

Continuum modeling of metal-sulfur batteries: Li-S and beyond

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The demand for electrochemical storage technology with high energy density increased rapidly in recent years. A major driving force is the electrification of the mobility sector. High expectations rest on the development of beyond Li-Ion battery systems [1]. Especially, lithium-sulfur batteries (Li-S) are believed to be a promising candidate already in the near future. However, degradation mechanisms like the polysulfide shuttle hinder a commercial breakthrough. A thorough understanding of the fundamental processes will be a key factor for future developments and the overall success of Li-S batteries.

In this talk we will present simulation results based on continuum models of Li-S batteries which were developed in our group [2], [3]. Our models describe the reaction and transport of solid and dissolved sulfur species in a single S/C particle as well as on cell level. A focus is set on the redistribution of the solid end-products S_8 and Li_2S upon cycling which includes a study of the transport mechanisms of polysulfides in solution.

An effect which is often neglected in battery modelling is the desolvation of ions prior to the electron transfer reaction. Recently, we developed a generic theory for charge and electron transfer reactions at electrified interfaces, which is able to describe charge accumulation at the electrode-electrolyte interface [4]. The model is derived from fundamental thermodynamic and electrostatic laws without any assumptions on structural properties of the double-layer. This concept is now transferred to the conversion reactions in Li-S batteries. This approach allows us to include surface effects, like adsorption and desolvation in our simulations. During discharge the overall concentration of charged species increases and raises the viscosity and cell resistance [5]. Therefore, surface effects are expected to be even more important in the case of Li-S batteries. Furthermore, these processes will be important for divalent ions like Mg^{2+} in magnesium-sulfur (Mg-S) batteries [6]. The concepts and transport mechanism of Li-S batteries described above were adopted for the modelling of Mg-S batteries and we will briefly present our latest results in this field.

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