Ordering in Na-ion Intercalation Materials Studied by TEM and DFT Computations

Xin Li, Alexandra J. Toumar, Xiaohua Ma, Shyue Ping Ong, Gerbrand Ceder

Department of Materials Science and Engineering

Massachusetts Institute of Technology

77 Massachusetts Avenue, Cambridge, MA 02139

In Na-ion rechargeable batteries, the Na*TM*O_2 (*TM* = 3d transition metals) materials with layered structures are of special interest because of their relatively high voltages and good capacity(*1*). Upon desodiation, Na ions can show different long or short-ranged ordering depending on transition metal or the oxygen stackings(*2*, *3*). Determining the nature of the ordering and understanding how it forms in these materials is important to the design of better Na-ion cathode materials.

In this work, we present results from a combined Transmission Electron Microscopy (TEM) experiment and Density Functional Theory (DFT) simulation on the Na ordering in Na*TM*O_2. TEM is used to characterize and determine the ground state Na orderings in several representative materials at certain de-intercalation levels, and DFT simulation is used to understand the basic interaction mechanism and also to investigate the ordering in the whole spectrum of Na_x*TM*O_2. Electrostatic, electronic and elastic driving forces are discussed in different systems, with special focus on the Jahn-Teller distortions. The implications of these findings to the battery design are also discussed.

References:

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