Zinc Batteries with Novel Electrolytes

Birger Horstmann, Tobias Schmitt, Max Schammer, Simon Clark, and Arnulf Latz

German Aerospace Center, Pfaffenwaldring 38–40, 70569 Stuttgart, Germany.

Helmholtz Institute Ulm, Helmholtzstraße 11 89081 Ulm, Germany.

Ulm University, Albert-Einstein-Allee 47 89081 Ulm, Germany.

*birger.horstmann@dlr.de

Zinc metal is a non-toxic, cheap, and abundant anode material with a great tradition in battery applications. Today, zinc batteries are among the few candidates for efficient and low-cost devices required for stationary grid storage. The development of rechargeable zinc batteries with high capacity and high cycling stability is challenging because of hydrogen evolution, dendritic growth of zinc, and low tolerance to ambient air. Commercial zinc batteries employ aqueous alkaline or aqueous neutral electrolytes. We investigate room temperature ionic liquids (RTILs) as alternative concepts for zinc-ion batteries and neutral aqueous solvents for zinc-air batteries. Previously, we presented a modeling framework for metal batteries and a methodology to derive a consistent description of electrolyte transport based on non-equilibrium thermodynamics. Here, we combine these approaches to model zinc batteries and compare our simulation results with experimental measurements. These models describe the cycling behavior of macroscopic battery cells as well as microscopic surface phenomena.

RTILs were shown to minimize dendrite growth and allow cycling of zinc ion batteries. We derive the first consistent electrolyte transport theory for RTILs that takes into account the important charge and volume constraints in these molten salts. We validate our transport theory against a Prussian blue battery cell. The electrochemical double layer of RTILs shows a quasi-crystalline surface structure. Based on the hardcore nature of molecules, our model explains this structure. To summarize, our model accurately describes macroscopic as well as microscopic effects.

Neutral aqueous solvents enable a long cycle life and homogeneous zinc deposition for zinc-air batteries, too. For coping with the variety of aqueous zinc-ligand complexes in the ZnCl₂-NH₄Cl electrolyte, we assume fast homogeneous reactions and introduce a quasi-particle concept. We find that the distribution of zinc-ligand complexes shifts significantly with small changes in concentration. Our model shows that the pH may strongly vary locally and accelerate cell degradation. We optimize the cell architecture to maintain a stable, neutral pH during cycling.

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