

Electrical Double Layer at the Air–Water Interface: A machine-learning interatomic potential simulation study

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The air–water interface plays a central role in atmospheric chemistry, particularly in aqueous aerosols and microdroplets, where it can strongly influence chemical reactivity and enable catalytic processes [1,2]. A key open question is whether an electric double layer (EDL) can form at this interface. Although the air–water surface is overall neutral, differences in the surface affinities of cations and anions may lead to charge separation, analogous to the EDL observed at charged electrodes.

Such a structure would generate an interfacial electric field, which could contribute to the enhancement of chemical reactions in confined environments. However, probing this phenomenon experimentally remains challenging, and existing measurements provide indirect and sometimes conflicting evidence regarding both the presence of an EDL and the magnitude of the associated electric field [3-5].

Here, we address this problem using machine-learned interatomic potentials (MLIPs) trained at the hybrid density functional theory level. An iterative active learning strategy is employed to construct a representative and reliable training dataset [6]. These potentials enable nanosecond-scale molecular dynamics simulations of the air–water interface in the presence of sodium chloride salts over a range of concentrations.

Our simulations reveal how ion-specific interfacial organization gives rise to a structured double layer and an associated electric field. We validate our approach by computing key observables, including surface tension, free energy profiles, and vibrational sum-frequency generation spectra, achieving good agreement with experimental and theoretical benchmarks.

These results provide new molecular-level insights into ionic distributions at aqueous interfaces and their role in interfacial reactivity. More broadly, this work highlights the potential of machine learning approaches to deliver near first-principles accuracy for complex interfacial systems at significantly reduced computational cost.

References

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Figures

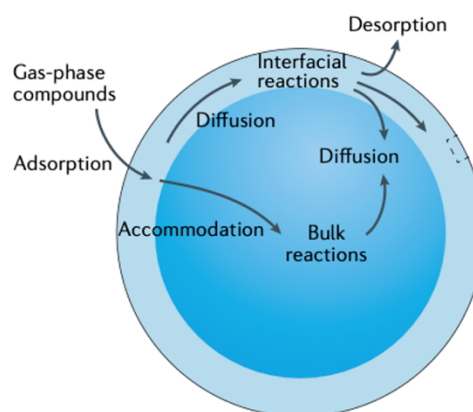


Figure 1. Processes at the air-water interface of water droplets [2]

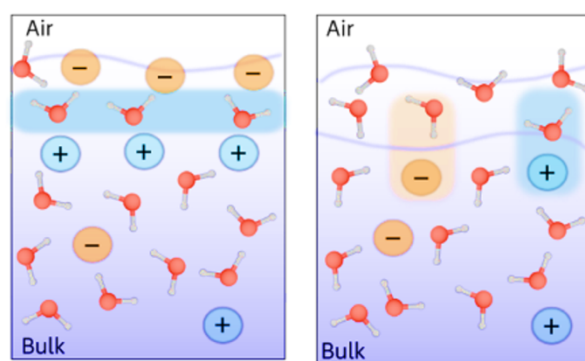


Figure 2. Schematic representation of proposed composition of ionic air-water interface [5]