

Modeling Ionic Liquids: Transport Theory for bulk and Interface

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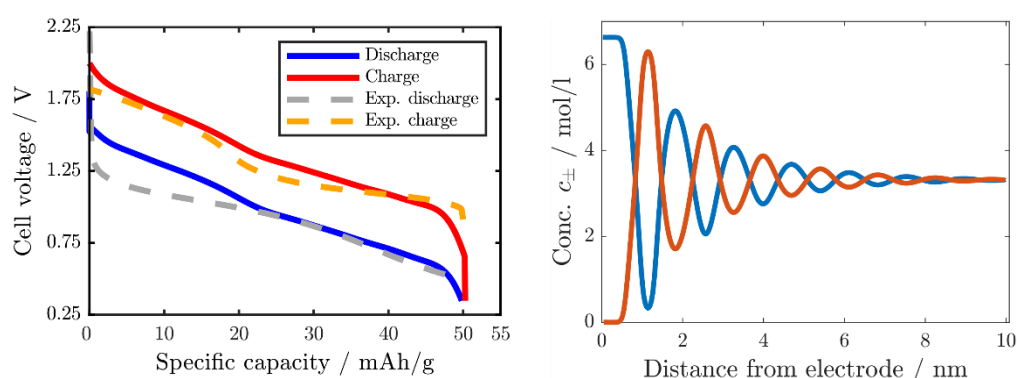
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Theoretical studies and simulations are efficient means for the evaluation of materials and for improving the design of electrochemical devices. Here, we present a thermodynamically consistent transport theory of ionic liquids (ILs). Our approach offers a holistic framework for the description of IL-electrolytes in the bulk phase and near electrified interfaces, thus spanning a wide range of length-scales from cell-level (micrometers) to microscopic interactions (nanometers).

The continuum theory is based on rigorous physical assumptions and provides coupled transport equations for ions, charge and heat, well-proved for lithium batteries [1].

Upon this framework we model a zinc-ion battery with a mixture of ionic liquid and water as electrolyte, described experimentally in [2]. This battery is simulated along one dimension and good agreement with the experimental observations is found (see Fig., left). We address the peculiar reference-dependence of the transport parameters using mass conservation and taking convection into account.

Integration of the particle nature of the medium into our theory leads to overscreening of neat ILs near electrified interfaces (see Fig., right). Recently, we examined the influence of a minor additive species (Ag[TFSA]) on the interfacial structure of Pyr[TFSA] in a joint experimental/theoretical study [3]. The results are validated by comparison with AFM-measurements. All theoretical predictions are in good agreement with experimental observations. Finally, we elucidate the numerical results by an analytical investigation, which reveals the influence of temperature and particle size on the interfacial structure. Thereby, we predict the emergence of three distinct phases of pure decay, damped-/ and undamped-oscillatory profile.



REFERENCES

1. Single, Fabian, Birger Horstmann, and Arnulf Latz. "Theory of Impedance Spectroscopy for Lithium Batteries." *The Journal of Physical Chemistry C* 123.45 (2019): 27327-27343.
2. F. Endres et al., *ACS Applied Materials and Interfaces*, 8 (2016), 12158–12164. doi:10.1021/acsami.6b01592.
3. V. Hoffmann, M. Schammer, F. Endres et al., *Physical Chemistry Chemical Physics*, 20(2018), 4760–4771. doi:10.1039/C7CP08243F.