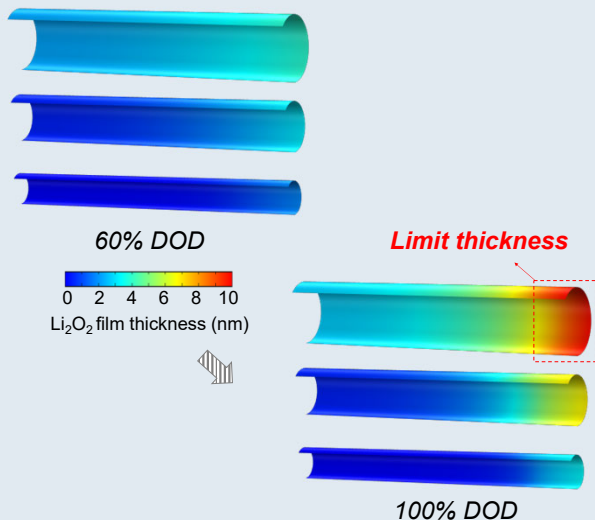


Mass Transfer Analysis in Lithium-Oxygen Batteries

A highly ordered electrode eliminates the interference from disordered pores and enables the study of the coupled mass and charge transfer mechanism.

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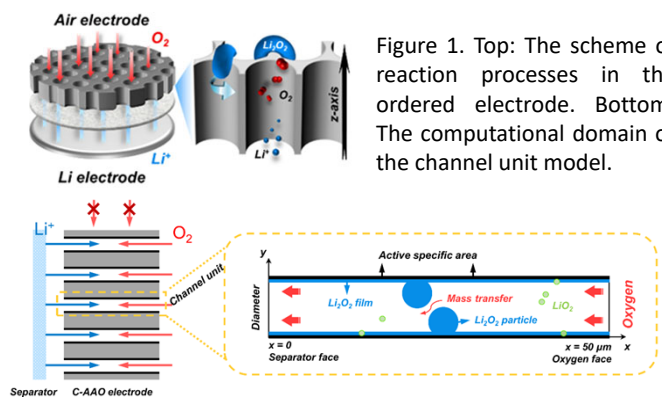
Abstract

The lithium-oxygen battery has the highest theoretical specific energy among all battery systems, while the actual value falls significantly short. The hindered oxygen and/or electron transport result(s) in limited utilization of the porous air electrode, while a quantitative understanding of the electrochemistry and mass transport coupling is challenging. A dynamic heterogeneous model is developed, providing the first spatio-temporal

distribution of LiO_2 and revealing its reversed diffusion trajectories at limited electron transport. The model identifies the crucial role of channel sizes on mechanisms that are divided into mass, hybridization, and electron transport control. The unit model offers a promising approach to quantitatively understand the reaction and transport mechanisms in other battery systems with porous electrodes.

Methodology

Based on the ordered electrode, the transport pathways of species during the discharge process become clear and simplified. As a result, an individual channel unit can be readily separated to represent the entire ordered electrode, since the transport pathways of active species, electrochemical reaction interfaces, and the maximum storage space for solid products can all be precisely determined. The influence of Li_2O_2 morphology (toroidal and film) on mass transfer and the generation/consumption and diffusion of the intermediate species LiO_2 are considered in this model.



Results

The critical channel size r_1 and r_2 ($r_2 > r_1$) divides the control mechanisms during discharge. In the smaller channels ($d < r_1$), the discharge is predominantly governed by oxygen transport, where oxygen depletion leads to a low capacity. With a larger channel size ($d > r_2$), the battery failure is entirely attributed to the loss of electron transport at the active sites caused by the deposition of ~ 10 nm Li_2O_2 film and independent of oxygen transport. When the channel size is between r_1 and r_2 , the discharge is controlled by a hybrid mechanism involving both mass and electron transport.

Figure 2. The distribution of resistance, Li_2O_2 film, O_2 , and LiO_2 with different channel size.

Reference

Z. Zhang, X. Xiao*, A. Yan, K. Sun, J. Yu, P. Tan*, A Quantitative Understanding of Electron and Mass Transport Coupling in Lithium–Oxygen Batteries. *Advanced Energy Materials*, 2023, 13, 2302816.



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