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Advancing Research through Computing 2021

A Neural Network and Lennard-Jones Hybrid Forcefield to Study Diffusion of Neon in UiO-66

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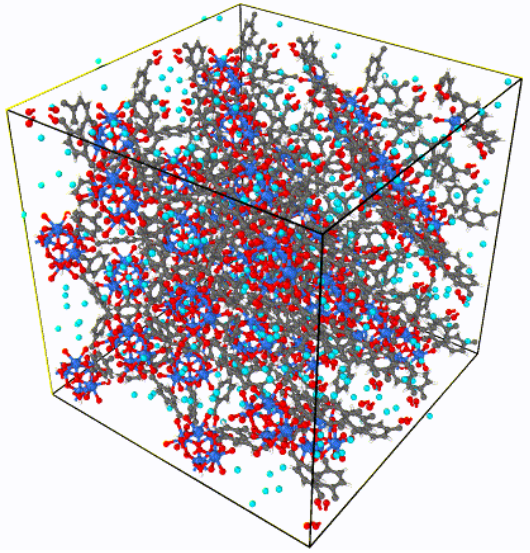

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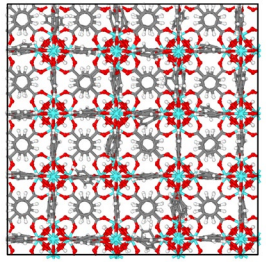
Overview

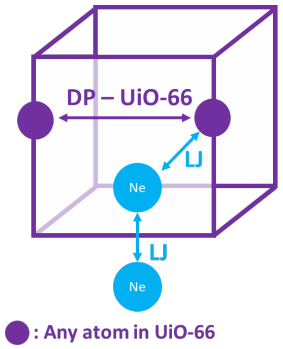
- We are using atomistic modeling to study UiO-66, a metal organic framework (MOF)
- Our goal is to model adsorption and diffusion of toxic molecules in UiO-66
- Density functional theory (DFT) simulations are very expensive for complex MOFs



- Classical forcefields have limited capabilities and often not very accurate
- We developed a neural network (NN) forcefield (DP) using optimized training techniques
- Out forcefield exhibits excellent scaling, both to number of atoms and expansion

VASP → TensorFlow → LAMMPS

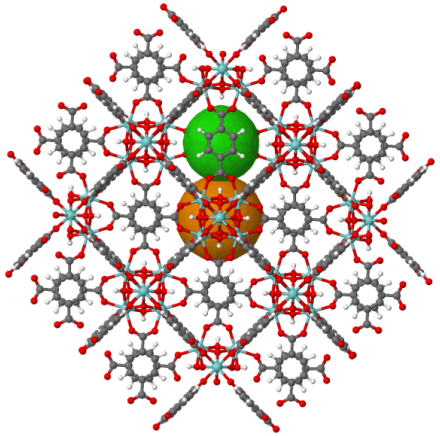
- We also demonstrate a technique to enable classical and DP to coexist in the same environment
- Diffusion of guest atoms was performed with this hybrid environment
- Future work will be to model diffusion for more complex molecules



UiO-66: Metal Organic Framework

UiO-66 is a robust and versatile MOF

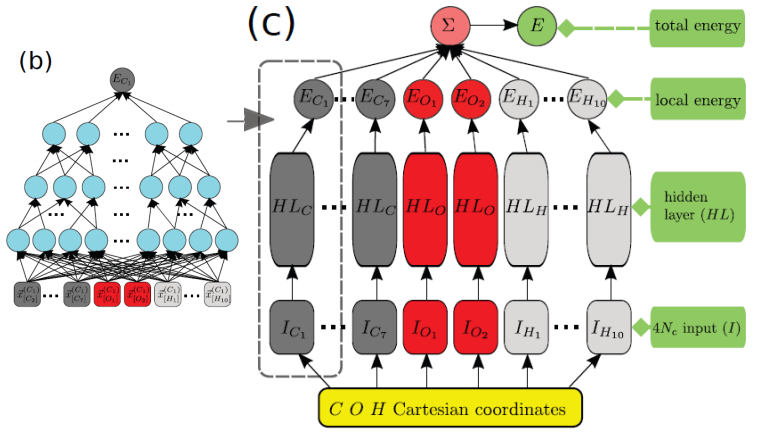
- Constructed of organic linkers: $C_8H_4O_4^{-2}$ and metal oxide nodes $[Zr_6(\mu_3\text{-}O)_4(\mu_3\text{-}OH)_4]^{+12}$
- Uses in adsorption, separations, catalysis
- Large **(octahedral)** and small **(tetrahedral)** pores connected by triangular windows



How we build a DP

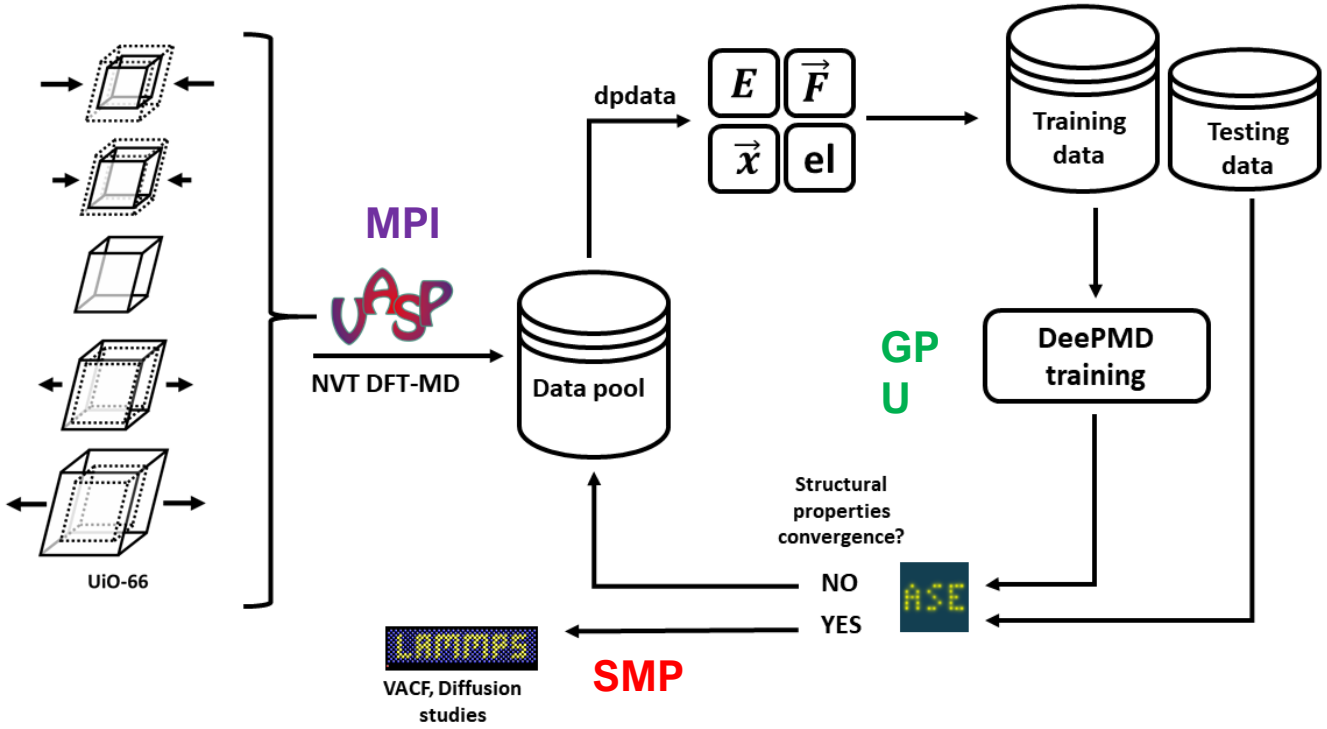
DeePMD:
Builds NN potentials for large potential energy surface

NN for **each atom** built by minimizing errors in energies and forces using the **loss function (*L*)**

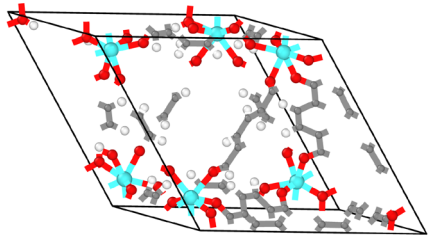


$$L(p_\epsilon, p_f) = \frac{p_\epsilon}{N}\Delta E^2 + \frac{p_f}{3N}|\Delta F_i|^2$$

Algorithm to build a DP

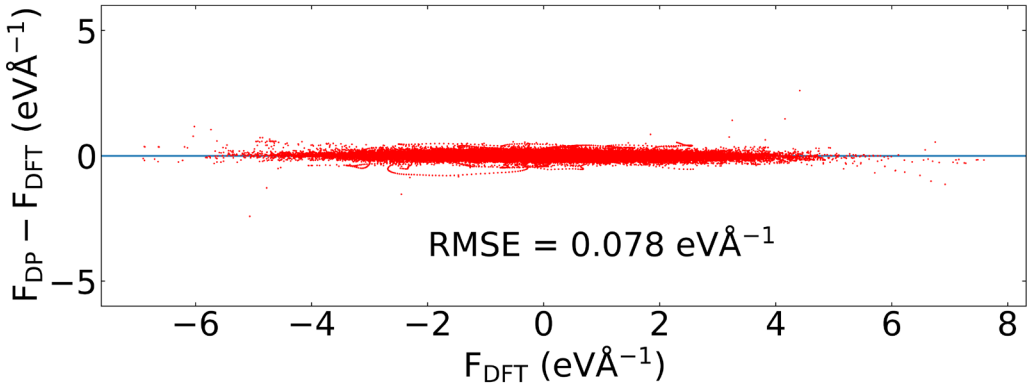


- All calculations and model building were performed at CRC: Mixture of MPI, SMP and GPUs
- NN Forcefield was built using small primitive cell
- Bulk modulus and lattice constants were convergence criteria



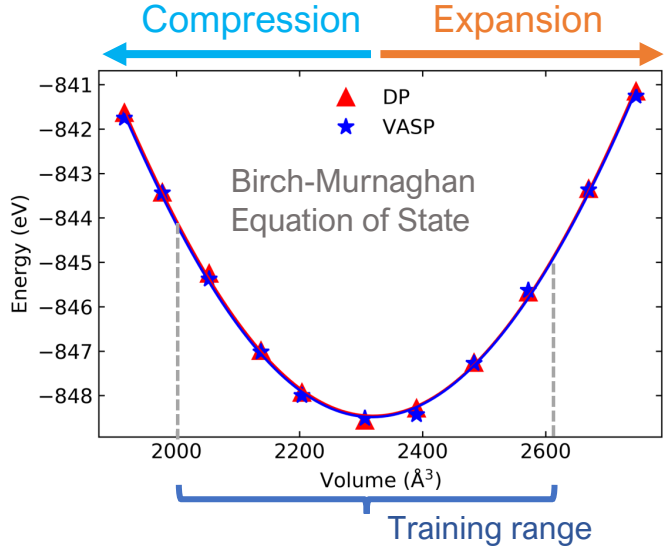
Primitive cell (114 atoms)

DP performance on primitive cell



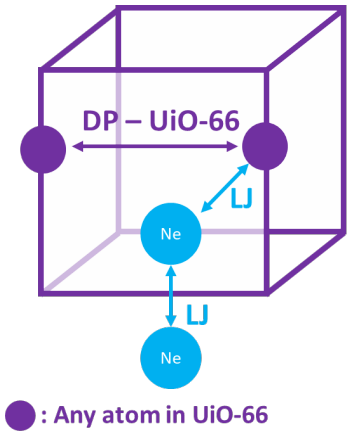
Force prediction accuracy on unseen data set

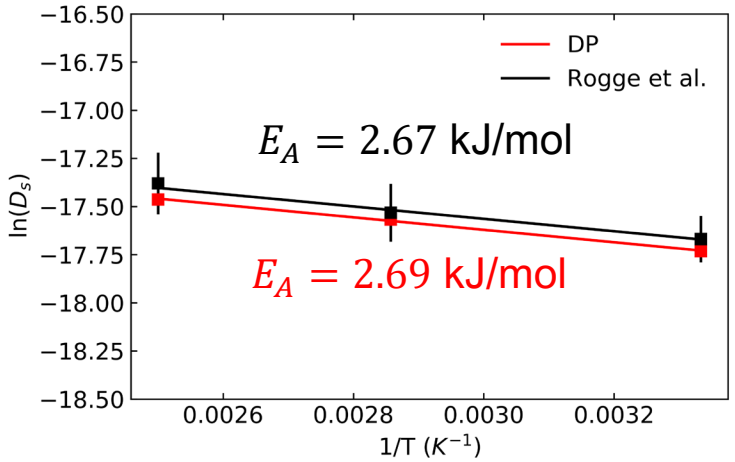
Evaluation of structural properties. Good agreement with experiments



Properties	DFT	DP	Experm.
Bulk Modulus	**32.11 GPa**	**32.41 GPa**	**-**
Lattice constant	**21.01 Å**	**21.01 Å**	**20.80 Å**

DP + Empirical Forcefield (EFF): Ne Diffusion





- 500+ Ne atoms in 3000+ atoms of UiO-66 cell
- Hybrid approach to computing diffusion
- EFF: Lennard-Jones (LJ) + DP-UiO-66
- EFF is applied to Ne-Ne and Ne-UiO-66 interaction
- DP is applied for interaction of atoms in UiO-66

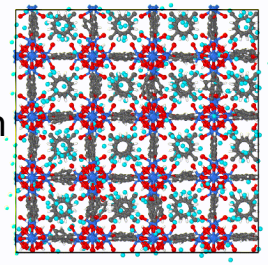


Table 1: Diffusion Constants of Neon at Zero Loading using (a) Rogge et al. Flexible UiO-66 and (b) DP for UiO-66

T (K)	(a) D_S (m^2/s)	(b) D_S (m^2/s)
300	2.12×10^{-8}	1.99×10^{-8}
350	2.43×10^{-8}	2.35×10^{-8}
400	2.83×10^{-8}	2.60×10^{-8}

Future work

- Performing diffusion for more complex systems:
  - Diatomic molecules (first step)
  - Chemical warfare agents like sarin (final goal)
- Using active learning techniques to reduce the number of DFT calculations for training a DP
- Train the potential on a smaller NN to make simulations faster

References

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2. Rogge *et al.*, *CMS* **28**, 1015 (2015)
3. Wang *et al., Phys Comm.* **228** 178 (2018)
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