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The Effect of O₂ Concentration on the Reaction Mechanism in Li–O₂ Batteries

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Abstract

The promising lithium-oxygen battery chemistry presents a set of challenges that need to be solved if commercialization is ever to be realized. This study focuses on how the O₂ reaction path is effected by the O₂ concentration in the electrolyte. An electrochemical quartz crystal microbalance system was used to measure current, potential, and change in electrode mass simultaneously. It is concluded that the mass reversibility is O₂ concentration dependent while the coulombic efficiency is not. The mass reversibility is higher at low O₂ concentration meaning that more of the deposited Li₂O₂ is removed during oxidation in relation to the amount deposited during reduction. The first step of the reduction is the formation of soluble LiO₂, which is then either reacting further at the electrode or being transported away from the electrode resulting in low current efficiency and low deposited mass per electrons transferred. During the oxidation, the first step involves delithiation of Li₂O₂ at low potential followed by bulk oxidation. The oxidation behavior is O₂ concentration dependent, and this dependence is likely indirect as the O₂ concentration effects the amount of discharge product formed during the reduction. The O₂ concentration at different saturation pressures was determined using a mass spectrometer. It was found that the electrolyte follows Henry's law at the pressures used in the study. In conclusion, this study provides insight to the O₂ concentration dependence and the preferred path of the O₂ electrochemical reactions in lithium-oxygen batteries.

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