Development of a robust resynthesis process for cathode active material (NCM-type) from the leachate of the COOL-process

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As a result of the energy transition, the demand for lithium as well as cobalt and nickel has been growing strongly in recent years due to a rapid increase in production capacities for lithium-ion batteries (LIB) as a key energy storage technology. According to a forecast, the EU-wide annual demand for lithium, cobalt and nickel for the production of lithium-ion batteries in 2030 will be 38,000 tons, 34,000 tons and 112,000 tons, respectively. A significant increase compared to 2018 where it was 2,000 tons (Li), 2,000 tons (Co) and 6,000 tons (Ni), respectively. (Cornet et al. 2019) The extraction and supply of the required raw materials is associated with a considerable ecological footprint due to the high water and energy consumption as well as the long transport routes.(Arshad et al. 2022) Therefore, the recycling of these raw materials is becoming increasingly important.

In the hydrometallurgical recycling process the discharged battery cells are being dismantled, crushed and leached. A promising approach for the leaching step is the so-called COOL process (Fig. 1), in which lithium is selectively extracted from the black mass and precipitated as a lithium carbonate. Subsequently, the lithium-free active material can be leached and hydrometallurgical refined to remove contaminations and recover the corresponding metal salts needed for the resynthesis of new usable cathode active materials (e.g., $LiNi_{0,6}Co_{0,2}Mn_{0,2}O_2$ known as NCM-622).

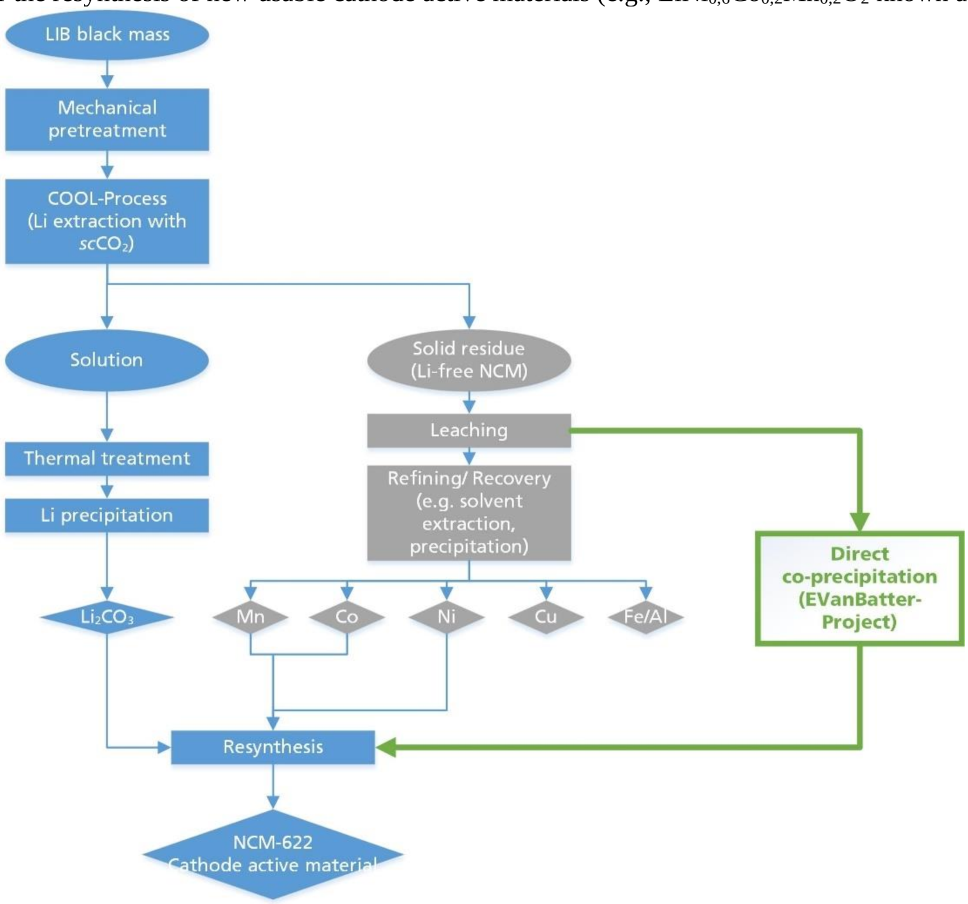

Figure 1: Schematic overview of hydrometallurgical black mass recycling using the COOL process to obtain lithium carbonate, the process of metal recovery via leaching and refining (grey) and the alternative approach being investigated in the EVanBatter project (green).

One of the goals is to develop a stable, robust resynthesis process that tolerates different reactant qualities. The focus lies on elements such as aluminum, iron, copper and other ions that originate from battery casings, current collectors or electronic circuits and are often present in the leaching solutions of hydrometallurgical battery recycling as impurities. These impurities may indeed be incorporated during the coprecipitation of NCM precursors leading to an irreversible contamination in the cathode active material. The effects however might vary. The impurities can have positive and negative effects on the active materials depending on the impurity itself and on its content in the NCM structure. Small contents of Fe^{3+} may improve the electro chemical performance due to increased lattice volume in the contaminated NCM allowing better (de)lithiation. The lithium (de)intercalation becomes less reversible if the Fe^{3+} content increases further leading to lower capacities. (Park et al. 2019)

In order to understand the influence of different impurity ions on the performance of resynthesized active materials, CAMs were prepared from both a synthetic and a real leaching solution from hydrometallurgical processing of lithium free black mass (product after COOL-process (Pavón et al. 2021)). Synthetic solutions of nickel sulfate, cobalt sulfate and manganese sulfate as the basic ingredients for NCM were prepared and contaminated with up to 1wt.% other metal salts like aluminum sulfate, iron sulfate and copper sulfate, respectively. Then, coprecipitation was carried out using sodium hydroxide and ammonia as chelating agent. The precursors obtained were subsequently mixed with lithium carbonate and calcined, resulting in the desired NCM-622 active material. The synthesized active materials were characterized with SEM to study the morphology and particle size. Particle size distribution was determined by using laser diffraction. EDX, XRF and XRD were used to determine the composition and purity of the calcined active material. Finally, the electrochemical performance of electrodes prepared with resynthesized materials was characterized and compared with cathodes made of pure NCM-622 active material.

With an in-depth understanding of the influence of impurities on the electrochemical properties of the cathode active material, it will be possible to optimize the recycling process in terms of process economy and ecology by adjusting the required purification steps. The first results of these investigations will be presented in the conference contribution.

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