Identifying and Closing Apparent Community Gaps in (Electro-)Chemistry

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This lecture comprises a case study of three examples, in which apparent community gaps between neighboring research fields in electrochemistry have been identified and resolved. The first example addresses the connection between experimental and theoretical electrocatalysis. In the past two decades, research in theoretical electrocatalysis has benefited tremendously from predictions on the free energy surface of electrocatalyzed reactions over single-crystalline model electrodes.[1] However, assessment and validation of the theoretical approach have been hampered by an apparent community gap in that experiments depict kinetic data mainly in the form of so-called Tafel plots, while theory provides free energy surfaces for the proposed reaction mechanism. This apparent community gap has been closed only recently by a dedicated evaluation scheme of experimental Tafel plots interpreted within the generalized Butler-Volmer formalism.[2] The evaluation of Tafel plots in terms of transition state free energies is a paradigm shift in electrocatalysis and allows constructing free energy diagrams along the reaction coordinate for any type of electrocatalytic reaction over single-crystalline model electrodes. This universal method is outlined and discussed at three prototypical electrocatalyzed reactions, namely chlorine evolution reaction (CER) and oxygen evolution reaction (OER) over RuO_2(110) as well as hydrogen evolution reaction (HER) on Pt(111).[3] Besides the dedicated Tafel analysis, the transition state free energy of the rate-determining reaction step can be obtained by temperature-dependent measurements, which is exploited for a kinetic study of gold nanoparticle synthesis according to a citrate-reduction route a la Turkevic.[4]

The second example tackles the connection between research in theoretical electrocatalysis and battery science. State-of-the-art research in electrocatalysis consists of three subsequent steps, namely first to resolve the surface structure of the electrocatalyst accurately by the ab initio thermodynamics approach in order to compile the thermodynamically most stable phases as function of the applied electrode potential that are employed in the second step for the investigation of the underlying kinetics by constructing the free energy landscape along the reaction coordinate. Based on the free energy landscape, microkinetic simulations can be performed in the third step.[5] Quite in contrast, the application of the ab initio thermodynamics approach in order to determine potential surface structures for subsequent kinetic investigations is omitted in lithium-ion battery (LIB) research. This finding is mainly related to the fact that hitherto a method that allows elucidating the concrete atomic surface structure of a LIB electrode material as function of the applied electrode potential is missing.[6] This apparent community gap has been closed by transferring the concept of surface Pourbaix diagrams from electrocatalysis to electrode materials in LIBs.[7] The underlying approach that relies on the concept of ab initio thermodynamics is exemplified and discussed at the example of a single-crystalline spinel lithium titanate (111) model electrode.[8]

A powerful approach for the identification of potential solid-state (electro-)catalysts for the application in practice based on *a priori* computational screening constitutes the construction of Volcano plots according to the Sabatier principle that allow quantifying the activity of a catalytic material as function of some property of the catalyst. However, Volcano plots have not found entrance into the investigation of battery materials yet. Within the third example, the concept of Volcano plots is transferred from catalysis to battery science by constructing Volcano curves for electrode materials in LIBs that consider both activity as well as stability.[9]

The lecture ends with a summary of recent advancements towards closing community gaps and provides an outlook of future research investigations.

References:

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