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Influence of the $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC 622) layered oxide morphology on the positive electrode/electrolyte interface

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The $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ layered oxide is a very attractive positive electrode material for Li-ion batteries and possibly in a near future for all-solid-state batteries, thanks to good capacity, stability and cyclability upon long range cycling in the former^{1,2}. To optimize its performances in both systems, a better knowledge of its reactivity, versus liquid and solid electrolyte, is needed. The aim of this work is to tailor its morphology, particle size and shape, in order to determine their impact on the reactivity. A series of samples with crystallographic structure close to the ideal 2D layered structure (less than 5% Ni^{2+} ions into the Li^+ sites) and the targeted composition were obtained by a two-step synthesis, consisting of a coprecipitation followed by a solid-state reaction.

Firstly, the primary particle size of a series of $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ layered oxide samples was tailored by playing on (i) the annealing temperature (from 780°C to 930°C), (ii) the atmosphere (air or O_2) and (iii) the excess of lithium carbonate added to the mixed transition metal precursor to prepare the lithiated composition ($\text{Li}/\text{TM}=1.05$ with 1 wt.% or 5 wt.%). Samples with primary particle sizes ranging from 200 nm to 2 μm were obtained, as shown in Figure 1. Electrochemical performances were determined in different conditions in coin cells versus Lithium metal (see Figure 1). At the end of these cycling tests, the electrode materials were recovered from the batteries in glove box under argon and washed with an excess of dimethyl carbonate. Then, they were analysed by X-ray Photoelectron and Auger spectroscopies in order to study the electrode/electrolyte interface and compare the reactivity of the materials as function of their morphology. The presentation will be focused on the study versus liquid electrolyte, the one versus solid electrolyte is just being started.

$\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ layered oxide samples with platelet-shaped particles were obtained by using a large amount of ammonium hydroxide as template during the coprecipitation step of the synthesis. By varying the annealing temperature of the following solid state reaction, it was possible to obtain platelets with thicknesses around 10 nm and 85 nm, as shown in Figure 2. The study of the electrode/electrolyte interface revealed a specific reactivity for the platelets, in comparison with that observed for conventional particles. The thinnest/smallest platelets were shown to be more prone to activate the degradation of the electrolyte salt. They also suffer more

important volume variations during cycling compared to conventional morphologies. Interestingly, the thickest platelets were shown to be less covered with the Solid Electrolyte Interphase while the conventional particle's one contained a higher quantity of degradation products from the electrolyte solvents.

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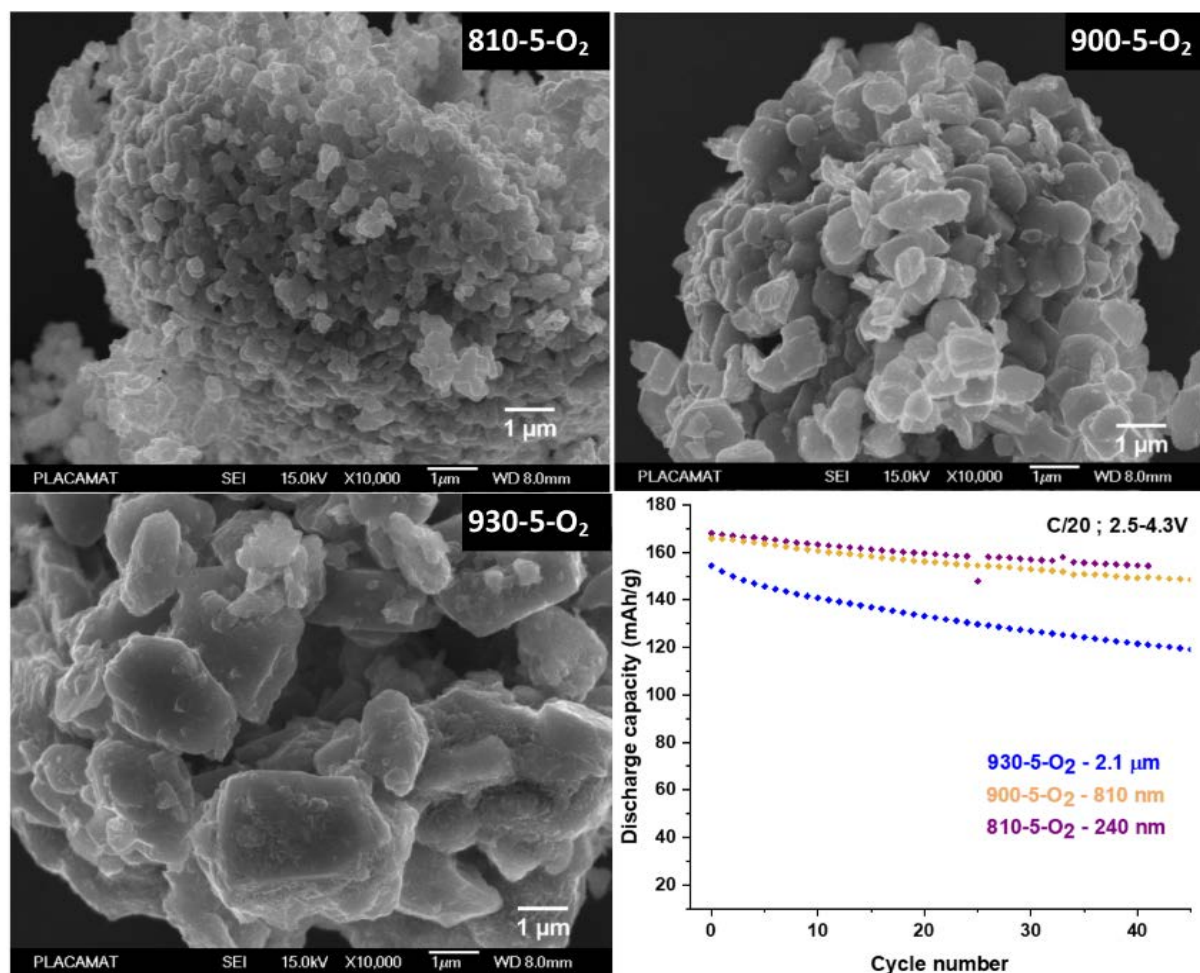


Figure 1: Morphologies of selected samples. They are named according to their synthesis conditions: lithiation temperature – wt.% Li₂CO₃ excess – lithiation atmosphere

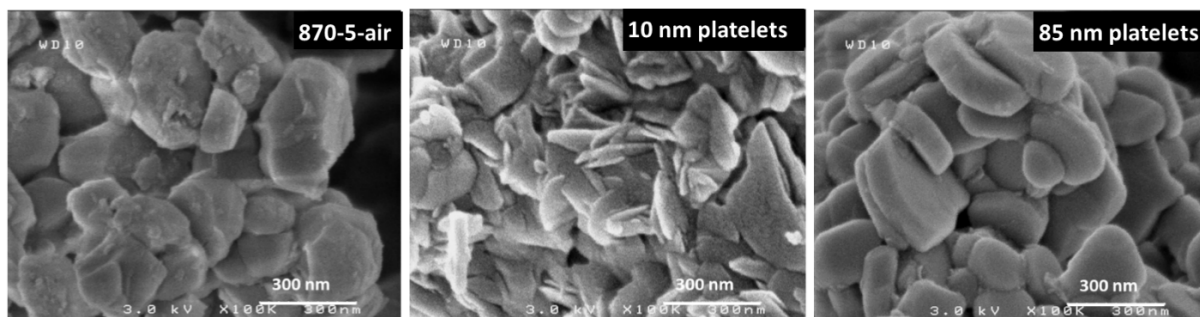


Figure 2: Morphologies of platelets-shaped particles compared with conventional ones

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