大阪大学 OSAKA UNIVERSITY

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

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A. OVERVIEW

Atomic-level description of **non-equilibrium states of catalysis** are investigated by **machine learning aided-atomic simulations.**

Multi-scale simulation enable big length-scale and long time-scale with **first-principles accuracy**.

Machine learning is used as a **bridge** between the first-principles calculations and higher scale simulations.

The simulations combines **physics**, **chemistry**, **material science**, **chemical engineering**, and **data science**.

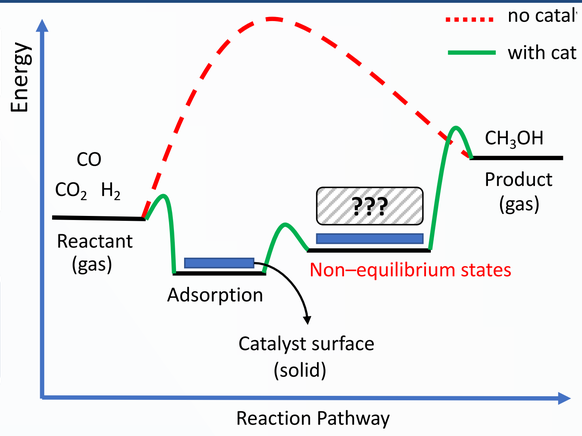
B. BACKGROUND

Catalysis is **chemical reactions** facilitated by external material known as "**catalyst**".

Heterogenous → multi-phases are involved (e.g., gas and solids)

CO_2 Hydrogenation to Methanol

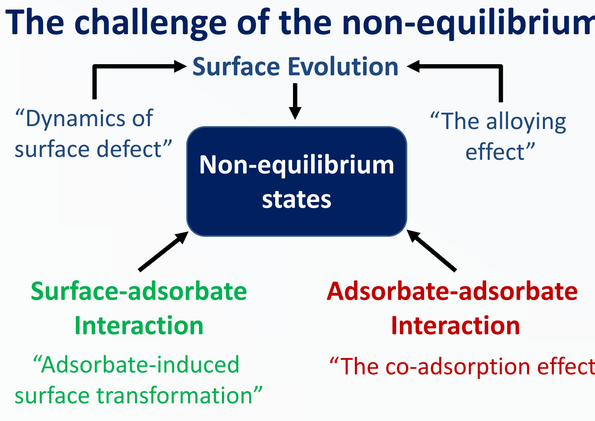
Synthetic Gas (CO / CO_2 / H_2) —[$Cu/ZnO/Al_2O_3$, 200 – 300 °C]→ Methanol (CH_3OH)

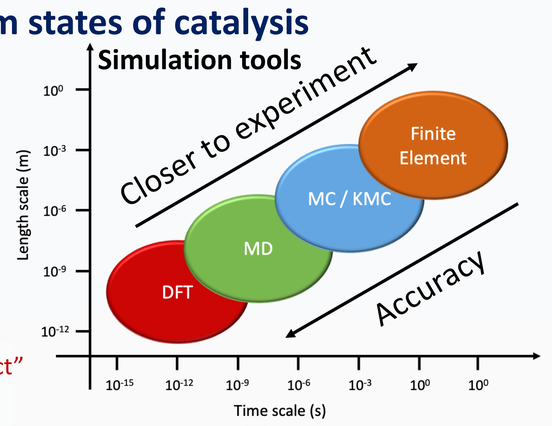


Better understanding in catalysis well contributes in energy & environments.

B. BACKGROUND (continued)

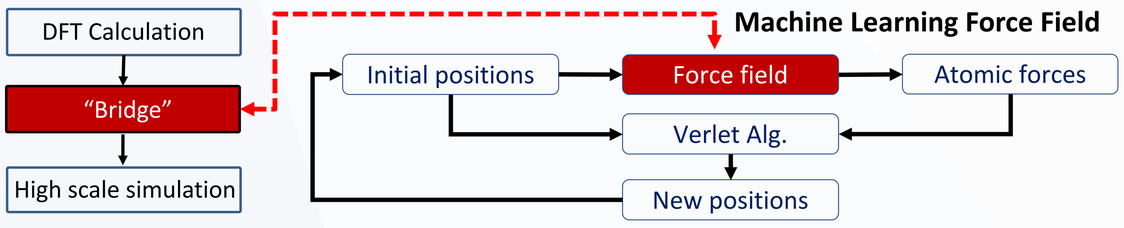
The challenge of the non-equilibrium states of catalysis





Objective: elucidate the behavior of catalysis at non-equilibrium states

❖ Machine Learning Aided Multi-scale Simulation



✓First-principles level of accuracy ✓Efficient for large system and long time scale

❖ The Research Plan in Elucidating Non-equilibrium States

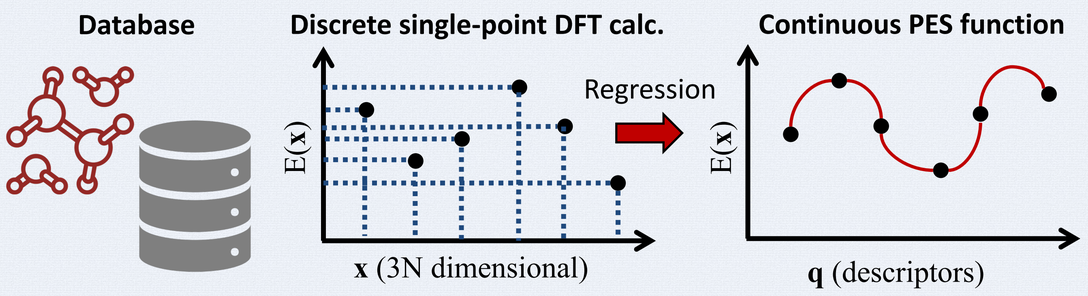
April-July '25	Aug '25	Sept-Dec'25	Jan-Mar '26
Build Training Database (DB) 1. Build DB for Clean Surface ($Cu/ZnO/Al_2O_3$) 2. Build DB for Surface + Adsorbates✿	**Train ML to Build Interatomic -Potential** 1. Use Gaussian Process (GP) 2. Run active learning scheme to generate more structures	**Launch First Multi-scale simulations** 1. Run MD and kMC Simulations based on MLIP 2. Perform path sampling to capture rare key events.	**Build Generative models** 1. Predefined properties used in objective function. 2. Optimize generative model to generate the target material.

✿ All adsorbates present in the methanol synthesis are included: CO_2, CO, HCO,CHO, HCOO, etc..

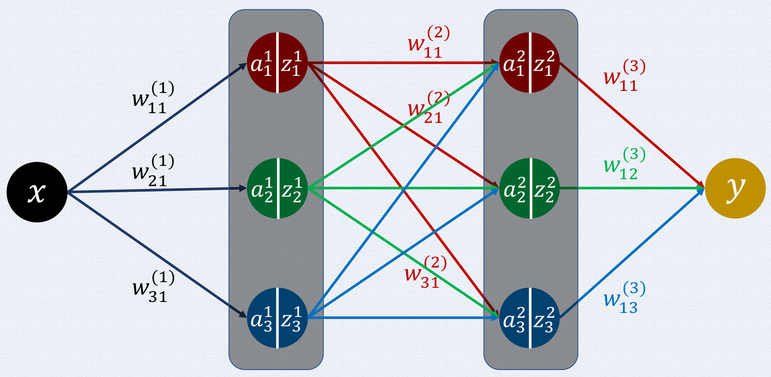
C. METHODOLOGY & PRELIMINARY (PUBLISHED) RESULTS

❖ Constructing Machine Learning Force Field

Generating database → Build Representation → Training → Validation



❖ Regression in Machine Learning by Artificial Neural Network

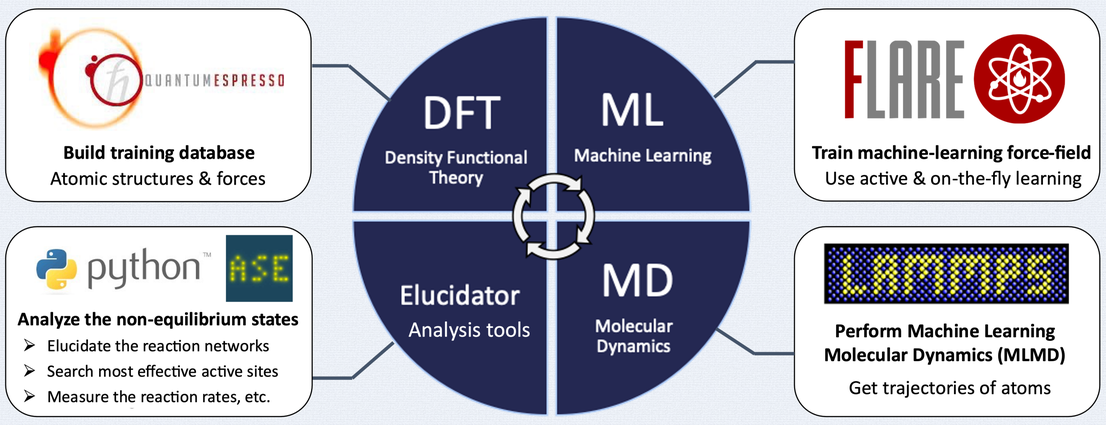


$$y(x,\boldsymbol{w}) = \sum_{j=1}^{3} w_{1j} z_j^{(2)} = \sum_{j=1}^{3} w_{1j} h\left(\sum_{i=1}^{3} w_{ji} h(w_{i1}^{(1)} x + b_i^{(1)}) + b_j^{(2)}\right)$$

ALGORITHM

- Forward pass — Calc. output based on **W**
- Compute Error — Based on error function
- Backpropagation — Finding gradients
- Update parameters — Iterative opt. algorithm

❖ The Implementation of Machine Learning Molecular Dynamics

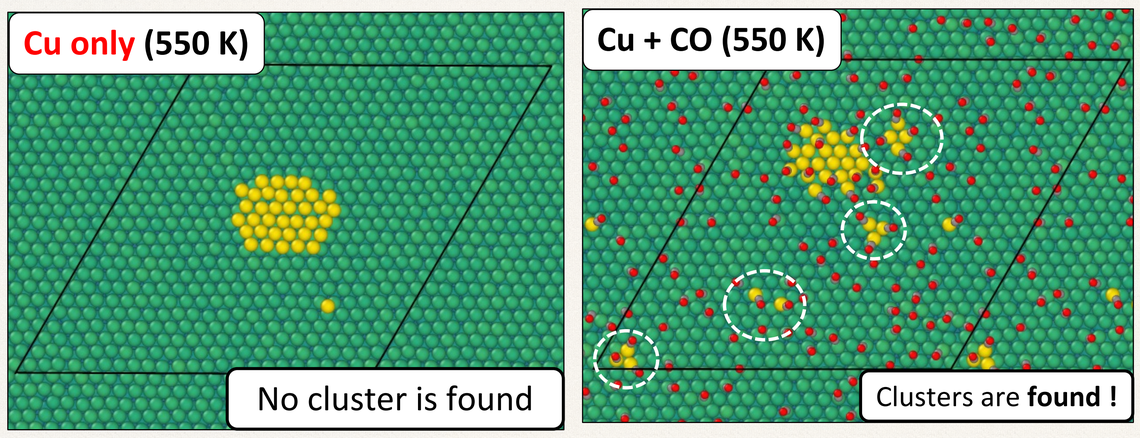


❖ Preliminary Results

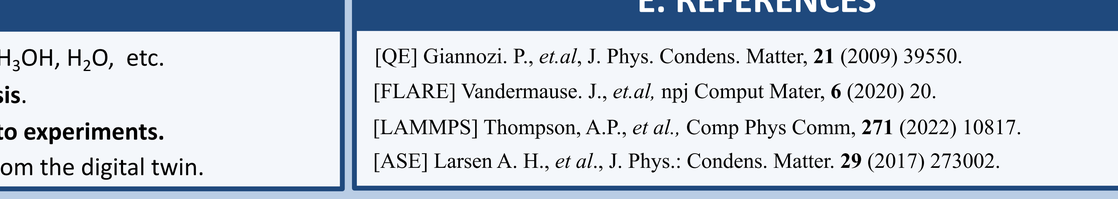
#1 The MLMD simulations of CO-induced formation of Cu clusters

Halim, H. H.; Ueda, R.; Morikawa, Y. *J. Phys. Condens. Matter* **2023**, *35* (49), 495001.

➢ The simulations at 450 K to 550 K show Cu clusters are formed within a hundred of ns when the Cu surface is exposed with CO, significantly alter the catalyst activity.

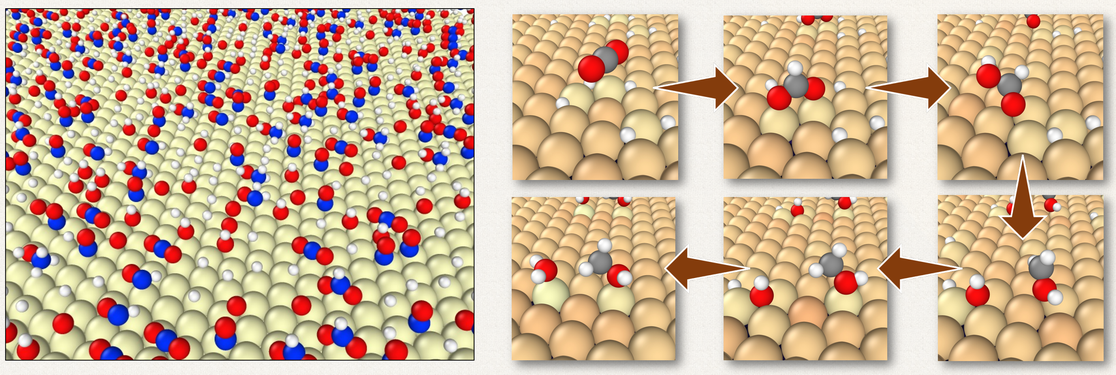


➢ **CO-decorated Cu clusters** are formed within 100 ns when the surface is ***exposed to CO.***



#2 The Digital Twin of the CO_2 Hydrogenation on Cu Surfaces

➢ A precise digital twin of catalysis enables inverse catalyst design in the digital world (i.e., simulations), which can then be transferred to the real world (i.e., experiments).



D. ONGOING WORK & FUTURE PLAN

- ❖ **Improve the database** to include the other species relevant to methanol synthesis: CO_2, HCOO, HCOOH, CH_3OH, H_2O, etc.
- ❖ Perform long-time and large-scale MLMD simulations to **provide explicit "molecular movie" of the catalysis**.
- ❖ Connect the MLMD to higher scale simulations such as **Kinetic Monte Carlo** to enable **better comparison to experiments.**
- ❖ **Design a new catalyst for the methanol synthesis** based on the inverse-design and knowledge obtained from the digital twin.

E. REFERENCES

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[FLARE] Vandermause. J., *et.al,* npj Comput Mater, **6** (2020) 20.

[LAMMPS] Thompson, A.P., *et al.,* Comp Phys Comm, **271** (2022) 10817.

[ASE] Larsen A. H., *et al*., J. Phys.: Condens. Matter. **29** (2017) 273002.