Operando modeling of materials as a function of reaction conditions using NNP

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" The identification and counting of active sites in heterogeneous catalysis became the ‘‘Holy Grail’’ of heterogeneous catalysis in 1925, and the situation remains the same today "

Operando modeling is one of the difficult aspects of computational chemistry, but it is crucial for understanding many important aspects of any reaction. This step, which involves transitioning from ideal simulations to more realistic simulations, is both theoretically challenging and computationally expensive. Traditional approaches to modeling high-coverage adsorbate-surfaces[1,2] often rely heavily on human intuition, which can lead to inefficient and incomplete sampling of the chemical space. Sampling in a desired chemical space can help in better understanding of a reaction .

The main focus of this project is to understand the reaction conditions and environment via surface coverage of mixed adsorbates on metal surfaces. Automatic surface reconstruction strategies will be used to achieve low-energy surface coverages .WE will use recent developments in Machine Learning Potentials (MLPs) and Message Passing Neural Networks (MPNNs) to overcome the computational challenges of classical DFT simulations for large systems. This is particularly true for machine-learning potentials, which offer a possible solution by building upon datasets obtained from numerous electronic-structure calculations ,and can be further improved with an active learning scheme. Enhanced sampling tools will be used to get better structures for active learning loop. Using these models to perform global optimizations like genetic algorithm or basin hopping will allow us to look at surface under the desired reaction conditions,without using any GCDFT reducing the computational cost. Another advantage lies in the automated workflow. With the help of workflow managing tools such as Aiiida[4] and Mkite[5]. We can automate the entire process of training a model and sampling for the active learning tool. Once these workflows are set up it can be employed with minimum human intervention reducing any possible human error.

During this project, we will leverage this new methodology to generate very large sets of structures for studying more realistic structures and replicating surface phase diagrams .Molecular dynamics will help us explore the desired chemical space for enhanced sampling .DFT simulations using GGA functionals like PBE enable detailed analysis of electrocatalysts' atomic-scale properties, offering insights into electronic structure, charge distribution, and reaction energetics. This aids in the design of more efficient catalysts for energy applications.

References

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