A new continuum model of Metal-Sulfurized Polyacrylonitrile (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 (3837 mAh/cm^3) [1]. This capacity is even higher than in Li-based systems (2062 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 bind the sulfur to a polymer backbone. Long cycle life and high specific capacities have been demonstrated for sulfurated poly(acrylonitrile) ("SPAN") cathodes in lithium-based batteries and, more recently, the proof-of-concept was also demonstrated for Mg-SPAN batteries [3,4].

In our contribution we present a novel continuum model for SPAN electrodes and demonstrate its application to Li-SPAN and Mg-SPAN batteries. Within our simulation framework [5] we are able to include both red/ox reactions of sulfur covalently bound to the polymeric backbone of SPAN and transport as well as electrochemical reactions of the polysulfides in solution. Additionally, we model side reactions on the negative electrode and precipitation of the solid discharge products. By comparing our simulation results to experimental data, such as the cell voltage during galvanostatic cycling, we are able to identify qualitative differences between the Li- and Mg-based systems.

The simulations provide insights on limiting factors for battery performance, which is the basis to guide new developments for Me-SPAN batteries.

***Keywords**:* Continuum modelling, kinetics, rechargeable metal-sulfurized polyacrylonitrile (SPAN) batteries

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