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Environmental Impacts of Hydrothermal Acid Leaching of Lithium-ion Battery Cathode Materials

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Lithium-ion batteries (LIBs) is a key technology for decarbonization, experiencing rapid growth in demand across diverse industrial applications, including electric vehicles, energy storage systems, and digital infrastructure. Cathode materials employed in LIBs encompass various lithium metal oxides, such as lithium nickel manganese cobalt oxide (NMC: LiNixMnyCo1-x-yO2). Nevertheless, the mining, smelting, and processing of lithium (Li), nickel (Ni), cobalt (Co), and manganese (Mn) remain highly concentrated in specific nations and regions, posing significant risks to supply stability. Furthermore, the accelerating extraction of mineral resources in regions characterized by inadequate governance frameworks has precipitated substantial environmental consequences, thereby underscoring the importance of advancing battery recycling as a cornerstone of sustainable resource utilization.

Hydromrallurgical processes represent current mainstream technologies for recovering critical metals (e.g., Li, Ni, Co, Mn) from spent LIB cathode materials and manufacturing scrap. Hydrometallurgical acid leaching processes typically require highly concentrated inorganic acids and hydrogen peroxide as a reductant to efficiently dissolve metal ions. However, the resulting high-salinity wastewater poses a significant environmental burden. In this study, we developed a continuous hydrothermal leaching process employing solely citric acid as a leaching agent under elevated temperature (> 100°C) and pressure conditions for the efficient leaching of critical metals from black mass derived from spent LIB cathodes, thereby reducing environmental impacts and shortening leaching times. A quantitative assessment of the environmental impacts for this proposed process is crucial for its technological improvement and commercialization.

This study applied a gate-to-gate life cycle assessment (LCA) to quantify the environmental impacts of the proposed hydrothermal organic acid leaching process for recovering critical metals from spent LIB cathode materials in Japan. The LCA was conducted in accordance with the international standard ISO 14040:2006, encompassing the four stages of goal and scope definition, inventory analysis, impact assessment, and interpretation of results. The goal of this study is to demonstrate the environmental impacts of the targeted process as applied to NMC111 cathode materials in Japan and to identify impact hotspots. The functional unit was defined as the leaching of 1 tonne of NMC111 cathode black mass. The system boundary encompasses only the targeted process, from slurry supply to leachate transfer to storage tanks. The inventory of the targeted process was created based on primary data for process inputs obtained from laboratory experiments. Emission factors for these inputs, along with secondary data for other material and energy inputs, were obtained using the Ecoinvent 3.8 database (allocation, cut-off by classification). The inventory analysis was conducted using SimaPro Craft 10.3 software (PRé Sustainability). The impact assessment method adopted ReCiPe 2016 Midpoint (H) V1.07/World (2010) H/Normalization, and 18 impact categories were selected.

As shown in Fig. 3, citric acid contributed over 98% in almost all environmental impact categories, making it the primary factor in the process's environmental burden. This is attributed to environmental burdens from nitrogen-based fertilizer runoff during agricultural raw material production for citric acid and emission of toxic substances from its chemical synthesis. Conversely, electricity contributed less than 1% across all categories but showed impacts in ionizing radiation, fossil resource scarcity, and global warming. Deionized water primarily impacted direct water consumption, contributing approximately 2% to that category, while its impact on other categories was negligible.

Effective approaches to minimize the environmental impacts of this process include reducing citric acid usage through optimization of leaching conditions, recirculating citic acid for leaching, and increasing consumption of renewable energy-sourced electricity. The LCA findings provide evidence-based insights for further improving the environmental performance of the proposed process for spent LIB cathode material recovery.

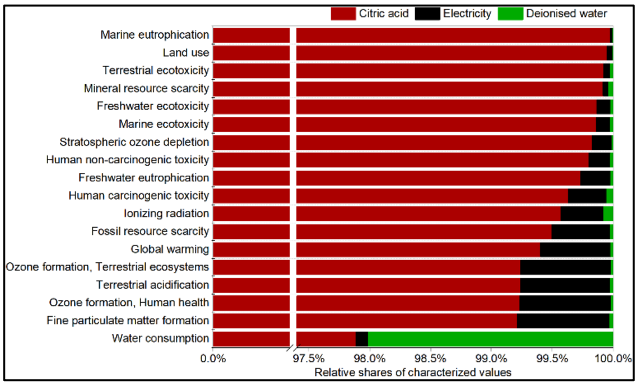

Fig.1 Relative contributions by process input to environmental impact categories in hydrothermal organic acid leaching of 1 tonne of NMC111 cathode black mass.

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