

Insights into the Sulfur Dissolution and Anode/Electrolyte Interface in Mg-S Batteries by operando UV/Vis and Impedance Spectroscopy

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Among the various post-lithium battery systems, the magnesium-sulfur battery represents a promising candidate due to its high energy density, improved safety and abundance of the applied raw materials. The underlying mechanisms are similar to those of a lithium-sulfur cell – analogously facing the challenge of sulfur retention in the cathode to mitigate the polysulfide shuttle. However, in contrast to lithium metal, reactions at the Mg metal surface – namely the reduction of sulfur or electrolyte species – might lead to anode passivation and localized stripping/plating behavior, inducing high overpotentials and dendrite formation, respectively.

To gain insights into the sulfur and polysulfide dissolution, operando UV/Vis spectroscopy and imaging were applied during OCV and cycling.¹ Instant sulfur dissolution was observed which leads to severe self-discharge by its reduction at the magnesium anode to polysulfides (S_6^{2-}/S_4^{2-}). The concentration of these polysulfide species in the electrolyte is reversibly declining during cycling with a certain amount being reduced to magnesium sulfides precipitating on the anode surface.² To analyze these reactions at the anode/electrolyte interface and identify the contribution of sulfur to the surface layer formation, galvanostatic electrochemical impedance spectroscopy was utilized during Mg-Mg and Mg-S cell operation. Thus, the high-ohmic adsorption layer is avoided enabling the investigation of the practical impedance response of a Mg anode during stripping and plating. To mitigate the self-discharge and active material loss, an artificial solid electrolyte interphase (SEI) was introduced and analyzed in addition to the native SEI of a bare magnesium foil to explore their contribution to the interfacial resistances.

References

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2. J. Häcker, C. Danner, B. Sievert, I. Biswas, Z. Zhao-Karger, N. Wagner, K. Andreas Friedrich, Electrochimica Acta 338, 135787 (2020)