In-situ Optical and Raman Studies of SEI Formation on Graphite in EC- and PC-based Electrolytes

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Current Li-ion batteries electrolytes use EC-based electrolyte systems. Although EC possess high viscosity they are preferred since they inflict less damage to the electrodes due to reasons that are not well identified. PC-based electrolytes despite having high conductivity and low viscosity cannot be productively implemented because they cause severe electrode exfoliation and fragmentation problem.

Using a turbostratic graphite electrode, installed in an electrochemical cell with an observation window, the sequence of graphite surface changes was observed during the course of experiments at $1000\times$ magnification. It was revealed that fragmentation and loss of graphite particles caused damage by formation of surface cavities during the first de-lithiation [1]. Microstructure and composition of SEI/graphite interfaces and the cracks morphologies were identified using focused ion beam milling (FIB) and SEM [2,3]. When an EC-based electrolyte was used, high-resolution (HR)-TEM found evidence of Li_2CO_3 and Li_2O co-intercalation compounds at the crack tips [3]. The SEI layers consisted of an amorphous matrix structure incorporating nanocrystalline domains (5-20 nm) of Li_2CO_3 and Li_2O.

Low voltage scan rate CV tests (~0.05 mV/s) produced a uniform and continuous SEI with a cellular morphology [4] and caused mild graphite damage [5]. Higher scan rates on the other hand resulted in the formation of an SEI layer with columnar morphology, and the graphite surface exhibited large regions devoid of SEI and thus graphite was prone to cavitation damage [4,5]. The effects of EC- and PC-based electrolytes on graphite degradation were examined at $50^{\circ}C$ as well including the role of vinylene carbonate additive in facilitating formation of protective SEI layers using both in-situ optical microscopy and Raman spectroscopy. Cross-sectional microstructures, obtained using FIB/SEM techniques, were used to compare the morphology of the SEI layers. CV curves were analyzed using Randles-Ševčík method to calculate Li-ion diffusion rates at the electrode/electrolyte interface and to gain insight into the formation mechanisms of the SEI layers.

Acknowledgements

References

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