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(I) The asymmetric charge-discharge kinetics in $Li_{1-x}Ni_{1+x}O_2$ from first principles

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The ever-increasing demand on Li-ion batteries requires the cathode materials to be inexpensive and environmentally friendly. $LiNiO_2$ is such a promising Co-free cathode. However, the presence of Ni in the Li layer (Ni_{Li}) becomes more common without Co, which limits its electrochemical performance. These excess Ni could randomly distribute in the bulk due to Li deficiency during synthesis, or/and form a surface densified phase due to oxygen loss during cycling. Their interactions with Li, on top of Li-Li interactions, further complicate the non-dilute Li diffusion. In this talk, I will combine the density functional theory (DFT), cluster expansion and kinetic Monte Carlo (KMC) simulations to identify the effects of Ni_{Li} on Li transport in realistic conditions. Interestingly, both types of Ni_{Li} impede Li transport at the end of charge and discharge, but not at the beginning. This asymmetry kinetics cannot be solely explained by the Li diffusivity as a function of Li contents but stems from the phase boundary orientation between Li orderings. Ni_{Li} from synthesis smooths the voltage plateaus and contributes to the 1^{st} cycle capacity loss. Ni_{Li} from degradation hinders Li transport more severely when the densified phase fully covers the particle surface. Moreover, this surface phase kinetically traps the last 25% Li for an extremely long time during charge but shows little impedance when Li%>25%. These understandings could open new ways to engineer the transport properties of $LiNiO_2$-based materials.

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