Machine Learning the Energetics of Electrified Solid-Liquid Interfaces

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In theory, accurate atomistic, computational studies of electrochemical systems in operando are necessary to understand fundamental processes at the heart of electrocatalysis. However, the large computational cost of ab initio methods limits the accessible time and size scales relevant for significant statistical sampling and thus comparison to experiments. While machine learning interatomic potentials (MLIPs) trained on ab initio data accurately predict energies and forces at a fraction of the computational cost, current architectures are unable to incorporate the biases introduced at applied potential conditions for typical electrocatalytic systems, where effects of the inner double layer play a critical role [1].

Here, we present the "Response Analysis in z-ORientation" (RAZOR) model [2], which includes the energetic effects of bias charges q via perturbation theory. While traditional MLIPs are trained on energies and forces in a bias-free environment, RAZOR also machine-learns the first-order energy change to bias charges q, the system's work function. Additional machine-learning of the atomic force derivative to q, a property related to Born effective charges, stabilizes the model. As a result, RAZOR is able to perform large-scale molecular dynamics simulations at different interfacial charges multiple orders of magnitude faster than ab initio methods. Furthermore, knowledge of the system's work function permits a straightforward translation of the electronically canonical "constant charge" ensemble to the experimentally relevant grand-canonical "constant potential" ensemble.

We demonstrate RAZOR's capabilities by investigating OH adsorption on Cu(100) in an implicit environment, showing the ability of "brute force" discovery of potential- and pH-induced adsorption site switching and explain the necessary thermodynamics for energetic comparisons [2]. Next, we show results for explicit Pt(111)-H_2O interfaces, highlighting important pitfalls to avoid when training RAZOR models with multiple layers of electrolyte [3]. Finally, as an outlook we discuss the physical importance of the atomic force-charge derivatives and the important information hidden within them.

References

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