The effects of Li2CO3 like species originating from reactions between CO2 and Li2O2 at the cathode of non-aqueous Li-air batteries were studied by density functional theory (DFT) and galvanostatic charge-discharge measurements. Adsorption energies of CO2 at various nucleation sites on a stepped (1\ 0\ 0) Li2O2 surface were determined and even a low concentration of CO2 effectively blocks the step nucleation site and alters the Li2O2 shape due to Li2CO3 formation. Nudged elastic band calculations show that once CO2 is adsorbed on a step valley site, it is effectively unable to diffuse and impacts the Li2O2 growth mechanism, capacity, and overvoltages. The charging processes are strongly influenced by CO2 contamination, and exhibit increased overvoltages and increased capacity, as a result of poisoning of nucleation sites: this effect is predicted from DFT calculations and observed experimentally already at 1% CO2. Large capacity losses and overvoltages are seen at higher CO2 concentrations. © 2014 AIP Publishing LLC.

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