Doctoral Dissertation

Doctoral Program in Civil and Environmental Engineering (36th Cycle)

Circular economy of Lithium-ion batteries

Material flow, environmental and economic analysis of Lithium-ion batteries in Europe and technical, environmental and economic optimization of recycling processes

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Summary

Lithium-ion batteries are crucial for Europe's goals on emissions control, due to their manifold applications as energy storage devices for renewable energy production and electric transport. The widespread use of Lithium-ion batteries leads to the increased demand for raw materials and efficient recycling processes for the recovery of secondary raw materials become essential to ensure the supply chain and limit the environmental impacts and costs associated with the extraction of raw materials. Current recycling methods involve pyrometallurgy and hydrometallurgy, However, the costs entailed by existing technologies are prohibitive to recycle batteries containing lower cobalt content, specifically lithium iron phosphate cathodes.

This Thesis tackles the circular economy perspective for lithium-ion batteries. Initially the environmental and economic implications of material flows associated with end-of-life batteries from electric vehicles in Europe have been analysed, considering the supply chain for primary raw materials and the recycling infrastructure providing secondary raw materials. Findings underline how the existing recycling infrastructure in Europe falls short in meeting the collection and recovery targets set by the European Union for batteries recycling. Furthermore, the prescribed requirements for materials circularity in the battery sector seems unattainable when relying solely on secondary raw materials sourced from European recycling plants.

The environmental implication of mining activities for batteries supply chain often overshadows the environmental benefits of electric transport systems, compared to traditional internal combustion engines. Enhancing the recycling capabilities in Europe is imperative in fulfilling regulatory mandates and ensuring a stable supply chain for essential raw materials sourced from regions beyond Europe, often entangled in geopolitical complexities and to provide secondary raw materials avoiding the environmental impacts due to mining activities. From an economic perspective, hydrometallurgical processes represent the best cost-benefit solution to address the proven inadequacy of European

recycling capacity. Nonetheless, the costs entailed by conventional hydrometallurgy are still prohibitive to recycle low-value batteries such as lithium iron phosphate batteries.

In the experimental section of this Thesis several steps of the lithium-ion batteries recycling process have been studied in order to limit their environmental and economic costs, considering cells electrochemical discharge, cathodes pre-treatments, and hydrometallurgical processes of end-of-life lithium iron phosphate cathodes and their production scraps. The results of the experimental activities conducted in this Thesis, proved the viability of carbonates solutions for the electrochemical discharge of lithium-ion batteries, without corrosion phenomena and the release of harmful gaseous contaminants and carbon dioxide. A physic-mechanical method to detach the active materials from the current collector of end-of-life and production scraps lithium iron phosphate cathodes have been proposed, by optimizing technical parameters and limiting environmental and economic costs. Besides, different hydrometallurgical processes have been tested for the recycling of end-of-life and production scraps lithium iron phosphate cathodes identifying the importance of selective leaching processes and the application of organic acids to ensure economic viability of the overall recycling process.