

Understanding electrified electrolyte/electrode interfaces from atomistic simulations

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Modelling the interfaces between electrolytes and electrodes has been traditionally a difficult problem. From the simple Helmholtz model for the double layer developed almost 150 years ago, the complexity of these interfaces has been incorporated in successive models. Currently, realistic simulations with atomic detail based on classical interatomic potential are feasible and relatively common. Nevertheless, first principles methods (such as DFT) have not been very useful in dealing with these systems, as the number of atoms and the time scales involved exceed the practical capabilities of these methods. Besides, dealing with electrified electrodes poses practical problems in DFT, adding complexity to the calculations. I will describe our recent work trying to overcome these difficulties and to address problems at the electrified interface from first principles but also combining DFT calculations with more simplified models in the spirit of hybrid QM/MM calculations. I will also show some promising ideas of how to use Machine Learning methods to estimate directly the charge density in these interfaces, avoiding the expensive calculations involved in the DFT and QM/MM approach.

Figures

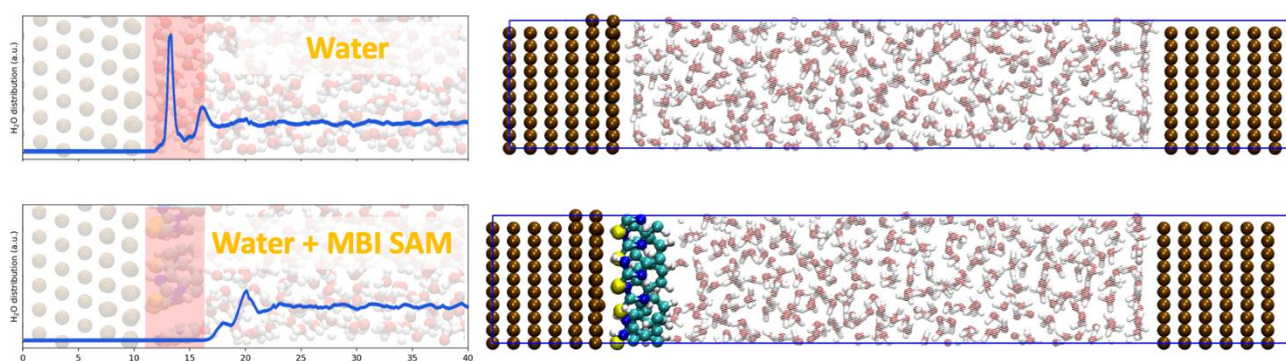


Figure 1: QM/MM simulation of water confined between two copper surfaces. Simulations without (top) and with a self-assembled monolayer (SAM) of corrosion inhibitor molecules (bottom) are shown. The inhibitor molecules are 2-mercaptobenzimidazole (MBI). The left panels show the distribution of water molecules as a function of position. The results in the lower panel show that water cannot penetrate the SAM, which therefore protects the copper surface from corrosion. The right panels show snapshots of the two molecular dynamics simulations. The SAM remained stable during the whole simulation time of 50ps.