Origins of irreversible capacity loss in hard carbon negative electrodes for potassium-ion batteries

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Hard carbon (HC) is a well-known negative electrode material for potassium-ion batteries, but it suffers from significant irreversible capacity loss at the first discharge cycle. We studied possible reasons of this capacity loss targeting the electrode/electrolyte interphase formation process in carbonate-based electrolyte with and without vinylene carbonate (VC) as an additive.

The studied HC consists of curved graphitic layers arranged into short packets and round cages, the latter acting as traps for K+-ions causing low Coulombic efficiency between cycling. Our comparative study of solid electrolyte interphase (SEI) formation in the carbonate-based electrolyte with and without VC additive revealed that in the pristine electrolyte SEI consists mostly of inorganic components whereas adding VC introduces a polymeric organic component to the SEI increasing its elasticity and stability against fracturing upon HC expansion/contraction during electrochemical cycling. In a pristine carbonate-based solution, the SEI layer starts to form when electrolyte components (solvent molecules and salt anions) decompose on the HC surface. We assume that the subsequent intercalation of K^+-ions results in the particle expansion and the SEI cracking. Freshly exposed HC surface react with the electrolyte components forming new SEI that further diminishes the CE values. When VC is added, its reduction shifts the SEI nucleation potential to lower values as compared with the pristine electrolyte solution and changes its composition.

The findings of this study suggest that the modified SEI layer with more polymeric components possesses higher elasticity and survives the HC expansion/contraction during K^+ intercalation/ deintercalation.

Study of metal-ion battery materials, surface layer formation at the electrode/electrolyte inteface and degradation processes during battery operation.

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