Ultrasound-Assisted Hydrometallurgy: A Novel Approach to Lithium-Ion Battery Cathode Recycling

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Abstract:

Recycling lithium-ion batteries is crucial for sustainability, driven by increasing demand and environmental concerns. Among the recycling methods, the hydrometallurgical process, involving acid leaching, stands out for high metal recovery and purity. However, it faces challenges like safety concerns and long dissolution times. This study explores ultrasonic process intensification to enhance leaching efficiency using cavitation. This results in a multiphase system consisting of the solid cathode material, the liquid leaching solution, and the vapor-filled cavitating bubbles. Experiments with lithium cobalt oxide and acetic acid demonstrate a significant reduction in leaching time, achieving high efficiency in less than ten minutes, compared to the few hours required by the conventional leaching process.

Keywords: Solid-liquid-vapor, Leaching, Battery recycling, Acoustic Cavitation.

Introduction:

Recycling metals from lithium-ion (Li-ion) batteries has become crucial due to the rise in battery use, especially in electric vehicles. Li-ion batteries, the top choice for energy storage, contain valuable metals like cobalt, manganese, and lithium in their cathodes, making up about a third of the battery's mass and over half its value. Recycling these metals reduces the need for mining, lowers environmental pollution, conserves resources, and supports a circular economy. With increasing battery demand and environmental concerns, efficient recycling is essential for a sustainable future. Nowadays, the most promising process to recycle wasted batteries is the hydrometallurgical process[1]. This process, favored for high metal recovery and purity, involves an acid-leaching step, currently, the bottleneck of the process, as it requires time scales in the ordered hours to dissolve the cathode material[2]. The ultrasonic intensification of the process might help in overcoming the limitation of the process making it more appealing for the scale-up to industrial scale. Ultrasonic process intensification leverages cavitation, where high-frequency acoustic waves in a liquid create alternating pressure zones, forming vapor-filled bubbles. These bubbles grow and collapse violently, releasing energy as shockwaves, heat, and liquid jets, which enhance the process efficiency. This results in a multiphase system consisting of the solid cathode material, the liquid leaching solution, and the vapor-filled cavitating bubbles. The system is here studied to optimize the leaching conditions and develop a model to describe its time evolution.

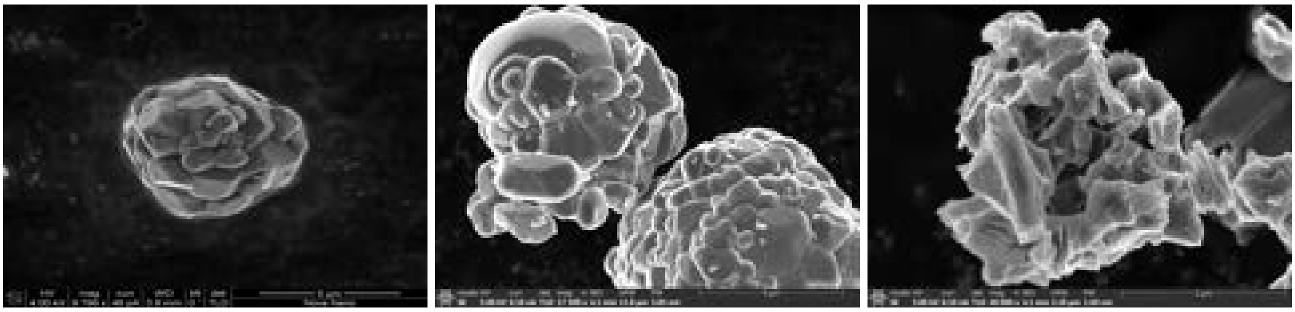
Experimental Facility:

This work experimentally investigated the effect of ultrasonically induced cavitation on the leaching process of cathodic material. Lithium cobalt oxide, purchased from Sigma Aldrich, was used as a surrogate to mimic the cathode material during the experimental campaign. The experiments were performed using about $200\,\mathrm{mL}$ ml of leachate solution placed in $400\,\mathrm{mL}$ quartz Becher. Acetic acid was utilized as the acidic medium for the leaching. The acoustic pressure field was established by a vibrating cylindrical metallic probe with a diameter of $1.4\,\mathrm{cm}$, a displacement of $125\,\mu\mathrm{m}$, and a vibrating frequency of $24\,\mathrm{MHz}$. A parametric study, involving more than 40 experiments, investigated the ultrasonically enhanced leaching at different operative conditions to understand the effect of parameters such as acid concentration, addition of a reducing agent, and initial solid-to-liquid ratio to optimize the cathode dissolution. Different state-of-the-art analytical techniques were adopted to investigate the leachate and the solid particle morphology after exposure to ultrasonically induced cavitation. In detail: Inductively coupled plasma-optical emission spectrometry (ICP-EOS) was adopted to quantify metals concentration in the leachate. Scanning electron microscope (SEM) permitted the visualization of solid particles before and after exposure to the ultrasonically induced cavitation. BET adsorption for surface area and porosimetry analysis.

Results and discusison:

Ultrasonically induced cavitation resulted in several beneficial effects, such as: (1) Enhanced Mixing: Nano-scale bubble formation and collapse create intense turbulence, improving the uniform distribution and contact between reactants. (2) Increased Surface Area: High-speed microjets and shockwaves from bubble collapse enhance transport properties and increase particle surface area through acid erosion. (3) Improved Mass Transfer: Turbulent flow and microstreaming boost diffusion and convection, accelerating reactant transport and system reactivity. (4) Disruption of Boundary Layers: Cavitation breaks down boundary layers, facilitating reactant penetration into confined spaces and improving efficiency.

Figure 1 presents SEM images of three particles: (a) the raw particle not exposed to leaching, (b) the particle subjected to ultrasonically enhanced leaching without the necessary reducing agent, and (c) the particle exposed to ultrasonically enhanced leaching in the presence of the reducing agent. The images prove that the cavitation activity alone does not directly benefit the process, as the energy released during bubble collapse is insufficient to erode and break solid LCO particles. Instead, cavitation serves as a catalyst, boosting the reactivity of an already active system.



(a) Not exposed to leaching (b) Leaching without reducing agent (c) Leached with reducing agent

Figure 1: SEM images of LCO particles

Conclusion:

The hydrometallurgical process has emerged as a promising method for recycling lithium-ion batteries due to its potential for achieving high recovery rates and purity of metals. However, this process faces challenges such as using strong acid solutions and lengthy residence times, highlighting the need for innovative solutions. Applying ultrasonic-induced cavitation in leaching processes shows promise in overcoming these challenges. The ultrasonic intensification resulted in a substantial reduction in the residence time needed for the leaching process. Specifically, ultrasonically enhanced leaching requires less than ten minutes, whereas conventional leaching typically takes a few hours.

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