A novel Modeling Approach for Metal-SPAN Batteries

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Metal-sulfur (Me-S) batteries present a promising class of next-generation batteries with very high theoretical capacity. In recent years, magnesium (Mg) was proposed as anode material for Me-S batteries due to negligible dendrite formation and high volumetric capacity (3,837 mAh/cm^3) [1]. This capacity is even higher than in the Li system (2,062 mAh/cm^3) which is very attractive for portable applications. However, similarly to Li-S batteries, Mg-S batteries show a low coulombic efficiency and fast self-discharge due to the polysulfide shuttle.
In order to reduce the polysulfide shuttle several mitigation strategies have been developed for Li-S batteries and some of these concepts have been also transferred to Mg-S batteries [2]. One promising approach is to covalently bond the sulfur to a polymer backbone. Long cycle life and high specific capacities could be shown for sulfurated Poly(acrylonitrile) (SPAN) cathodes in lithium-based batteries and more recently the proof-of-concept was also demonstrated for Mg-SPAN batteries [3].
In our contribution, we will present a novel continuum model for SPAN electrodes and demonstrate its application in Li and Mg-SPAN batteries. Within our simulation framework [4] we are able to include both red/ox reactions of covalently bond sulfur on PAN as well as transport and electrochemical reactions of polysulfides in solution. By comparing our simulation results to experimental data, we are able to identify qualitative differences in the sulfur reduction mechanism between the Li and Mg based system. In collaboration with our experimental partners, we aim to provide more insights on degradation mechanisms and limiting factors for battery performance which are able to guide new developments for Me-SPAN batteries.

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