Computational Screening of Metal Organic Frameworks (MOFS) for Carbon Capture

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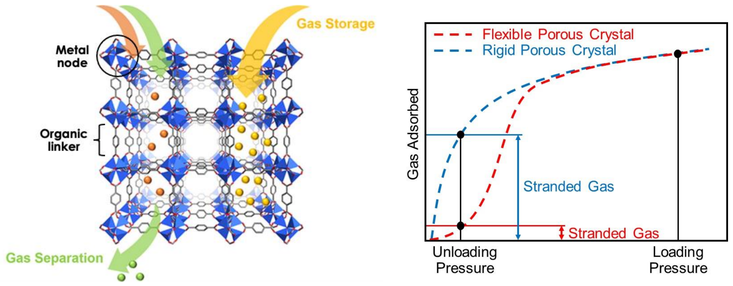
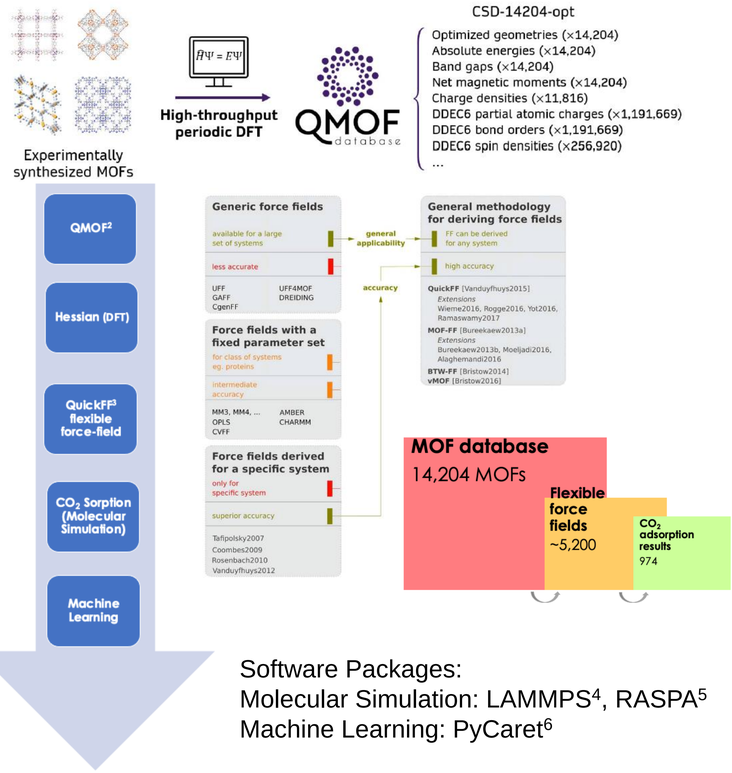
Motivation

- MOFs are organic inorganic crystalline materials
- MOFs have excellent CO_2 sorption
- MOF design space is enormous due to a wide variety of constituents



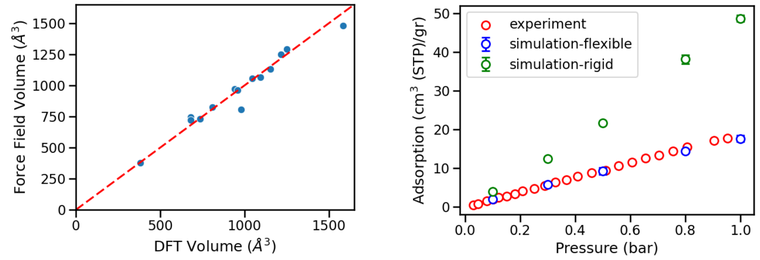
- Subtle flexible modes and large framework volume changes can influence MOF performance regarding adsorption-based applications[1]
- Most of the studies have focused on using fixed-atom force-fields (MOF atoms do not move), which are less accurate
- Flexible force-field describes movement of the MOFs environment, which is a more realistic model

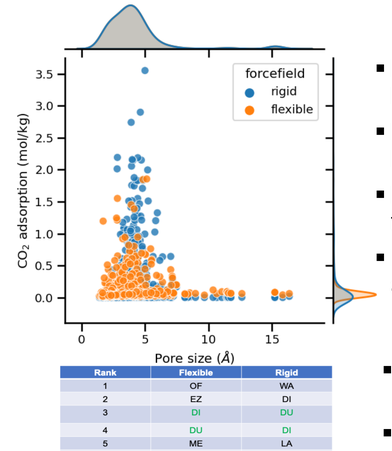
Approach



Software Packages:
Molecular Simulation: LAMMPS[4], RASPA[5]
Machine Learning: PyCaret[6]

Results: Flexible force-field validation and CO_2 sorption at ambient condition

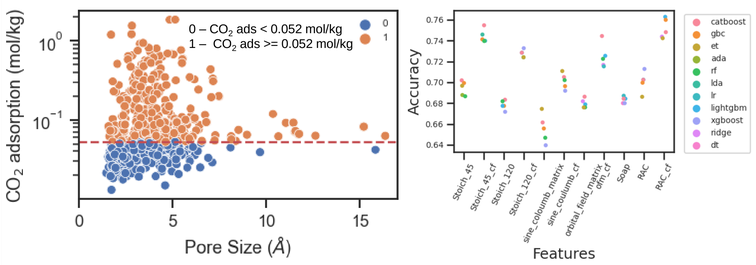




Rank	Flexible	Rigid
1	OF	WA
2	EZ	DI
3	DI	DU
4	DU	DI
5	ME	LA
6	HO	PI
7	DI	EP
8	UJ	PI
9	ME	NE
10	PE	DI

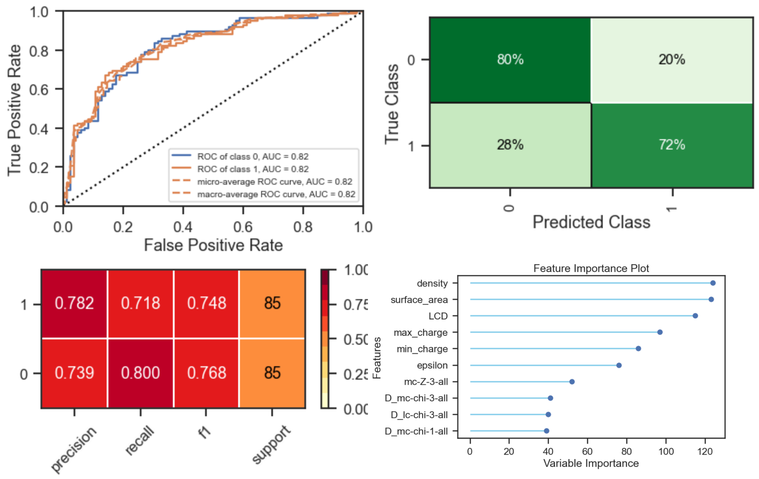
- Flexible force-field yields CO_2 adsorption in MOFs with small pore sizes
- Rigid force-field yields no CO_2 adsorption for MOFs with pore size less than 3 Å
- For some MOFs, rigid force-field overestimates the CO_2 adsorption
- In general, flexibility enhances the CO_2 adsorption in MOFs
- Spearman's ranking correlation coefficient is 0.021
- This is a different ranking of top MOFs for flexible and rigid MOF force-fields

Machine Learning (ML) Classifier



- 11 different classification ML models were evaluated for different MOF featurization techniques
- All the features were customized (denoted by '_cf' in plot) with geometric features such as surface area, pore size, electrostatics, and dispersion term
- Customized RAC_cf (revised auto-correlation) featurization yielded the best accuracy of all the models
- Light Gradient Boosting model (LGBM) algorithm performed the best

ML Results



- ML model has an average accuracy of 75%, which is how often the model correctly identifies a MOF as 0 (low CO_2 adsorbing or 1 (high CO_2 adsorbing)

Conclusions

- Flexible force-fields yielded CO_2 sorption for MOFs with pore size < 3 Å
- Ranking of MOFs based on CO_2 sorption was different for flexible models and rigid models, which underscores the importance of incorporating flexibility for estimation of CO_2 sorption
- LGBM yielded about 75% accuracy in identifying low CO_2 adsorbing MOFs and high CO_2 adsorbing MOFs
- MOF geometric features (pore size, density, surface area) along with electrostatic and dispersion terms were found to be the most important features for the ML model

References

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