively used.<sup>[172]</sup> Solvometallurgy offers a complementary set of tools parallel to hydrometallurgy, and aims to reduce the waste and environmental impact of



**Figure 4.** Overview about traditional hydrometallurgical processing.

hydrometallurgy.<sup>[172]</sup> Currently, the recovery capacities of hydrometallurgy are able to cover the needs and benchmarks set by European Policy in terms of metal recovery. Nevertheless, this does not address lithium, electrolytes and future water consumption regulations. The organic phases reduce waste by limiting water consumption, performing the leaching and solvent extraction in a single step, and increasing selectivity. An ideal organic phase must be efficient, low cost, easily recoverable or biodegradable.<sup>[172]</sup> Low transition temperature mixtures (LTTM) can offer all of these properties. LTTM refers to a mixture of two or more compounds: one hydrogen bond donor (HBD) and one hydrogen bond acceptor (HBA) that undergoes a second phase transition resulting in a glass transition temperature.<sup>[173]</sup> Deep eutectic solvents (DES) refer to a sub-category of LTTM in which there is a significant drop in the melting point and which remain liquid at specific molar proportions.<sup>[174]</sup> There is a broad nomenclature used depending on the composition of the solvent.<sup>[175]</sup> “Hydrophobic DES” is used when both components are hydrophobic<sup>[176]</sup>, and “Brønsted acid DES” implies that the HBA is a Brønsted acid instead of the more commonly used quaternary ammonium salt.<sup>[177]</sup> These solvents have been recently introduced to the metal and battery recycling flow-sheets as successful media for electrochemical recovery,<sup>[178,179]</sup> direct leaching of metals (Li<sup>[180]</sup>, Ni, Co) from LIB cathodes, and even the dissolution of the polymeric binders in cathodes.<sup>[181]</sup>

Ethylene glycol,<sup>[179]</sup> urea<sup>[182–184]</sup> based DES have the physical properties required to act as leaching agent and electrolyte for the electrochemical recovery of metals from LIBs. The studies by Wang<sup>[181]</sup> and Landa-Castro<sup>[182]</sup> discuss the role of urea and ethylene glycol as reducing agents of the leaching reaction. This redox process requires high temperatures (above 150 °C) and longer leaching times than those using mineral acid leaching. Electrooxidation, a process in which a DES/metal oxide paste coats the anode and the cell is held at constant

voltage (2.7 V), was introduced by Pateli et al.<sup>[185]</sup> as alternative using lower temperature and shorter processing times. The formation of superoxide that allows for the reaction of oxides into organometallic and chloro complex species is suggested as the driving force but further studies are required. However, the low proton activity and poor ligand capabilities of ethylene glycol and urea limits their applicability for direct leaching when compared to DES with more acidic HBD.<sup>[186]</sup> Solvometallurgical approaches for the leaching of cobalt from LiCoO<sub>2</sub> have been reported using p-toluenesulfonic acid and citric acid as HBDs.<sup>[178,184,187]</sup> Those studies report higher S/L ratio, shorter leaching times and lower temperatures than urea, ethylene glycol and glycerole. Recent work on the thermodynamics of the process have shown that overcoming the lattice energy and Gibbs energy of the metal oxides is an important factor in controlling solubility.<sup>[186,188]</sup> These findings point to acid-based DES as the most promising category for the direct leaching of metals from LIBs in the near future, but the kinetics and solvation mechanisms need to be studied in further detail before scaling. LTTMs are the latest entry into the wet (hydro, solvo, iono) metallurgy arsenal. They are part of the efforts to lower the entry barrier of room temperature ionic liquids and their analogues into the metallurgy flowsheet. Nevertheless, the fundamental knowledge of aqueous solutions cannot be applied to these new solvents and their specific kinetics and speciation remains to be determined. Other challenges like the role and means of pH variations on the extraction of specific metals, and the effect of additives and co-solvents are also at early stages of research. As our understanding of LTTM and DES increases, so will their importance for the recycling of LIBs.

The main advantage of hydrometallurgy is the possibility to produce new battery precursors from waste with sufficient purity. Despite the large demand for chemical reagents, hydrometallurgy allows the re-utilization of many solvents



and by-products for several years, minimizing the overall secondary waste generation.<sup>[4]</sup> The integration of solvometallurgy into critical hydrometallurgical steps, like leaching could boost the overall efficiency of the approach with citric acid DES being able to achieve effective leaching at 55 g L<sup>-1</sup> S/L versus 25 g L<sup>-1</sup> of the aqueous solution.<sup>[143]</sup> Novel organic media use less volume and shorter leaching times with higher average loading capacities. With future battery legislation and demands for higher material recovery rates, hydrometallurgy is one of the most promising approaches to meet the requirements but also to create a path to circular economy in the battery market.

### 3.3. Pyrometallurgy

Pyrometallurgical approaches use a high-temperature furnace to reduce the component metal oxides from LIBs to form an alloy. In general, smelting and roasting/calcination steps are the main procedures during processing.<sup>[189–192]</sup> Further classification is possible with regard to the applied extraction mechanisms and the processing atmosphere. During this process, the organic and volatile parts of the battery (electrolyte, separator, and binder) are evaporated. While high temperatures for metal recovery cause more chemical reactions, low temperatures lead to phase transitions during the process.<sup>[193]</sup> Therefore, pyrometallurgical techniques are depending on factors like temperature, processing time, types of purge gas, and flux addition.<sup>[194]</sup> Higher temperatures are used to incinerate the plastics and all remaining organic compounds.<sup>[114]</sup>

The process is aided by the metal current collectors and therefore relatively mature compared to other techniques.<sup>[72]</sup> In addition, the evaporation and burning of the electrolytes, binders and plastics is exothermic, which reduces the energy consumption required for the process.<sup>[195]</sup> The products from this process are slags, metallic alloys, and gases. The alloys can be further refined by hydrometallurgical processes. While the lost fraction (electrolyte, salts, plastics, etc.) can significantly reduce the overall recycling efficiency, the process itself contains only little risks. However, the production of toxic gases and the requirement for hydrometallurgical post-processing poses an environmental drawback, while the robustness of the process is well suited for imperfectly sorted feedstock of cells and no special pre-treatment of cells is necessary.<sup>[116,191,194,196,197]</sup>

The pyrometallurgical approach is applied in several industrial processes like the ultra-high-temperature smelting-technology (UHT) of Umicore, the roasting–smelting process of Glencore, the high temperature melting recovery (HTMR) process of Inmetco and the calcination processes of Sony–Sumitomo and Accurec.<sup>[197,198,199]</sup>

Research works mainly focus on the variation of the above-mentioned classification parameters on a laboratory scale. Several methods have been reported for roasting processes involving exothermic processes such as gas-solid reactions at elevated temperatures. Carbothermic reduction roasting uses a reducing agent such as carbon along the cathode active material to form alloys.<sup>[200]</sup> NMC-based cathodes were roasted at a temperature of 650 °C, a roasting time of 30 min, and a carbon dosage of 10% of raw material input resulting

in Li<sub>2</sub>CO<sub>3</sub>, MnO, Ni, and Co with high recovery rates.<sup>[201]</sup> Roasting of LCO cathodes was reported under a nitrogen atmosphere at 1000 °C for 30 min. The resulting residue was leached with water and after a wet magnetic separation step, the following recovery rates were achieved: Li 98.93%, Co 95.72%, and graphite 91.05%.<sup>[190]</sup> This approach was further applied for LMO cathodes with the following parameters: roasting for 45 min at a temperature of 800 °C. Afterward the residue (Li<sub>2</sub>CO<sub>3</sub> and MnO) was leached with water and was mechanical separated. 99.13% of the Li was recovered, while the filter residue was calcined to remove carbon. This resulted in a 95.11% recovery rate for Mn<sub>2</sub>O<sub>4</sub>.<sup>[202]</sup> In contrast, oxidizing conditions were used to treat LFP cathodes. Here, an effective temperature control is important. At temperatures above 650 °C the oxidizing roasting leads to the release of fluorine-containing gas. Furthermore, the recovery of iron is reduced due to the formation of Fe<sub>2</sub>O<sub>3</sub>.<sup>[203]</sup> Microwave-assisted carbothermic reduction was recently reported as a development for roasting. The reactivity of carbon in a microwave environment results in faster reaction times.<sup>[204]</sup>

In order to cope with the low solubility of Li<sub>2</sub>CO<sub>3</sub> more recent advances in pyrometallurgical approaches included the application of salt assisted roasting. Depending on the salt, classification is normally described as chlorination, sulfation, and nitration roasting. During the roasting, different agents are applied to produce better soluble lithium compounds. For chlorination, agents like NH<sub>4</sub>Cl were reported.<sup>[189]</sup> In comparison, SO<sub>2</sub>(g), MgSO<sub>4</sub>, NH<sub>4</sub>SO<sub>4</sub>, NaHSO<sub>4</sub>·H<sub>2</sub>O, or Na<sub>2</sub>SO<sub>4</sub> were applied during sulfation to obtain Li<sub>2</sub>SO<sub>4</sub>.<sup>[198,205–208]</sup> Nitration roasting aims at the formation of LiNO<sub>3</sub>.<sup>[192]</sup>

Smelting on the other hand uses temperatures above the melting points of the respective battery components. Here, the metals are separated in the liquid phase.<sup>[209,210]</sup> The advantage of this process is its robustness against different cell chemistries and that no passivation/deactivation step is needed.<sup>[196]</sup> The electrolyte is evaporated at lower temperature before the actual smelting. The burning of the remaining organic components add additional energy to the process.<sup>[36]</sup> At higher temperatures, alloys, gases, and slags are produced which can be further optimized.<sup>[197,211,212]</sup>

Nevertheless, pyrometallurgical approaches need a refining step (e.g., hydrometallurgy) after the initial treatment for the subsequent separation and recovery of the metals.<sup>[213]</sup> Furthermore, the lithium recovery must be enhanced.<sup>[39,214]</sup>

### 3.4. Recycling of Specific Components—Short Overview

#### 3.4.1. Anode (Graphite)

While not established as cathode recycling, anode recycling has gained more attention in the last years. Anodes can contain natural and artificial graphite, carbonaceous materials or even silicon. However, graphite is the commercially most applied material for now.<sup>[7,215,216]</sup> Several approaches have been reported in literature, including pre-treatment, pyrolysis, hydrometallurgy, supercritical, and water treatment. The research is still ongoing, with reviews and research papers appearing in a more frequent manner.<sup>[216–224]</sup>

### 3.4.2. Cathode

Hydro- and pyrometallurgical approaches dominate the recycling of metals from cathode materials.<sup>[96,116,130,150,160,163,170,194]</sup> Bioleaching on the other hand is relatively new in the field of LIB recycling and seldom reported so far. Some literature reports the successful application of bacteria to obtain metal fractions from spent batteries.<sup>[146,225–227]</sup> Phytoremediation was also reported as one way to extract spent cathode material from storage and handling sides.<sup>[228]</sup>

### 3.4.3. Electrolyte

Similar to the anode, the electrolyte gains more attention in terms of recycling. While usually evaporated or burnt during thermal processes, several approaches have been reported to recover at least the lithium from this component.<sup>[72,229,230]</sup> The first approach was the recovery of the electrolyte via liquid extraction.<sup>[229]</sup> Afterward, extraction via sub- and supercritical media was proposed with relatively good recovery rates,<sup>[219,231,232,233–240]</sup> which was also applied for binder recovery.<sup>[113]</sup> However, other valorisation approaches were reported as well.<sup>[241,242]</sup>

## 3.5. Differentiation between Academic Approaches and Industrial Reality

Despite the recent increase in publications with regard to the topic of LIB recycling, most of the proposed methods are still based on lab-scale experiments. While under research conditions, most parameters, chemicals, or processes can be chosen freely, but the applicability transfer into larger processes may not be possible or at least rather difficult. Especially in the upscaling of chemicals, for example, for leaching, cost and safety must be considered. On the other hand, high energy demands, due to either high temperatures, long processing times, or high pressures can interfere with a successful upscaling of a process. The same is true for high operating costs related to gas consumption or special components needed for extraction. Therefore, a critical view on lab-scale processes and suggested procedures should always accompany these developments. At the moment, the main industrial recycling technologies can be divided into three categories: pyrometallurgy, hydrometallurgy, and a combination of both methods.<sup>[40]</sup> Therefore, without the research of new and out of the box approaches, the recycling of existing and upcoming battery generations will only be developed slowly.

## 4. Recycling of Future Batteries—Current Approaches and Challenges, and Their Critical Assessment

### 4.1. Future Generation Battery Chemistries

#### 4.1.1. Li-Metal Systems

The emergence of electric vehicles and of new developments in the field of portable electronics are driving the constantly increasing demand for batteries with higher energy densities,

faster charging capabilities, and longer cycle lifetimes.<sup>[2,57]</sup> As the graphite-based lithium-ion technology slowly approaches its theoretical capacity limits, alternative anode materials are receiving more and more attention. With a more than ten times higher theoretical specific capacity compared to graphite (372 mAh g<sup>−1</sup> in the unlithiated state), lithium metal anodes (3860 mAh g<sup>−1</sup>) are one of the most promising options to meet these increasing performance requirements. The development of safe, high-energy lithium metal batteries (LMBs) is based on several different approaches, including for instance Li–sulfur batteries (Li–S), Li–oxygen batteries (Li–O<sub>2</sub>), and Li–intercalation type cathode batteries. The commercialization of LMBs has so far mainly been hampered by the issue of high surface area lithium metal deposits (so-called “dendrites”) and the associated safety concerns.<sup>[69,86,243,244]</sup> However, continuous progress in research indicates that LMBs will be one of the dominant battery technologies among next generation battery systems. Therefore, an early consideration of possible recycling methods for this battery type is important.

In comparison to commercial LIBs, LMBs have significantly higher lithium contents. In view of rising lithium prices and the lower material value of alternative cathode materials, such as sulfides, high recycling efficiencies for lithium will be an important prerequisite for future recycling processes.<sup>[245]</sup> As one of the most mature and robust recycling techniques, pyrometallurgy is widely used.<sup>[71]</sup> However, since non-noble metals such as aluminum and lithium are slagged in this process, further refining steps, for example, through hydrometallurgy, are required to achieve high recycling efficiencies.<sup>[71,72,246]</sup> Hydrometallurgy will therefore play a particularly important role in the recycling of the batteries of the future.

As described in Table 2, there are different approaches for the hydrometallurgical recovery of lithium from used LIBs. Typically, these processes consist of the dissolution of metallic components followed by various separation processes, such as sieving, solvent extraction, ion exchange, and precipitation.<sup>[71,247]</sup> In addition, thermal pre-treatment may be applied. For the dissolution of metallic lithium, water is a suitable solvent.<sup>[248]</sup> From aqueous solution lithium recovery can eventually be achieved through precipitation as carbonate, sulphate, phosphate, chloride or hydroxide.<sup>[71,246,248]</sup> A well-known method, for instance applied by Retriev Technologies and Recupyl, is based on the precipitation of Li<sub>2</sub>CO<sub>3</sub> from aqueous solution by adding CO<sub>2</sub>.<sup>[249]</sup>

A major challenge in the recycling of LMBs results from the high reactivity of metallic lithium. This particularly includes the high energy release (≈32 MJ kg<sup>−1</sup> Li) and the formation of hydrogen (≈1.6 L H<sub>2</sub> kg<sup>−1</sup> Li) during the dissolution in water.<sup>[248]</sup> In addition, dendrite formation during aging of LMBs still increases the risk of short circuits and subsequent thermal run-aways.<sup>[69,86,243,244]</sup> Special safety precautions must therefore be taken into account, especially when handling EOL LMBs. Possible approaches to address these issues in the recycling process include cryogenic treatment with liquid nitrogen prior to cell crushing, mechanical crushing in an inert atmosphere, or thermal oxidation of Li prior to dissolution in water.<sup>[71,72,247,248]</sup> Beyond that, deactivation of the cells as a pre-treatment could be beneficial to reduce potential safety risks. One method for the deactivation of battery cells is the discharge through electrical circuits, immersion in salt solutions or placement in

containers filled with steel chips.<sup>[107,250,251]</sup> However, since many cells are protected against over discharge, they usually retain a residual charge after treatment.<sup>[252]</sup> Corrosion processes at the contacts, as well as different cell designs, can also hinder deactivation by discharging. Another deactivation method is based on the removal of liquid electrolytes by evaporation or extraction.<sup>[232,236,253]</sup> Thereby, potentially toxic, and flammable substances can be removed from the cells and the hazard potential of thermal runaways can be reduced.<sup>[238,254,255]</sup> Potential safety risks associated with the reactivity of lithium metal, however, are not eliminated. Alternative approaches for the deactivation of LIBs and LMBs are based on the treatment with supercritical CO<sub>2</sub>. One strategy could be the removal of the metallic lithium in the form of CO<sub>2</sub>-soluble complexes from the battery cells before the actual recycling process. Similar procedures are known from the extraction of cobalt from LIBs, heavy metals from soil or radioactive metals from nuclear waste.<sup>[256–260]</sup> As complexing agents, organophosphates,  $\beta$ -diketones or crown ethers could be used. Besides the deactivation, an effective extraction would enable the early separation of different material streams and could make subsequent recycling processes more efficient. In a different approach by Sloop et al., the metallic lithium from LIBs is converted to Li<sub>2</sub>CO<sub>3</sub> using supercritical CO<sub>2</sub>.<sup>[261]</sup> The inert nature of Li<sub>2</sub>CO<sub>3</sub> facilitates the handling and transportation of treated cells and can contribute to significant cost savings throughout the recycling process. In addition, through treatment with supercritical CO<sub>2</sub>, liquid electrolytes can be partially extracted, increasing the overall recycling efficiency.<sup>[261]</sup>

Further challenges for the recycling of LMBs arise from other cell components, such as the cathode material or the type of electrolyte. Therefore, in the following, Li–S batteries and all-solid-state batteries will be discussed in more detail.

#### 4.1.2. Li–S

Using the high theoretical capacity of sulfur (1675 mAh g<sup>−1</sup>), lithium sulfur batteries (Li–S) are among the most promising future batteries. The cell chemistry of Li–S is quite different to LIBs. Typically, the Li–S system consists of a lithium metal anode and a sulfur/conductive carbon composite cathode with a liquid organic solvent-based electrolyte.<sup>[262,263]</sup> Since several disadvantages still hamper a broad practical application of these batteries, no recycling processes are established so far.<sup>[264–266]</sup> However, some approaches were reported in the literature.<sup>[267]</sup> While the challenges with regard to the Li metal are discussed in the section above, the separator, binder and electrolyte can be subject to similar recycling procedures which are used or investigated for LIBs. Nevertheless, it is necessary to carry out further studies on the applicability and possible upscaling for industrial processes. Regarding the sulfur-containing cathode, several points need to be taken into account. During operation (charging and discharging), intermediate Li-polysulfides are formed, which makes the recycling process challenging.<sup>[268]</sup> Furthermore, toxic gases like H<sub>2</sub>S can be formed during operation, opening and processing of the cells. In best cases, these gases can be converted to elemental sulfur or precipitated as sulfides, for example with copper.<sup>[267]</sup> However, in combination

with the highly reactive Li metal anode, separation and handling of the electrodes will complicate future recycling.

#### 4.1.3. All Solid State

The development of all-solid-state batteries (ASSBs) is driven by several factors, including the need of high-energy batteries, improved battery safety and also new applications. Some types of solid electrolytes (SE) are now able to achieve ambient-temperature (RT) ionic conductivities in the order of 1 mS cm<sup>−1</sup>, which makes them comparable to conventional liquid electrolytes.<sup>[269,270]</sup> Employing solid-state electrolytes also enables the safe application of Li metal anodes and the operation at high voltages.<sup>[270]</sup> Solid electrolytes are generally classified as:<sup>[271,272]</sup>

##### 1) Inorganic/Ceramic:

- Sulfides: for example, Li<sub>2</sub>S–GeS<sub>2</sub>–P<sub>2</sub>S<sub>5</sub>, Li<sub>10</sub>GeP<sub>2</sub>S<sub>12</sub>, Li<sub>6</sub>PS<sub>5</sub>X (X = Cl, Br, I), Li<sub>7</sub>P<sub>2.9</sub>Mn<sub>0.1</sub>S<sub>10.7</sub>I<sub>0.3</sub>
- Oxides: for example, Li<sub>7</sub>La<sub>3</sub>Zr<sub>2</sub>O<sub>12</sub>, lithium phosphorus oxynitride (LiPON), NaSICON type LiM<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub> (M = Ti, Ge, Zr, Hf), perovskite-type Li<sub>3</sub>xLa<sub>2/3</sub>–xTiO<sub>3</sub> (LLTO), etc.
- Phosphates: for example, LiTi<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>, LiGe<sub>2</sub>(PO<sub>4</sub>)<sub>3</sub>, γ-Li<sub>3</sub>PO<sub>4</sub>, LiPON, etc.
- Others: for example, LiBH<sub>4</sub>, LiBH<sub>4</sub>–LiX (X = Cl, Br, or I), LiBH<sub>4</sub>–LiNH<sub>2</sub>, Li<sub>3</sub>AlH<sub>6</sub>, Li<sub>6</sub>PS<sub>5</sub>X (X = Cl, Br, I), etc.

##### 2) Solid polymers:

- Polyether based
- Polyester based
- Nitrile based
- Polysiloxane based
- Polyurethane

In the future, several aspects of the recycling will be affected by solid electrolytes in spent batteries. There will be less safety issues related to the evaporation of the liquid electrolyte and the accumulation of harmful fumes. The formation of the corrosive hydrofluoric acid can also be eliminated. The mechanical pre-treatment can be expected to be easier since solid electrolytes have been reported to be very brittle. For example, sulfide-based SE are reported to be elastic and have plastic properties but display low fracture toughness compared to polymer and oxide electrolytes. Also oxide-based SE are in general very hard particles, resulting in brittle layers.<sup>[273]</sup> The brittle and low-porous media in ASSBs can be damaged when mechanical force is applied.<sup>[274]</sup>

More challenges can be expected during the mechanical separation if very small, brittle particles are formed during the crushing processes. Today battery components are more diverse and easier to be separated by conventional methods. However, if very fine particles are formed the black mass for hydrometallurgical treatment can be more homogeneous.

The most significant effects can be expected for hydrometallurgical treatment due to the diversification of the chemical compositions. Most challenging will be the recycling of the ASSBs using inorganic resp. ceramic electrolytes, which contain metals that are not present in current cell chemistries.

New elements such as Ge, Ti, Zr, Sn, or rare earths elements (REEs) will interfere with the solvent extraction process of current recycling technologies. For example, if the acidic extractant D2EHPA is used for Mn recovery, REEs and Zn will interfere with the extraction process.<sup>[275]</sup> However, some adjustment to the recycling processes can be made, and those metals can be recovered prior the extraction of Mn. The REEs, Al, Fe, and Zn can be separated from Co and Ni from an HCl leachate at pH 2 using 25% D2EHPA in kerosene.<sup>[276]</sup> From nitric acid leachates, Fe and Zr ions can also be extracted using D2EHPA and then stripped with oxalic acid.<sup>[277]</sup> Ti will also react with D2EHPA as it can be extracted from both hydrochloric and sulfuric media.<sup>[278]</sup> As D2EHPA is usually one of the first extractants used, more extraction stages will be required before the extraction of Mn, if current and future batteries are recycled together. Same phenomena can be expected for Cyanex 272 used for Co extraction. In case that “new” metal ions are present in the leachate after extraction with D2EHPA, they will affect the extraction of Co and Ni. For example Cyanex 272 shows a greater affinity for Ti over Fe at very lower solvent concentration.<sup>[279]</sup>

Another issue will be the presence of different anions in the system such as Cl<sup>-</sup>, B<sup>-</sup>, or F<sup>-</sup> as they also affect solvent extraction processes and the formation of organo-complexes.<sup>[277]</sup> However, this phenomena is not very well documented even for current recycling technologies.<sup>[131]</sup> Major challenges of future recycling will result from an extremely diverse chemical composition of batteries and low concentrations of valuable elements. Today the main motivation to recycle is based on the high Co/Ni/Li prices and government regulations (especially in EU). This should be considered while developing ASSBs for real applications and markets. In order to create incentives for recycling, a reasonable range of different metals in SE should be selected and concentrations of valuable metals should be sufficiently high to make recycling profitable.

## 4.2. Future Generation Battery Recycling

### 4.2.1. Design for Recycling

For a sustainable transformation of the energy industry and the transport sector, it is necessary to enable cost-efficient battery production. While production costs are expected to decrease in the medium to long term due to economies of scale, learning curves and technical developments, this should be accompanied by the establishment of a circular production chain.<sup>[280]</sup> In this context, battery recycling is in constant competition with suppliers of virgin battery materials, both in terms of prices and material performance.<sup>[281,282]</sup> Several publications have indicated that better separation of material streams at the beginning of recycling can lead to higher product purity and recycling efficiency, contributing to a more competitive battery recycling industry.<sup>[281–284]</sup> Recently, Thompson et al. evaluated the recycling costs for ten hydrometallurgical recycling processes from the literature and found that recycling based on shredded starting material offered cost savings of <20% for battery production compared to using virgin materials. In comparison, recycling processes based on disassembled batteries as starting material showed cost savings of 40–80%, without considering additional disassembly costs.<sup>[282]</sup>

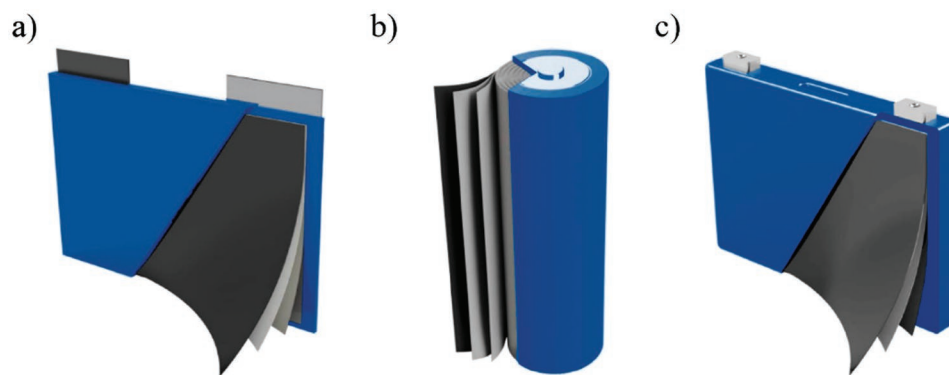
As of today, the development of an efficient battery disassembly on an industrial scale still faces several challenges. On the one hand, the current recycling network is not designed to process many different material streams, but rather focuses on treating a wide variety of feedstocks in a single process. This is especially true for pyrometallurgical recycling approaches, which have very low battery pre-treatment requirements but offer low recycling efficiencies and material values in comparison to hydrometallurgy. On the other hand, as the demand for energy storage continues to grow, more and more players are getting involved in the development, production, marketing and application of batteries, leading to the continuous development of new material concepts and cell designs. This leads to an increasing inhomogeneity and complexity of the feedstock for battery recycling, posing significant challenges to the applied processes.

An important concept to facilitate the development of an efficient circular battery economy is called design for recycling. The concept is based on considering the eventual treatment of the battery cells during recycling already in the design phase. The design principles are mainly focused on three levels, pack and module design, cell design, and material design.

Larger batteries (e.g., EV batteries) typically consist of many individual cells arranged in modules which are then assembled into a pack. The manual disassembly of such batteries is associated with high costs, long lead times and significant safety risks for workers.<sup>[196,285]</sup> There are several publications discussing the automation potential of battery disassembly.<sup>[285–288]</sup> For instance, Hellmuth et al. identified a 46-step disassembly sequence for a 2017 Chevrolet Bolt battery and found that about one-third of the steps could be automated based on an assessment of the technical feasibility and necessity of automation. At present, fully automated disassembly by self-learning robots remains unrealistic due to the high complexity of some disassembly steps. A more promising approach are hybrid systems in which human workers perform the disassembly together with automated robotic arms (human-robot collaboration).

Regarding battery design, the types of connections between the modules and the individual cells are of particular importance. These include, for example, threaded, screwed, bespoke, or push-fit connections, but also various types of welded joints.<sup>[289]</sup> While the mechanical connections are in principle easy to detach, the different sizes and shapes of the connections pose a challenge for both human workers and robots. Standardizing these connections could significantly reduce the amount of work involved in the disassembly by minimizing the number of tools and steps required. Permanent connections based on welds or adhesives, which are often used to assemble several cells into a module, are much more problematic for disassembly. In order to remove adhesives, organic solvents can be used, which increase costs and pose an additional safety risk to human workers.<sup>[282,290]</sup> Alternatively, adhered cells can be detached by heating. However, external heat exposure can increase the risk of thermal runaways.<sup>[291]</sup> Flexible cables pose another problem, especially for robotic disassembly, as they can be positioned in unpredictable ways reducing the robots' freedom of movement and adding complexity. A potential design alternative is a rigid busbar to which individual cells can be connected via mechanical connections to form a battery pack





**Figure 5.** Commonly used cell geometries for battery pack construction: a) Pouch cell, b) cylindrical cell, c) prismatic cell. Reproduced according to the terms of the CC-BY license.<sup>[293]</sup> Copyright 2020, The Authors, published by MDPI.

in a so-called cell-to-pack design. Such a design reduces the amount of adhesive and cables in the battery pack and simplifies disassembly into individual cells. A corresponding solution was presented by the Chinese battery manufacturer BYD with the so-called “blade battery.”

Disassembly at the cell level is even more challenging since manufacturers use different cell geometries, including prismatic, cylindrical, and pouch cells (**Figure 5**). The cell housings are usually permanently sealed by welding or crimping to reduce the risk of leakage. Built-in designs for an easy opening of the cells could significantly facilitate the processing. Features such as vacuum seals and screw caps could also facilitate recycling by maximizing the recovery of electrolyte, thereby improving the recovery of lithium and fluoride compounds.<sup>[292]</sup>

In order to separate electrodes and separator, the jellyrolls of prismatic and cylindrical cells must be unwound. Pouch cells contain stacks of electrodes, which must also be separated. Li et al. reported a disassembly system for pouch cells with Z-shaped folded electrode-separator compounds consisting of a vacuum conveyor, multiple pinch grippers, and skimmers.<sup>[294]</sup> The system allows automatic separation of anodes, cathodes, and separators and should also be adaptable for processing prismatic and cylindrical cells. In general, standardization of cell geometries would contribute to a simplified disassembly. However, such a severe restriction of design freedom would significantly limit future innovations in pack and module design, including new cooling and battery management systems.

Another central challenge for disassembly is the liberation of the active materials from the current collectors. A typical binder material for cathodes is polyvinylidene fluoride (PVDF), which has high electrochemical stability and good adhesive properties. However, PVDF only dissolves in very few solvents, one of which is *N*-methyl-2-pyrrolidone (NMP). The use of NMP as a solvent to separate the active materials is associated with high costs and represents a safety risk because it is hazardous, teratogenic, and irritating.<sup>[295]</sup> Recently, alternative methods for dissolving the PVDF binder, such as supercritical extraction with dimethyl sulfoxide (DMSO) as co-solvent<sup>[113]</sup> or the use of green solvents, such as dimethyl isosorbide (DMI)<sup>[296]</sup> have been published. Another chemical method for the liberation of the cathode active material is based on the selective etching of the aluminum current collector using alkaline solutions.<sup>[133]</sup>

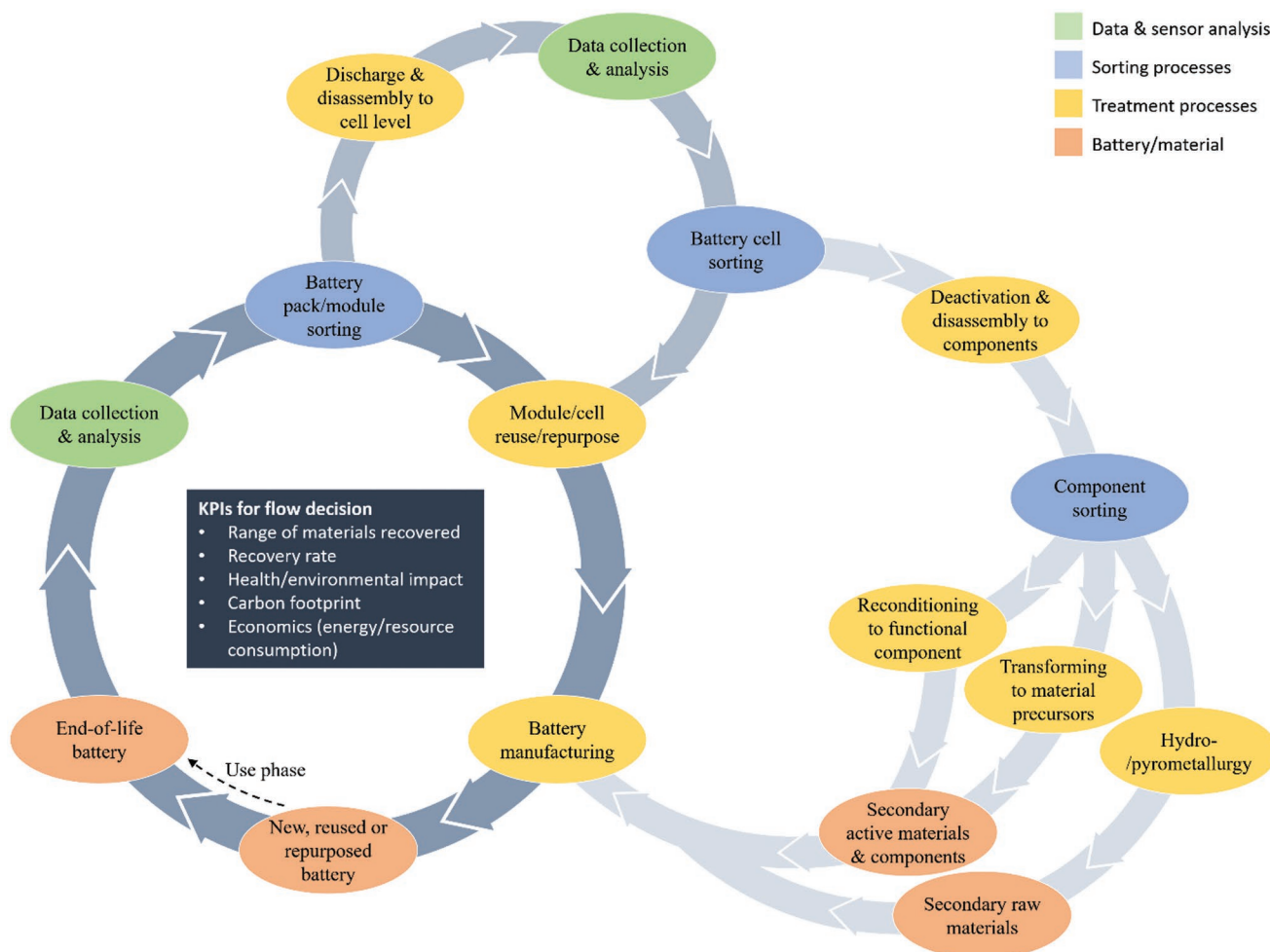
Currently, also high power ultrasonic pulses are being studied as technology to ease liberation of cathode materials, regardless of binder types.<sup>[297]</sup> The cavitation from the ultrasonic pulses separates the active material from the current collectors in cathodes. Other methods include thermal processing, high voltage treatment, or mechanical crushing.<sup>[111]</sup>

The issue of active material liberation can also be addressed from a material design perspective. The development of new binders that can be processed in aqueous solutions or other green solvents like ethanol is of particular interest. Various potential alternatives for PVDF have already been proposed, for example, polyacrylic acid (PAA), styrene-butadiene rubber (SBR), carboxymethylcellulose (CMC), polyethylene (PE) or chitosan. While CMC and SBR are already state of the art as binders for anodes, aqueous processing of cathodes is still a major constraint due to the high reactivity of commercially available cathode materials with water. The most promising results so far have been achieved for LFP cathode materials, which have comparatively low sensitivity to water.

#### 4.2.2. Direct Recycling

An effective disassembly of batteries also offers opportunities for the use of new recycling approaches. Especially the concept of direct recycling has received a lot of attention in recent years. The fundamental idea behind direct recycling is to refresh or reactivate active materials in order to restore the capacity and property losses through cycling, instead of first breaking them down into their individual components and then re-synthesizing them.

By using standardized cell designs and specific battery chemistries, the resetting of the lifecycle of batteries becomes a possibility via relithiation and direct recycling. As the battery ages, one of the most common indicators of aging is the loss of lithium from the layered oxide in the cathodes. After successful disassembly, the high purity cathode material can be exposed to high lithium moieties and resintered. A multitude of approaches have been explored, but the presence of remained impurities such as different rock salt phases on the material affects its final performance unless a higher temperature process is used.<sup>[298]</sup> A thermal reactivation method for LFP cathodes was presented by Li et al. who treated the cells



**Figure 6.** Proposed framework for the development of future recycling infrastructures.

at temperatures of 600–800 °C with the addition of a reducing gas and  $\text{Li}_2\text{CO}_3$  as Li precursor.<sup>[299]</sup> Similar approaches were also published elsewhere.<sup>[300,301]</sup> Another method based on hydrothermal relithiation was investigated by Sloop et al. and Shi et al. The cathode materials were treated in LiOH and  $\text{Li}_2\text{SO}_4$  solutions and then thermally annealed.<sup>[302,303]</sup> Electrochemical methods for relithiation were also investigated.<sup>[298]</sup> The methods include, for example, the treatment of LCO in an aqueous solution with LiOH and KOH with a small current of 1 mA cm<sup>-2</sup> applied.<sup>[250]</sup> In a different procedure, short high-voltage pulses were applied at a frequency of 150 Hz.<sup>[304]</sup> It is essential to remember that the works on direct relithiation synergize with a disassembly approach and its efficiency suffers when coupled with the current shredding approach in commercial recycling.

More recently, there has been an uptick on the production of batteries with recycled materials. Ma et al. have published the effects of their recycling process on the performance of recycled batteries.<sup>[305]</sup> While the details of their leaching process are not public yet, it is important to notice how the recycling process affected the final morphology of the cathode material. The changes in structure are assigned as one of the main drivers on the improved performance of the material.

To the best of our knowledge, no research has been published on the “upcycling” of battery cathodes from high to low cobalt chemistries. Nevertheless, changes in battery chemistry such as cobalt-free cells or sodium-based cells are promising within the design for recycling approach. The recycling of such cells is still a new topic with very limited studies such as a recent report of a fully recyclable Na-ion battery using  $\text{Na}_3\text{V}_2(\text{PO}_4)_3$  as the cathode material.<sup>[306]</sup> The cells have an innovative design of stacked electrodes that uses Al foil as a common current collector between anodes and cathodes. These batteries can be separated with aqueous based solutions with NaOH produced as a by-product. Such designs are ideal conditions in design for recycling, but the overall electrochemical capacity and stability is still missing significant improvements to properly address the needs of the market. Some of these limitations are being addressed by the design of new sodium battery chemistries that are more environmentally benign from the start such as: new fluorine free electrolytes,<sup>[307]</sup> and biomass based hard carbons<sup>[308]</sup> among other innovations. Nevertheless, the constantly changing battery compositions are a major limitation for direct recycling, because the reactivation of cathode materials requires a single-type material input. At the same time, direct recycling can

offer advantages especially for the recycling of cathode materials with low intrinsic material value, such as LFP or LMO. The prerequisite for a practical application of direct recycling will therefore be an efficient sorting of battery types based on a convenient battery labelling.

#### 4.2.3. Future Battery Recycling Infrastructure

As a result of the above discussion of the regulatory framework, available recycling methods, and future developments, we propose the recycling flowchart shown in **Figure 6** as a foundation for the development of a future recycling infrastructure.

The framework is based on three interconnecting cycles. In the first cycle, newly produced batteries first go through a use phase. The resulting end-of-life batteries are sorted and can either be used as a module in second-life applications or are broken down to the cell level in the second cycle. The individual cells are then sorted again and, according to their condition, can be directly reused or end up in the final recycling cycle. Here the cells are first deactivated and disassembled. The cell components can then be converted into secondary active materials through direct recycling or into secondary raw materials for battery production through classical recycling approaches. An important component of the framework is the data collection and analysis, which allows the determination of flow directions within the system based on several key performance indicators (KPIs), including the range of materials recovered, recovery rates, health and environmental impact, carbon footprint, and economics.

## 5. Conclusions

The recycling of Li ion batteries is an emerging field that will likely undergo severe changes as the process updates itself to fix the different challenges presented in this review. In the early stages due to the mix of chemistries and traceability issues, hydro and pyrometallurgy offer the best routes for the recovery of the metals of interest. Hydrometallurgy has recovery rates over 99% for Ni, Co, Mn, and can handle the mixed waste streams being produced at present. The precursors produced via hydrometallurgy will be partially reintroduced into the market mixed into new battery components. Alternative approaches to hydrometallurgy such as solvometallurgy need to be tested beyond the laboratory scale. Their performance on the increasingly complex metal matrices and tighter regulations on water consumption will be the most likely drivers for their adoption. Newer approaches like direct recycling are highly dependent on the efficient sorting of battery types based on a convenient battery labelling with regard to the cell chemistry. For Li-metal and Li-S batteries, the reactivity of the materials and side reactions will bring up some additional safety concerns during recycling. Less safety concerns need to be addressed when dealing with ASSBs. However, mechanical handling and hydrometallurgy will be more difficult compared to current state of the art batteries.

As traceability and collection schemes are refined, the separation of metals prior to the actual recycling is likely to become more important because companies will shift toward direct

recycling approaches having lower environmental footprints. This transition will be highly dependent on a stabilization of battery chemistries and an increased ability to recover electrolytes.

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## Conflict of Interest

The authors declare no conflict of interest.

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