Machine Learning Potentials for Material Science

Construction of a Neural Network Potential for Supported Copper Clusters on Zinc Oxide Surfaces

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In Short

- Our group utilizes machine learning based neural network potentials to reproduce the potential energy surface of systems and perform simulations.
- These potentials are trained on thousands of single-point reference *ab-initio* calculations, and are able to approach the accuracy of the reference method with a computational efficiency close to classical force fields.
- Here we apply the potential to a model catalyst system, composed of copper and zinc oxide nanoparticles, utilized in the industrial synthesis of methanol.
- As a first approximation, we study structure and energy properties of copper clusters on zinc oxide surfaces. With the original project proposal, we obtained enough structures to construct a NNP to optimize up to ternary (Cu-Zn-O) clusters on different ZnO supports
- Computational time is required to extend the current potential to new configurations of the system, using as a reference method DFT calculations with the VASP code.
- These new configurations will allow us to study the formation of brass (Cu-Zn alloy) in large supported clusters, study the interface of coincident Cu and ZnO interfaces, and introduce H to our systems to form a 4-element neural network potential.

Methanol is one of the main base products in the chemical industry. The catalyst used for methanol production in industrial plants from synthesis gas (a mixture of H_2, CO and CO_2) consists of copper nanoparticles, which are supported by zinc oxide. The investigation of the structural properties of the catalyst by computer simulations is very challenging for the presently available theoretical methods, since a direct application of methods such as DFT is prohibitively expensive due to the large system size.

In order to study the large systems involved, efficient interatomic potentials are required. A possible solution to this dilemma is the application of artificial neural networks [1,2], which have been shown to provide potentials with close to DFT quality for the Cu/ZnO system [3]. Neural network potentials (NNP) are able to reproduce the potential energy surface (PES) of a system, combining the high accuracy of electronic structure methods with the efficiency provided by a simple but flexible functional form. NNs have several properties that make them ideal candidates for constructing PESs: they are smooth and continuous, they do not require any knowledge about the underlying functional form, they allow to make and break bonds, and analytic derivatives are available for the calculation of forces. The NNP needs to be trained on hundreds of single point *ab-initio* calculations, using the calculated energies and forces to learn the PES of the system.

An example of one of our neural networks can be seen in figure 1. An atomic neural network calculates the forces on one atom, and the energy contribution of that atom to the total system energy; multiple atomic networks are then added together to obtain the total system energy. The atomic neural network is divided into an input layer, one or more hidden layers, and an output layer. Each layer is composed of nodes, functions of different types, with parameters (multiplicative weights and additive biases) connecting the nodes. In the input layer, the nodes are composed of symmetry functions, which process the environment around the atom of interest to provide a unique numerical fingerprint. In the hidden layers, the nodes are non-linear activation functions (usually hyperbolic tangent) that provide flexibility to the network.

The network is trained by adjusting the parameters connecting all the different nodes, until the outputs of the network (energies and forces) match those in the reference dataset (single-point calculations using a given *ab-initio* method). For this, thousands of single-point calculations are required, each giving a sample of the PES of the system. Once so trained, the network is able to reproduce energies and forces accurately but at a fraction of the computational cost of an *ab-initio* calculation. This allows for longer simulations in larger systems to be performed, but keeping the accuracy of the reference method.

The currently available NNP is able to accurately reproduce many simple structural and energetic parameters for both bulk and slab structures of copper and zinc oxide. In the original proposal, structures were generated that allowed us to run genetic algorithm [4] searches for the global minima structure of clusters with up to 30 atoms, on two ZnO supports ($(10\bar{1}0)$ and $(11\bar{2}0)$), and with different compositions (pure Cu, binary brass Cu-Zn alloy, and ternary Cu-Zn-O clusters simulating the incorporation of sub-

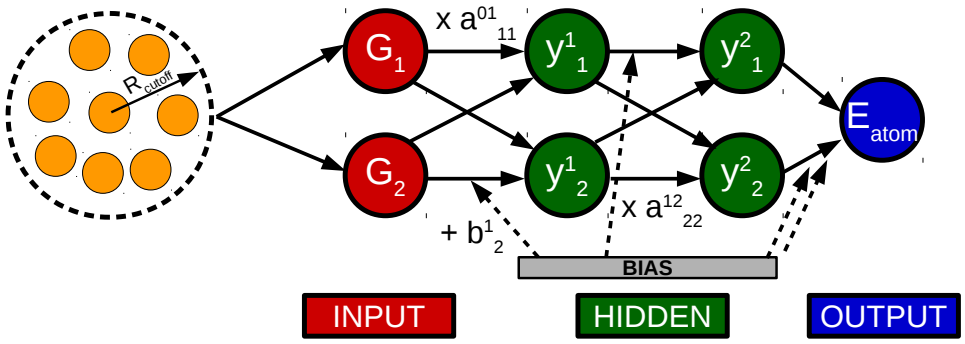

Figure 1: *Scheme of a single atomic neural network. a are multiplicative weights, b are additive biases, G are input symmetry functions, y are activation function nodes.*

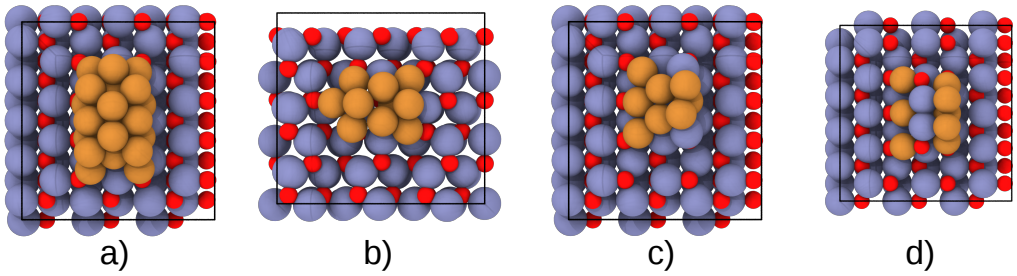

Figure 2: *Putative global minima for a) 20 atom Cu cluster on ZnO $(10\bar{1}0)$ b) 10 atom Cu cluster on ZnO $(11\bar{2}0)$ c) brass cluster (8 Cu, 2 Zn atoms) on ZnO $(10\bar{1}0)$ d) ternary cluster (6 Cu, 2 Zn, 2 O atoms) on ZnO $(10\bar{1}0)$*

strate atoms). A selection of putative global minima is shown in fig. 2.

In this continuation project, the computation time at the HLRN will be utilized to extend the potential by calculating further structures utilizing as a reference DFT calculations with the program VASP. The goal is to treat three new challenges: the formation of brass on large supported clusters (up to 1000 atoms), the interaction between large coincident Cu and ZnO interfaces, and the adsorption of H on the previously globally optimized clusters. We estimate that a total of 12700 structures will be required to accurately simulate all of these systems, the equivalent to 203k NPL hours.

With these extra structures, we will be able to perform a variety of simulations utilizing our NNP. For the brass clusters, we will study the proportion of brass in the cluster under different temperature and chemical potential conditions using Monte Carlo, and the structure of the clusters themselves at ambient temperature utilizing simulated annealing. From the systematic study of coincident interfaces, we hope to explain the behavior of the interface with the supporting ZnO for smaller globally optimized clusters, and estimate the influence of copper strain in the formation of these interfaces. Finally, introducing a fourth element into our NNP will allow us to start studying reaction conditions, and identify suitable H adsorption sites on clusters.

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https://www.uni-goettingen.de/de/556198.html

More Information

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