

## NMC cathode materials: effect of processing parameters on their functional properties

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Lithium-Nickel-Manganese-Cobalt-Oxide (so-called NMC) based cathode materials have proven to yield enhanced specific capacity and thermal stability [1] thus, can outperform lithium iron phosphate (LFP) cathodes in many areas, particularly in terms of operational voltage in which LFP-based Li-ion batteries (LIBs) can only output voltages below 3.4 V [2] and suffer from high rates of self-discharge. Within NMC compositions, Nickel provides high energy density and increased storage capacity at lower cost and contributes to the circular economy due to the durability, recyclability and possible second life. However, the majority of degradation mechanisms of Ni-rich NMC-based cathodes are linked to their morphology and crystal structure related mechanisms as well as being dependent on the Ni/Mn/Co ratios. Increased nickel content can result in an increased cation mixing due to the bulk diffusion of Ni<sup>2+</sup> causing increased parasitic reactions as the valence of surface Ni increases toward highly reactive Ni<sup>4+</sup>. Due to the issues with their thermal instability at the fully charged state and insufficient cycle life, researchers are forced to explore approaches to improve these for enhancement of the overall performance of cathodes.

In this work, Ni-rich NMC cathode particles have been synthesized via a cost efficient and easy process-controlled co-precipitation method using different co-precipitation agents such as oxalic, succinic and malonic acids. As precursors, aqueous solutions of the metal salts (Ni, Mn, Co) have been employed in the required ratio. The precursor solutions have been co-precipitated by dropwise addition of the selected co-precipitating agent under continuous stirring. The resulting precipitates have been dried and heat-treated at various temperatures. SEM, EDX and XRD characterizations of the obtained powders have been performed in order to study their morphology and phase composition. It was found that a more ordered NMC ( $R\bar{3}m$ ) structure is achievable at the temperature range of 700-800 °C. Besides, the co-precipitation agent influences slightly the phase composition, leading to the higher quantity of the NMC phase formed for the oxalic acid samples in comparison to succinic and malonic once. Furthermore, the effect of co-precipitation agent on electrochemical performance has been studied by means of CV, CCD and EIS measurements of the powders heat treated at 750°C. The differences in the phase characteristics in the NMC powders, produced using various co-precipitation agents, affect primarily the electrochemical performance of the obtained cells. Particle morphology appears to have secondary effect. Accordingly, the powders produced with succinic and malonic acid displayed compressed CV profiles, which signifies the sluggish kinetics and aggravating obstacle of lithiation/delithiation, probably because of the lacking NMC phase order in these. On the other hand, NMC powder, produced using oxalic acid, provides higher capacitance value and promising battery performance than these two that were produced using the other co-precipitating agents.

[1] J. Phys. Chem. 2017, 121, 21865–21876.

[2] Inorganics 2014, 2, 132–154.