A NOVEL CONTINUUM MODEL AND SIMULATION STUDY OF MAGNESIUM-SULFUR BATTERY WITH SULFURIZED POLYACRYLONITRILE (SPAN) CATHODES

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Magnesium-Sulfur (Mg-S) batteries are considered as promising contender for the next generation batteries due to its set of advantages: high gravimetric energy density (3.837 mAh/cm^3), its abundancy, absence of scarce elements such as nickel or cobalt, and its reduced tendency to dendrite formation [1]. Unfortunately, sulfur-based batteries suffer from low coulombic efficiency and fast self-discharge due to the polysulfide shuttle. Several mitigation strategies to reduce the polysulfide shuttle effect have been developed and 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 for Mg-SPAN batteries [3,4].

We present a novel continuum model for Mg-SPAN battery, which includes both red/ox reactions of sulfur covalently bound to the polymeric backbone of SPAN, species transport, and electrochemical reactions of the polysulfides in solution. The model parameters are extracted from structural and electrochemical characterization, such that the measured discharge curves are reproduced. In our study, we discovered that for a case of denser electrode, the capacity faded quite significantly as the tortuosity value increases and hinders the diffusion process. This shows that the morphology study of electrode essential for upscaling an SPAN cathode based battery. In collaboration with our experimental partners, 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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References:
[1] Z. Zhao-Karger, M. Fichtner: “Magnesium–sulfur battery: its beginning and recent progress”, MRS Commun. 2017, 770 - 784.
[2] P. Wang, M. R. Buchmeiser: “Rechargeable Magnesium–Sulfur Battery Technology: State of the Art and Key Challenges”, Adv. Funct. Mater. 2019, 1905248.

[3] P. Wang, M. R. Buchmeiser: “High-Performance Magnesium-Sulfur Batteries Based on a Sulfurated Poly(acrylonitrile) Cathode, a Borohydride Electrolyte and a HighSurface Area Magnesium Anode”, Batteries & Supercaps 2020, 10.1002/batt.202000097.
[4] P. Wang, M. R. Buchmeiser: “Characteristics of magnesium-sulfur batteries based on a sulfurized poly(acrylonitrile) composite and a fluorinated electrolyte”, Electrochimica Acta 2020, 361, 167024.
[5] R. Richter, A. Latz: “Insights into Self-Discharge of Lithium– and Magnesium–Sulfur Batteries”, ACS Applied Energy Materials. 2020, 3, 9, 8457-5474.