Design of catalysts for CO_2 hydrogenation with fine-tuned machine learning potentials

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Significance and Relevance

We show the development and application of methodologies based on density functional theory and machine learning (graph Gaussian process regression and graph neural network potentials) for the modeling of surface reactions on nanoparticle catalysts. We demonstrate applications of the methodologies on CO_2 hydrogenation processes on metals, single-atom alloys, and oxide materials.

Preferred and 2nd choice for the topic: Multiscale modeling and advanced simulation aspects, CO2 utilization and recycling.
Preferred presentation: Oral preferred or Short Oral

Introduction and Motivations

Heterogeneous catalysis is nowadays expected to solve the challenges related to the diversification of energy sources and the reduction of greenhouse gases. The processes of CO_2 hydrogenation have the potential to sustainably produce chemicals and fuels from green hydrogen and waste CO_2 [1]. To make such processes economically advantageous, R&D in catalysis relies on the discovery and optimization of catalytic materials based on experimental testing and computational analyses. The in-silico modeling and design of catalyst materials must tackle the extreme complexity of chemical reactions at catalytic surfaces. This makes the direct application of density-functional theory (DFT) methods computationally prohibitive, especially when targeting a wide combinatory space of elements of the periodic table. This problem can be addressed with machine learning (ML) techniques, which can significantly reduce the number of DFT calculations required.

In this contribution, we apply DFT and ML models to study CO_2 hydrogenation reactions on a wide range of bi-metallic catalysts, including single-atom alloys (SAAs), i.e., materials made of single metal atoms dispersed within the surface layer of another metal. Particular attention is given to SAA materials and their ability to break the Brønsted-Evans-Polanyi relations that limit the performances of conventional catalysts.

Materials and Methods

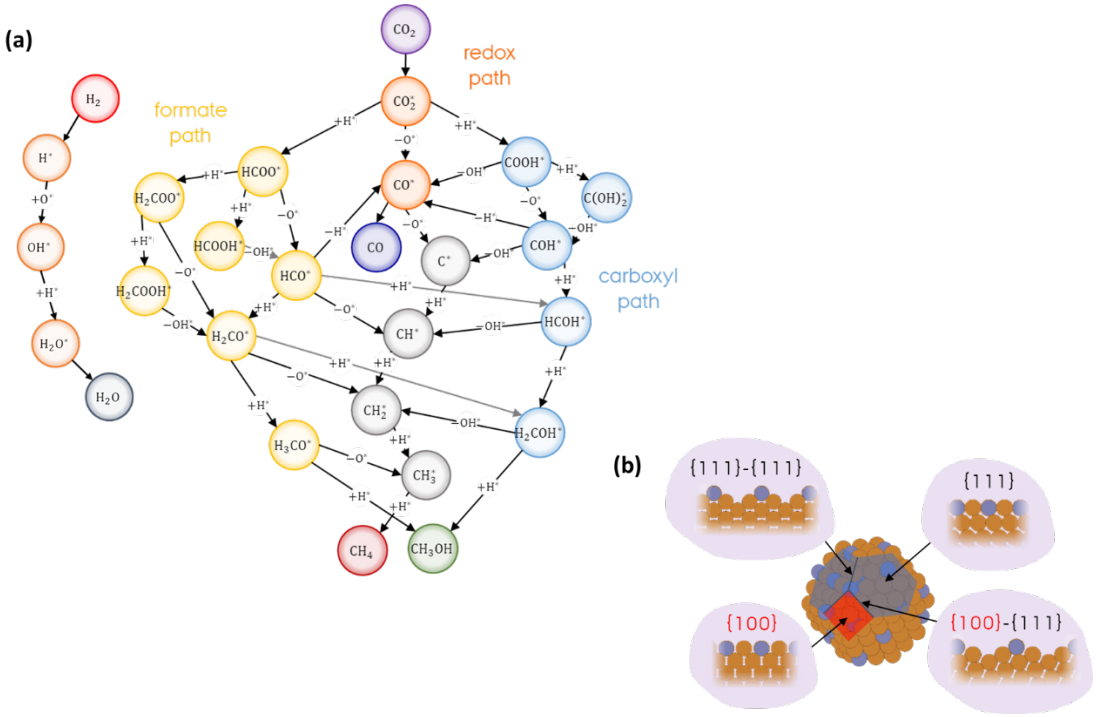
We use a graph-based Gaussian Process Regression (WWL-GPR) model [2] and machine learning potentials (MLP) from the Open Catalyst project [3-4] to calculate the adsorption energies and activation energies of the reaction mechanisms. We include in the study multiple crystal facets that catalyst nanoparticles can expose under reaction conditions. The use of pre-trained MLP models from the Open Catalyst project [4] allows us to substitute most of the DFT calculations with much faster ML evaluations in the search for minima and saddle points of the potential energy surface. Eventually, we apply structure-dependent mean-field microkinetic modeling [5] and finite differences reactor modeling to calculate the catalytic activity of the materials under investigation, accounting for the contribution of the different active sites of the catalyst nanoparticles.

Results and Discussion

The application of the framework to CO_2 hydrogenation reactions (reverse water-gas shift, methanation, and methanol production) allows us to rationalize how reaction mechanisms and catalytic performances (i.e., activity and selectivity) change with the catalyst composition, paving the way toward the design and nano-engineering of bi-metallic and SAA catalysts. On top of that, for the different materials under analysis, we evaluate the contribution of the different reaction paths

(reaction mechanism in Figure 1a) and active sites (crystal facets in Figure 1b), and we identify the energies of adsorbates and elementary steps that limit the catalytic performances of each material.

Figure 1. (a) Reaction mechanism of CO_2 hydrogenation to CO, methane, and methanol. (b) Nanoparticle shape and crystal facets considered in the study, corresponding to catalyst nanoparticles with a diameter of about 2 nm.



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