



Canadian Association
of Physicists

Association canadienne
des physiciens et physiciennes

Contribution ID: 3428

Type: **Invited Speaker / Conférencier(ère) invité(e)**

(I) The asymmetric charge-discharge kinetics in $\text{Li}_{1-x}\text{Ni}_{1+x}\text{O}_2$ from first principles

Thursday 9 June 2022 09:15 (30 minutes)

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 1st 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 $\text{Li}\% > 25\%$. These understandings could open new ways to engineer the transport properties of LiNiO_2 -based materials.

Author: Prof. XIAO, Penghao (Dalhousie University)

Presenter: Prof. XIAO, Penghao (Dalhousie University)

Session Classification: R1-7 Materials for Energy Applications (DCMMP) | Matériaux pour applications en énergie (DPMCM)

Track Classification: Technical Sessions / Sessions techniques: Condensed Matter and Materials Physics / Physique de la matière condensée et matériaux (DCMMP-DPMCM)