Progress in Disordered Rocksalt Cathode Materials

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Since the discovery that cation-disordered rocksalts can have large reversible Li-intercalation capacity when their Li-excess is above the percolation limit [1], many different compounds in this class have been synthesized and tested. Releasing the requirement that the cathode stays in the layered structure enables the use of a much broader chemistry of elements than is typical for NMC-style cathodes.

I will present some of the scientific and technical advances that have been made in disordered rocksalts. Using a novel electronic structure analysis, we will show the importance of cations with a d^0 oxidation state in promoting disorder [2]. Due to their lack of filled bonding states, these cations can most easily accommodate the distorted octahedral environments created in cation-disordered rocksalts. Hence, most disordered rocksalt cathodes contain elements such as Ti^{4+}, Nb^{5+}, Zr^{4+}, Mo^{6+}, etc. However, the Li-excess and high-valent inactive cation needed, all act to reduce the theoretical capacity that can be derived from the redox-active transition metal. As a result all disordered rocksalts use a large amount of bulk oxygen redox [3] to achieve high capacity, leading in many cases to oxygen loss at the surface and impedance growth.

We recently showed that oxygen-loss can be minimized or even eliminated in disordered rocksalts by their surprising ability to incorporate fluorine [4]. I will demonstrate how this can be used to create stable high capacity cathode materials with specific energy contents approaching 1000Wh/kg.

References:

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