DFT Study on Effects of CO_2 Contamination in Non-aqueous Li-Air Batteries

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Abstract

Density Functional Theory (DFT) studies on the effects of carbon dioxide (CO_2) contamination at the cathode of rechargeable non-aqueous Li-O_2 batteries, where the insulating material Lithium peroxide (Li_2O_2) is the main discharge product. The Li_2O_2 growth mechanism and overpotentials are investigated with and without CO_2 at different nucleation sites such as steps, kinks and terraces of valley and ridge of the stable (1-100) Li_2O_2 surface. Though their corresponding binding energies show that CO_2 binds weakly at the surface, it binds preferentially at steps and kinks of the vally (1-100) Li_2O_2 surfaces, forming a type of Lithium coordinated carbonate species (Li_nCO_3), which reduces the effective equilibrium potential by 0.2 V. Small amounts of CO_2 can also affects the morphological growth directions of Li_2O_2 due to blocking of nucleation centers; why may enhance the electronic conduction and result in an increased battery capacity. However, CO_2 contamination on the Li_2O_2 surface confirms an asymmetric increase in the overpotentials; particularly the charging overvoltage exhibits sustantial increase, which would reduce the efficiency of the Li-air battery.

Key Words: Li_2O_2, CO_2, Li-air battery, rechargeability, overvoltages