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Supercritical ionothermal relithiation of $\text{Li}_{1-x}(\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2})\text{O}_2$ positive electrode materials for Li-Ion Battery Direct Recycling

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Supercritical fluid assisted ionothermal relithiation of $\text{Li}_{1-x}(\text{Ni}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2})\text{O}_2$ positive electrode materials for Li-ion battery direct recycling

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In response to the escalating demand for Li-ion batteries, the management of their end-of-life has emerged as a critical concern. While conventional recycling methods, such as hydrometallurgy and pyrometallurgy, have traditionally been employed, they are associated with drawbacks, including high energy consumption, total destruction of valuable components with incomplete critical raw materials recovery and emission of greenhouse gases contributing to environmental concerns [1]. A growing interest is observed in an alternative approach known as direct recycling [2]. This innovative method involves the retrieval of various battery components, potential repairs, and subsequent reuse, aiming to extend the lifecycle of the batteries and complying with current Regulations [3, 4] without resorting to destructive processes.

The present work deals with $\text{Li}_{1-x}(\text{Ni}_{1-y-z}\text{Mn}_y\text{Co}_z)\text{O}_2$ positive electrode materials, in particular, with NMC622 ($y=z=0.2$), which are used predominantly in commercial Li-ion batteries nowadays. Once aged, these layered phases become lithium-deficient and exhibit changes in their surface and bulk structure [5]. This project proposes a novel regeneration scheme, comprising an ionothermal route [6] combined with supercritical fluid technology in mild conditions (Figure 1.). The main goal is to shed light on the structural evolution behind relithiation for further understanding and optimization. Delithiated samples were prepared chemically in a glovebox under argon using nitronium tetrafluoroborate as the oxidant, the cutoff delithiation state being set to 50% to ensure full relithiation in real-life settings. The ionothermal reaction was carried out in an unstirred custom reactor setup employing supercritical carbon dioxide, argon, and nitrogen. Lithium bromide was utilized as the lithium source and reducing agent, with EMIM TFSI serving as the ionic liquid medium. The influence was tested of several parameters on efficiency. Identifying the optimal relithiation conditions led to the production of a material whose structure is indistinguishable from that of the pristine phase, all while reducing both reaction time and temperature significantly. The versatility of the process was evaluated by regenerating successfully in a single batch NMC622 with varying degrees of delithiation (10%, 30%, 50%), an unprecedented outcome not reported previously in the literature.

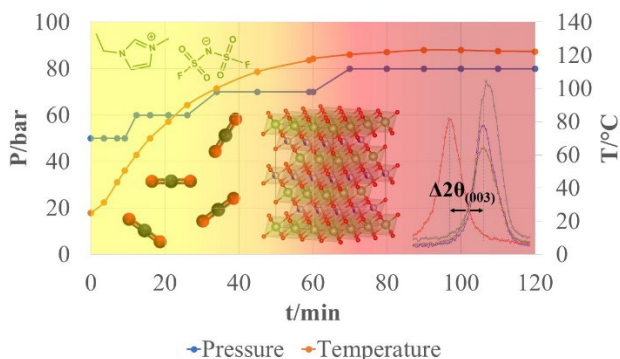


Figure 1: T/P profiles during ionothermal relithiation and evolution of the (003) peak.

This presentation outlines the ionothermal relithiation process, emphasizing the impact of optimized variables such as pressure and gas nature in achieving complete regeneration of the lithium content in NMC622. Multiple techniques are combined to characterize the structure, chemical composition, and physical properties of the

regenerated active material. The proposed procedure can be considered an effective and convenient approach to the direct recycling of layered oxide positive electrode materials.

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