A Common Framework for the Simulation of Next-Generation Metal-Sulfur Batteries

Raphael Richter[1,2], Joachim Häcker[1] ,Timo Danner[1,2] , Norbert Wagner[1], Andreas Friedrich[1,3] and Arnulf Latz[1,2,4]

[1]German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Stuttgart Germany
[2]Helmholtz Institute Ulm for Electrochemical Energy Storage (HIU), Ulm, Germany
[3]University of Stuttgart, Institute for Energy Storage, Stuttgart, Germany
[4]University of Ulm, Institute of Electrochemistry, Ulm, Germany
Helmholtzstraße 11, 89081 Ulm, GermanyAddress
E-Mail: raphael.richter@dlr.de

Modern Lithium-ion batteries based on intercalation chemistry hold more than twice as much energy by weight and are ten times cheaper than the first commercial versions sold by Sony. But today they are near its limits. Metal anodes for 'beyond Li-Ion' batteries, such as Lithium-sulfur or Lithium-air, promise higher energy density and lower cost. However, the formation of dendrites is a major security risk and prevented a commercialization of the technology in large scales.

Magnesium in term can be directly used as anode material due to its dendrite-free deposition and thus increases the safety as well as energy density. Two electrons are stored per Mg atom which compensates the generally lower discharge potential of magnesium-based cell chemistries. In recent years magnesium-sulfur batteries are discussed as an attractive next-generation energy storage system and provides a high capacity of 3832 mAh/cm^3 and 2230 mAh/g with an energy density of over 3200 Wh/l [1]. Such an energy density is beyond that of lithium-sulfur batteries and is, therefore, very promising for automotive and stationary applications. Furthermore, magnesium and sulfur are both naturally abundant, low in price and non-toxic.

However, magnesium-sulfur batteries are in a very early stage of research and development. The system suffers from similar problems like the early Li-S batteries which are a low coulombic efficiency and cycle life, mainly associated with the well-known polysulfide shuttle. Moreover, the reactions at both the positive and negative electrode are not yet fully understood but similar sulfur reduction mechanisms are generally assumed [2].

In order to harvest the conceptual similarities between lithium and magnesium sulfur batteries we formulate a common framework for metal sulfur batteries. In a multiscale approach we describe the processes in sulfur host materials (e.g. meso/mricoporous carbons) and on cell level (1+1D). The transport of dissolved species is modeled by the Nernst-Planck equation and sulfur red/ox kinetics are described by a reduced mechanism which is able to reproduce the key experimental results for Me-S batteries [3]. Therefore, the model is able to capture the kinetics of sulfur redistribution in the cell during cycling driven by the polysulfide shuttle. By taking into account side reactions at the negative electrode we are able to describe the experimentally observed decrease in coulombic efficiency and capacity.

In our presentation we focus on the investigation of common properties of Li-S and Mg-S batteries, and more importantly key differences between the two. The simulation results will be compared to in-house experimental data measured on sulfur/carbon composite electrodes, such as galvanostatic cycling data. For these experiments the Mg-S and Li-S cells under investigation are identical with respect to dimensions, cathode material and electrolyte solvent. The insights gained from these are used to focus the simulations on aspects specific for Mg-S batteries. In close collaboration with the experimental groups at DLR and HIU we aim at guiding new developments of the Mg-S system.

References:

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