Fluoroethylene Carbonate Induces Ordered Electrolyte Interface On Silicon And Sapphire Surfaces As Revealed By Sum Frequency Generation Vibrational Spectroscopy And X-Ray Reflectivity

The cyclability of silicon anodes in lithium ion batteries (LIBs) is affected by the reduction of the electrolyte on the anode surface to produce a coating layer termed the solid electrolyte interphase (SEI). One of the key steps for a major improvement of LIBs is unraveling the SEI's structure-related diffusion properties as charge and discharge rates of lithium-ion batteries are diffusion-limited. To this end, we have combined two surface sensitive techniques, sum frequency generation (SFG) vibrational spectroscopy and X-ray reflectivity (XRR), to explore the first monolayer and to probe the first several layers of electrolyte, respectively, for solutions consisting of 1M lithium perchlorate ($LiClO_4$) salt dissolved in ethylene carbonate (EC) or fluoroethylene carbonate (FEC) and their mixtures (EC:FEC 7:3 and 1:1 wt%) on silicon and sapphire surfaces. Our results suggest that the addition of FEC to EC solution causes the first monolayer to rearrange itself more perpendicular to the anode surface while subsequent layers tend to maintain their initial, on average, surface-parallel arrangements. This fundamental understanding of the orientation of the electrolyte molecules can modify operational strategies for designing high-performance LIBs.