Analysis of the Interphase on Carbon Black Formed in High Voltage Batteries

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Carbon black (CB) additives commonly used to increase the electrical conductivity of electrodes in Li-ion batteries are generally believed to be electrochemically inert additives in cathodes. Decomposition of electrolyte in the surface region of CB in Li-ion cells at high voltages up to 4.9 V is here studied using electrochemical measurements as well as structural and surface characterizations. $LiPF_6$ and $LiClO_4$ dissolved in ethylene carbonate:diethylene carbonate (1:1) were used as the electrolyte to study irreversible charge capacity of CB cathodes when cycled between 4.9 V and 2.5 V. Synchrotron-based soft X-ray photoelectron spectroscopy (SOXPES) results revealed spontaneous partial decomposition of the electrolytes on the CB electrode, without applying external current or voltage. Depth profile analysis of the electrolyte/cathode interphase indicated that the concentration of decomposed species is highest at the outermost surface of the CB. It is concluded that carboxylate and carbonate bonds (originating from solvent decomposition) and LiF (when $LiPF_6$ was used) take part in the formation of the decomposed species. Electrochemical impedance spectroscopy measurements and transmission electron microscopy results, however, did not show formation of a dense surface layer on CB particles.

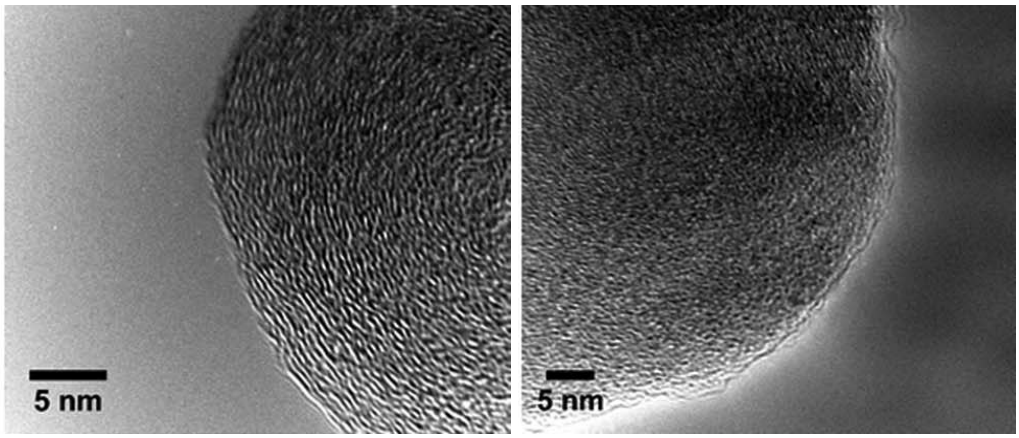

Figure 1: High-resolution TEM images of pristine (left) carbon black particles and after charge to 5.2 V (right).