

Developing Relationships between FW/OSDA orientation and AI siting

Craig Waitt,[†] Xuyao Gao,[‡] and William F. Schneider^{*,¶,†}

[†]*Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame,
Indiana 46556, United States*

[‡]*Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre
Dame, Indiana 4656, United States*

[¶]*Department of Chemical and Biomolecular Engineering, University of Notre Dame, Notre
Dame, Indiana 46556, United States*

E-mail: wschneider@nd.edu

Workflow

Objectives

1. Develop models to capture/describe the orientational preference a framework (FW) has for a particular organic structure directing agent (OSDA)
2. Identify FW/OSDA combinations that could influence Al ordering through preferential OSDA orientations.
3. Compute and compare organic and inorganic structure directing agent interactions with frameworks to identify consequences on Si/Al ratios

Plan of Attack

1. **What is the preferential orientational of a specific OSDA/Zelite combination**
 - (a) Develop a model to capture OSDA Orientations
 - Classical molecular dynamics (CMD) at very high temperatures ($T \geq 5,000$) to sample different possible orientation
 - (b) Compute interaction energy (IE) of all FW and OSDA combinations (maybe free energies):
 - CHA and AEI with TMADA, DMDMP (isomers), and DEDMP (isomers)
 - include LTA and TMA?
 - Using CMD at 343 K (rigid framework)
 - Using density functional theory with dispersion (DFT-D3) (flexible framework)
 - Rigid Framework as well?
 - (c) Evaluate orientation and positioning dependence of OSDA in FW

- Measure the orientation of an OSDA relative to some axis which encompasses the cage the OSDA is in
- Measure the location of the OSDA center of mass is relative to the center of the zeolite cage

2. What is the impact of Al siting with respect to OSDA orientation

(a) Using some preferred orientation(s)

- Using the same technique developed by Sichi, run CMD on Al substituted FW and charged OSDA as in 1(a)
- Refine using techniques in 1(b)

(b) Compare relative energies (or IE?)

- Is there a difference between AEI and CHA?

3. What is the impact of co-caging (Relevant?)

(a) Charged or chargeless model