O 24: Solid-liquid interfaces: Reactions and electrochemistry – Poster

Time: Monday 18:00–20:00 Location: P2

O 24.1 Mon 18:00 P2

Real-Time Electrochemical AFM Observation of Lithium Plating and Dissolution on HOPG: Surface Morphology and Mechanistic Insights — •Luca Kaufer[1,2], Daniel Ebeling[1], André Schirmeisen[1], and Jürgen Janek[2] — [1]Institute of Applied Physics, Justus-Liebig-University, Gießen, Germany — [2]Institute of Physical Chemistry, Justus-Liebig-University, Gießen, Germany

This investigation demonstrates the utilisation of atomic force microscopy (AFM) to examine the interactions between liquid electrolytes and Highly Ordered Pyrolytic Graphite (HOPG) electrodes. In particular, the focus is on the deposition of lithium on HOPG surfaces, a crucial process in lithium batteries. Atomic force microscopy measurements facilitate precise analysis of the surface structure, topography, and mechanical properties. In addition, they permit observation of dynamic changes during lithium deposition. This, in turn, provides a deeper insight into the mechanisms of electrode surface reactions and the quality of the electrode surface. Such measurements are of great importance to improve the efficiency and lifetime of batteries by helping to understand and control processes such as dendritic growth or non-uniform deposition.[1,2] We will demonstrate the use of Asylum Research's Fast Force Mapping mode in relation to the mechanical changes in HOPG during lithium deposition. [1] J. Phys. Chem. C 2011, 115, 25484*25489. [2] ACS Appl. Mater.Interfaces 2015, 7, 25441*25447

O 24.2 Mon 18:00 P2

Influence of electrolyte and electrode parameters on the current voltage characteristics during high voltage and plasma electrolysis — •Benjamin Schilling, Lukas Forschner, Justus Leist, Timo Jacob, and Albert K. Engstfled — Ulm University, Ulm, Germany

Electrolysis at gas evolving electrodes at high voltages, up to several hundreds of volts, leads to vivid bubble formation at the electrode, which ultimately leads to the ignition of a plasma in a gas sheath forming around the significantly smaller driving electrode. [1] Depending on the applied voltage, the approach can be used to tailor the electrodes structural properties or to modify the electrolyte, to form nanoparticles or generate H2O2. Systematic studies to elucidate the fundamental physical properties are still scarce [2], which are, however, highly relevant to understand the aforementioned changes on the electrode and electrolyte. In this work we focus on the influence of the electrolyte concentration and composition, voltage scan rate, working electrode geometry and material, electrode distance and electrolyte convection. The electrode structural properties are studied by SEM imaging, the dynamics of the gas film around the electrode are monitored using a high-speed camera and the temperature is proved locally by multiple temperature probes. By correlating these results, we discuss the parameters influencing the temporal changes of the system's properties and their consequences for the application of high-voltage electrolysis. [1] Yerokhin et al., *SURF COAT TECH* **122** (1999) 73-93. [2] Forschner et al., *J. Phys. D: Appl. Phys* **58** (2025) 215204.

O 24.3 Mon 18:00 P2

Predicting Potential-Dependent Binding Energies at the Electrified Metal/Water Interface — •Ann Kathrin Tran, Elias Diesen, Karsten Reuter, and Vanessa J. Bukas — Fritz-Haber-Institut der MPG, Berlin

Density-functional theory studies of electrified metal/water interfaces present many challenges and uncertainties. Even the methodology for computing the thermodynamics of adsorption is not yet well-established, albeit crucial to e.g. heterogeneous electrocatalysis. Here, we discuss common approaches and approximations in a systematic study of electrochemical binding energies. We choose Au(111) as our model system and focus on the adsorption of key intermediates during O_2 electro-reduction. Firstly, we benchmark predictions from three increasingly advanced models of the double layer: an applied saw-tooth potential in vacuum, an implicit solvent model, and explicitly modelled H_2O via *ab initio* molecular dynamics (AIMD). This comparison disentangles the purely electrostatic energy contribution from stabilization due to explicit H-bonding with the solvent. The latter contribution can be sizeable, yet is only captured by atomistic H_2O in expensive AIMD. We next address the issue of predicting grand-canonical energetics, that is, energies under realistic constant-potential conditions. In this context, we present workfunction information within constant-charge AIMD data, the emerging workfunction-charge relation, as well as different transformation techniques toward constant-potential results. This analysis reveals notable caveats for each approach and highlights the need for further systematic benchmark studies.

O 24.4 Mon 18:00 P2

molecular dynamics simulations of belite-water interactions using HDNNPs — •Usman Tafida[1,2], Maite Böhm[1,2], Henry Wang[1,2], Bernadeta Prus[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

Advances in the development of machine learning potentials (MLPs), such as High-Dimensional Neural Network Potentials (HDNNPs), have enabled *ab initio*-level accuracy for molecular dynamics (MD) simulations of high dimensional systems at greatly reduced computational cost. In this work, HDNNPs are trained on DFT-D3 energies and forces data of beta-belite (Ca_2SiO_4) clusters in water, which serve as model systems for complex oxide-water interfaces. Here, we present the dataset construction scheme, training and validation of the potential. Moreover, MD simulations have been performed to study the structural and dynamical properties of interfacial water at these clusters.

O 24.5 Mon 18:00 P2

High-Dimensional Neural Network Potentials for Molecular Dynamics Simulations of Mineral-Water Interfaces — •Maite Böhm[1,2], Bernadeta Prus[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

In recent years, High-Dimensional Neural Network Potentials (HDNNP) have emerged as a popular tool for atomistic simulations of complex systems such as mineral-water interfaces. In this work, we present the construction of an HDNNP based on density functional theory energies and forces for tricalcium aluminate (C_3A)-water interfaces. We discuss the development of a dataset that includes the relevant solid-liquid interface interactions by active learning and propose approaches for an accelerated extension of the dataset. The trained potential is applied in Molecular Dynamics simulations of the (100) surface of C_3A in contact with water, which are further investigated with respect to the structure and dynamics of water on the surface.

O 24.6 Mon 18:00 P2

Developing High-Dimensional Neural Network Potentials for Studying Tricalcium Silicate-Water Interfaces — •Henry Wang[1,2], Bernadeta Prus[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

The emergence of machine learning potentials (MLP) trained on energies and forces from electronic structure calculations has revolutionized the simulation of solid-liquid interfaces by molecular dynamics (MD). For instance, High-Dimensional Neural Network Potentials (HDNNP) have shown excellent accuracy for describing the interaction of water with several solid minerals. In this study, we investigate interfaces of liquid water with alite (Ca_3SiO_5), a calcium silicate forming several polymorphs, which is characterized by a particularly high reactivity with water. Insights into the behavior of water at calcium silicate surfaces at an atomistic level are crucial to reach a better understanding of general hydration reactions of this class of materials. An approach for realizing high-accuracy, large-scale MD simulations is presented.

O 24.7 Mon 18:00 P2

Exploration of the Pt(111)-water interface by high-dimensional neural network potentials — •Daniel Trzewik[1,2], Moritz R. Schäfer[1,2], Alexander L. Knoll[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

In-depth knowledge of solid-liquid interfaces is essential for understanding numerous catalytic and electrochemical processes. However, accurately describing these interfaces with first-principles methods is computationally demanding, limiting the system sizes and complexities that can be explored. Modern machine learning potentials offer a powerful alternative, delivering near-first-principles accuracy at a fraction of the cost. In this work, we use high-dimensional neural network potentials (HDNNPs) to examine the Pt(111)-water interface in detail. Trained on DFT reference data, these models enable molecular dynamics simulations which reveal the structural and dynamical behavior of water molecules at the interface.