

# Lithium manganese oxyfluoride as a new cathode material exhibiting oxygen redox

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The next generation of high performance Li-ion rechargeable batteries relies on the discovery of new positive electrode materials with a greater capacity to store charge. Generally speaking, highly ordered layered materials, such as LiCoO<sub>2</sub> (LCO), LiNi<sub>0.85</sub>Co<sub>0.1</sub>Al<sub>0.05</sub>O<sub>2</sub> (NCA) and LiNi<sub>0.13</sub>Mn<sub>0.54</sub>Co<sub>0.13</sub>O<sub>2</sub> (NMC), are considered to be the best-in-class<sup>1</sup>. One strategy to increase the capacity has been to explore the ordered Li-Rich materials, Li<sub>1+x</sub>M<sub>1-x</sub>O<sub>2</sub>, where x~1.2, where the greater number of Li ions increases the total theoretical capacity. However, an optimal candidate system for commercialisation within this space has yet to be identified.

In contrast to ordered materials, disordered structures have been largely ignored by the scientific community as they are perceived to exhibit poor Li<sup>+</sup> ion mobility. However, recent research has overturned this understanding and several disordered rock salt structures with competitive energy densities have been proposed<sup>2-5</sup>. Could these highly disordered Li-rich materials offer better structural stability and performance than their layered counterparts?

Here, we present the discovery of a new Li-rich disordered rocksalt material, lithium manganese oxyfluoride (LMOF), XRD and structure presented in Figure 1, whose structure appears to remain stable even over 100 charge-discharge cycles. Furthermore, LMOF exhibits a large, reversible capacity to store charge and, interestingly, this appears to be unexplained within the limits of transition metal redox capacity alone, raising the question of how such a large charge storage capacity can be achieved. In this work we fully characterise this new material and specifically we examine the role that oxygen plays in this anomalous charge compensation process.

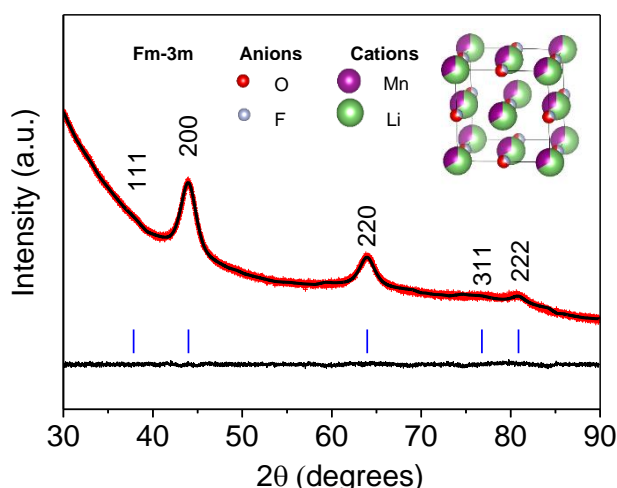


Figure 1. Powder X-ray Diffraction pattern obtained using a glass capillary sealed under argon and crystal structure (inset)

## References:

- [1] G. E. Blomgren, J. Electrochem. Soc. 164 (2017) A5019–A5025
- [2] M. Freire, et al., Nat. Mater., 15 (2015), 173–177.
- [3] R. Chen, et al., Adv. Energy Mater. 5, (2015), 1401814
- [4] N. Yabuuchi, et al., Nat. Commun 7, (2016), 13814.
- [5] J. Lee, et al., Energy Environ. Sci., 8, (2015), 3255–3265.