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Li-ion Battery Electrode Materials Design from First-Principles Calculations

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First-principles calculations can provide a powerful tool for investigating and optimizing electrode materials. While the strength of computations lies in the ability to control what is being calculated, the challenge is to ensure that the calculation is relevant for the physical processes that dominate the performance of the material. We will discuss this balance and show examples of how computations can aid in the design of current Li-ion rechargeable battery electrode materials by identifying and understanding the performance bottlenecks on the atomistic level. As the most commonly used anode in today's Li-ion batteries, graphite shows poor rate capability at lower temperatures, leading to over-potential and Li plating. Using first-principles calculations, coupled with a cluster expansion of Li interactions and kinetic Monte Carlo we were able to show that *intrinsic* Li diffusion in graphite can be very fast, providing guidance towards designing higher-rate carbonaceous anode materials. On the cathode side, we have studied the layered $Li(Ni_{1/3},Mn_{1/3},Co_{1/3})O_2$ material, which is an interesting candidate if Co is partially substituted by the cheaper Al. Li migration in this material is influenced by several factors such as Li slab space, cation ordering and interlayer mixing. We present ab initio calculations of Li diffusivity as a function of Al content and slab spacing in the layered material, which elucidates the intrinsic rate performance effect of the Al substitution in the bulk material.