Selective recovery of Li from leaching solution coming from the processing of Lithium-Ion Batteries (ULiège)

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Abstract

Due to its high consumption and unequal distribution of primary lithium resources such as minerals and brines, lithium, a key component in LIBs, is experiencing an economic and supply crisis. Thus, it has been added to the EU critical raw material list since 2020. And a target of 50% lithium recycling from LIBs is set by the end of 2027 in Directive 2006/66/EC. This research aims to develop an economical and environmentally friendly process for recycling lithium from the leachate after LIB recycling. It will benefit in meeting the EU lithium recycling target, industrial growth, sustainability in the lithium value chain, the circular economy, and industrial wastewater regulations.

In the methodology, crystallization experiments were carried out by cooling the Li solution at 1 °C for 20 to 48 hours to remove Na impurities. Moreover, chemical precipitation experiments were conducted to precipitate Li_2CO_3. The pH of the solution was adjusted with NaOH and heated to the desired temperature, followed by the saturated sodium carbonate addition. The reaction time was 60 minutes to reach equilibrium. The slurry was filtered, and the wet precipitates were heated at 100 °C for 1 hour. Moreover, the same methodology was applied to the solution with chloride chemistry after ion exchange resin.

From the results, crystallization removed 49.67% Na from the Li solution in 20 hours. In chemical precipitation, temperature, pH value, Li concentration, and CO3/Li+ ratio are observed to be influential parameters on recovery and grade. The Li recovery of 80.91% with 96.40% Li_2CO_3 purity was obtained at 95 °C, CO3/Li+ 1.075, and 10.45 g/L Li concentration after crystallization. The purity can be improved to 99.56% by repulping precipitates with hot water with minimal loss of Li recovery and less than 0.42% Na content. On the other hand, the solution after ion exchange resin obtained 80.44% recovery at 70 °C, CO3/Li+ 1.0, and 8.32 g/L Li with 99.40% purity without repulping and 0.26% Na. Thus, the solution with less Na impurity and chloride chemistry needs less temperature to precipitate lithium carbonate.

Nevertheless, during the economic analysis, the ion exchange resin phase appeared costly. As the elution step gives lithium concentration of 0.9 g/L, it requires a substantial amount of energy to concentrate lithium. Conversely, the experimental outcomes after the crystallization phase revealed cost-effectiveness and fewer energy requirements across the entire process. Consequently, an economically viable flowsheet for the targeted recovery of lithium as lithium carbonate from the leaching solution after the LIB recycling is formulated and presents a potential industrial proposition.

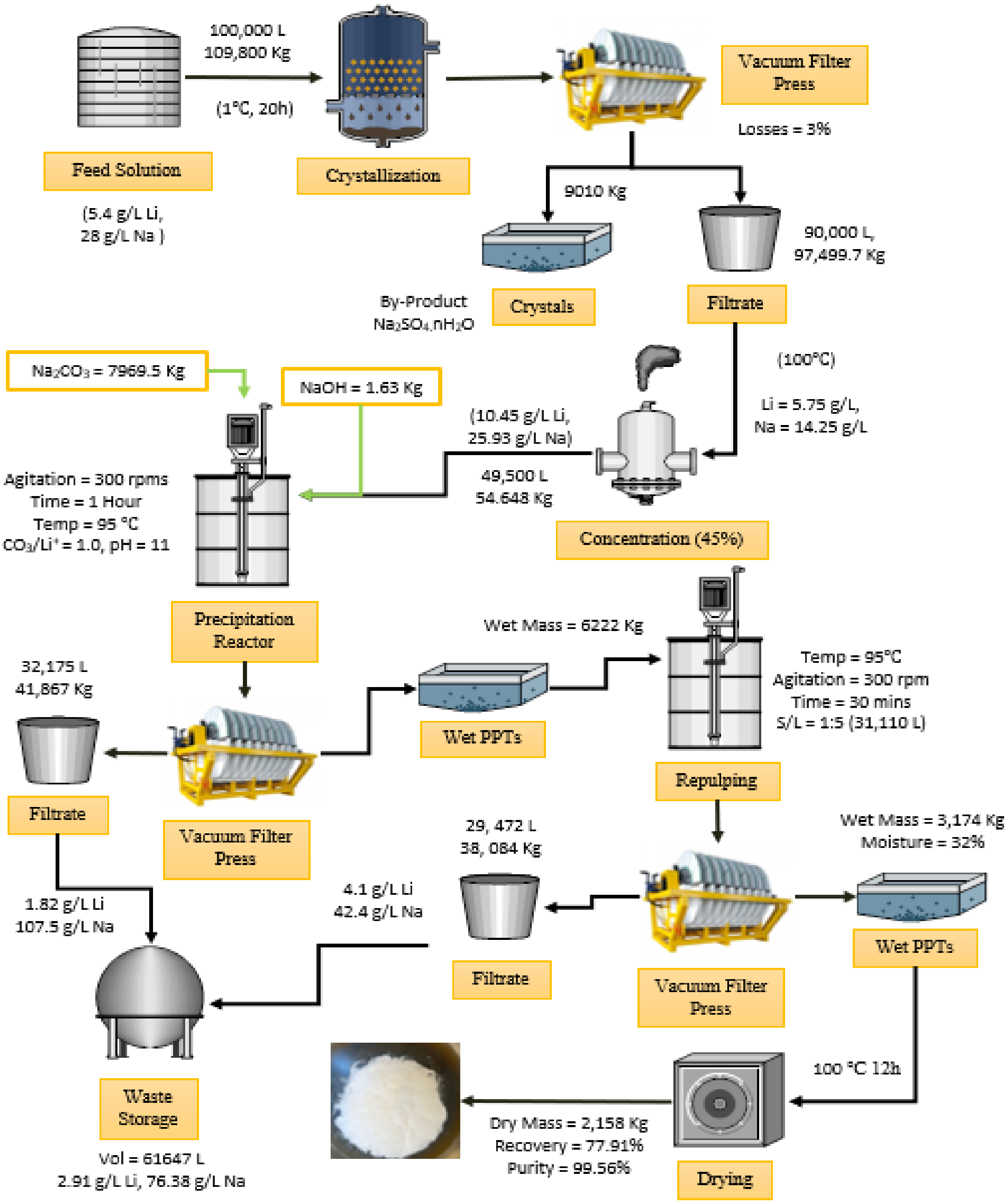

Figure: The Flowsheet 1 which is scaled up based on the experiment 11 (Uliege) operated in sulfate chemistry. The process is designed based on the 100,000L feed solution per day to reach around 50,000 L in the chemical precipitation step to produce nearly 2 tons of Li_2CO_3 per day.