

Solid electrolyte interphase evolution in the presence of vinylene carbonate and vinyl ethylene carbonate electrolyte additives in Li-ion battery anodes

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We report on the effect of vinylene carbonate (VC) and vinyl ethylene carbonate (VEC) electrolyte additives on the composition and structure of the solid electrolyte interphase (SEI). VC and VEC additives are known to facilitate SEI formation through an electrochemically induced polymerization. Carbonate based solvents, e.g. propylene carbonate and ethylene carbonate-dimethyl carbonate, are also known to form SEI oligomers during cycling. Electrochemical quartz crystal microbalance (EQCM) measurements of VC containing electrolytes show less mass accumulation during Li deposition and SEI formation, while electrolytes containing VEC show a mass increase. These mass changes in the presence of the VC and VEC additives are concentration dependent. Matrix assisted laser desorption ionization (MALDI) mass spectrometry (MS) results show increased oligomerization in the presence of the electrolyte additives. The lengths of the SEI oligomers are determined by the additives concentration. Additionally, surface stress measurements show that SEI oligomers induce a residual tensile stress at the end of delithiation. The residual tensile stress to some degree can be controlled via additives concentration and structure. These data show that the SEI formed in the presence of VC is different from the one forming in VEC containing electrolytes. VC containing electrolytes show less mass accumulation during SEI formation, while increased oligomerization is observed. Interestingly VEC causes increased oligomerization while increasing the deposited mass during SEI formation and Li deposition. This may likely originate from a more packed SEI formed in VC containing electrolytes, while VEC provides a stable SEI without hindering Li transport. We will discuss the interplay between surface stress, SEI composition and SEI mass in the presence of VC and VEC additives.