Unbiased Monte-Carlo Approach to Study Discharging in MnO_2 Ramsdellite

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Transition metal oxides serve as key cathode materials in modern lithium-ion batteries. Among these, MnO_2 stands out as a widely utilised option due to its environmentally friendly and cost-effectiveness. Its significance has surged recently, driven by the escalating demand for mid- and large-range batteries in electric vehicles. MnO_2 has diverse polymorphs with distinct crystal structure, stability, and lithiation capacity. Notably, γ-MnO_2 demonstrates exceptional electrochemical activity. This polymorph is formed by an intergrown crystals of ramsdellite and pyrolusite.

In this study, our primary focus has been on ramsdellite MnO_2 (R-MnO_2), a cathode material that has been less explored. We have investigated the discharging reaction, or lithiation process, within this material. Using simulations based on the interatomic potential method, we initially ensured the accurate reproduction of various MnO_2 polymorphs and their stability, validating the applicability of our model. Our refined interatomic potentials reproduce the important MnO_2 polymorphs. Using the in-house Monte-Carlo approach, we investigated $Li_xMn_{24}O_{48}$ structures, where x ranges from 1 to 24, employing a (1×2×3) supercell of R-MnO_2. In my talk, I will present both details of the method and the results obtained.