Modelling Zinc Batteries with Ionic Liquid Electrolytes

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Room temperature ionic liquids are promising candidates for stable battery electrolytes [1]. Their advantages comprise a large electrochemical window (up to 6V), chemical and thermal stability, non-flammability (as safety asset) and low vapor pressure [2]. Ionic liquids minimize dendrite growth during electrodeposition, have high ionic conductivity and exhibit a plethora of tunable realizations [3]. In particular, ionic liquids possess beneficial properties to overcome technical problems associated with zinc air batteries which have yet to be resolved [4]. Zinc-air batteries with ionic liquids are potentially stable towards moisture as well as carbon dioxide and can support a reversible oxygen electrochemistry [5].

Theoretical studies and simulations are efficient means for the evaluation of materials and for improving the design of zinc-air batteries [6]. Here, we present a thermodynamically consistent transport theory of room temperature ionic liquids. Our theory describes ionic liquids in a continuum framework. The focus lies on the systematic derivation of a set of fully coupled transport equations for the ions, charge and heat based on the development of modeling methodologies for lithium-ion batteries [7].

Upon this framework we model a zinc ion battery with a mixture of ionic liquid and water as electrolyte. This setup is described experimentally in Ref. [8]. The battery comprises a zinc anode and a Prussian blue analogue cathode in combination with a choline-acetate-water mixture, [Ch]OAc+30%wt water. [Ch]OAc, is a biodegradable, biocompatible, and inexpensive ionic liquid. The battery is simulated along one dimension and good agreement with the experimental observations described in Ref. [8] is found.

For the understanding of microscopic structures, we incorporate the hardcore particle nature of the medium into our transport theory. In the vicinity of an electrified interphase, we find a quasi-crystalline structure with crowding and overscreening for a neat ionic liquid [9]. Recently, we examined the influence of a minor additive species (Ag[TFSA]) on the interfacial structure of the neat ionic liquid Pyr[TFSA] in a joint experimental/theoretical study [10]. Our theory predicts the minimal minor salt concentration which is needed to destroy the layered structure. The theoretical predictions are in good agreement with experimental observations.

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