Modeling Bulk and Surface Dynamics of Ionic Liquids in Batteries

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The quest for safer and cheaper batteries motivates research on novel electrolytes. Room temperature ionic liquids (RTIL) promise improved electrolyte stability [1]. The smooth electrodeposition of metals from RTILs constitutes an important step towards their application.

The breakthrough of RTILs as electrolytes is hindered by their complex speciation in the bulk and their interfacial structure formation. Continuum theories predict the multilayer structure of the electrochemical double layer in ionic liquids [2]. These models highlight the fundamental relations behind the multilayer structure by neglecting microscopic details. In this way, they connect meso-scale surface and macro-scale bulk properties of RTILs.

We derived a rigorous physics-based model from non-equilibrium thermodynamics [3]. Our model goes beyond previous continuum models as it takes into account the full hardcore interaction between molecules and describes the effect of additives, e.g., water or salt. Firstly, we derive transport equations for RTILs in the bulk taking into account mixtures with water or silver ions. Secondly, we take into account hardcore interactions between ions as a strong repulsion between particles at atomistic. These microscopic details become relevant in the nano-sized electrochemical double layer.

We validate our transport theory against a zinc-ion battery [4] considering the speciation in the bulk [5]. Based on the hardcore nature of molecules, our model explains the effect of additives on the multi-layer surface structure as observed with atomic force microscopy [3]. Speciation takes a central role in this context.

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References

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