A novel continuum model and simulation study of metal (Mg & Li) -sulfur battery with sulfurized polyacrylonitrile (SPAN) cathodes

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Magnesium-sulfur (Mg-S) batteries are considered promising contenders for the next generation of batteries due to a set of advantages: high gravimetric energy density, high abundancy of both Mg and S, absence of scarce elements such as nickel or cobalt, and a reduced tendency to dendrite formation [1]. Unfortunately, similar to Lithium-Sulfur batteries, Mg-S batteries show a low coulombic efficiency and fast self-discharge due to the polysulfide shuttle. Several mitigation strategies to reduce the polysulfide shuttle effect, have been developed for Li-S batteries and some of these concepts have been also transferred to Mg-S batteries [2]. One of the promising approaches 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 shown for Mg-SPAN batteries [3,4]. We present a novel continuum model for both Li & Mg-SPAN batteries, which includes redox reactions of sulfur covalently bound to the polymeric backbone of SPAN, species transport, and electrochemical reactions of the polysulfides in solution. The model requires a number of parameters which however can beextracted from structural and electrochemical characterization. Using our model we performed simulation studies screening the influence of different parameters on the cell performance. We demonstrate that the morphology of the electrode is essential for Mg-SPAN battery performance. In our contribution we aim on providing more insights into the degradation mechanisms and limiting factors for battery performance, which are able to guide new developments for Mg-SPAN batteries

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