Modeling Electrochemical Systems: From Fundamentals to Batteries

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The characterization of solid-liquid interfaces is of broad interest to electrochemistry since it is the place where electrochemical reactions occur. Several factors are influencing the nature of a given electrode/electrolyte interface such as the chosen metallic electrode, the solvent, the electrode potential, or the electrolyte. Also co-adsorbates from the electrolyte affect the physicochemical processes, *e.g.* adsorption and desorption, metal deposition, corrosion, and the kinetics of electrochemical reactions.
In this talk we will discuss how theoretical investigations that combine methods describing different length- and time-scales can be useful in determining the interfacial morphology and composition as well as ongoing electrocatalytic processes. In the first part, examples will be presented where *ab initio* studies combined with thermodynamic considerations helped in revealing the nature of the electrode under reaction conditions. Afterwards, electrode processes such as the electrochemical oxygen reduction reaction and the formic acid oxidation will be considered [1,2]. Based on extensive first principles-based calculations on the reaction intermediates and transition states, we will discuss the ORR mechanism on Pt(111) electrodes. After benchmarking our QM calculations to experimental data, we report a multi-pathway electrochemical ORR mechanism that is sensitive to reaction conditions, and specifically to the applied electrode potential. A simple analysis based on calculated rate constants remarkably reproduces experimentally known factors concerning the electrocatalytic ORR.
Finally, I will discuss our recently formulated continuum approach to investigate an all-solid-state lithium-ion battery [3], consisting of transport equations and intercalation reactions.

[1] J. A. Keith, T. Jacob, *Angew. Chem. Int. Ed.* **49**, 9521-9525 (2010).
[2] W. Gao, J. E. Mueller, Q. Jiang, T. Jacob, *Angew. Chem. Int. Ed.* **51**, 9448-9452 (2012).
[3] M. Landstorfer, T. Jacob, *Chem. Soc. Rev.* **42**, 3234-3252 (2013).

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ICP, Allmandring 3