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Commentary

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Better Batteries – Better Recycling?

1 Introduction

Lithium ion batteries (LIBs) are used in modern day portable consumer electronics like laptops, smartphones or tablets due to their high energy density and high specific energy. Furthermore, as the most interesting battery technology for pure and hybrid electric vehicles, they offer a widespread applicability for private and industrial processes. In addition, the increasing energy consumption, due to the world population growth and the depletion of fossil-fuel resources lead to a strong demand for more renewable energy sources. However, many renewable energy sources produce electricity very unsteadily. To compensate these fluctuations, energy storage solutions are needed which is another important application of LIBs. Overall, this is directly related to the recycling of LIBs and the respective components and materials. However, the recycling of lithium ion batteries is still under development and has not reached its full potential so far. The complexity of the batteries with varying chemistries interferes with the development of a single robust recycling procedure for all kinds of LIBs. Furthermore, the next generations of batteries will further increase the diversity of cell chemistries and components. Therefore, the current processes and technologies needs to be further investigated and adapted to handle the upcoming stream of new batteries.

2 Current State of the Art – Regulations and Economic Aspects

Legislative and economic aspects mainly drive the recycling of LIBs. The legislative aspect is encouraged on the one hand due to hazardous battery components that can pose a threat for the environment and human health if released. On the other hand, mass homogenization for a functioning European market, exclusion of market distortions as well as independence from mining sites and countries is also of great importance. Present regulations include the *Battery Directive* (Directive 2006/66/EC) and the *Waste Electrical and Electronic Equipment* (WEEE) *Directive* (Directive 2012/19/EU). These regulations are precisely defined targets for collection rates and recycling efficiencies. The directives also set disposal responsibilities and safety requirements. Therefore, government authorities can and should contribute to the establishment of an effective circular economy. The Extended Producer Responsibility (EPR), an important concept for recycling, is also defined by these guidelines. Due to the EPR, the physical (e.g. collection, handling and recycling) and financial responsibilities (e.g. internalization of the related cost and incorporation to the prices) are distinguished and assigned for the treatment of spent LIBs. According to the EPR, the costs for collection, treatment, recycling and disposal must be financed by the battery producers. Furthermore, they are obligated to take back portable, automotive and industrial batteries free of charge. Industrial, automotive and collected portable waste batteries must undergo a treatment and recycling using the best available techniques to protect health and the environment before residual compounds can be landfilled or incinerated. The directives also 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 (Neumann et al., 2022). These directives were evaluated regarding their effectiveness showing that only 14 member states could achieve the 45 % goal in 2016 and over 50 % of the portables batteries were not collected in

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the mentioned period. However, the targets were considered insufficient, since only portable batteries were considered and the varying lifetimes for different battery types were neglected. In 2020, as a part of the green deal of the EU, a replacement of the Battery Directive was proposed. New collection targets for portable batteries, which now include light means of transport e.g. E—bikes/E-scooters, were set: 45 % by 2023, 65 % by 2025 and 70 % by 2030. Nevertheless, targets for LIBs from EV are still missing; however, the legal framework was established. The overall weight of the recycled LIBs was raised as well as single targets were set for several materials, including lithium. This refers directly to the economic part of the recycling due to the price of the different constituents. The transition metals, but also lithium and graphite are considered bottleneck materials for LIB production. However, graphite is so far not included in the target materials and should be mandatory implemented in further revisions of the Directive. In average, state-of-the-art high-energy LIBs normally contain 5 %–20 % cobalt (Co), 5 %–10 % nickel (Ni), 5 %–7 % lithium (Li), 5 %–10 % other metals (copper (Cu), aluminium (Al), iron (Fe), etc.), 15 % organic compounds, and 7 % plastic (Ordonez et al., 2016). To maximize the overall LIB recycling efficiency the recovery of none of these materials can be neglected. In addition, the European Commission should include a uniform labelling 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. Furthermore, the heterogeneity of battery types available on the market could be managed by implementing a battery passport which should provide all necessary information (Neumann et al., 2022). To overcome the issues, the legislative should not only include targets but also encourage to achieve them by establishing even a system of penalties in the worst case.

3 New Chemistries, new Challenges

Overall, recycling processes are the only option to re-introduce spent batteries and their components into the economic cycle, reducing the need for primary raw materials and promoting an improved acceptance of pure and hybrid electric vehicles and other battery electric transportation applications. However, a LIB is composed of several components: typically a graphite based anode, a lithium transition metal oxide cathode and an electrolyte soaked polyolefin-based separator that is placed between anode and cathode. Furthermore, while around 2005 only one cell chemistry was applied (lithium cobalt oxide, LCO), nowadays several different cathode materials such as varying stoichiometries of lithium nickel manganese cobalt oxides (NMC) or materials like lithium nickel cobalt aluminum oxides (NCA) or lithium iron phosphate (LFP) are applied. Additionally, mixed streams of materials are caused due to the differences in the lifetime depending on the application, e.g. cell phones 2 years, other consumer electronics 3-4 years and electric vehicles 10 years or more. This complexity and diversity offers quite a challenge for the recycling of LIBs. Nevertheless, recycling processes are already applied to handle the rising stream of spent cells. Nowadays, mostly pyro- and hydrometallurgical processes, or a combination of both, are established to deal with the current cell chemistries. However, so far, a completely closed loop was not achieved. A major obstacle to a completely closed loop are the high requirements for the purity of battery materials. After recycling, LIB materials are often used for other applications not requiring very high purities. In addition to this so-called downcycling, the recovery of low-cost battery materials is another obstacle for high recycling rates in a closed loop. Components like the binder, the anode or the electrolyte which contains for example lithium, are mostly not recovered but have recently gathered more attention. With this in mind, the development of future generation materials will intensify this situation. In comparison to the numerous reports in literature about the recycling of LIBs (whether as a pilot process or new processes for single components), nearly no reports or industrial activities can be found with regard to the upcoming cell chemistries. Therefore, an early consideration of possible recycling methods for these types of upcoming batteries is important. One development, the application of Li-metal electrodes will introduce much higher contents of lithium into the system so that much higher recycling efficiencies for lithium will be needed. During pyrometallurgy, lithium normally ends up in a slag, so refinement by hydrometallurgy will gain more and more importance. But because of the higher energy densities and reactivity, another challenge not only for the actual recycling but also for the handling, transportation and storage of spent Li-metal batteries arises. In this regard, a deactivation of the Li-metal batteries as an early step of the recycling procedure, e.g. by extraction of the metallic lithium would increase safety and it would allow for a subsequent transportation of the deactivated batteries.

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Especially in case of damaged batteries with an unclear hazard potential, long transport routes should be avoided, which could be achieved by a decentralized distribution of small deactivation facilities. A promising candidate for a future battery generation is the lithium sulfur battery. For the recycling of lithium sulfur batteries, most of the technical challenges are attributed to their metallic lithium anodes. However, toxic gases like H2S can be formed when the compounds inside a lithium sulfur battery get in contact with moisture, which can further complicate the recycling of future batteries. In comparison, less safety concerns need to be addressed when dealing with all-solid-state batteries (ASSBs). However, mechanical handling will be more difficult compared to current state-of-the-art batteries. Furthermore, due to the introduction of new chemical compositions the hydrometallurgy will be affected as well. There is also a variety of non-Li chemistries including batteries based on sodium, zinc, magnesium or calcium currently investigated. The main motivation is the development of new batteries based on naturally abundant elements. Among those non-Li batteries, sodium-ion battery technology is most similar to commercial LIBs. From a recycling point of view, however, battery chemistries with low-cost elements such as sodium or sulfur are accompanied with little economical interest for the recycling industry. Therefore, the recycling of the new batteries in general needs to be supported by legislation with specific regulations for the efficiency of the new processes.

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