

Ionic transport properties and structural stability of high-capacity silicate cathode materials for Li-ion batteries

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Using density-functional theory (DFT) methods, we have investigated the structural, electronic and Li transport properties of several silicate polymorphs, some of them already synthesized and characterized in the laboratory. Our study includes the lithiated and several delithiated phases, in order to accurately describe the voltage profiles of these compounds and the effect of charge/discharge process on their structural stability. We also describe the electronic structure of these tetrahedral silicates, to examine the most favorable mechanisms for both ionic and electronic conductivities. Our kinetic studies of vacancy migration barriers show that, depending on the polymorph, some of these materials have higher diffusion barriers for Li motion than other cathode materials, but we also show a way to control the composition of the cathode to decrease such barrier and increase the ionic conductivity. Finally, we show that, with a suitable combination of transition metals (TM) in the cathode, we can tailor the position of the valence band maximum of the silicate and the *d* orbitals of the TM redox couple. This finding will facilitate to increase the intrinsic limit voltage of the cathode and, hence, the energy density of the Li-ion battery.

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