CPP 33: Modeling and Simulation of Soft Matter IV

Time: Thursday 11:45–13:00 Location: H34

CPP 33.1 Thu 11:45 H34

Understanding COF/Electrode interfaces for electrocatalysis using DFT and molecular simulations — •Henrik Stooss[1], Philip Stärk[1,2], and Alexander Schlaich[1] — [1]Institute for Atomistic Modeling of Materials in Aqueous Media, Hamburg University of Technology, Hamburg — [2]SC SimTech, University of Stuttgart, Stuttgart

This study explores the complex dynamics at electrode/electrolyte interfaces under constant potential, crucial for advancing electrocatalysis and designing efficient energy systems, by combining advanced computational techniques to gain insights into mechanisms at these interfaces. We perform Density Functional Theory (DFT) simulations while maintaining a constant electrode potential. Despite challenges and computational costs, the DFT simulations provide insights into the electronic structure and behavior of electrode surfaces. We then parameterize a classical model based on the DFT data, enabling simulations of larger systems over longer timescales for comprehensive comparison with experimental ATR-SEIRAS data. This approach offers a detailed understanding of adsorption and transport phenomena at the electrode interface, potentially leading to better electrolyte compositions and improved electrode designs. This work advances the connection between first-principles calculations and experimental observations for material design.

CPP 33.2 Thu 12:00 H34

Porous microstructure of fibrous sheets in two transport regimes — Alexandra Serebrennikova[1], Phillip Gräfensteiner[2], Matthias Neumann[1], Volker Schmidt[2], Andoni Rodriguez[3], Peter Leitl[3], Werner Napetschnig[1], Ekaterina Baikova[1], Maximilian Fuchs[1], and •Karin Zojer[1] — [1]Graz University of Technology, Graz, Austria — [2]Ulm University, Ulm, Germany — [3]bionic surface technologies, Graz, Austria

In many applications, a porous material serves multiple functions. For example, paper sheets in packaging bags should allow excess air to escape quickly while minimizing moisture migration. Although the underlying physics are different, both transport processes depend on porosity. However, each function it is likely to be supported by additional, possibly different, microstructural properties. Can these microstructural properties be optimized for all functions or is this not possible due to inherently dependent properties? To answer this question for Stokes flow and reactive diffusive transport through paper, we simulate flow through μ-CT-determined microstructures using physics-informed neural networks, computational fluid dynamics, and pore network modeling. We combine these simulations with statistical morphological analysis including dependency quantification to provide the relevance and dependence of structural properties in both transport processes. Our study suggests that the two transport scenarios do not rely on the same set of structural properties, even when fiber swelling due to moisture transport is considered.

CPP 33.3 Thu 12:15 H34

Nuclear Quantum Effects in Clays — Sam Shepherd, Pawan Kurapothula, Nataly Realpe, Gareth Tribello, and •David Wilkins — Queen's University Belfast, Belfast, United Kingdom

Clay materials consist of layers, whose structure is heavily influenced by hydrogen-bonding interactions. Given the importance of nuclear quantum effects such as zero-point energy in water, a hydrogen-bonded liquid, a natural question to ask is how important these effects are in clays themselves.

I describe some work done by my group to understand the importance of nuclear quantum effects in clays and clay-water systems, and to interpret these effects in the same terms used to understand water. I also demonstrate a fully quantum-mechanical description of the interactions and dynamics in kaolinite clay.

CPP 33.4 Thu 12:30 H34

Protonated water clusters by stochastic approaches: probing machine learning resilience against quantum Monte Carlo noise — •Matteo Peria, Antonino Marco Saitta, and Michele Casula — Sorbonne Université, 4 place Jussieu Paris, France

A complete understanding of the hydrogen bond and proton transfer mechanism in water is still lacking, since it requires an accurate potential energy surface (PES) and very expensive quantum mechanical simulations of the nuclear part. Reproducing this high-dimensional surface with current high-level computational chemistry methods is infeasible for the largest clusters. We test gradient-based kernel ridge regression methods and neural networks to reproduce the PES starting from a dataset of energies and forces of the protonated water clusters obtained via simulations combining classical molecular dynamics (MD) for the nuclei and quantum Monte Carlo (QMC) for the electrons. The QMC+MD approach yields very accurate results for the classical dynamics, which are however affected by the intrinsic noise inherent in the stochastic sampling of both nuclear and electronic phase space. We prove that QMC multivariate noise is not necessarily detrimental to the learning of energies and forces and we determine under which conditions one can derive accurate and reliable MLIPs from QMC data.

CPP 33.5 Thu 12:45 H34

Effect of solid fillers on the thermodynamics and electrical properties of diblock copolymers and polymer blends. — •Alexander Chervanyov — University of Münster, Münster, Germany.

By making use of the developed theory we study the effect of solid fillers on the thermodynamics, phase behaviour and electrical properties of diblock copolymers (DBC) and polymer blends (PB). The theory relies on the combination of the liquid state approach, phase-field model for polymers, Monte-Carlo simulations, and the resistor random network model for fillers. Using the developed approach, we prove that the correlations imposed by the variations of the composition of PB cause a significant non-osmotic contribution to the polymer mediated interaction between fillers immersed in this blend. The effect of fillers on the stability and miscibility of compressible PB is studied in detail. We show that the presence of non-adsorbing fillers can be used to enhance the stability of a PB that shows low critical solution temperature (LCST) behavior. Finally, as an important practical application of the developed theory, we study the electrical response of an insulating DBC filled with conduictive fillers. In particular, the order-disorder transition in the host DBC system is found to be accompanied by the conductor-insulator transition in the filler network. The order-order transition between the lamella and cylindrical microphases of DBC proves to co-occur with a spike of the composite conductivity caused by restructuring of the conductive filler network.