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Citation	サイバーメディアHPCジャーナル. 2026, 16, p. 64-67
Version Type	VoR
URL	https://doi.org/10.18910/106237
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Integrating Multi-scale Simulations and Inverse Design for Heterogeneous Catalysis Materials at Operating Conditions by Machine Learning Interatomic Potentials

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1. Introduction

Heterogeneous catalysts play an essential role in energy conversion, chemical production, and environmental applications. In particular, the catalytic conversion of carbon dioxide into valuable chemicals is an important research topic toward the development of renewable energy technologies and a sustainable carbon cycle. However, catalytic reactions under operating conditions involve complex atomic-scale processes, including molecular adsorption, surface diffusion, bond breaking and formation, co-adsorption effects, and surface reconstruction. Therefore, understanding and designing catalytic materials requires simulation methods that can connect atomic-scale reaction mechanisms with long-time and large-scale dynamical behaviour.

Density Functional Theory (DFT) is a powerful method for evaluating adsorption structures, reaction energies, and activation barriers on catalyst surfaces with high accuracy. Nevertheless, the computational cost of DFT makes it difficult to directly simulate large systems or long-time dynamics under realistic reaction conditions. To overcome this limitation, machine learning interatomic potentials (MLIPs) have attracted increasing attention. By learning energies and forces from first-principles calculations, MLIPs can reproduce DFT-level atomic interactions at a much lower computational cost, enabling large-scale molecular dynamics and other multi-scale simulations [1].

In this study, we integrate DFT-based database, MLIP- training, multi-scale simulations, and inverse design to establish a computational framework for

heterogeneous catalysis materials under operating conditions. As a target application, we focus on catalysis related to methanol synthesis through CO₂ hydrogenation. DFT, Molecular dynamics, metadynamics, and microkinetic modelling are combined to investigate dynamic surface behaviour under actual operating conditions. Through this approach, we aim to move beyond conventional material screening based only on static properties, such as adsorption energies and reaction barriers, and to develop a design strategy that accounts for the dynamic nature of catalytic processes under realistic operating conditions.

2. Computational Model

The computational model used in this study consists of Cu-based catalyst surfaces with adsorbed reaction species relevant to CO₂ hydrogenation and the water–gas shift reaction. Several surface configurations, adsorbate states, and reaction intermediates were considered to describe catalytic processes under realistic reaction conditions. In particular, the model was designed to capture not only ideal flat/pristine surfaces, but also dynamically reconstructed surfaces and cluster-like Cu structures that can appear during the reactions.

3. Large-scale Atomistic Simulations Using Machine Learning Interatomic Potentials

3.1 Construction of DFT Training Datasets and MLIPs

To perform large-scale atomistic simulations with first-principles accuracy, we constructed a workflow that combines DFT database generation, atomic representation, machine learning, and multi-scale simulations, as illustrated in Fig. 1.

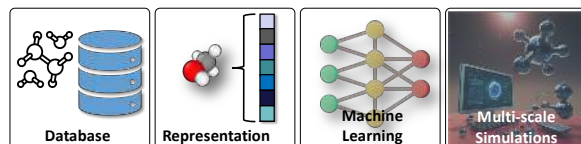


Fig. 1 : Schematic workflow for constructing machine learning interatomic potentials and applying them to multi-scale atomistic simulations.

The training datasets were constructed based on Density Functional Theory (DFT) calculations. The datasets included various Cu surface configurations, adsorbate states, and reaction intermediates relevant to CO₂ hydrogenation and the water-gas shift reaction. These DFT calculations provided reference atomic structures, total energies, and atomic forces for training machine learning interatomic potentials.

The machine learning interatomic potentials were constructed using DFT-generated data and improved through an active learning procedure. In this procedure, preliminary molecular dynamics simulations were performed using the current potential, and newly sampled atomic configurations were evaluated according to their relevance and uncertainty. Important configurations were then added to the training dataset, and the potential was retrained. This iterative procedure allowed us to efficiently improve the accuracy and transferability of the potential while avoiding an unnecessarily large database. High-performance computing resources were essential for this workflow because many DFT calculations and repeated training cycles were required. The resulting machine learning interatomic potentials enabled

molecular dynamics simulations of Cu-based catalytic systems at much larger length and time scales than direct DFT calculations, while retaining accuracy close to the first-principles reference data.

3.2 Large-scale Molecular Dynamics Simulations

Using the trained machine learning interatomic potentials, we performed large-scale molecular dynamics simulations of Cu-based catalytic systems. These simulations enabled us to investigate surface dynamics that are difficult to capture using static surface models or direct DFT calculations. In particular, we focused on adsorbate-induced surface restructuring and the formation of dynamic active sites under reaction conditions.

For CO₂ hydrogenation, the simulations revealed that hydrogen adsorption can significantly modify the structure of the Cu(100) surface. The time evolution of the lateral coordination number of Cu atoms showed that increasing hydrogen coverage results in the formation of the threefold hollow sites through hydrogen-induced surface restructuring. Such dynamic changes in the local coordination environment are important because they can create active sites that are absent in ideal flat-surface models.

We also investigated the effect of Cu cluster formation on the water-gas shift reaction. The simulations showed that Cu clusters formed on Cu(111) can enhance the catalytic activity compared with the flat Cu(111) surface. The calculated turnover frequency exhibited good agreement with experimental trends, suggesting that dynamically formed Cu clusters act as highly active catalytic sites. These results demonstrate that MLIP-based molecular dynamics simulations are useful for identifying active sites that emerge only under realistic reaction conditions.

4. Dynamic Active Sites in Cu-based Catalysts

The MLIP-based molecular dynamics simulations revealed that active sites in Cu-based catalysts are not always fixed structures. Instead, they can dynamically emerge under reaction conditions through adsorbate-induced surface restructuring and cluster formation. This finding is important because conventional static surface models often cannot describe the structural evolution of catalyst surfaces during reactions.

4.1 Hydrogen-induced Surface

Reconstruction on Cu(100)

First, we investigated the effect of hydrogen adsorption on the Cu(100) surface. Exposure to hydrogen adsorbates induces surface reconstruction, forming a rotated Cu(100) structure, denoted as R-Cu(100), and a line-shifted Cu-row structure, denoted as LS-Cu(100). The snapshots of the pristine and reconstructed surfaces are shown in Fig. 2. Quantitatively, the time evolution of the lateral coordination number of Cu atoms was analyzed at 300 K under different hydrogen coverages. As the hydrogen coverage increased, the average coordination number shifted toward approximately 6 (Fig. 3). This indicates that hydrogen adsorption induces significant restructuring of the Cu surface and promotes the formation of three-fold hollow sites [2].

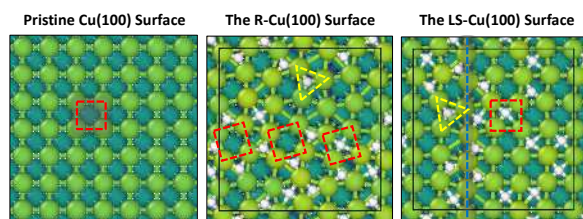


Fig. 2 : Atomic structures of the pristine Cu(100), R-Cu(100), and LS-Cu(100) surfaces formed by hydrogen-induced surface reconstruction.

This result suggests that hydrogen does not only participate as a reactant, but also modifies the surface morphology of the catalyst. The dynamically formed three-fold hollow sites may provide local

environments that are different from those on the ideal Cu(100) surface. Therefore, the catalytic properties under reaction conditions can differ from those predicted by static surface models.

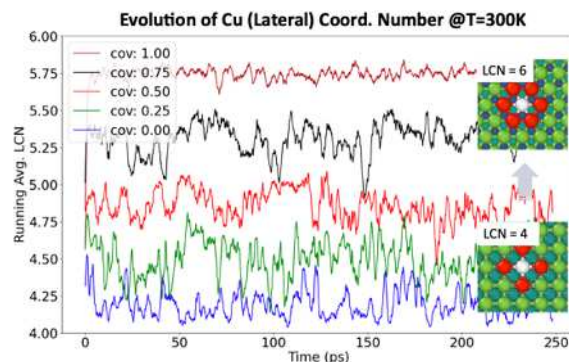


Fig. 3 : Time evolution of the lateral coordination number of Cu atoms on Cu(100) at 300 K under different hydrogen coverages. Increasing hydrogen coverage shifts the average coordination number toward approximately 6, indicating hydrogen-induced surface restructuring and formation of three-fold hollow sites similar with Cu(111) surface.

4.2 Cu Cluster Formation and Water-Gas Shift Reaction Activity

Second, we studied the effect of Cu cluster formation on the water-gas shift reaction. The simulations showed that Cu-cluster-modified surfaces exhibit higher catalytic activity than the flat Cu(111) surface. In particular, the calculated turnover frequency was significantly enhanced by the presence of Cu clusters [3].

The temperature dependence of the calculated turnover frequency showed good agreement with experimental trends (Fig. 4). This indicates that dynamically formed Cu clusters can act as highly active catalytic sites for the water-gas shift reaction. These results demonstrate that the catalytic activity of Cu surfaces is strongly influenced by structural changes that occur under reaction conditions.

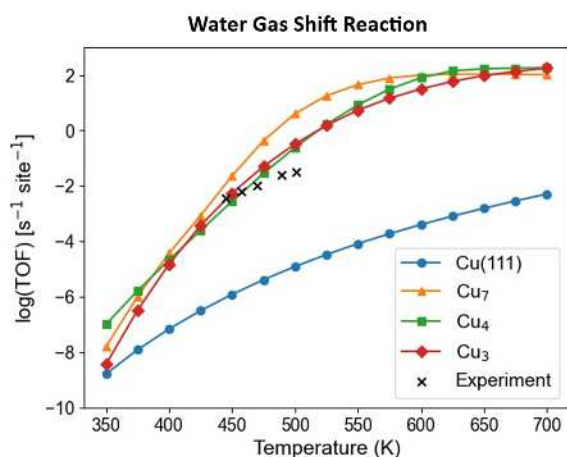


Fig. 4 : Temperature dependence of the turnover frequency for the water–gas shift reaction on flat Cu(111) and Cu-cluster-modified surfaces. The Cu-cluster surfaces show significantly higher activity than flat Cu(111), and the calculated trend agrees well with experiment.

Overall, the results show that dynamic active-site formation is a key factor in Cu-based catalysis. By combining DFT calculations, MLIPs, and large-scale molecular dynamics simulations, it becomes possible to capture surface evolution that is difficult to observe using static models alone.

5. Conclusion

In this study, we performed large-scale atomistic simulations of Cu-based catalysts using DFT calculations, machine learning interatomic potentials, and molecular dynamics simulations. By constructing DFT-based training datasets and using active learning, we developed machine learning interatomic potentials that enable efficient simulations of catalytic surfaces beyond the length and time scales accessible by direct first-principles calculations.

The simulations revealed that active sites in Cu-based catalysts can dynamically emerge under reaction conditions. Hydrogen adsorption induced significant reconstruction of the Cu(100) surface and promoted the formation of three-fold hollow sites. In addition, Cu clusters formed on Cu(111) enhanced the catalytic activity of the water-gas shift reaction, showing good agreement with experimental trends. These results

demonstrate that dynamic surface structures play an important role in determining catalytic performance.

Overall, this work highlights the importance of treating catalyst surfaces as dynamic systems rather than ideal static models. The combination of high-performance computing, DFT, MLIPs, and molecular dynamics provides a powerful framework for understanding catalytic reactions under realistic conditions. In future work, this approach will be extended to kinetic Monte Carlo simulations, rare-event sampling, and inverse catalyst design for discovering improved catalytic materials.

References

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