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"Calculations of product selectivity in electrochemical CO_2 and N_2 reduction"

In this presentation I will give an overview of the latest results from our lab where we both use density functional theory calculations and experiments to understand electro-catalytic processes and to search for new catalysts for electrochemical reduction of molecules such as N_2 and CO_2. Firstly, I will present a detailed investigation of electrochemical reduction of CO_2 to hydrocarbons and alcohols on metal and metal oxide electrodes [1,2,3]. The calculations explain why copper is the only metal tested so far that can form significant yields of hydrocarbons and alcohols in the electrochemical CO_2 reduction while other metal electrodes mainly form H_2, CO or formate. We find a very good agreement between simulations and experiments by modeling explicitly the electrochemical interface between the charged electrode and solvated ions in the electrolyte where activation energies of all proton-electron transfer steps are calculated. This agreement between theory and experiments cannot be obtained when using simpler models, where only the thermodynamics of intermediate species are included and/or the electrode potential is modeled implicitly with the computational hydrogen electrode. From this new insight we have identified descriptors that can be used in a high-throughput screening for new materials where higher efficiencies towards hydrocarbons and alcohols can be achieved compared to H_2, CO and formate. Secondly, I will present a detailed computational screening study of metal nitride and metal oxide surfaces to search for electro-catalysts to reduce N_2 to ammonia in aqueous electrolyte at ambient conditions [4,5,6]. Promising results have been obtained and we have recently confirmed these predictions experimentally on one of the most promising candidates.

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