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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.

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, the lithium-ion battery (LIB), as an established technology, is a promising candidate for the energy-storage of the future.[1] 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.[2–4] 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.[5,6] Currently, the market is dominated by $LiPF_6$-based organic solvent electrolytes due to the excellent properties of $LiPF_6$ regarding ion conductivity, supporting solid electrolyte interphase (SEI) formation at the anode, and protection of the Al current collector at cathode.[7–12] Additionally, the state-of-the-art LIB cell consists of a graphite anode, and in most cases a layered lithium metal oxide ($LiMO_2$, M =, e.g., Co, Ni, Mn, Al) with varying metal contents or a lithium transition metal phosphate as a cathode.[13–19]

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.[20] 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.[21–26] 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[27,28] 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.[29,30].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.[31] 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.[32–35] Furthermore, in contrast to the lead acid battery, only high amounts of metals such as nickel or cobalt provide financial viability for LIB recycling.[36] And last, so far no standardizations for cell designs of LIBs exists.[37]

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.[32,38,39] 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.[40,41]

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.[42,43] Physical responsibility refers to ensuring the treatment of waste products, including collection, transport, sorting, reuse, recycling, and disposal.[42] These tasks can usually be delegated to third parties.[42–44] 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.[42,43] 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.[44,45]

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).[45] 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.[46]

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.[44]

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 (SSLA) 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.[47] The Universal Waste Rule prohibits the disposal of certain hazardous wastes and sets standards for the collection, storage, and transportation of these wastes.[48]

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.[49–52] 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.[53] However, until the 2010s, there was a significant lack of regulation governing the collection and treatment of waste LIBs.[42,53] 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.[42,53]

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.[54–57] Three of the main markets for LIBs are consumer electronics, stationary battery energy storage (SBES), and EVs.[55,58,59] 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.[58–60] 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.[61]

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.[62] 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.[63,64] 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.[60,65]. Currently, only a small portion of the electronic waste generated each year is collected and properly recycled.[66] 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.[66] 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.[54,57] 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 $LiCoO_2$ (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