Modeling of Bulk and Surface Effects in Ionic Liquids

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Theoretical studies and simulations are efficient means for the evaluation of materials and for improving the design of electrochemical devices [1]. Here, we present a thermodynamically consistent transport theory of ionic liquids. Our approach is based on a continuum framework and rigorous physical assumptions and we derive a set of coupled transport equations for the ions, charge and heat, based on modeling methodologies for lithium-ion batteries [2]. It describes the bulk phase of ionic liquid electrolytes. In addition, we incorporate microscopic interactions into our transport theory.

Upon this framework we model a zinc-ion battery with a mixture of ionic liquid and water as electrolyte, described experimentally in Ref. [3]. The battery is simulated along one dimension and good agreement with the experimental observations is found (see Fig. 1, left). We integrate the particle nature of the medium into our transport theory. Near electrified interfaces, this leads to crowding and overscreening [4] for neat ionic liquids (see Fig. 1, right). Recently, we have 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 [5]. 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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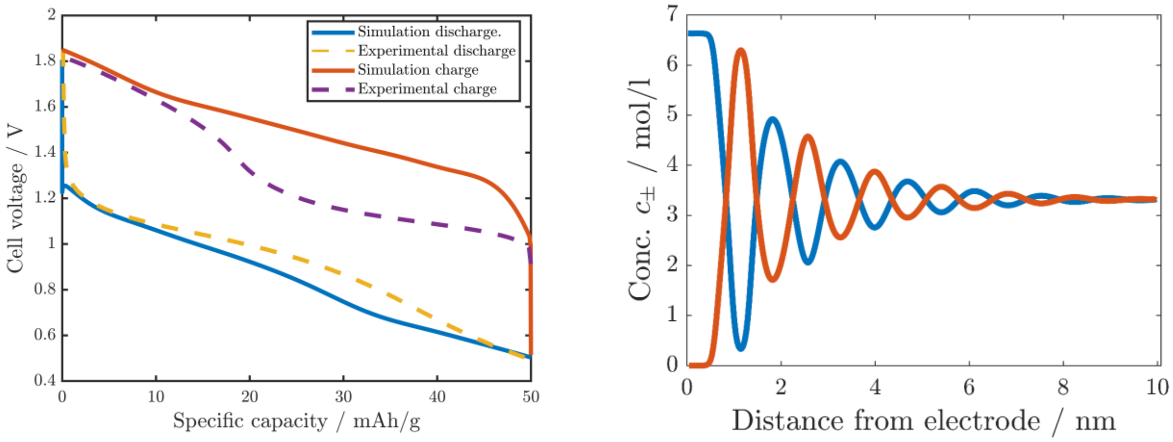

Figure 1. Left: Discharging and Charging of zinc ion battery. Right: Alternating layers of ions near electrified interface (overscreening).

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