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► **To cite this version:**

François Weill, Gozde Oney, Jacob Olchowka, Arnaud Demortière, Laurence Croguennec. Influence of local order on electrochemical properties of high voltage spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$. 24th International Conference on Solid State Ionics, International Society for Solid State Ionics, Jul 2024, Londres, United Kingdom. hal-04651796

HAL Id: hal-04651796

<https://hal.science/hal-04651796v1>

Submitted on 17 Jul 2024

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Influence of Local Order on Electrochemical Properties of High Voltage Spinel LiNi_{0.5}Mn_{1.5}O₄

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The spinel material LiNi_{0.5}Mn_{1.5}O₄ is a good candidate as a positive electrode material for Li-ion batteries, thanks in particular to its high energy density and the absence of cobalt. But the performance of this material is highly dependent on the stoichiometry and order of the two transition metals. These parameters are closely linked to the conditions under which the material is synthesized.

Molten salt synthesis has enabled us to obtain a multi-faceted platelet material for which, by varying the temperature and atmosphere of the heat treatment, it is possible to control the order of the transition metals in the absence of impurities detectable by diffraction. The electrochemical properties of this platelet material are fully comparable to those of a material with a classical octahedral morphology.

Raman and NMR spectroscopies were used to explore the local order in these platelet materials. 4D-STEM was used to observe transition metal order at the particle level with a spatial resolution of 10 nm. By comparing two globally ordered materials with a difference in order homogeneity at the particle level, we were able to show that heterogeneity in transition metal order was beneficial to lithium mobility, enabling a globally ordered material to achieve electrochemical performance comparable to a disordered material.

References:

[1] Oney G., Olchowka J., Demortière A., Weill F., Croguennec L., [ACS Applied Energy Materials 2023, 6, 15, 8189–8196](#)

Acknowledgments:

The authors thank Région Nouvelle Aquitaine, as well as the French Research National Agency (ANR) for the funding of GC's PhD, the Projet DESTINa-ion_operando (ANR-19-CE42-0014-02) and the projet Labex STORE-EX (ANR-10-LABX-76-01).