Li-rich layered oxides: towards more sustainable and high energy cathode materials for Li-ion batteries

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Li-rich transition metal layered oxides (LRLO) are a valuable alternative to current commercial cathode materials, thank to their higher specific capacities (>250mAhg^{-1}), larger operating voltage (>3.6V) and high energy density (900 WhKg^{-1}).

LRLOs have a generic formula $Li_{1+x}M_{1-x}O_2$, in which M is a mix of redox active transition metal as Ni, Mn or Co. Lithium is over-stoichiometric and partially occupy atomic sites of transition metals. The large capacity values reversibly exchanged by these types of materials originates either from the transition metal redox couples and the anionic redox activity (O^{2-}/O^{-}). This oxygen-redox reaction takes place at potential above 4.2-4.3V and could leads to the formation of molecular oxygen, making it only partially reversible. Therefore, the main problems that a researcher has to face for the development of LRLOs includes the improvement of low initial Coulombic efficiency, the mitigation of voltage decay and the increasing of the rate performance.

Nonetheless, to make these materials effectively competitive in the battery market the real challenge is to reduce also their production costs through more sustainable materials supply and through the integration of their synthetic processes into the manufacturing infrastructure already existing. In this respect, the removal of cobalt from the structure of LRLOs is a key-issue to improve their environmental compatibility, minimize the need of critical raw materials in the manufacture and reduce the costs.

Doping is one of the most used strategies not only to improve the structural stability, mitigate the voltage fading and reduce the capacity loss of LRLOs. But also, this strategy has been adopted to reduce or completely remove the cobalt from the structure.

Here, we propose a series of LRLOs by tackling the challenge of the simultaneous limitation of cobalt and nickel. These materials proved to maintain the layered structure characteristic of this type of materials and, especially, good electrochemical performance in lithium cell. The materials here proposed have been characterized in terms of structure and morphology, while the electrochemical properties have been demonstrated in lithium half-cells by galvanostatic cycling.