Joint Usage/Research Center for Interdisciplinary Large-scale Information Infrastructures
Final Report for JHPCN Joint Research of FY 2025

jh250072

Machine Learning-Driven Multi-Scale Simulations for Green Catalysis Design

Yoshitada Morikawa（Graduate School of Engineering, The University of Osaka）

Understanding the nature of heterogeneous catalysis is a challenging task, particularly when the catalyst is sensitive to operating conditions. The difficulty comes from the complex and elusive states which occur at atomic scales in both space and time. A significant challenge is identifying the active sites of Cu-based catalysts for methanol synthesis. In this study, we apply molecular dynamics driven by machine learning force fields (MLMD) to elucidate the dynamical states of Cu surfaces under the influence of relevant adsorbates. Specifically, we investigated the surface evolution involving defect sites, adsorbate-induced surface transformation, and the co-adsorption effects of the adsorbates. Our findings reveal that Cu surfaces undergo considerable reconstruction under the influence of CO and hydrogen. The force fields are trained using first-principles calculations to capture the interactions between surfaces and adsorbates. In the case of CO interacting with the surface, the simulations indicate the formation of small active clusters, which enhances the water-gas shift reaction. Further, we observed that adsorbed hydrogen also induces surface reconstruction, particularly on pristine Cu(100) surfaces, where new active sites of three-fold hollow sites emerge. These results help to elucidate the true active sites of Cu catalysts and its effect to catalytic properties under reaction conditions, contributing to realising the catalytic systems that enable energy-efficient, low-emission, and sustainable chemical transformations.

1. Basic Information

(1) Collaborating JHPCN Centers

Osaka University

(2) Theme Area

Data science/data usage area

(3) Project Members and Their Roles

(ア)Yoshitada Morikawa: Conceptualise the research and supervise the project.

(イ)Harry H. Halim: realise the research plan, supervise the creation of the machine-learning potential (MLP), do active learning for data generation, perform Density Functional Theory calculations, design the atomic models, perform and analyse Molecular Dynamics (MD) simulations, and create the generative model.

(ウ)M. Fadhlan Anshor: create and evaluate the MLP, calculate DFT data, and perform multi-scale simulations such as MD, meta-dynamics, and microkinetic modelling.

(エ)Trinh Bao Anh: create and evaluate the MLP, calculate DFT data, and perform multi-scale simulations such as MD, meta-dynamics, and microkinetic modelling.

2. Purpose and Significance of the Research

Global warming remains a critical challenge that demands urgent technological solutions. One promising strategy is the conversion of greenhouse gas CO_2 into value-added chemicals such as methanol, which can serve as a sustainable energy carrier and chemical feedstock. Achieving this goal requires the rational design of highly efficient **green catalysts** (i.e., the catalytic systems that enable energy-efficient, low-emission, and sustainable chemical transformations under realistic operating conditions). However, achieving this in trial-and-error fashion by experiment and first-principles calculations is inherently inefficient.

In the era of data science, Machine Learning (ML) has transformed materials science, accelerating catalytic material design and optimisation. **ML Potentials (MLPs)** provide accurate atomic-scale interaction predictions at a fraction of the cost of traditional methods. By learning from first-principles data, MLIPs enable large-scale catalytic simulations, and when combined with multi-scale modelling, they connect atomic phenomena to macroscopic behaviour. Combining MLIP-driven simulations with multi-scale modelling helps researchers efficiently explore materials, reduce discovery time, and improve catalytic performance. This research aims to develop MLPs integrated with multi-scale simulations to optimally elucidate and design the heterogeneous catalysts, focusing on reactions relevant to the CO_2 hydrogenation to methanol.

3. Significance as JHPCN Joint Research Project

This project leverages large-scale high-performance computing (HPC) resources to enable data-intensive and computationally demanding simulations that are essential for next-generation catalyst design. The development of MLIPs requires extensive first-principles datasets, iterative active learning cycles, and large-scale molecular dynamics simulations, all of which demand huge computational power and efficient parallelisation. By utilising JHPCN resources, this project not only advances the methodology for ML-driven materials design but also demonstrates an effective framework for combining data science with physics-based simulations. The outcomes will contribute to the broader HPC community by establishing scalable workflows for inverse catalyst design and by promoting the use of supercomputing resources in addressing critical challenges in sustainable energy and green catalysis.

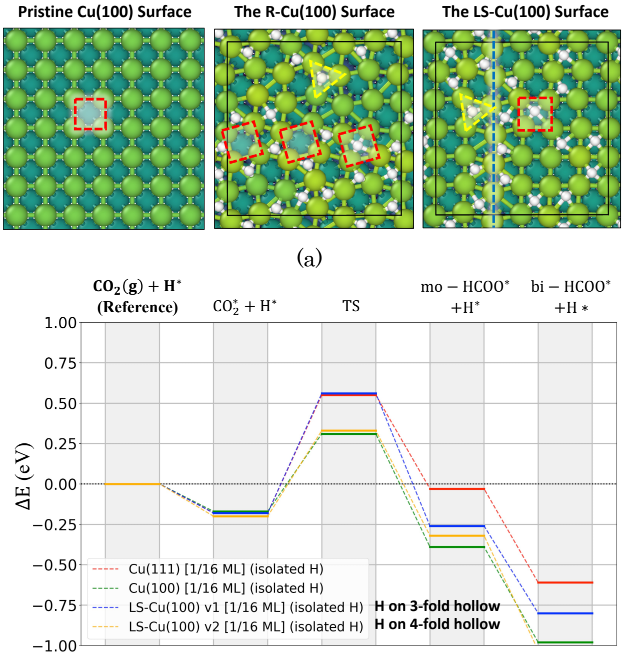
4. Outline of Research Achievements until FY2024

In the FY2024 of this JHPCN joint research project, we have demonstrated the feasibility and accuracy of machine-learning molecular dynamics (MLMD) for CO_2 hydrogenation on Cu surfaces: By combining MLMD with Transition Interface Sampling (TIS), we accurately reproduced mode-specific reaction probabilities and obtained hydrogenation rates that closely match experimental trends under surface temperatures of interest.

5. Details of FY2025 Research Achievements

In FY2025, we completed the calculations and published the results that we started in FY2024, as well as extended the research to more complex surfaces, such as flat and stepped Cu(110) and CuZn surface alloy.

a) For the CO_2 hydrogenation to formate on the Cu surfaces, we have successfully evaluated the kinetics of the reaction under consideration of the surface dynamics. We found that the exposure of the hydrogen on the Cu surface induces the surface reconstruction in the form of formation of the three-fold hollow sites, which then alter the hydrogenation barrier (Fig. 1). The paper has been published in the highimpact journal, namely, ***ACS Catalysis*****.** 2026, 16, 5, 5068–5079.



(a)

(b)

Fig 1. (a) The surface reconstruction of Cu(100) induced by hydrogen adsorption. (b) The energy barrier of hydrogenation on the pristine, line-shifted (LS) reconstruction, and Cu(111) surfaces.

b) In the case of the WGSR reaction, we have included the lateral interaction of the co-adsorbed species. This is achieved with the help of the MLP to efficiently evaluate the energetics of many samples of different arrangements of the co-adsorbed species. The result was then tested on the Cu clusters that dynamically formed on the Cu surface under the exposure of the CO adsorbates. The results show significant improvement in the catalytic activity of the WGSR compared to the typical flat Cu(111) surface (Fig. 2). The research is complete and has been published in ***Phys. Chem. Chem. Phys.*****,** 2026, (Advance Article).

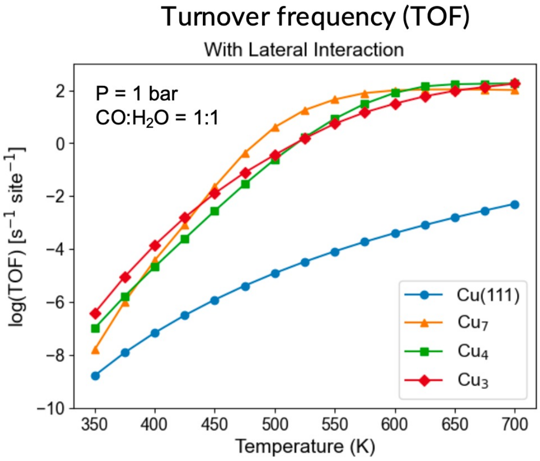


Fig 2. The lateral interaction is non-negligible in high-coverage condition. (b) The change of Turn Over Frequency due to cluster formation on the surface. The clusters on Cu(111) surface increase the rate of the WGSR reaction that would be beneficial for methanol synthesis.

Furthermore, as the extension to the project in FY2024, we are now investigating the following. c) H-induced surface reconstruction on the flat and stepped Cu(110) surface. Given the less stable Cu(110), we found that the H also induces the formation of the three-fold hollow sites, similar to what we observed on the Cu(100) surface (Fig. 3). We expect this work to clarify the facet insensitivity of the CO_2 hydrogenation to formate on the Cu surface. The manuscript is under preparation.

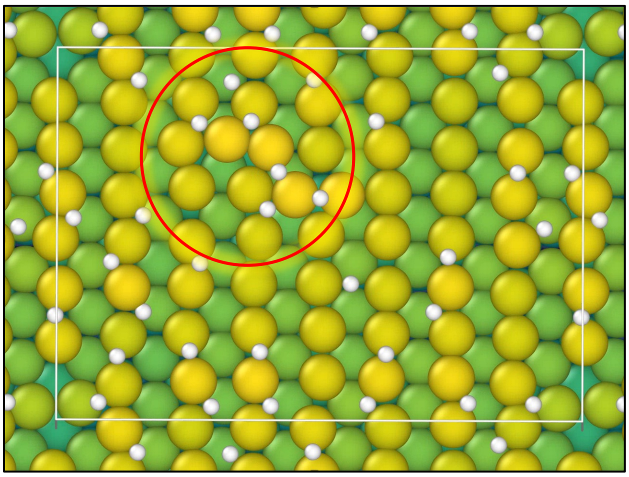


Fig 3. The hydrogen on the Cu(110) surface and the formation of the three-fold hollow sites.

d) The CO-induced Zn aggregation on the CuZn surface.

We found that the adsorbed CO induced the surface aggregation of the Zn atoms, resulting in the formation of Zn clusters, which is significantly different from the CuZn under vacuum conditions. The change in the state of the Zn atoms play important role on the kinetics of the intermediates of the methanol synthesis.

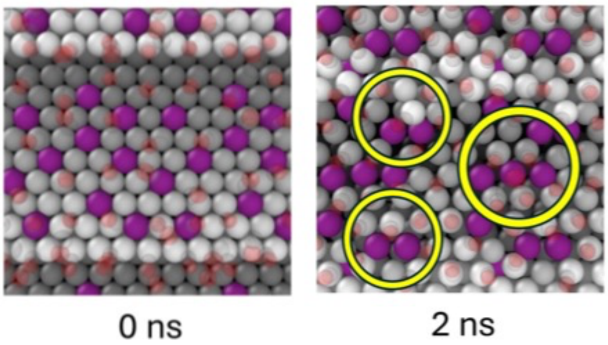


Fig 4. The CO-induced aggregation of Zn atoms observed in the MD simulations.

6. Self-review of Current Progress and Future Prospects

In FY2025, we have successfully established a machine learning–driven simulation framework that integrates MLIP-based molecular dynamics with kinetic analysis to investigate catalytic reactions on Cu-based systems. The developed MLIPs achieve sufficient accuracy to reproduce key reaction energetics while enabling the exploration of dynamic surface phenomena, including hydrogen-induced surface reconstruction and adsorbate-driven cluster formation under reaction conditions. These results demonstrate the capability of the framework to capture catalytic behavior beyond conventional static models.

Future work will focus on completing the study on the H-induced reconstruction on the Cu(110) surface, CO-induced Zn segregation, and systematically expanding the DFT training dataset through active learning to improve model robustness and transferability. We will integrate MLIP-derived energetics into higher-scale simulations such as Kinetic Monte Carlo simulations to explicitly capture co-adsorption effects and reaction networks at experimentally relevant timescales. Finally, we will develop a generative modelling framework to enable inverse design of catalyst structures optimized for targeted reactions.

These developments will require extensive use of high-performance computing resources for large-scale data generation, model training, and multi-scale simulations. Through these efforts, we aim to establish a scalable and predictive framework for the rational design of green catalysts under realistic operating conditions.

#Section 7, achievements of this FY, should be input on the JHPCN website.