Controlling the cation ordering on a $LiNi_{0.5}Mn_{1.5}O_4$ thin-film model system by modifying the cooling rate during annealing

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The introduction of cathode materials that have both intrinsically higher capacities as well as higher operating potentials can help to further move towards the usage of electric vehicles and renewable energies. In this regard, high energy density cathodes which are cobalt free are of high interest in the field of batteries, mainly due to their lower cost, as well as for environmental reasons. The high cost of cobalt [1], as well as the environmental conditions under which it is extracted are the main determining facts which rise major concerns in the continuing use of cobalt containing cathodes.

In this regard, cobalt free cathodes such as $LiNi_{0.5}Mn_{1.5}O_4$ (LNMO) are of high interest due to its high operating potential (4.7 V vs Li^+/Li) as well as its high specific capacity (147mAh/g). However, regardless of the intensive research done over the years, this material has not yet really made it into the market. One of the reasons being the contradictory results reported in literature trying to correlate the composition, material properties and electrochemical performance with the crystal structure as well as differentiating its bulk and interface properties when in contact with electrolytes [2].

For this reason, in this work, we would like to present the potential of using a thin film $LiNi_{0.5}Mn_{1.5}O_4$ model system to address this issue. So, it is possible to relate more accurately the composition, crystallinity and cation ordering of this material with its electrochemical performance.

Thin film LNMO has been previously reported as model system for study of cation ordering in LNMO. Yet only the effect of the pressure during RF sputter deposition was investigated so far, which, however, can affect both the cation ordering and the composition of the thin film [3],[4]. As such the correlation between the electrochemical performance with the cation ordering was challenging, as it is also dependent on composition. Herein, we present an approach to control the cation ordering of a thin film LNMO while maintaining the composition constant based on controlling the cooling rate during the annealing step after sputtering at the same deposition pressure.

[1] Materials Today, Volume 18, Issue 5, 2015, Pages 252-264
[2] Journal of Power Sources 467 (2020) 228318
[3] Chem. Mater. 2017, 29, 14, 6044-6057
[4] Energy Storage Materials 15 (2018) 396-406