

Computational Study on the Properties and Kinetic Behavior of Carbon-based Li-ion Batteries Electrolytes Solvents, Unraveling their Differences.

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For a long time the differences in the cycling behavior for different electrolytes used in Li-ion batteries has intrigued researchers. For example, ethylene carbonate (EC)-based electrolytes can be cycled successfully without catastrophic degradation, however charging a similar cell based on a propylene carbonate (PC)-based electrolyte results in continuous decomposition of PC and exfoliation of the graphite anode. In this work, the differences in the reactivity between EC and PC is investigated, with a focus on the energetics controlling the kinetics in gas phase and in the graphite inner phase. Simulations based on quantum mechanics and classical force-fields are carried out using the Schrödinger Materials Science Suite, in order to understand details of the charge/discharge process in Li-ion batteries and to predict atomistic details and energetics of the reaction mechanisms involved.

Density Functional Theory (DFT) within the pseudo-spectral approximation, as implemented in the Jaguar code, has been used to gain insight into the differences between EC-based and the PC-based electrolytes in lithium-ion battery cells. To understand the differences in kinetics, the $2e^-$ reduction potential for both EC and PC solvents was calculated. The comparison was made between reduction potentials in gas phase and for the solvents intercalated between two graphite layers. This comparison sheds light on the differences in cycling behaviors of these two solvents.