Designing Aqueous Electrolytes for Next-Generation Zinc-Air Batteries

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In the on-going pursuit of high-performance and sustainable energy storage, zinc-air batteries are among the most promising systems under development. With a theoretical energy density over 6000 $Wh*L^{-1}$, ZABs could play a pivotal role in grid-scale renewable energy storage and electric mobility applications. Unfortunately, both the electrical rechargeability and calendar lifetime of ZABs are limited. New electrolyte materials are needed to overcome these challenges. The application of physics-based numerical modeling and simulation can accelerate the development of ZAB materials and cell architectures [1].

We have derived and implemented 0D thermodynamic models and 1D continuum models to evaluate the performance of ZABs with a range of electrolyte compositions. Our models allow us to determine the equilibrium composition of aqueous electrolytes and the solubility of various precipitates. We then simulate the dynamic and spatially resolved 1D concentration and multi-phase volume fraction profiles that develop in a ZAB cell during operation. This method can be applied to better understand the performance of current alkaline and near-neutral ZABs and predict next-generation designs.

Industry-standard primary ZABs currently contain aqueous alkaline KOH electrolytes. These batteries have good discharge characteristics, but the Zn electrode changes shape when cycled and the electrolyte slowly degrades as it absorbs CO_2 from air. The lifetime of these cells is limited to just a few months [2]. Aqueous near-neutral electrolytes have been proposed to address these challenges. ZABs with pH-adjusted NH_4Cl-$ZnCl_2$ have shown good cycleability and are not subject to carbonation [3, 4, 5]. But the strongly oxidizing nature of chlorine and the precipitation of unwanted solids hinders the development of a robust and high energy-density battery [6]. In spite of these challenges, aqueous near-neutral ZABs are beginning to be commercialized (most notably by *Eos Energy Storage* [7]) for some large-scale stationary applications.

In this contribution we present a theory-based method for evaluating the performance of current systems and designing new aqueous electrolytes for next-generation ZABs. Beginning with fundamental thermodynamics and moving up to cell-level engineering, we identify the important characteristics required for new electrolyte materials and highlight an example of electrolyte design in action. By applying a rational design method, we show the promising potential for long-life rechargeable ZABs based on abundant and non-toxic materials and indicate topics for future research.

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