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Density Functional Theory Studies on the Role of Electrolyte Additives in Lithium-ion Batteries

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To further progress the adoption of electric vehicles and other high power energy storage applications, it is desirable to develop lithium-ion cell chemistries that offer longer lifetimes at high temperatures and cell voltages, without significantly increasing the cost. The introduction of sacrificial electrolyte additives on the order of a few weight percent is a practical method to form protective solid-electrolyte interphase (SEI) layers that limit electrolyte decomposition during cell storage and operation. In recent years, significant efforts have provided new understanding of the underlying chemistry of several such additives, including sulfur-containing heterocyclic compounds and species that contain a Lewis acid-base adduct.

This work will present how density functional theory (DFT) calculations have been used to explore the underlying chemical reactions leading to SEI formation. Two sulfur-containing additives, prop-1-ene-1,3-sultone (PES) and ethylene sulfate (DTD), and two Lewis adducts, pyridine boron trifluoride (PBF) and pyridine phosphorus pentafluorophosphate (PPF), will be discussed. The DFT results offer new insight into the onset potential and reaction products of electrochemical reduction. By pairing DFT with a diverse set of experimental techniques, including X-ray photoelectron spectroscopy, isothermal microcalorimetry, solid-state nuclear magnetic resonance spectroscopy, gas volume measurements, and electrochemical techniques, new SEI components are proposed for each additive.

In general, the results in this work confirm previous recommendations that a wide variety of experimental techniques, coupled with computational methods such as density functional theory, can offer new insights into the underlying chemistry of SEI formation in lithium-ion cells. It is hoped that future work can apply the results of this work to understand what makes a 'good' electrolyte additive and, ultimately, to design new and improved electrolyte cell chemistries.

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