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Holistic Research for Lithium-Ion Battery Recycling as Basis for a Sustainable Industrial Business

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An understanding of the global climate and technological progress are driving e-mobility forward worldwide. As the production of electric cars and mobile devices increases, so does the demand for rechargeable batteries. As a result, the demand for rare elements is also rising sharply, leading to a shortage and increase in the price of raw materials. For cost reasons, but above all in view of the waste streams generated, a shift towards e-mobility is therefore only possible if batteries can be safely recycled on an industrial scale, thus enabling a circular and sustainable economy [1].

For a successful and long-term existence in the market, the "big five" factors for a sustainable industrial business in lithium-ion battery (LIB) recycling should be taken into account (see Figure 1). As a result, the company structure is an important factor and requires continuous research and development activities, a highly flexible concept, a broad expertise base and a logistically optimized location in order to be able to react to further changes and challenges. As LIBs pose a great potential risk in recycling, the issue of safety is also important: problematic waste must be managed and safe storage ensured. Due to the risk of explosion, handling damaged cells is also problematic. Components that are harmful to health require an elaborate safety concept for all employees. In terms of profitability, a good knowledge of the market and ideally a scrap supply on a contract basis are required in order to remain marketable in the long term. Quality management and a profitable cost structure are also essential. Environmental friendliness is essential when processing critical materials. Optimized life cycle assessment performance, low heavy metal emissions and a low consumption of additives are helpful here [2]. The most important factor is to ensure compliance with EU regulations. To this end, at least 65% of the total mass and more than 50% lithium as well as more than 90% nickel, cobalt, and manganese must be recovered from year 2028, raising to 80% lithium and 95% nickel, cobalt, and manganese from 2032 [3].

The five must-haves illustrate how complex and extensive the necessary conditions are and that extensive research is essential along the way. This special issue provides an insight into current research work in various areas of the battery recycling process. All sub-areas of the entire process are addressed (see Table 1). In addition, a wide range of battery types is covered (see Table 2). Starting with a battery cell, depending on the process, it has to be crushed at first and fractionated in a mechanical pre-treatment. In this context, Kaas et al. investigated a zig-zag air classifier, which is used for the separation of current conductor foils of different types of batteries (Contribution 1). Richter et al. used different reflectance sensors with visible to long-wave infrared to identify for sensor based sorting of components (Contribution 2). The influence of different types of batteries on the typical steps of mechanical pretreatment was analyzed by Wilke et al. (Contribution 3).



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Figure 1. The "big five" factors for a sustainable industrial business in lithium-ion battery recycling [2].

Additionally to mechanical pretreatment, cells need a thermal pretreatment to remove organics and to crack several compounds. For this, Stallmeister et al. examined the influence of process parameter of thermal pretreatment on NMC 622 battery black mass (Contribution 4). The focus of Biswas et al. was on the sulfuric roasting of LCO rich blackmass (Contribution 5). Schwich et al. took a look on thermal pretreatment as a part of a safe process for li recovery of LiS battery cells (Contribution 6), while Qui et al. investigates the influence of the pretreatment on graphite and cathode material of NMC 622 blackmass (Contribution 7). Balachandra et al. varied in their studies the temperatures of pyrolysis and incinerations by using NMC 111 (Contribution 8), whereas Kuzuhara et al. did this for calcination of LCO material to see the influence on following processes (Contribution 9).

The two possible pretreatments are usually followed by hydrometallurgical treatment, which is highly represented in this special issue. The great variability of hydrometallurgy starts with optimization of already evaluated process steps. For example, Munchen et al. set the focus on raising the leaching efficiencies of Li and F (Contribution 10), while Gerold et al. look for a solution by analyzing parameters to prevent silicon based process-disrupting viscosity (Contribution 11). Due to the environmental aspect, a further point is alternative leaching agents. This brings organic acids into focus. Here, Schmitz et al. used organic leaching solution to recover Co, Ni, and Mn of NMC 111 pouch cells (Contribution 12), whereas Lerchbammer et al. try to recover the same elements of NMC 622 batteries by using gluconic acid (Contribution 13). With the intention to create an environmental friendly process, Zorin et al. applied organic acids to select black mass of LMO and NCA batteries (Contribution 14). Sahu et al. analyzed the use of acetic acid to recover valuable materials of LCO batteries (Contribution 15). As an alternative to the classic batteries, Schneider et al. have investigated LLZO based SSB and tested various leaching reagents (Contribution 16). In addition to leaching, there are also other processes that can be investigated: For example, Prasetyo et al. focusses the isolation of Cu, Co, Mn, and Ni of black mass by using solvent extraction and precipitation (Contribution 17), while the work of Kutzer-Schulze et al. deals with the separate electrochemical recovery of transition metals from black liquor of batteries (Contribution 18).

A combination of hydrometallurgy and pyrometallurgy was core of the investigation of Rinne et al. They selected electrode particles with a froth flotation and used them in high-temperature Cu-slag reduction (Contribution 19). In contrast to hydrometallurgy, pyrometallurgy can also be carried out completely without pretreatment. This is how Holzer et al. developed a novel reactor for a pyrometallurgical process to recover Li and P of various types of batteries in the gas phase (Contribution 20). Beside all parts of metallurgical battery recycling, simulation and thermodynamically investigations bring

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detailed insights of elemental behavior in the different processes. For example, Luo et al. examined the volatilization of valuable metals like Ni, Co, Mn, Li, Al, and Cu of LIBs by vacuum chlorination (Contribution 21).

Additionally, this special issue also includes three reviews. Petzold et al. provides an insight into the use of LIBs as a part of a sustainable energy solution with a focus on presorting and its optimization potential (Contribution 22). Going back to the hydrometallurgy, Marcinov et al. thematize the recycling methods of LIBs, the losses in this steps and possibilities to fix them (Contribution 23). In the last review, Botelho et al. give a critical insight in Co-recovery of LIBs (Contribution 24).

Table 1. Articles in the different fields of sience.

| Flied of Research | Source |
|-------------------------|---------------------------|
| Mechanical Pretreatment | (Contributions 1–3) |
| Thermal Pretreatment | (Contributions 4–9) |
| Hydrometallurgy | (Contributions 5–18) |
| Pyrometallurgy | (Contributions 19 and 20) |
| Simulation | (Contribution 21) |
| Review | (Contributions 22–24) |

Table 2. Treated types of batteries and their source.

| Type of Battery | Source |
|-----------------|---|
| NMC | (Contributions 1, 3, 4, 7, 8, 11–13, 20, 22 and 24) |
| NCA | (Contributions 1, 3, 14, 20, 22 and 24) |
| LFP | (Contributions 1, 3, 20, 22 and 24) |
| LCO | (Contributions 3, 5, 9, 15, 19, 20, 22 and 24) |
| LMO | (Contributions 3, 14, 22 and 24) |
| LiS | (Contribution 6) |
| SSB/LLZO | (Contribution 16) |

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Abbreviations

The following abbreviations are used in this manuscript:

| LCO | Lithium cobalt oxide |
|------|--|
| LFP | Lithium iron phosphate |
| LIB | Lithium-ion battery |
| LiS | Lithium-sulfur |
| LLZO | Lithium lanthanum zirconium oxide |
| LMO | Lithium manganese oxide |
| MDPI | Multidisciplinary Digital Publishing Institute |
| NCA | Lithium nickel cobalt aluminium oxide |
| NMC | Nickel cobalt manganese |
| SSB | Solid-state battery |

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