PhD Thesis Defense Presentation

Ethylene/Ethane Separation in Metal-Organic Frameworks by Computational Modeling

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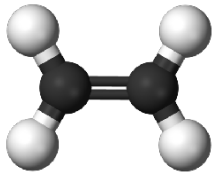
Dr. Matthew J. Realff
Dr. Ryan P. Lively
Dr. Eric Vogel

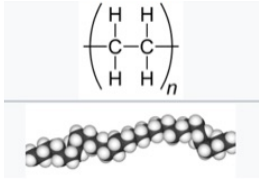
Sholl Group @GT

Chapter 1: Motivation for C_2H_4/C_2H_6 Separation



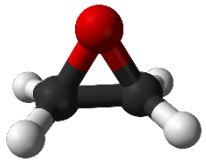
C_2H_4
<u>>150 million tonnes in 2016</u>
(Raw material)

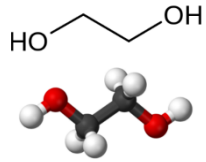
Polyethylene: plastic



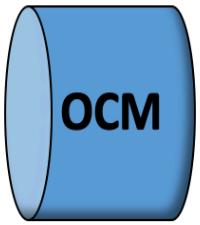
Plant hormone: ripen fruits

Ethylene oxide: Ethylene glycol



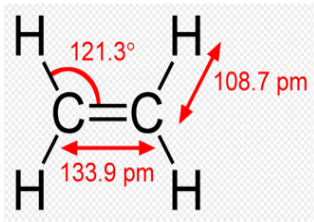


Stream Cracking

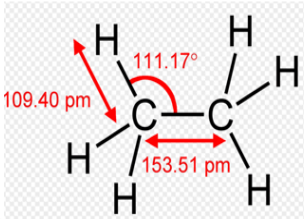


C_2 products

Ethylene: C_2H_4



Ethane: C_2H_6



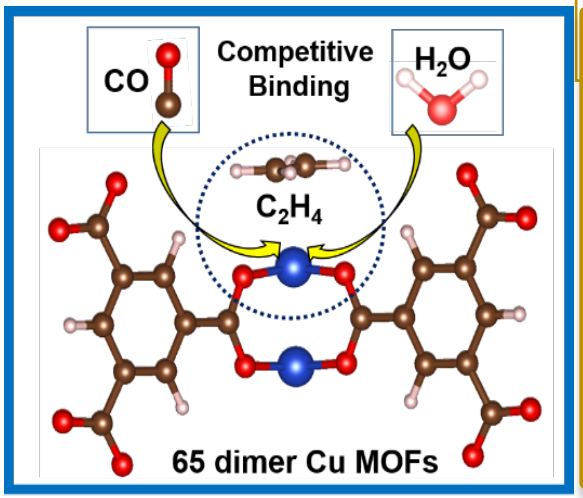
Kinetic diameter	**4.163 Å**	**4.443 Å**
Boiling point	**-103 °C**	**-89 °C**

A key step in producing ethylene is the separation of ethylene from ethane

MOFs with OMS: Adsorption-based C_2H_4/ C_2H_6 Separation

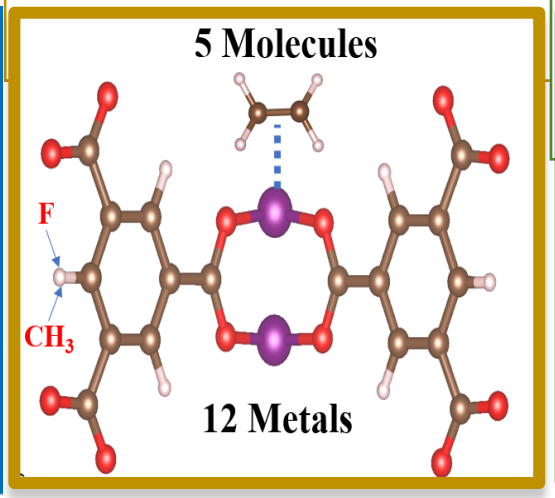
Chapter 2 (Published)

Competitive binding of H_2O, CO, and C_2H_4 in 65 dimer Cu MOFs[1]



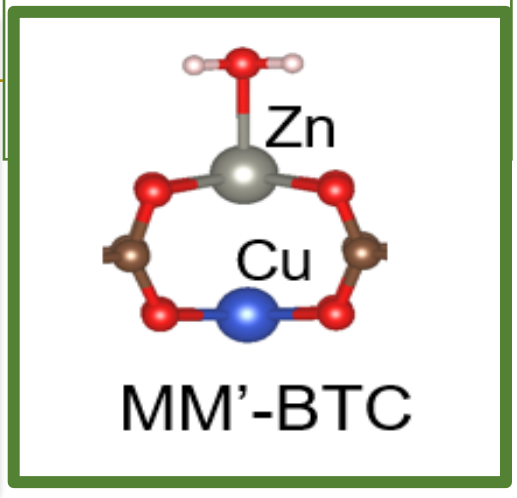
Chapter 3 (Published)

Tune the binding tendencies by metal substitution and linker functionalization[2]



Chapter 4 (Done)

Performance of the mixed-metal MOFs



[1] **You, W.**; Liu, Y.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018,** *122* (16), 8960-8966.

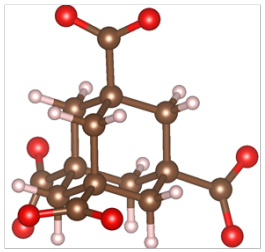
[2] **You, W.**; Liu, Y.; Dai T.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018**, 122(48),27486-27494

Chapter 2: Two Top Ethylene-Selective MOFs

[1]

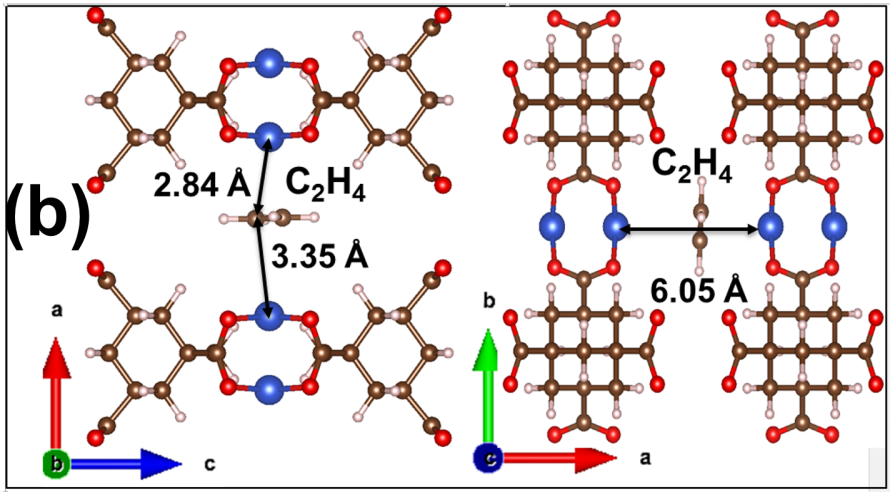
BIMDIL: MOF-11

(a)

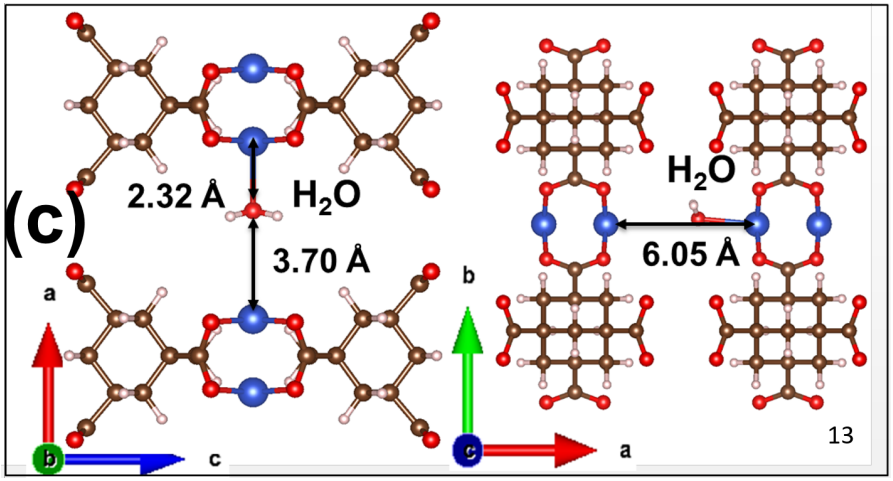


Adamantane cluster

[2]



Notable dual site binding



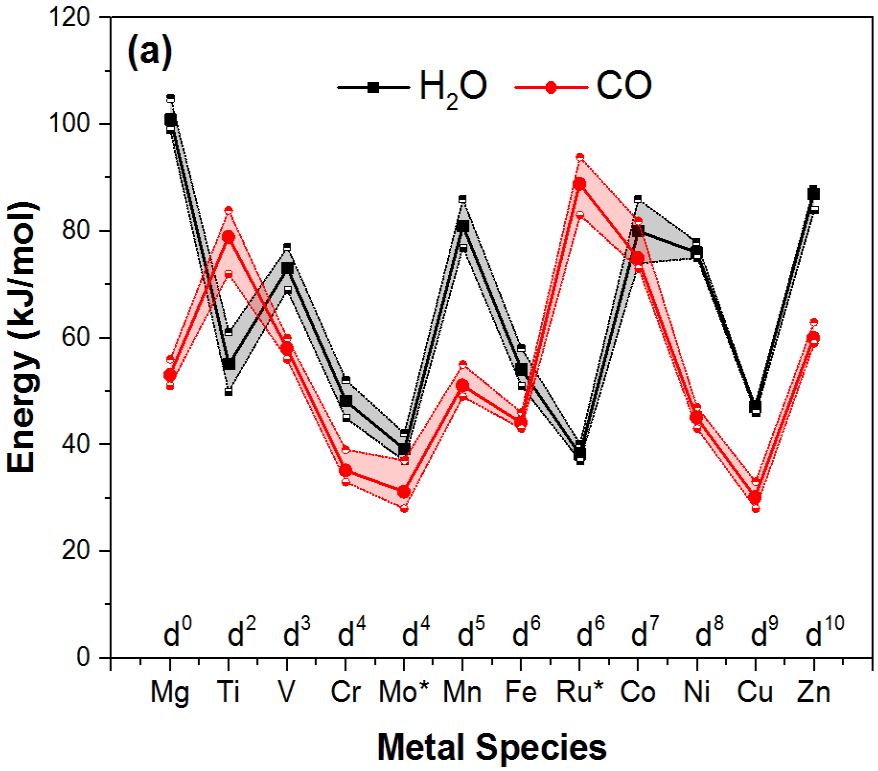
Negligible dual site binding

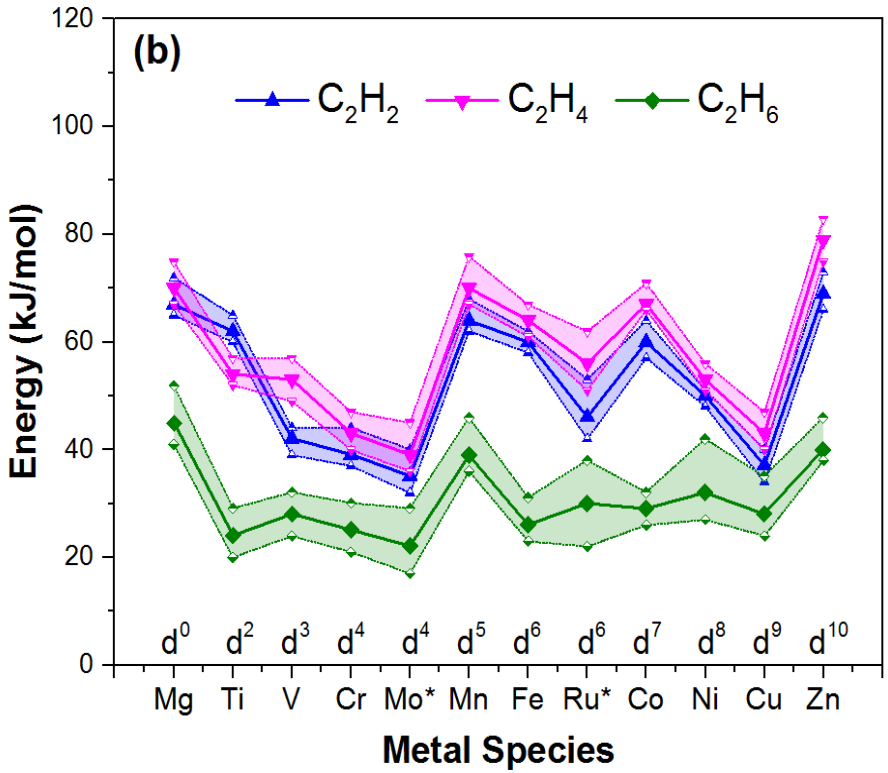
Finding 4: Suggests a possible avenue for developing other MOFs in which the $BE_{C_2H_4}$ is higher than the BE_{H_2O}

[1] Chen, Banglin, et al. *Journal of the American Chemical Society* 122.46 (2000): 11559-11560.

[2] **You, W.**; Liu, Y.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018,** *122* (16), 8960-8966.

Chapter 3: Metal-Substitution on Isostructural MOFs

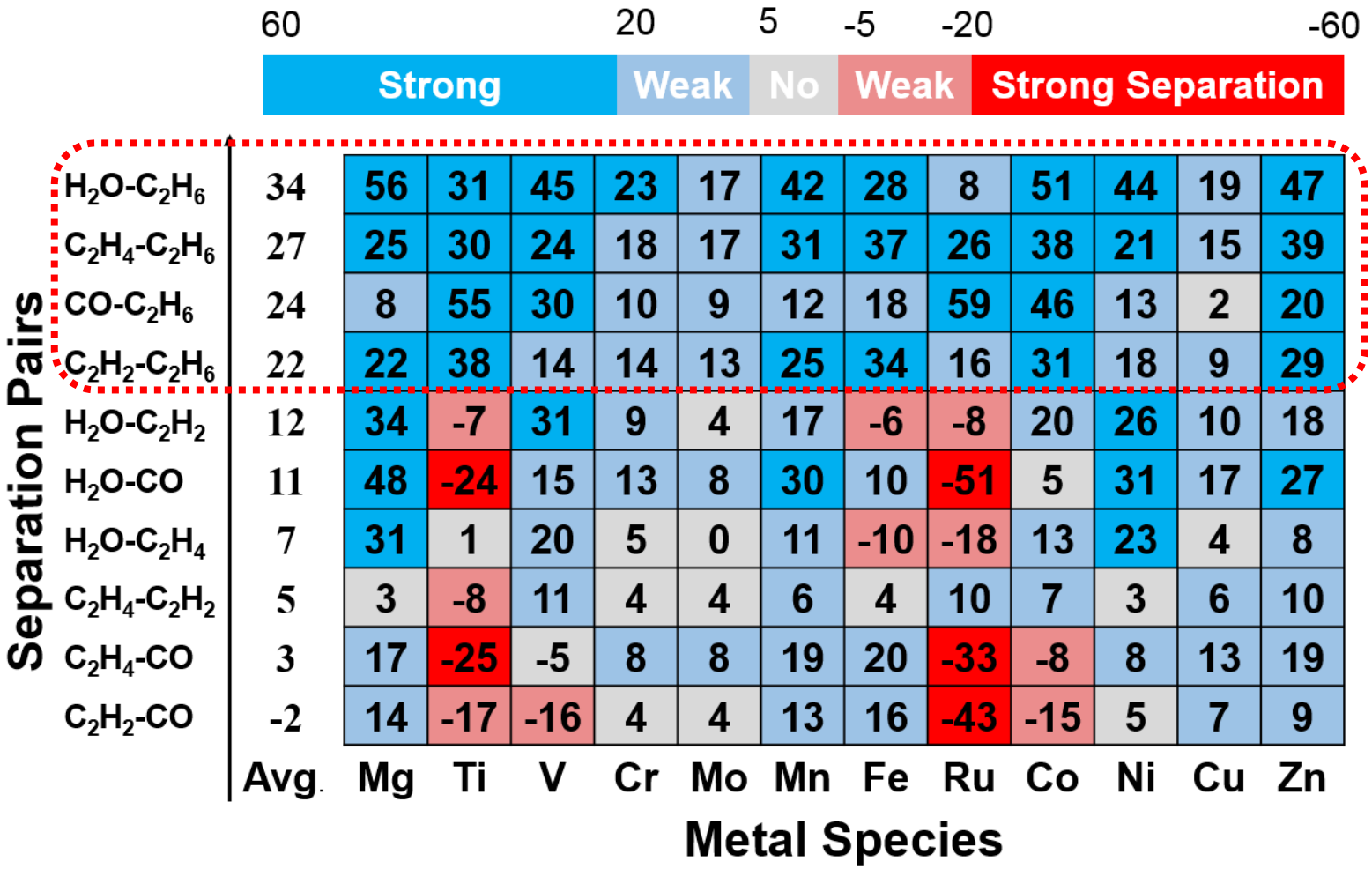




- Each data point shows average of 5 MOFs.
- The energy variation due to metals is stronger than the identity of linker
- C_2H_6 has the lowest binding affinity in all cases.

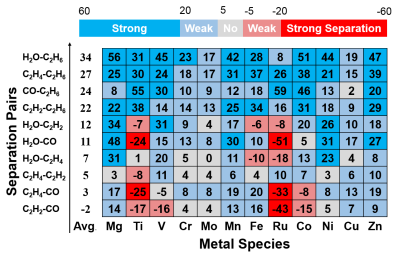
You, W.; Liu, Y.; Dai T.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018**, 122(48),27486-27494

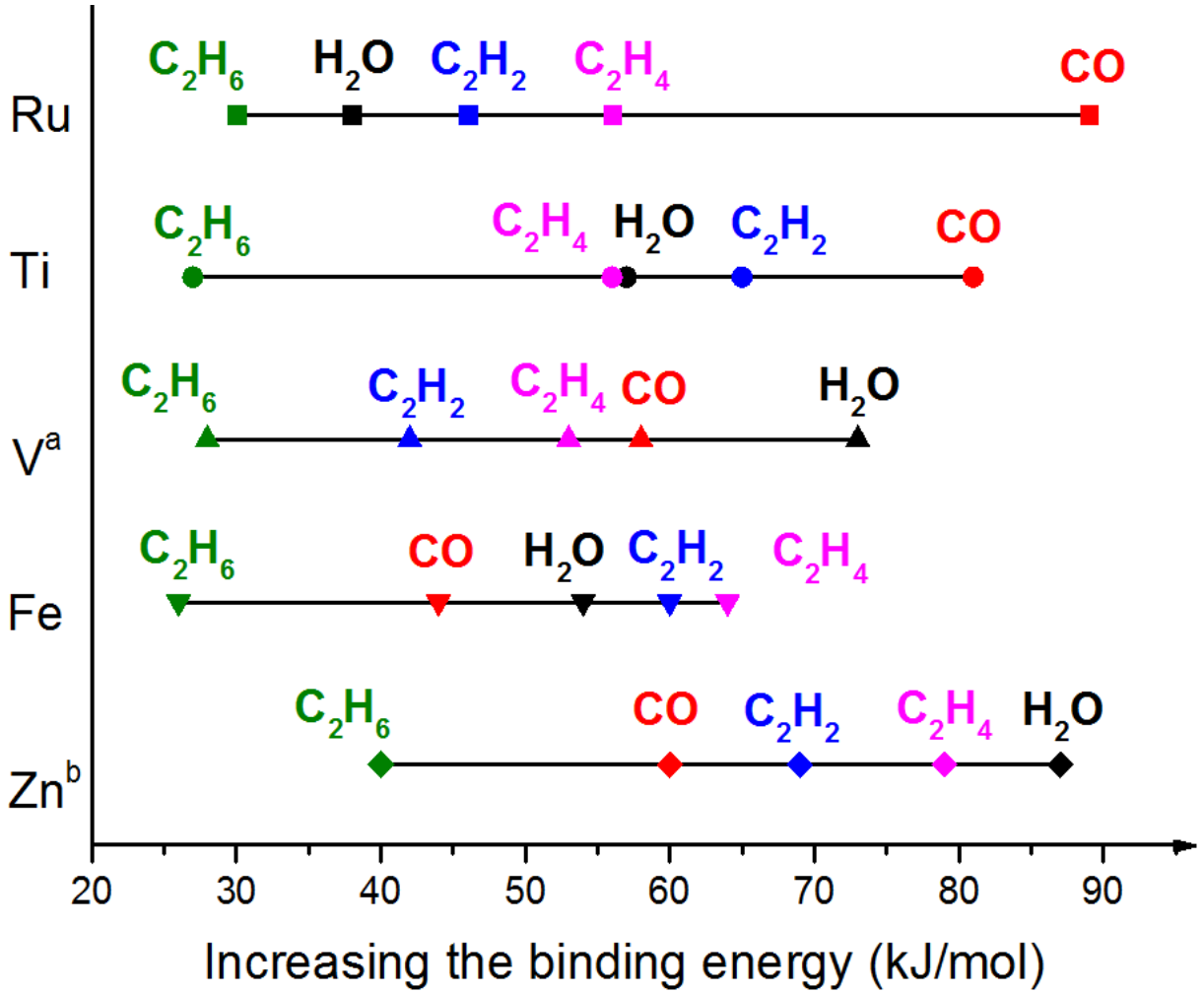
Chapter 3: Relative Binding Energies in 12-metal MOFs



You, W.; Liu, Y.; Dai T.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018**, 122(48),27486-27494

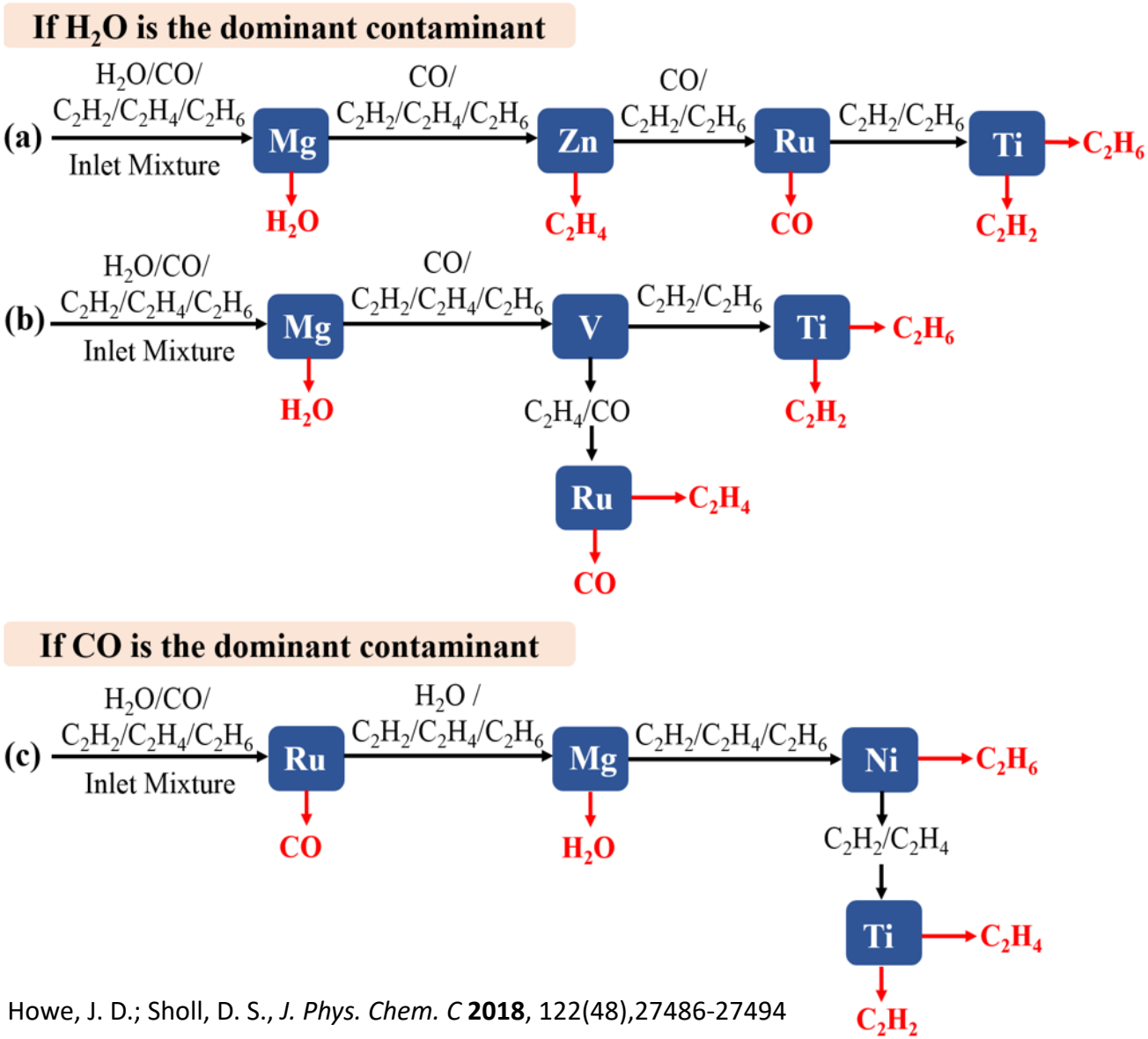
Chapter 3: Five Types of Binding Tendencies



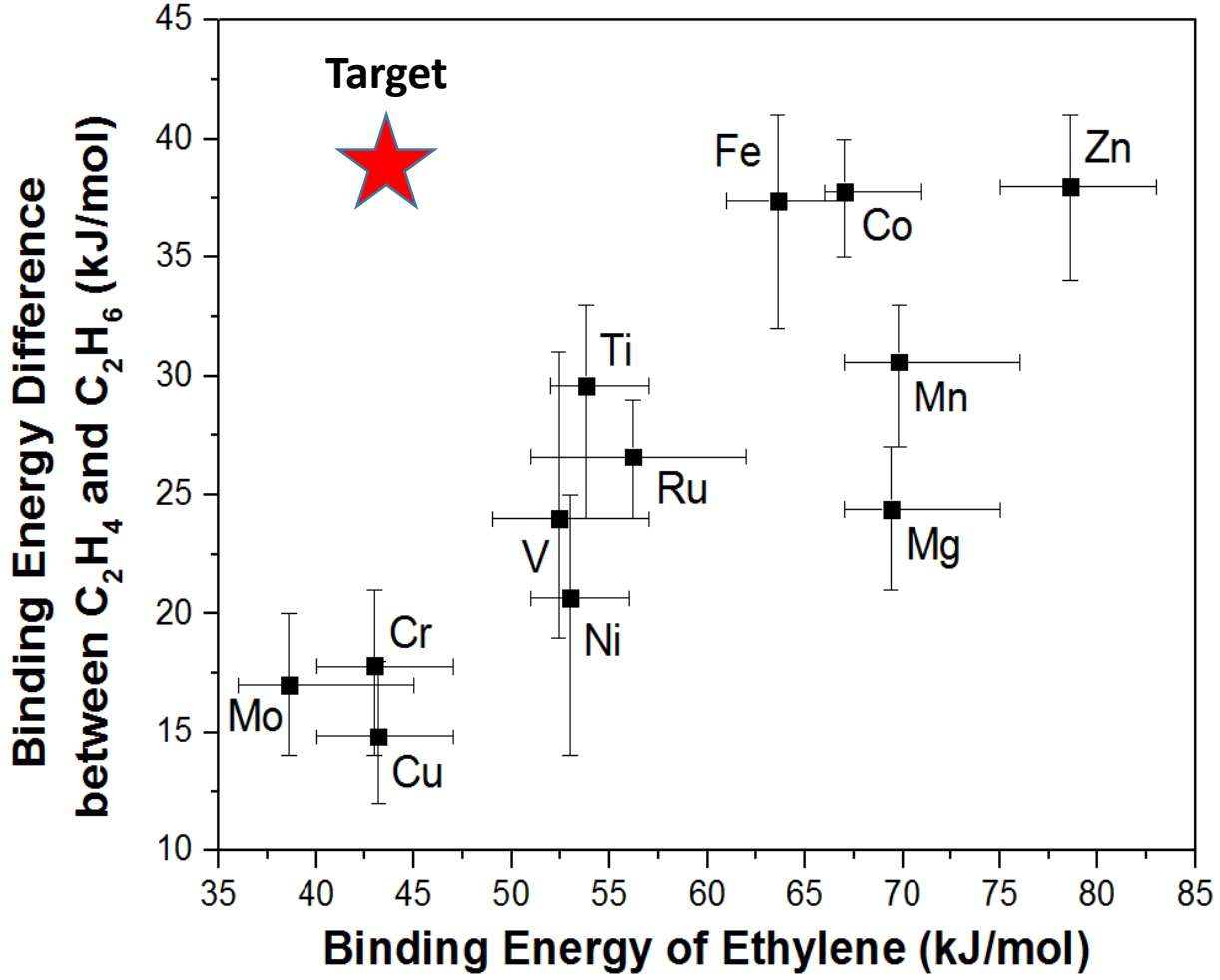


You, W.; Liu, Y.; Dai T.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018**, 122(48),27486-27494

Chapter 3: Separation Strategies



Chapter 3: Ethylene/Ethane Selectivity



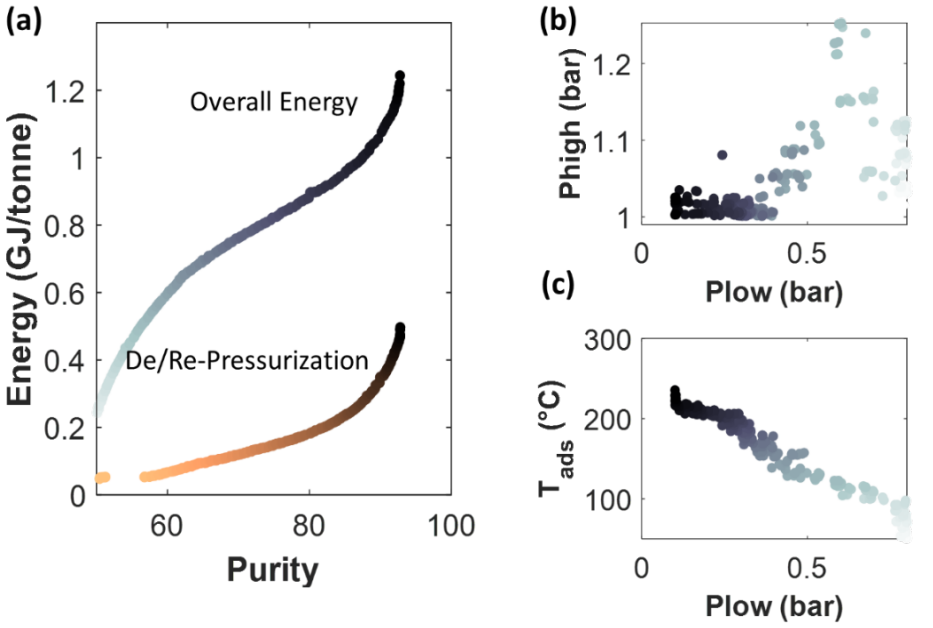
Trade off between larger $\Delta E(C_2H_4 - C_2H_6)$ and smaller $E\_(C_2H_4)$

You, W.; Liu, Y.; Dai T.; Howe, J. D.; Sholl, D. S., *J. Phys. Chem. C* **2018**, 122(48),27486-27494

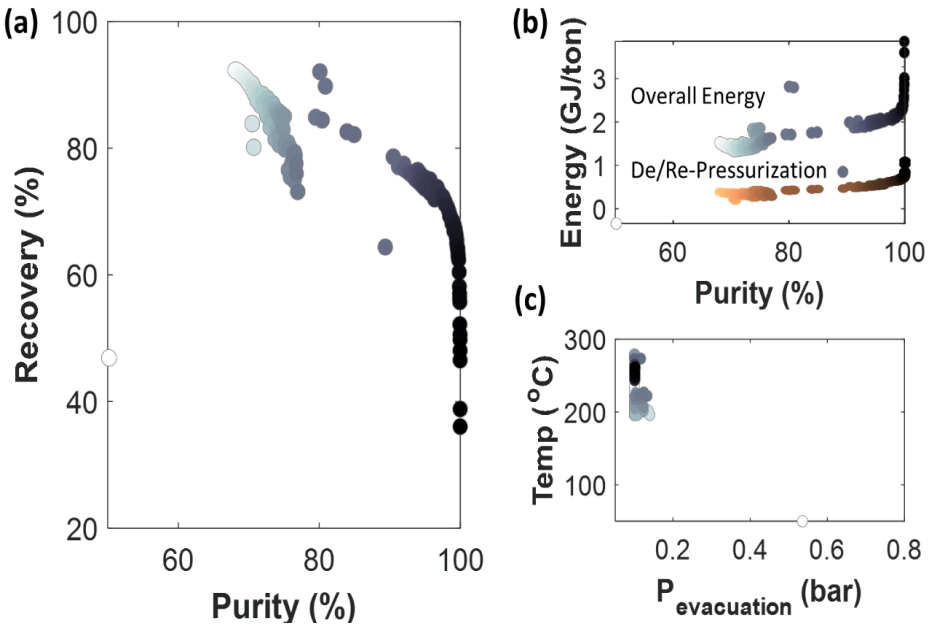
Chapter 4: Optimization of the Adsorption Process

Case Study for the Zn-BTC

Simplified Model



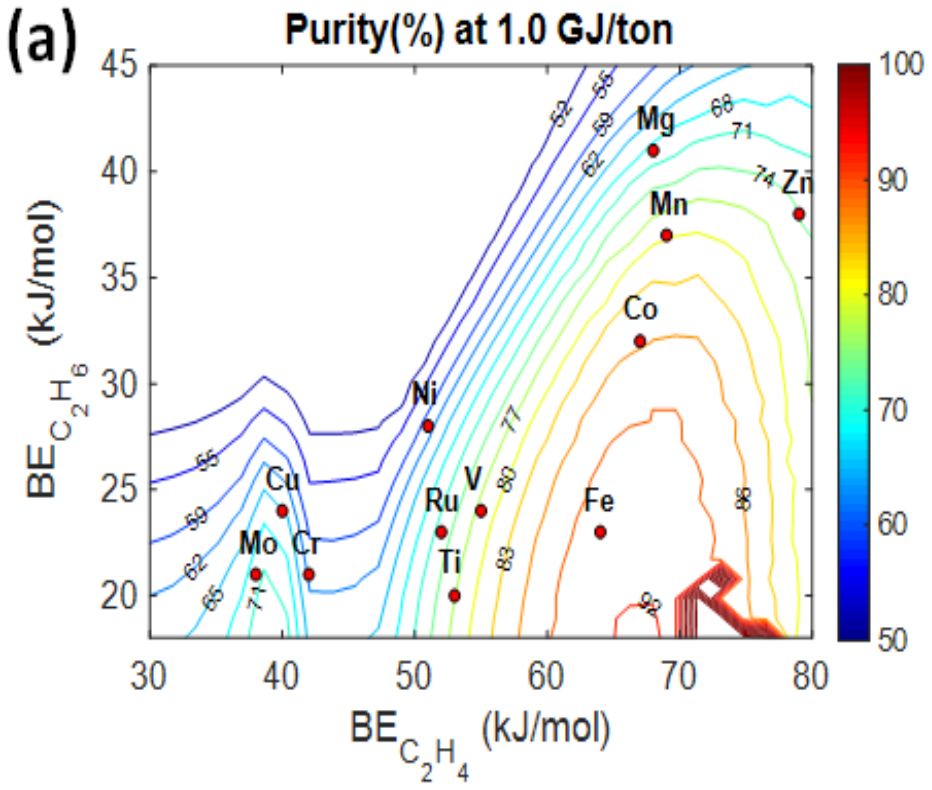
Full Process Model



Finding: Although the simple model underestimates the maximum possible purity and the energy consumed, it still provides a useful general estimate of optimal operating conditions and process performance.

Chapter 4: Performance of Pure Metal MOFs

Simplified Model



Full Process Model