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Experimental Study and Process Modeling of Closed-loop LIB Recycling with Lithium Sulphate Electrodialysis

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Lithium-ion batteries (LIBs) play a crucial role in urbanization and human life, providing essential energy storage capabilities distinguished by their high energy density, long cycle life, and low weight [1,2]. The hydrometallurgical method for recycling of spent LIBs is of great importance due to its remarkable features, including high metal recovery rates, their capacity for Li+ and Al3+ recycling, extraction of high-purity metals, operation at relatively low temperatures, and reduced energy consumption and emissions [3,4]. The conventional hydrometallurgical processes generate a sodium-enriched wastewater, requiring treatment prior to safe discharge. This study concentrates on a closed-loop recycling process for spent Lithium Nickel Manganese Cobalt Oxide (NMC) cathode material, depicted in Fig. 1. This innovative approach also aims to eliminate sodium ions through the deviation from conventional hydrometallurgical methods, concurrently repurposing dissolved ions within the disposal slurry, resulting in a more eco-friendly and cost-effective approach. Lithium hydroxide (LiOH) is utilized as the precipitating reagent in this process, while lithium recovery is conducted employing electrodialysis (ED) to regenerate LiOH and sulfuric acid (H2SO4) from the lithium sulfate solution. A portion of the LiOH and H2SO4 reagents is subsequently employed in the leaching and precipitation steps, establishing a closed-loop recycling system. To validate this approach, an experimental setup was established to study leaching, impurity removal, and metal extraction processes. The closed-loop recycling process was further investigated by the simulation of this process using Aspen Plus. Therefore, the Aspen Custom Modeler was employed to create the ED module in both continuous and batch configurations. Subsequently, this ED module was incorporated into Aspen Plus to integrate with the recycling process under experimental operational conditions. The minimal deviations of 3.34% and 2.38% within the precipitation and co-precipitation processes indicated the accuracy and validity of this work. Continuous and batch-mode EDs were integrated with the recycling LIBs process to extract LiOH and H2SO4 from the Li2SO4 solution resulting from the metal extraction procedure. Based on the results, a 40% greater recovery of LiOH solution from the Li2SO4 solution using batch-mode ED indicates the better performance of this mode compared to continuous configuration. The effect of time and temperature on the leaching efficiency is also investigated. As illustrated in Fig. 2, it was found that the enhancement of reaction time from 5 to 30 min resulted in the 33, 39, 45, and 73% increment on Li+, Ni2+, Co2+, and Mn2+ leaching efficiency, receptively. Also, the 53% increment of Li+ was observed by raising the temperature from 10 to 70℃.

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