DFT Study on Effects of CO₂ Contamination in Non-aqueous Li-Air Batteries

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Abstract

Density Functional Theory (DFT) studies on the effects of carbon dioxide (CO₂) contamination at the cathode of rechargeable non-aqueous Li-O₂ batteries, where the insulating material Lithium peroxide (Li₂O₂) is the main discharge product. The Li₂O₂ growth mechanism and overpotentials are investigated with and without CO₂ at different nucleation sites such as steps, kinks and terraces of valley and ridge of the stable (1-100) Li₂O₂ surface. Though their corresponding binding energies show that CO₂ binds weakly at the surface, it binds preferentially at steps and kinks of the valley (1-100) Li₂O₂ surfaces, forming a type of Lithium coordinated carbonate species (LiₙCO₃), which reduces the effective equilibrium potential by 0.2 V. Small amounts of CO₂ can also affects the morphological growth directions of Li₂O₂ due to blocking of nucleation centers; why may enhance the electronic conduction and result in an increased battery capacity. However, CO₂ contamination on the Li₂O₂ surface confirms an asymmetric increase in the overpotentials; particularly the charging overvoltage exhibits substantial increase, which would reduce the efficiency of the Li-air battery.

Key Words: Li₂O₂, CO₂, Li-air battery, rechargeability, overvoltages