

Continuum Modelling as Tool for Optimizing the Cell Design of Magnesium Batteries

Janina Drews^{1,2}, Piotr Jankowski^{3,4}, Joachim Häcker¹, Rudi Ruben Maça Alaluf⁵, Liping Wang^{2,6}, Zhenyou Li^{2,6}, Johannes Wiedemann^{1,2}, Juan Maria García Lastra³, Tejs Vegge³, Norbert Wagner¹, K. Andreas Friedrich^{1,7}, J. Alberto Blázquez⁵, Zhirong Zhao-Karger^{2,6}, Maximilian Fichtner^{2,6}, Timo Danner^{1,2}, and Arnulf Latz^{1,2,8}

¹German Aerospace Center (DLR), Institute of Engineering Thermodynamics, Stuttgart, Germany

²Helmholtz Institute Ulm for Electrochemical Energy Storage (HIU), Ulm, Germany

³Technical University of Denmark (DTU), Department of Energy Conversion and Storage, Lyngby, Denmark

⁴Warsaw University of Technology (WUT), Faculty of Chemistry, Warsaw (Poland)

⁵CIDETEC Energy Storage, Basque Research and Technology Alliance (BRTA), Donostia-San Sebastián (Spain)

⁶Karlsruhe Institute of Technology (KIT), Institute of Nanotechnology, Eggenstein-Leopoldshafen, Germany

⁷University of Stuttgart, Institute of Energy Storage, Stuttgart, Germany

⁸University of Ulm, Institute of Electrochemistry, Ulm, Germany
Wilhelm-Runge-Straße 10, 89081 Ulm, Germany
janina.drews@dlr.de

For a successful commercialization of magnesium batteries there are still some challenges to overcome. The high charge density of the bivalent cation causes strong coulomb interactions with anions and solvent molecules. Therefore, Mg salts are prone to form ion pairs and bigger clusters – especially at high concentrations, which may adversely affect the transport in the electrolyte and the electrochemical reaction at the electrode.^[1] Moreover, energetic barriers for desolvation and solid-state diffusion of the double-charged magnesium ion are usually very high, which can have a crucial impact on the battery performance. Former can significantly hinder the electron-transfer reaction,^[2] whereas latter makes the choice of suitable cathode materials very challenging. For instance, it is well-known that the morphology of an intercalation material can strongly influence the battery performance and smaller particles as well as thinner electrodes are common strategies for avoiding adverse effects of transport limitations. However, high mass loadings as well as suitable separators are still essential bottlenecks for commercialization of magnesium-ion batteries.

Up to date Chevrel phase (CP) Mo₆S₈ is considered as benchmark intercalation cathode and Mg[B(hfip)₄]₂ / DME is seen as most promising chloride-free magnesium electrolyte. In our contribution we carefully study this model system of a Mg-ion battery to get a better understanding of how to overcome undesired limitations. Therefore, we present a newly-developed continuum model, which is able to describe the complex intercalation process of Mg²⁺ into a CP cathode. The model considers not only the different thermodynamics and kinetics of the two intercalation sites of Mo₆S₈ and their interplay but also the impact of the desolvation on the electrochemical reactions and possible ion agglomeration. The parameterization and validation of the model is based on DFT calculations and experimental data. All in all, the combination of different modelling techniques with experimental measurements provides important insights into the operation of Mg-ion batteries and enables an optimization of the cell design.

References

- [1] J. Drews, T. Danner, P. Jankowski et al., ChemSusChem, 3 (2020), 3599-3604.
- [2] J. Drews, P. Jankowski, J. Häcker et al., ChemSusChem, 14 (2021), 4820-4835.