Process Development in Recycle of Lithium-Ion Battery Cathode Materials

By

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Abstract

The rapid expansion of the lithium-ion batteries (LIBs) market not only raised the price of raw materials but also attracted increasing concern of the environmental hazards from the inappropriate disposal of spent LIBs. It is thus desirable to recycle the spent LIBs from both the economic and environmental perspectives. This thesis aims at designing the recycle processes for different cathode materials (LiNiₓMnᵧCo₁₋ₓᵧO₂, and LiNiₓCo₀Al₁₋ₓᵧO₂). Oxalic acid was used to recover lithium from LiNiₓMnᵧCo₁₋ₓᵧO₂ of various ratios selectively. Using LiNi₀.₅Mn₀.₅Co₀.₂O₂ (NMC-532) as the base case cathode materials, 96.3% of lithium was dissolved along with 2.16% of manganese. An integrated process based on chemical and anti-solvent precipitation was synthesized to separate and recover manganese and lithium with high recovery and high purity from the liquid after dissolution. Ammoniacal dissolution was utilized to recover Ni, Co, and Mn in the mixed oxalates produced from selective lithium dissolution. By controlling the formulation of the ammonium solution, transition metal ions were selectively dissolved in the order of Ni, Co, and Mn. When NH₂(NH₄)₂C₂O₄ was used, 97.2% of nickel and 0.6% of cobalt was dissolved, no manganese was detected in the solution. Then 96.2% of cobalt was recovered using NH₂(NH₄)SO₄·K₂SO₃. In order to better connect to the synthesis route of cathode materials, another possibility to handle the mixed oxalates was explored. The mixed oxalates (Ni, Co, and Mn) could be converted to mixed sulfates through an oxidative, thermal treatment. A process was developed to use the recycled mixed sulfates as the metal feed for LiNiₓMn₀.₅Co₀.₂O₂ precursor synthesis. The influence of different factors, including pH, the concentration of NH₄OH, inner atmosphere, and reaction time, on the quality of the precursor was investigated. A casual table has been developed to guide the synthesis of high-quality LiNi₀.₅Mn₀.₅Co₀.₂O₂ precursors. In addition, the developed separation processes were also shown to work for LiNi₀.₅Co₀.₁₅Al₀.ₐ₅O₂ (NCA) cathode materials.

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