Multi-scale simulation of transport processes in metal-oxygen batteries

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Li-Ion batteries are commonly used in mobile electronic devices. However, for some applications, such as electric vehicles (EVs), the energy density is not sufficient [1]. Due to their high theoretical capacity metal-oxygen batteries, e.g. Li-O_2, Zn-O_2, Si-O_2, Mg-O_2, Na-O_2, have the potential to replace Li-ion batteries in next-generation battery powered EVs. In this context systems employing aqueous electrolytes received a renewed interest [2,3]. One of the advantages of aqueous systems is the use of the well-developed concept of gas diffusion electrodes (GDEs) at the cathode. In this kind of electrode a network of hydrophobic binder ensures the coexistence of gas and liquid phase which guarantees a sufficient supply of O_2 from the gas phase even at high discharge rates.

In our contribution we present results bridging the length scales between pore-scale transport and full-cell battery simulations. In a first step 2D and 3D lattice Boltzmann simulations [4,5] are conducted on tomographic reconstructions of Ag model GDEs. The 3D simulations yield information about the distribution of the liquid electrolyte in the porous structure of the GDE. This result can be used to extract effective transport parameters and specific active surface areas at various saturations. Moreover, the simulations are used to obtain p_c-s_w characteristics of the porous media. We demonstrate that computationally less demanding 2D simulations are an efficient tool for the screening of the wetting properties of different new electrode materials and geometries.

The LBM simulations deliver important input parameters for physics-based continuum simulations on the electrode and cell level [2,6]. The continuum models take into account the dissolution, transport and reduction of O_2 in the gas and liquid phase as well as the formation of solid reaction products. A 1D transport model was validated against IV curves and electrochemical impedance spectra recorded in in-house half-cell measurements. We can report excellent agreement between simulation and experiment for various conditions and electrode structures. Finally, the transport model can be inserted at the cathode of virtual metal-O_2 cells for a simulation of discharge characteristics [7].

The multi-scale simulation methodology presented in this work allows predictions of cell performance based on the structural information of the electrode. Therefore, the presented work constitutes an important contribution for a systematic improvement of aqueous metal-O_2 batteries.

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