Towards a fundamental understanding of forced liquid intrusion for shock absorption applications using machine learning potentials

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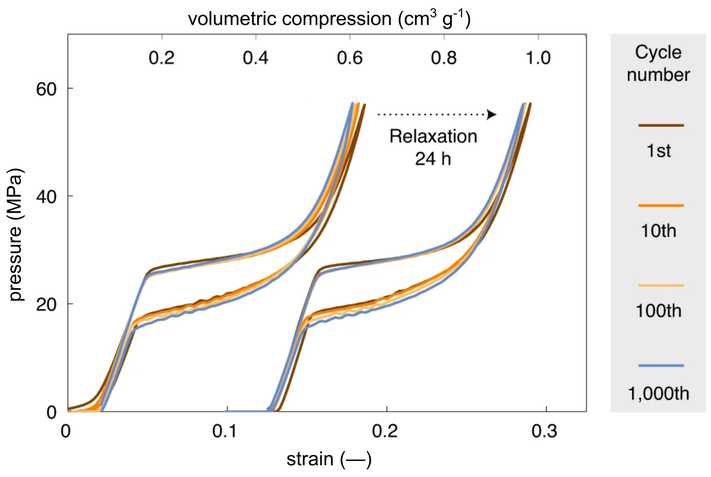
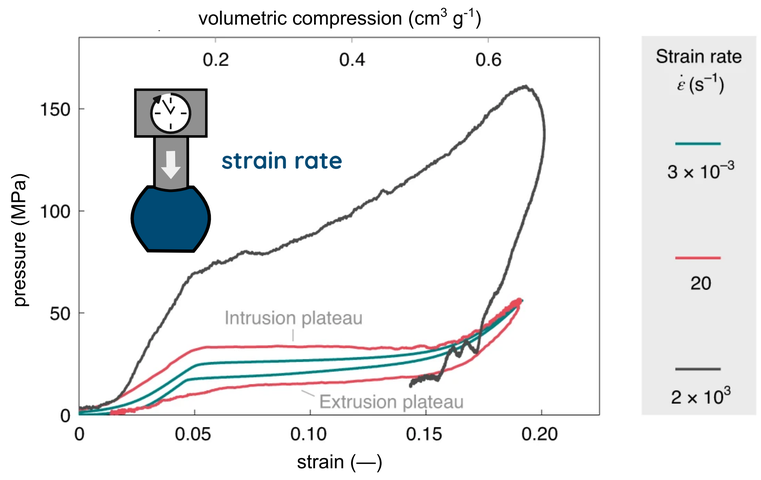
Liquid-ZIF intrusion...

Zeolitic imidazolate frameworks (ZIFs) are **efficient** and **reusable** high-rate **shock absorbers**. The amount of energy dissipated during the intrusion-extrusion process is determined by the **hysteresis** between the intrusion and extrusion curves.

repeated intrusion-extrusion cycles of water in ZIF-8[1]



strain-dependent intrusion-extrusion cycles of water in ZIF-8[1]



...to create the ideal shock absorber?

Depending on the choice of ZIF, electrolyte, and crystallite size, different macroscopic behaviours can be realised ranging from **springs** over **shock absorbers**, to **bumpers**.

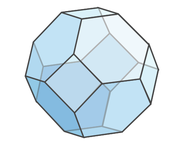
By screening **many ZIFs** and **aequous solutions**, we aim to examine and understand their behaviour under different **strain rates** and find the best **crystallite size**

Main goal:
to engineer the best shock absorber in silico by understanding...

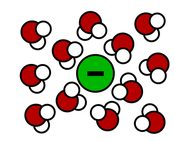
...how does intrusion work on the nanoscale?

...how to couple water hopping (ns timescale) with intrsuion.extrusion (ms timescale)?

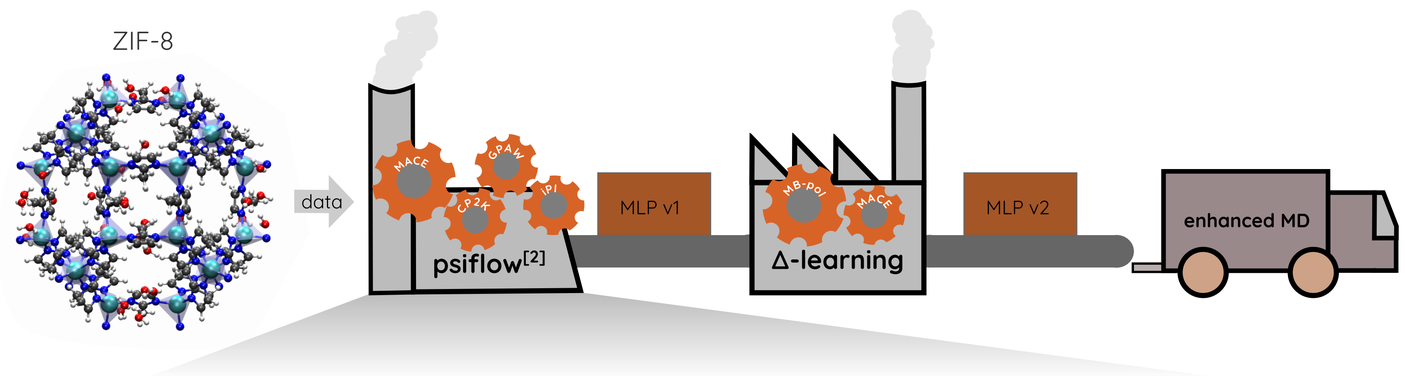
...how to model the strain rate?

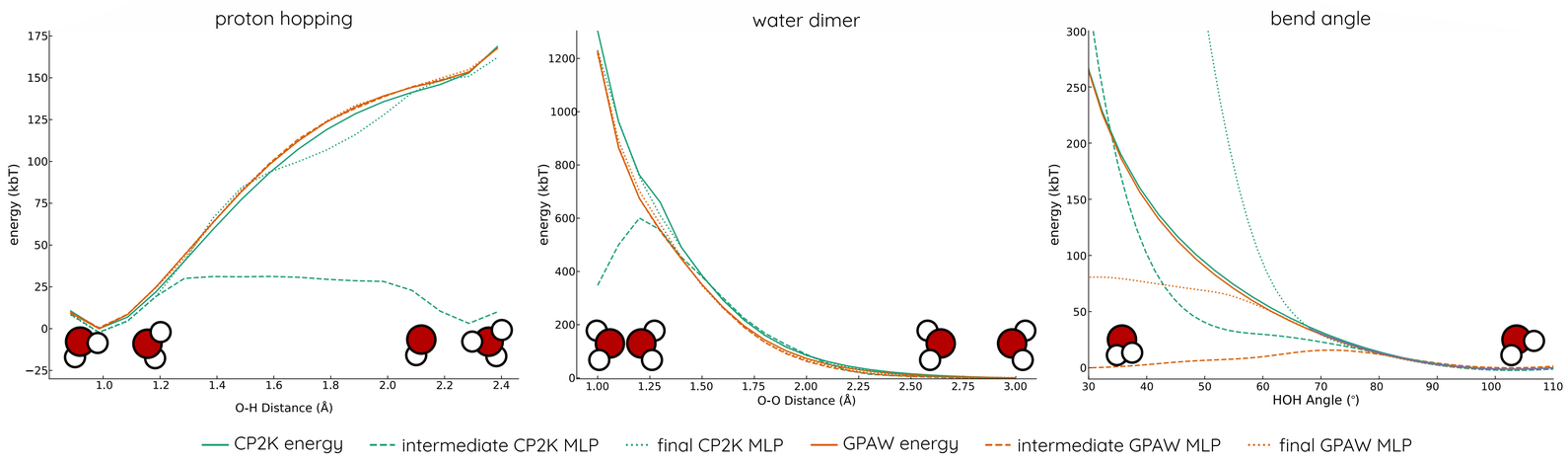

pore geometry


crystallite size


liquid composition

Connecting the nanoscale: A machine learning potential (MLP) to model fundamental intrusion events





Why MLP enhanced with MB-pol?[3]

low computational cost with ab initio accuracy for the ZIF framework (improving on force field derived results)

accurate modeling of water-water interactions due to MB-pol

ability to expand to other ZIFs and aequous solutions

Current challenges training the current MLP

removing BSSE errors with GPAW

finding and correcting non-physical events

efficient data generation to reduce error metrics

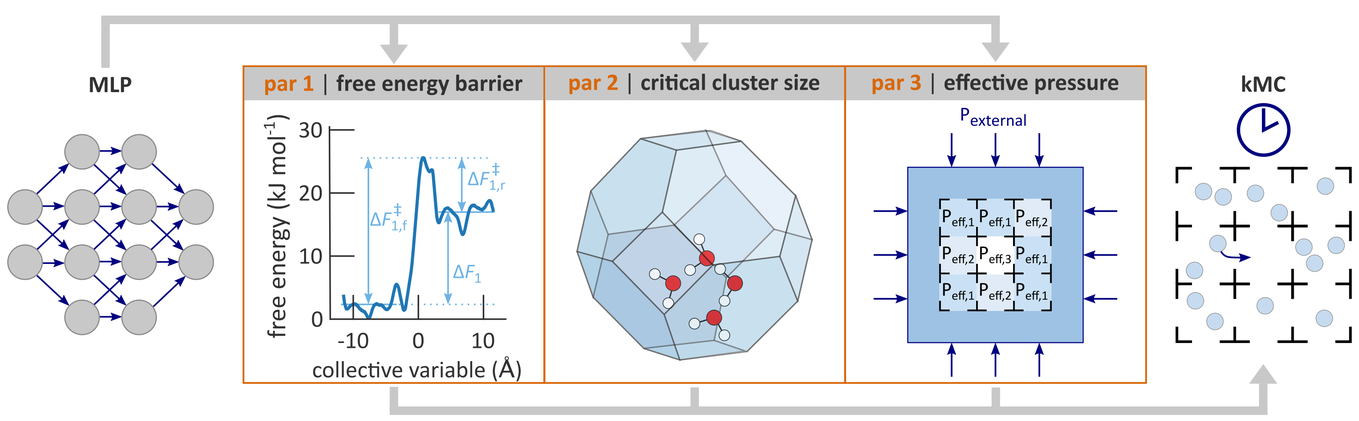
Future challenges

efficient dataset generation for Δ-learning

finding an approach to make this MLP transferable to other ZIFs and liquids

To the macroscale: Kinetic Monte Carlo (kMC) as a multiscale method to model water intrusion/extrusion

Connecting the **nanoscale** to the **macroscale** by deriving **three key nanoscale parameters** with my MLP to parametrise a **lattice kMC**, maintaining **quantum accuracy**.



Capacity of this model

faster than molecular dynamics

predicts intrusion/extrusion pressures and timescales

explains crystallite size effects

can be expanded to other topologies and liquids

Remaining challenges

how to describe effective pressure?

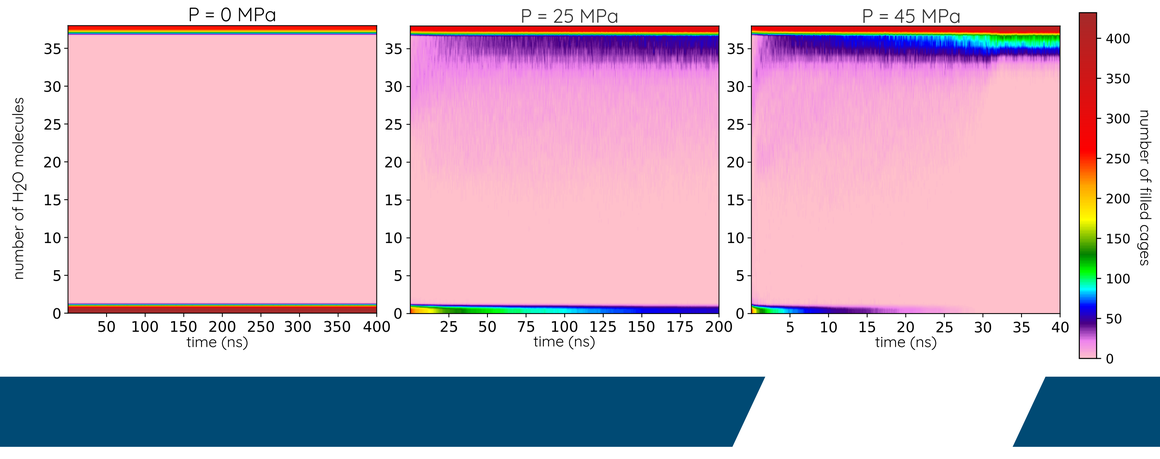
how to model strain rate?

impact of the crystal surface?

predicts intrusion...

example:
intrusion in a 7x7x7 unit cell (12 nm x 12 nm x 12 nm)

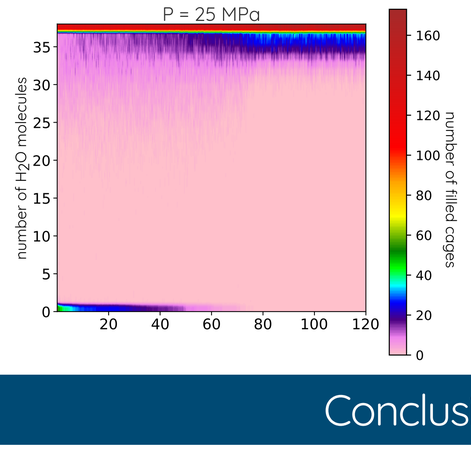
edges are being filled
FF derived energy barriers



...and crystal size effects

example:
intrusion in a 5x5x5 unit cell (9 nm x 9 nm x 9 nm)

edges are being filled
FF derived energy barriers



What's next?

improve **energy barriers** with the Δ-learned MLP to understand and model the physics of water intrusion

improve the **effective pressure model** and **implement strain** to correspond with experiments

expand the MLP and kMC workflow toward **different ZIF-liquid systems**, e.g., ZIF-71, MAF-6... combined with ions or alcohols, which will lead to the choice of most optimal shock absorber

combining all of the above **to model and understand intrusion**

Conclusions

The ab-initio trained MLP should be **enhanced to model water-water interactions** more accurately to improve on the earlier derived force field results. **MB-pol** looks promising for this goal, but still has to be verified.

The kMC model can describe the **multiscale** intrusion properties. The accuracy is mainly determined by the **free energy barriers** associated with the **hopping frequency**, the **critical cluster size** and **effective pressure**.

The major challenge with the kMC model is the **implementation of pressure and strain**.

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strain engineering to design functional 4D polymorphism in nanostructured materials
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[1] Nat. Mater. **20**: 1015, **2021**
[2] npj Comput. Mater. **9**: 19, **2023**
[3] J. Chem. Theory Comput. **9**: 5395, **2013**

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