$LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ Cathode active material recovery from ethaline

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Abstract

Lithium-ion batteries are used everywhere from computer technology to drones and small household appliances. The transition to electric mobility, currently in progress, is also based on the massive use of these batteries. Although we use the definition of "green energy/mobility" they have a limited life span. Their poor end-of-life management will have negative consequences on the environment and thus on human health. Moreover, most of the metals that compose a battery are classified as critical and/or strategic (lithium, cobalt and recently nickel and copper) by the E.U. [1]. Also considering the current geopolitical situation, recycling is a major issue to get out of the dependence on large producers located outside the E.U. The development of efficient and environmentally friendly recycling processes is therefore a major challenge for the energy transition and sovereignty. This work proposes a novel method for $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$ cathode material closed-loop recycling by leaching in choline chloride/ethylene glycol (ethaline) deep eutectic solvent, which has been doped with a small amount of HCl. It was demonstrated that the addition of 4 molar equivalents of protons per one molar equivalent of metals (Co, Ni …) guarantees the full dissolution of the active material at 80 °C and 2h. When a liquid/solid mass ratio of 50 is used, the final concentration of the acid in DES is about 0.82 mol. L^{-1}. The co-precipitation was then induced by the addition of 0.2 M Li_2CO_3 with 99% efficiency. The recovered mixed carbonate was mixed with Li_2CO_3 and then transformed into oxide by thermal annealing. The transformation was monitored using *in situ* X-ray diffraction. The batteries made from the recycled powder showed a good discharge capacity at 0.2C rate of about 130 $mAh.g^{-1}$, however, this value tended to decrease after 100 charge/discharge cycles. An XPS analysis has been performed to understand this capacity loss, revealing the presence of carbonate species on the surface of the recycled material, so an additional washing step was added at the end of the annealing process. The batteries, in this case, exhibit a discharge capacity of 170 $mAh.g^{-1}$, which is higher than the commercial material (155$mAh.g^{-1}$).

Reference

[1] Study on the Critical Raw Materials for the EU 2023 Final Report

doi.org/10.2873/725585.