

PRODUCTION OF NICKEL-COBALT-MANGANESE MIXTURES WITH TAILORED COMPOSITIONS FROM COBALT-RICH LITHIUM-ION BATTERY LEACHATES BY SOLVENT EXTRACTION

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ABSTRACT

Recovery of metals from end-of-life lithium-ion batteries (LIBs) promotes sustainability and resource efficiency. The metals in spent LIB cathodes are typically liberated by reductive acid leaching, which produces heterogeneous multi-metal solutions (LIB leachates). The LIB leachates contain mainly lithium, nickel, cobalt, and manganese, together with significantly lower amounts of aluminium, iron, and copper.

Solvent extraction (SX) is an established technique for separating the metals from the LIB leachates. In conventional SX processes the metals are separated into their own purified solution fractions, from where they can be precipitated or crystallized as pure metal salts, or electrowon. Because of the increasing demand of new batteries and shift towards nickel-rich LIB cathode chemistries, an alternative process concept in which a mixture with a specific stoichiometric ratio of nickel, cobalt, and manganese (*e.g.*, 8:1:1) (NCM mixture), is obtained directly from the SX process. The NCM mixture can then be used as such, or after polishing treatment, in the co-precipitation synthesis of new precursor cathode active materials (PCAM) without additional input of nickel, cobalt, or manganese to the synthesis solution. Furthermore, purified lithium sulphate, manganese sulphate, and cobalt sulphate solutions are obtained simultaneously from the SX process.

The excess manganese and cobalt were extracted from a synthetic cobalt-rich LIB leachate by 0.8 M bis(2ethylhexyl) hydrogen phosphate (D2EHPA) and 0.8 M bis(2,4,4-trimethylpentyl)phosphinic acid (Cyanex 272), respectively. The remaining cobalt and nickel were extracted together from the leachate by 0.8 M D2EHPA. Mixing of the cobalt- and nickel-bearing D2EHPA with manganese-loaded D2EHPA yields NCM-D2EHPA, from which the NCM sulphate mixture is produced by stripping.

A non-optimized flowsheet was designed based on the measured liquid-liquid equilibrium data, and the extraction characteristics of D2EHPA and Cyanex 272. The key separations of the flowsheet were demonstrated and studied further in continuous counter-current mode in industrial-type laboratory mixer-settlers (MEAB MSU0,5; MEAB Metallextraktion AB, Sweden).

An NCM mixture with n(Ni):n(Co) = 14.16 and n(Ni):n(Mn) = 8.06 was produced by continuous stripping of NCM-D2EHPA. The stoichiometric ratio of nickel, cobalt, and manganese can be adjusted to, for example, 8:1:1 using the cobalt-rich raffinate obtained from the same process. The raffinate from stripping of cobalt-loaded Cyanex 272 contained 102.7 g dm⁻³ cobalt at 99.8 % relative purity without intermediate scrubbing.

Keywords: solvent extraction, lithium, nickel, cobalt, manganese, co-extraction, recycling, lithium-ion batteries