

Conversion Reactions and Transport Paths in Li-FeF₂ Cathodes

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Newly developed conversion materials such as iron fluorides enable access to the multiple valence states of the cathode cation in high capacity Li-ion batteries. While post-reaction TEM shows nanoscale clusters of LiF and Fe-metal in FeF₂ exposed to Li, the detailed mechanisms and reaction pathways are not known. To address this problem, we have developed a variable charge potential for molecular dynamics simulations that accurately describes FeF₂, FeF₃, and LiF crystal structures, elastic constants, and surface energies consistent with experiment and ab-initio calculations, with charges on the ions in the different phases consistent with Bader charges calculated from ab-initio calculations. A single potential allows for a change in charge on Fe from the +2 state in FeF₂ to Fe-metal and formation of LiF based on Li exposure. The charge variations observed in the simulations are similar to ab-initio calculations. The effect of FeF₂ surface orientation on the conversion reaction and Li transport and the role of rapid transport paths will be presented, as well as the effect of electronic conduction on additional transport paths along phase boundaries. Additional results involving charging and the role of oxygen will be discussed.