

## Modeling Electrochemistry and Transport in Aqueous Lithium-Oxygen Batteries

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We will present results for metallic lithium-oxygen (Li-O) batteries within a multi-phase modeling framework for batteries and fuel cells. Li-O batteries offer a very high theoretical energy density (11.680 Wh/kg) and are a promising alternative to conventional combustion technologies in the mobility sector. However, the development of Li-O cells is still at an early stage and a list of issues needs to be solved. Most importantly, a stable electrolyte must be found, since the standard carbonate-based solvents for lithium intercalation batteries decompose during discharge of Li-O cells [1]. Systems based on aqueous LiOH electrolytes offer an interesting alternative. As previous research on alkaline fuel cells provides a good basis for modeling oxygen reduction electrodes in alkaline media [2], it is surprising that only non-aqueous systems are tackled in the modeling literature so far [1,3].

The model includes a multi-phase description of the cathode, where solid (graphite, reaction products, catalysts), liquid (electrolyte), and gaseous (oxygen/air) phases coexist. We take into account molecular oxygen dissolution in the electrolyte, oxygen reduction and lithium oxidation at the two electrodes, formation of dissolved LiOH, and precipitation of solid LiOH•H<sub>2</sub>O in the porous electrode and separator. Simulations show how and where the discharge products accumulate in the porous structure, leading to transport and therefore capacity limitations by clogged pore space.

The performance of the system can be greatly improved by gas diffusion electrodes (GDE). Oxygen transport to the active sites is enhanced and allows for higher power densities, which are needed in automotive applications. We model capillary-pressure driven transport of the liquid electrolyte in the GDE and discuss the dynamic equilibrium between the liquid electrolyte and gaseous oxygen. Capacity limitations due to the inhomogeneous precipitation of LiOH•H<sub>2</sub>O are predicted by the simulations. Model parameters are calibrated and validated against in-house experimental data of half-cell experiments at model GDEs in an aqueous one molal LiOH solution.

The simulation methodology provides detailed insight into capacity and rate limitations in aqueous Li-O cells, allowing for a knowledge-based optimization of the system.

### Literature:

[1] J. Christensen, P. Albertus, R.S. Sanchez-Carrera, T. Lohmann, B. Kozinsky, R. Liedtke, J. Ahmed, A. Kojic, J. Electrochem. Soc., 159 (2012) R1-R30. [2] F. Bidault, D.J.L. Brett, P.H. Middleton, N.P. Brandon, Journal of Power Sources, 187 (2009) 39-48. [3] S.S. Sandhu, J.P. Fellner, G.W. Brutchon, Journal of Power Sources, 164 (2007) 365-371.