Recycling of Lithium-Ion Batteries – the UPRAISE Project

Franziska Klein-Punt[1,*], Harald Zetzener[1] and Arno Kwade[1]

[1]Institute for particle technology, TU Braunschweig, Germany

Abstract. The rapid growth in the production of electric vehicles and the stricter European Union (EU) recycling targets of 65 % (2025) and 70 % (2030) mean that end-of-life and production scrap from lithium-ion batteries are becoming an important source of secondary raw materials. In order to achieve these recycling targets in hydrometallurgy, efficient mechanical pre-treatment is particularly important. This work focuses on optimising mechanical pre-treatment using a combined vacuum grinding and drying unit. The material is dried in a mixing vessel, where the mixing blade applies additional stress to the shredded material, in particular the electrode foils. By optimising the stress intensity on the electrodes, the recovery rate of the black mass can be maximised whilst keeping impurities to a minimum. The aim of the UPRAISE project is to achieve black mass impurity levels of less than 3 wt %.

1. Introduction

The importance of recycling lithium-ion batteries is growing as the popularity of electric vehicles increases, production at the Gigafactories expands and regulations affecting the recycling industry become increasingly stringent due to the demand for greater independence from global raw materials markets. End-of-life (EoL) batteries and manufacturing waste now represent a major secondary supply of critical raw materials such as lithium, cobalt, and nickel.

A variety of recycling methods are already employed in industry, and for end-of-life batteries the valuable metals can be reclaimed through mechanical, hydrometallurgical or pyrometallurgical routes. The present study focuses on the mechanical pathway, which comprises a series of comminution and separation operations. A representative example is the LithoRec process [1] (see Fig. 1). The key stages are:

- Inert shredding – cells or modules are shredded in a nitrogen-purged chamber to suppress ignition
- Drying of the shredded fragments
- Mixing to increase the delamination of the electrodes
- First air classification – a zig-zag sifter removes heavy components (e.g., casings)
- A second grinding stage can be carried out to increase the black mass yield
- Sieving – Separation of the black mass fraction
- Second air classification – Separate the electrode foils from the separator

While this mechanical chain efficiently recovers the bulk of valuable materials, it shares the typical limitations of conventional mechanical recycling like high inert-gas consumption and a black-mass product that remains contaminated with copper and aluminum. Thus, the described mechanical process provides a practical pathway for extracting critical raw materials from EoL batteries, but further optimization - particularly in electrolyte recovery and contaminant reduction - is required to meet the European Union's (EU) expanding recycling demand.

The UPRAISE project will provide a detailed understanding of the interdependencies between the individual stages of the recycling and recovery processes – from diagnosis and safe dismantling, through the processing of the black mass, to the production of high-performance battery materials. Within this project the mechanical pre-treatment of NMC- and LFP batteries will be investigated regarding black mass yield and purity, electrolyte recovery and energy consumption. The EU has set targets for overall recycling rates of 65% by the end of 2025 and 70% by the end of 2030. The objective of the UPRAISE project is to achieve an overall recycling rate of 75% for NMC processes and 77% in the LFP process.

Figure 1. Mechanical process chain LithoRec [1]

2. Methods and Objectives

The pre-treatment concept in UPRAISE aims to generate a high-purity black-mass from NMC and LFP batteries while lowering the environmental footprint. In conventional mechanical recycling the entire module is shredded and then dried whereby the volatile electrolyte solvents are often completely burned off. The yield of black-mass commonly contains Cu- and Al-impurities. In contrast, the approach which is used in UPRAISE employs a rapid, vacuum-based shredding and drying stage that separates the shredded materials from the

* Corresponding author: franziska.punt@tu-braunschweig.de

volatile fraction in a single sealed chamber. The aim of process optimization in UPRAISE is to reduce the black mass impurities (Al and Cu) to < 3 wt %, increase the black-mass yield by roughly 15 % through optimized delamination from the electrode foils, and increase the recovery of the volatile electrolyte components by up to 80 % compared to non-vacuum processes.

For LFP batteries the process is refined further: crushing is performed on electrode stacks alone, leaving the remaining module parts, which can account for half the module weight, out of the feed stream. This selective comminution shall enable the production of a LFP black mass fraction with about 90 % purity.

In order to optimize the alignment of the mechanical pre-treatment with the subsequent thermal and hydrometallurgical processes, flow diagrams are first developed to define the key input and output parameters required for specific process steps. Therefore, cell analyses of the used modules are carried out first to determine the input composition; during the process, the recovered material fractions are analyzed and, where necessary, the processes are adjusted and optimized accordingly to ensure that the target parameters are achieved.

The mechanical pre-treatment process starts with the shredding of the discharged and short-circuited battery modules or electrode stacks to separate the individual components of the batteries. The use of an integrated vacuum shredding and drying unit offers a simultaneous fragmentation and drying of the modules under vacuum in a sealed chamber. This results in a reduced process time and energy consumption. In this process the shredded material falls into the mixing vessel where it is dried whilst being mixed. In this combined pre-module (Fig. 2), the volatile electrolytes are evaporated and condensed, which delivers a separate liquid stream with an efficient electrolyte recovery of 90–96 %. The use of a vacuum drying process also reduces the inert gas demand. Only a small, continuous nitrogen purge is required to protect the vacuum pump and manage vapors. This dramatically lowering nitrogen consumption and cost.

This combined pre-module also improves the separation and purity of the black mass. Real-time drying combined with optimized mechanical parameters (stress intensity) produces a black-mass with far fewer contaminants. The stress applied to the shredded material in the mixer ensures that, during this drying step, further stress is applied to the coated electrode foils thereby increasing the detachment of the coating from the foils. A high recovery rate of the black mass is particularly important here, as this is where the valuable components are found.

Figure 2. Used pilot plant, with pre-module (left) and product module (right)

A deeper understanding of the process is intended to identify the optimum stress conditions for comminution and mixing. Among other things, the kinetics of the detachment of the black mass from the electrode foils are to be investigated. The stress intensity must be sufficient to detach the coatings from the foils, but not so high that the foils themselves are further comminuted to sizes that fall within the size range of the black mass. Another factor in the mixing step is the evaporation of the electrolyte; it must therefore also be ensured that the material is thoroughly mixed to facilitate complete drying. Furthermore, the mixing time must be sufficiently long to guarantee complete drying. Preliminary investigations have already shown that a large proportion of the black mass is detached after a relatively short mixing time, and that further mixing only leads to the formation of additional impurities. Therefore, various requirements must be balanced against one another in this process step.

Following the pre-module, the various battery materials can then be separated from one another through subsequent separation steps in the product module (Fig. 2). The complex material system, comprising hard/compact materials, foil-like components, powders and liquid, makes this process especially complex. Once at least the volatile battery components have been removed from the shredded material in the mixer, the heavy components – such as parts of the module casing – are then separated using a zig-zag sifter. The remaining material is then passed through an air-jet tumbling sieve to separate the valuable black mass. A 500 µm sieve is planned to be used for this purpose. The use of an additional 2 mm sieve results in a total of three fractions. The black mass fraction (< 500µm) is then analyzed using XRF or ICP to determine the proportion of impurities, which mainly originate from crushed Cu and Al components. The aim is to achieve the highest possible yield of the black mass present in the modules, with the lowest possible level of impurities (< 3%).

In addition to the black mass, the copper and aluminum electrode foils are also valuable materials for which recovery is worthwhile. This recovery can be achieved, for example, using a zig-zag sifter. Once the heavy components, as well as the electrolyte and the black mass, have been separated, the remaining shredded material consists mainly of copper, aluminum and separator foils. As the separator foils have a very low density, it is possible to separate them using a second sifting process. This as well requires appropriate parameter studies to be carried out to achieve the best possible separation of the materials for the given material system, in accordance with the modules used.

3. Summary

In order to recover as much of the critical raw materials from a battery as possible during battery recycling – and thereby also meet EU targets – efficient mechanical pre-treatment is especially important. The used mechanical pre-treatment consists of several comminution and separations steps like a combined shredder and mixer pre-module with a single vacuum unit. This offers a number of advantages in this respect such as the recovery of the electrolyte.

Key challenges in the process are determining the optimum energy input to detach the coating, as well as handling and separation of this complex material system, which consists of compact materials, foils, powders and liquid. If the electrode material does not receive sufficient stress, the black mass does not detach completely from the electrode foils; if the stress is too high, foil material is fragmented and contaminates the resulting black mass. The objective in UPRAISE is to produce a black mass with a contamination level of less than 3 wt %.

Through a cascade of different separation steps, it is possible to recover not only the electrolyte and the valuable black mass, but also the copper and aluminium components of the batteries in order to achieve a high overall recycling rate.

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