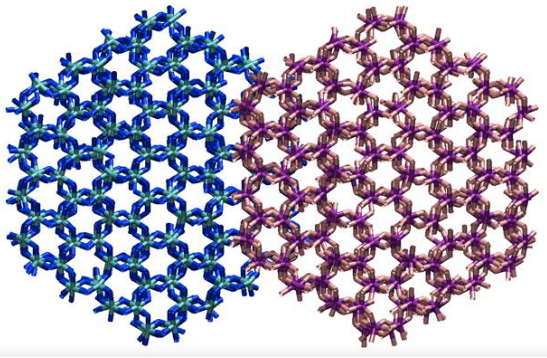
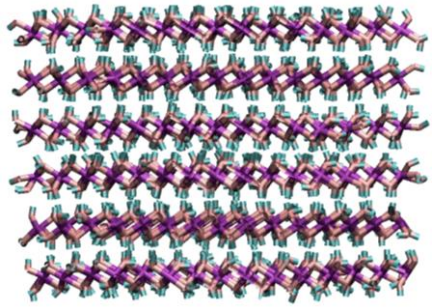
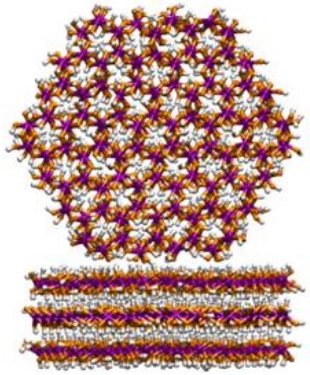
SAND2022-2899C

Sandia National Laboratories

Energetics of Gibbsite Particle Oriented Attachment







Tuan Ho and Louise Criscenti

This work was funded by the DOE Office of Science, Office of Basic Energy Sciences, Chemical Sciences, Geosciences, and Biosciences Division

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

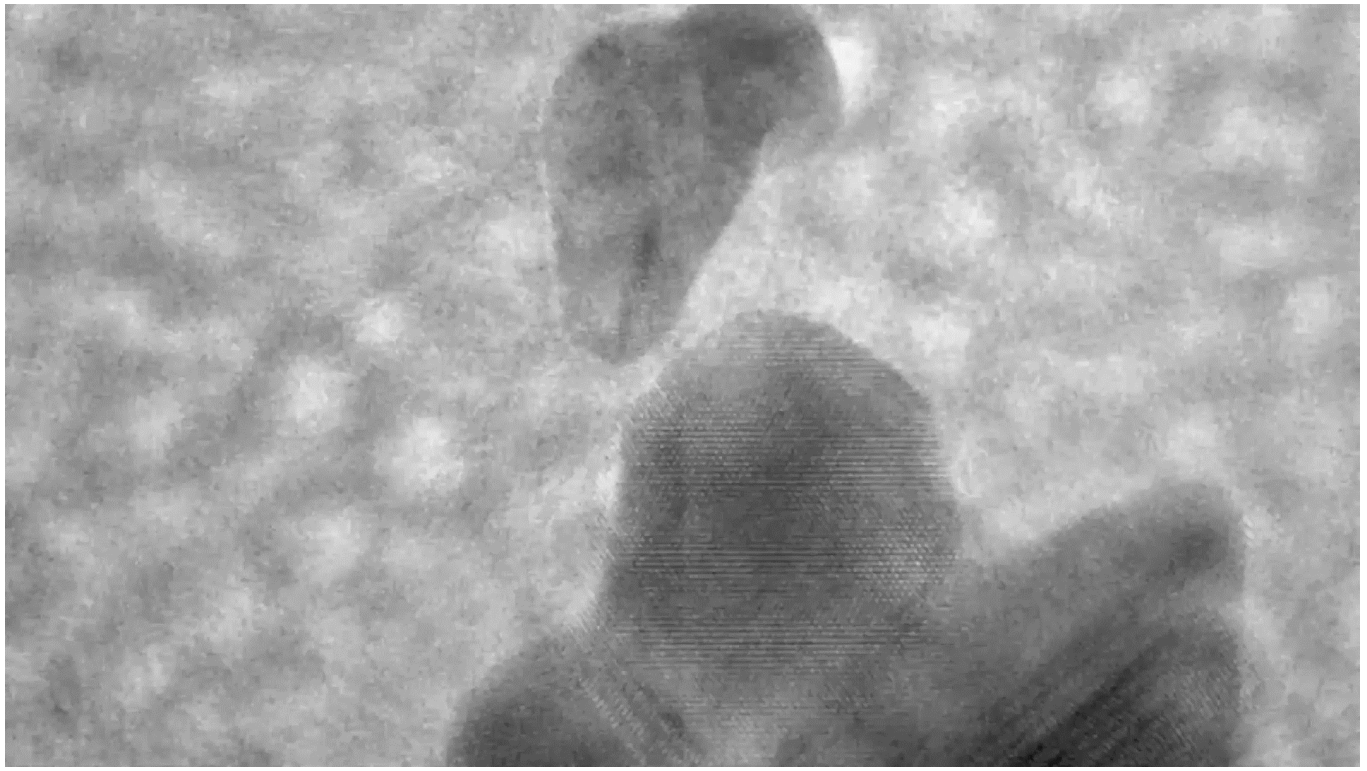
Introduction: particle aggregation and oriented attachment

- Nuclear waste management of Hanford and Savanah River nuclear waste tanks:

Gibbsite and boehmite particles form complex aggregates

- Oriented attachment is a special case of particle aggregation:

Crystalline particles assemble into a larger particle by attaching on specific crystal faces that are lattice-matched.



iron oxyhydroxide nanoparticles

(Li et al, Science 2012, 336, 1014)

Introduction: oriented attachment

- Thermodynamics: particle-particle, particle-solution, and solution-solution interactions
- Kinetics: Brownian motions and experimental conditions (e.g., dehydration)

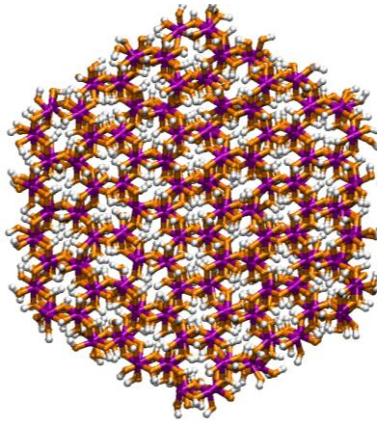
Particle motions: approaching, translating, rotating

Energy-structure relationship?

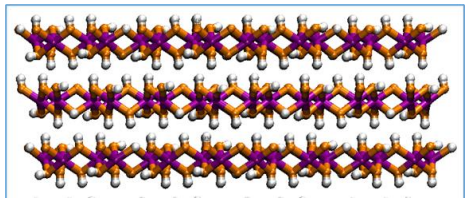
Gibbsite particle oriented attachment

Molecular dynamics simulations

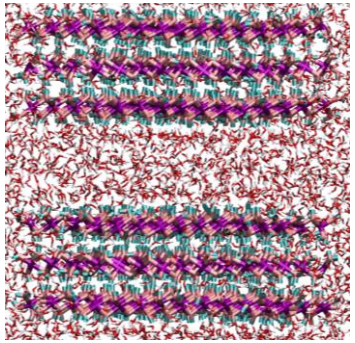
Top view



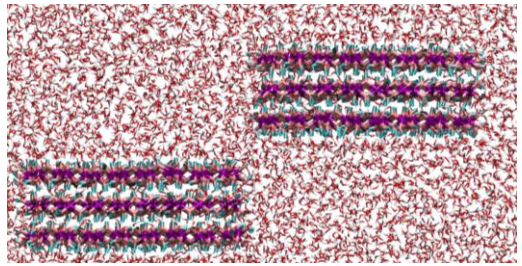
Side view



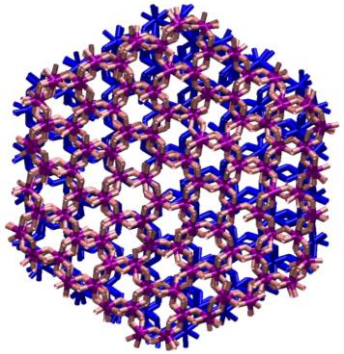
Approaching

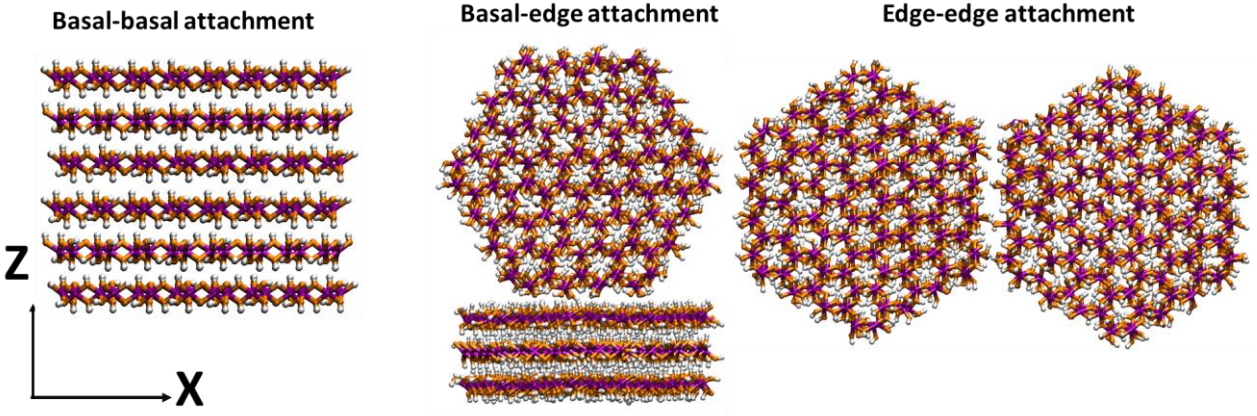


Sliding



Rotating

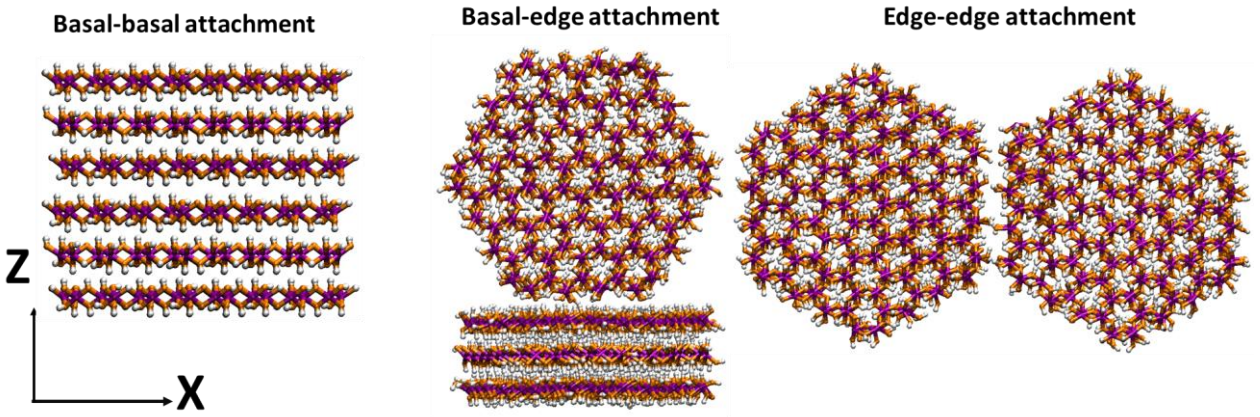


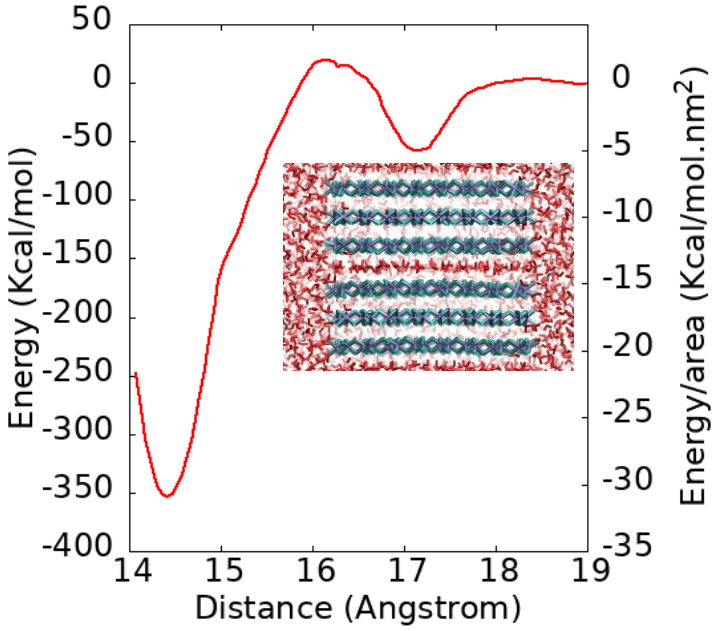


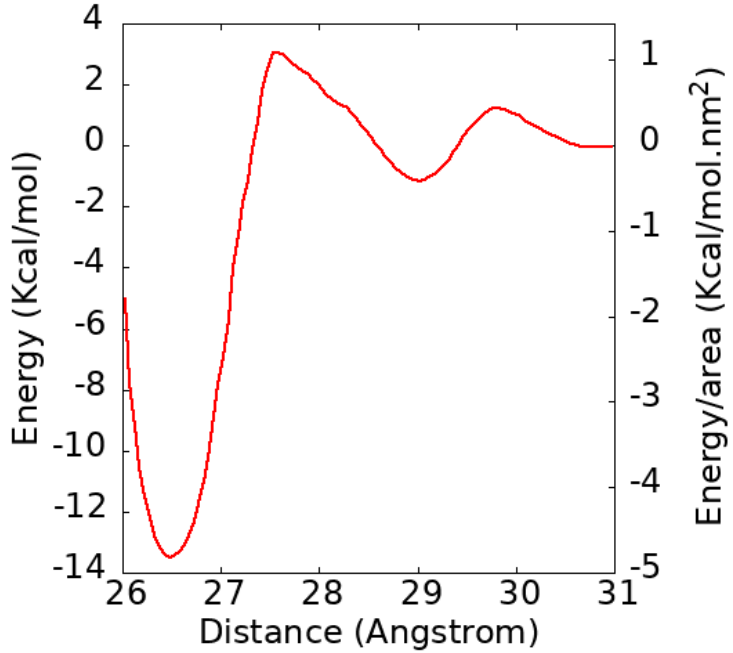
Basal-basal surfaces interactions

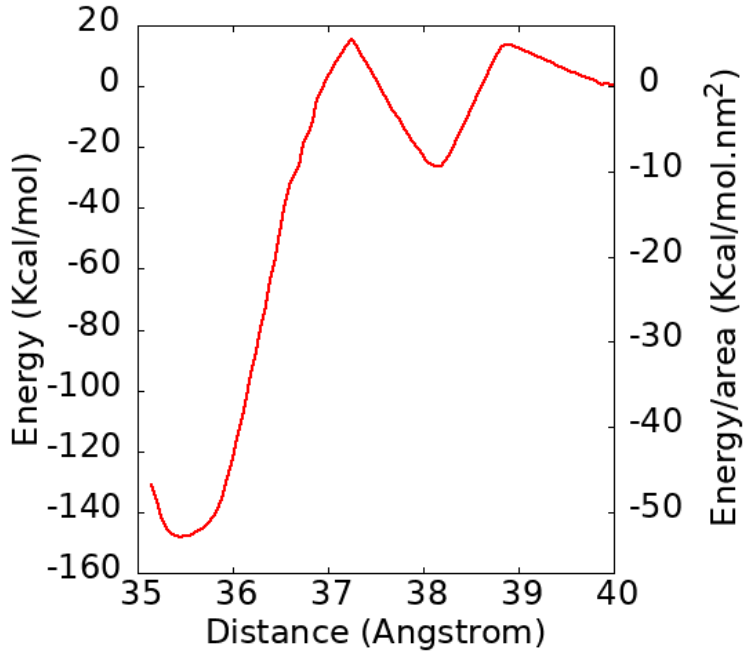
Energy-structure relationships:
Potential of mean force calculations
(very expensive)

Approaching motion





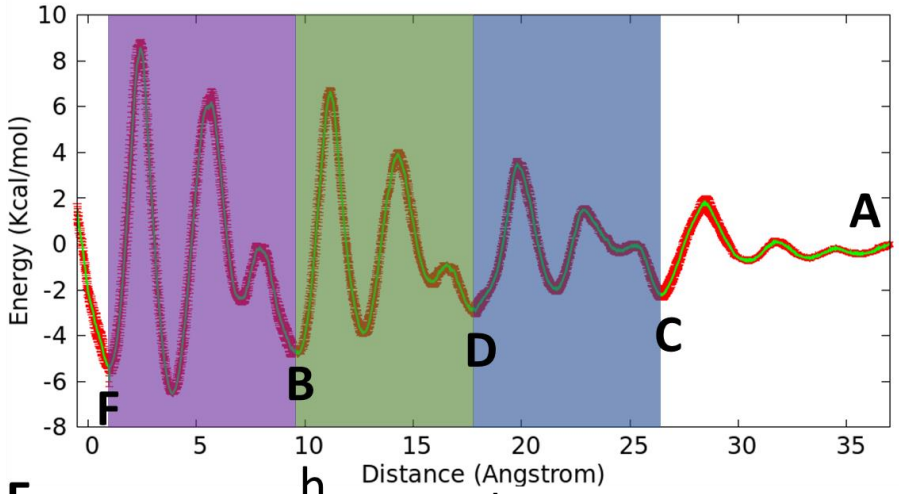


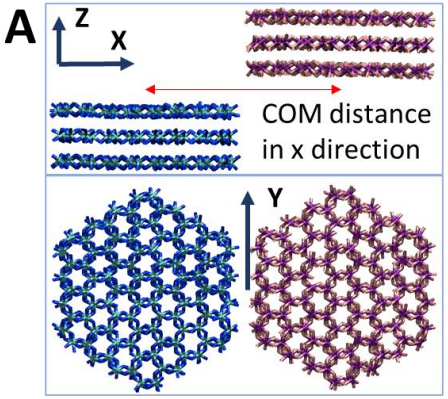


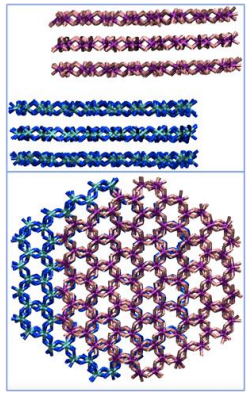
Per surface area: edge-edge attachment is more favorable
Large particle: basal-basal attachment is more favorable

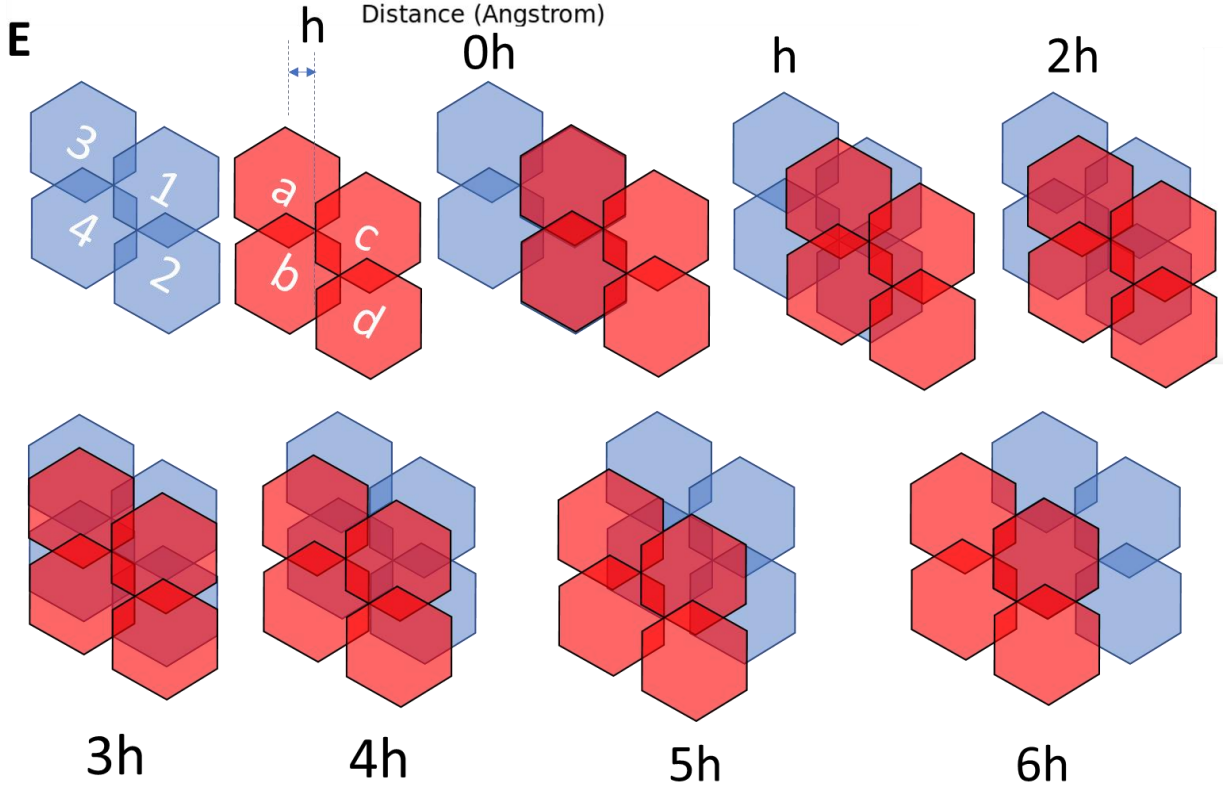
Journal of Colloid and Interface Science 600, 310-317

Sliding motion

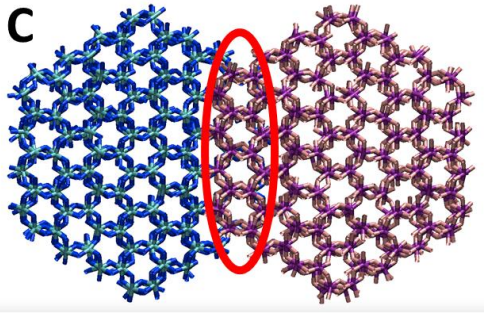


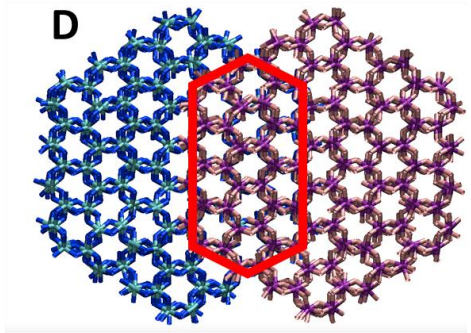


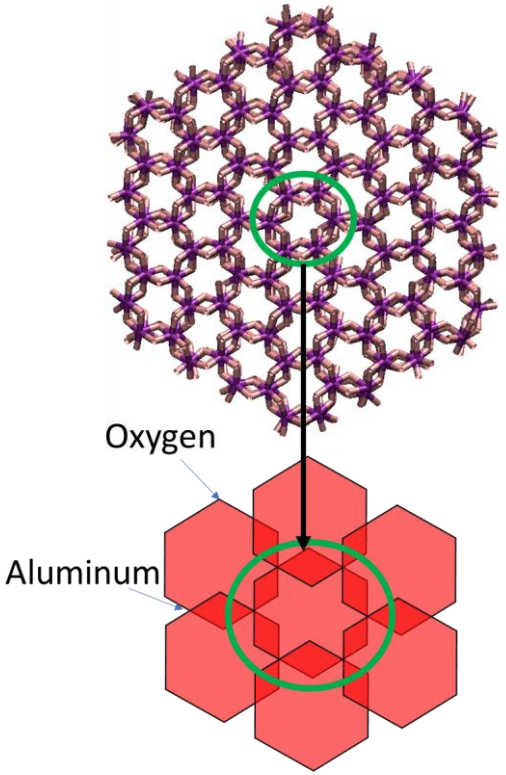




Lattice alignment

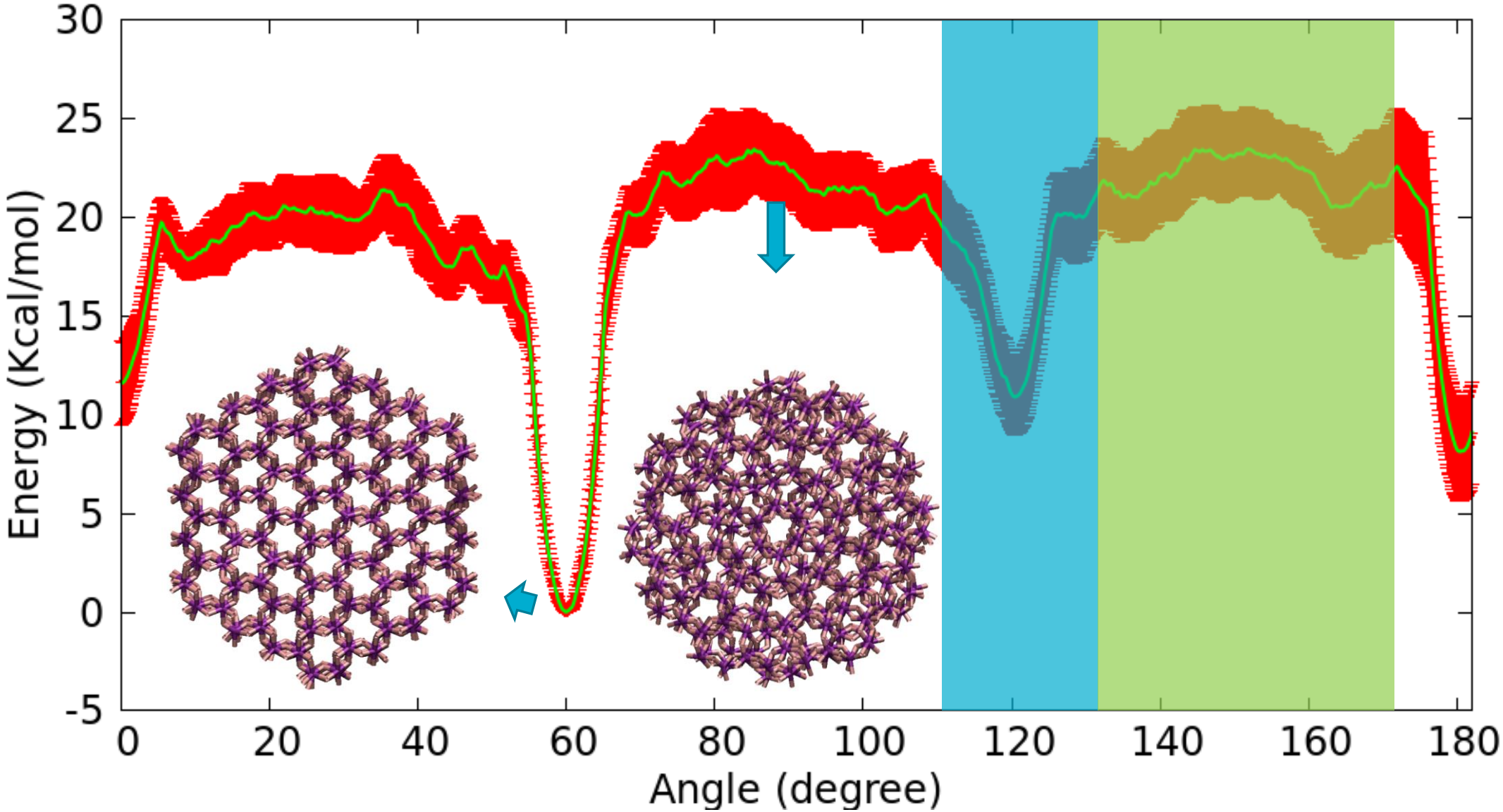






Importance of atom-by-atom mismatch

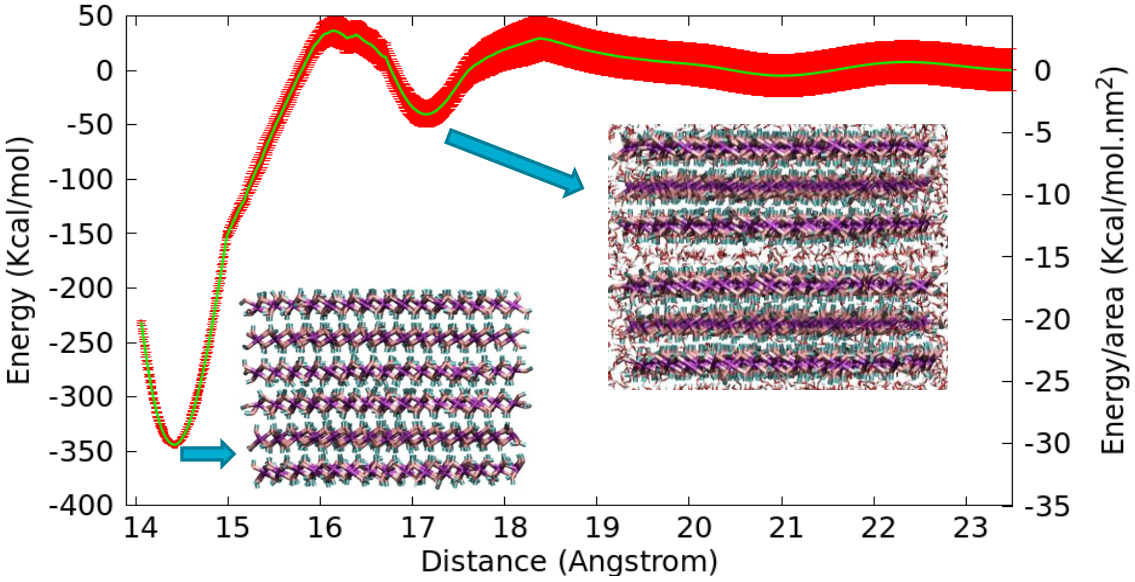
Rotating motion



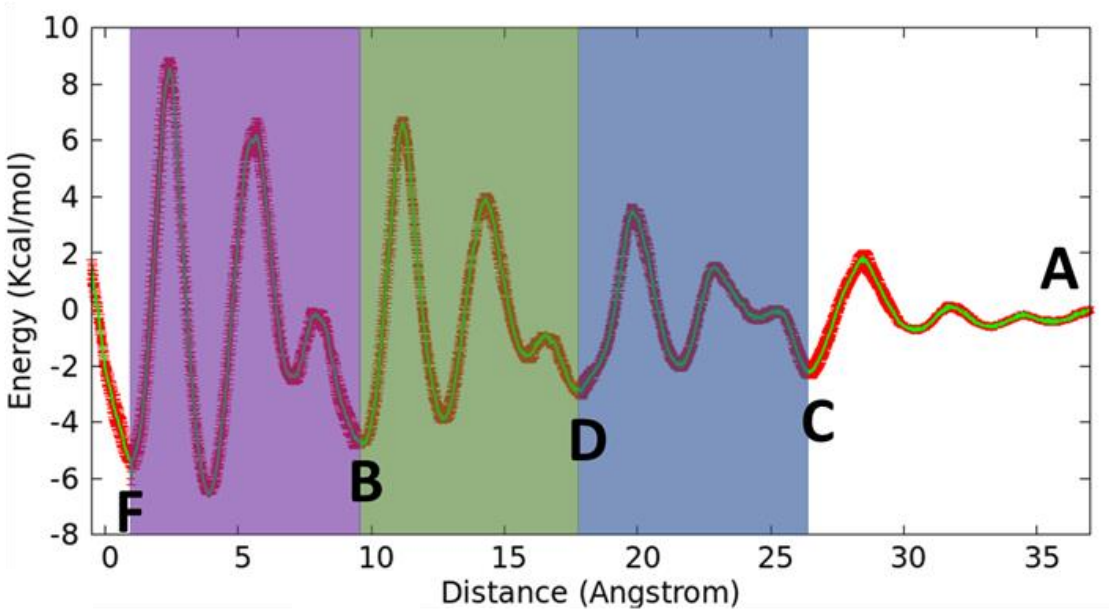
60º periodicity

Energy barriers

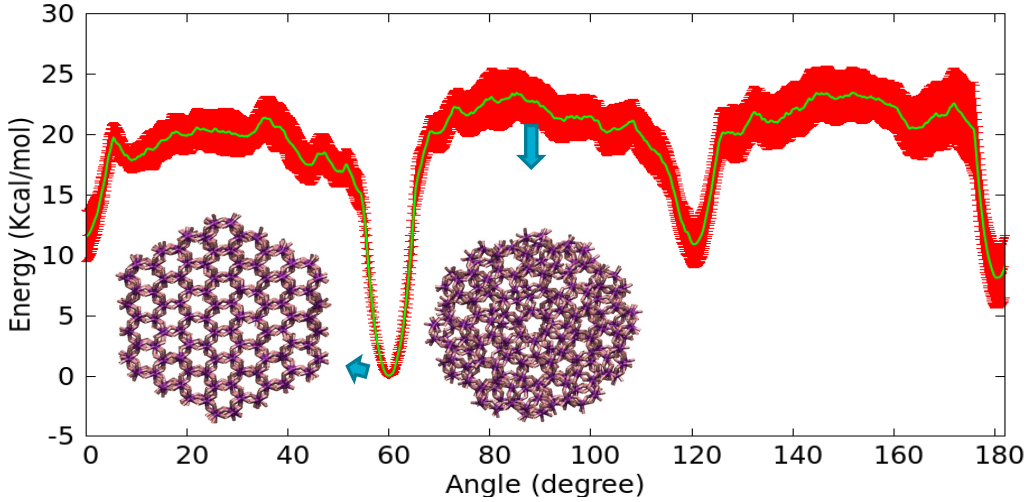
Approaching motion



Sliding motion



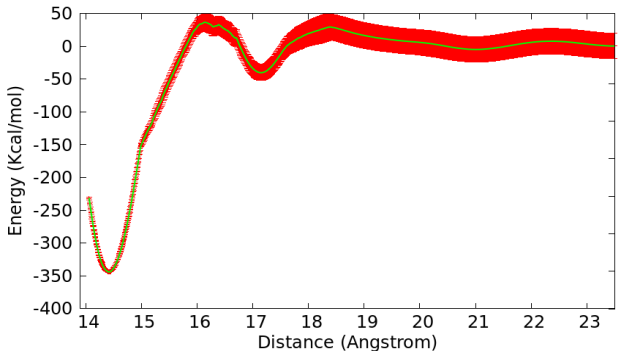
Rotating motion

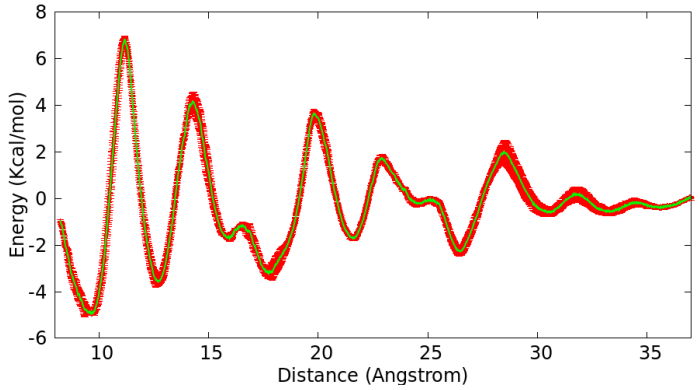


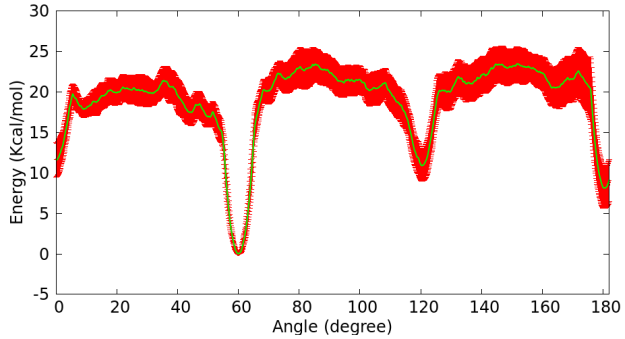
Approaching motion encounters the highest energy barrier

Roles of water

Interaction in water





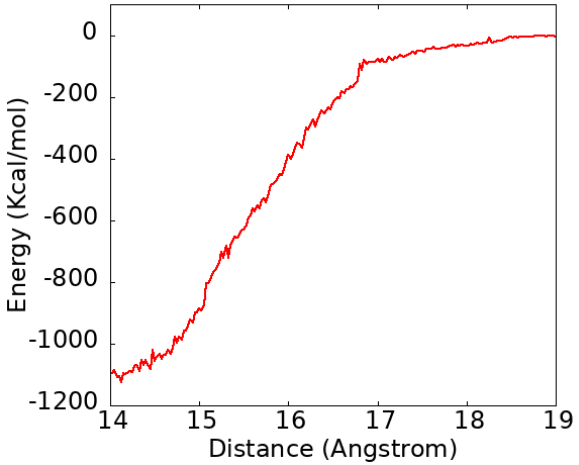


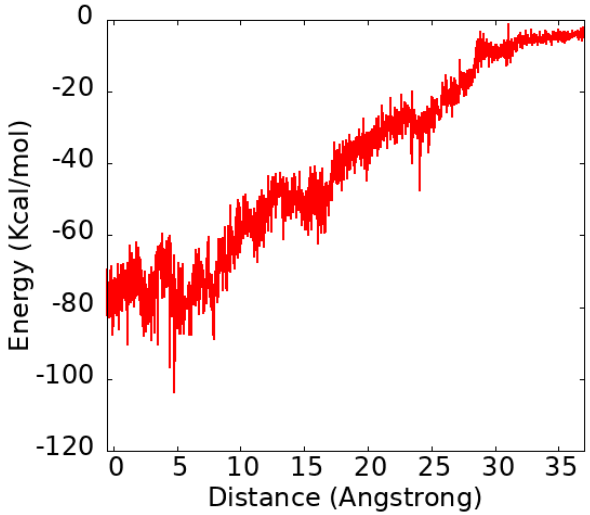
Approaching motion

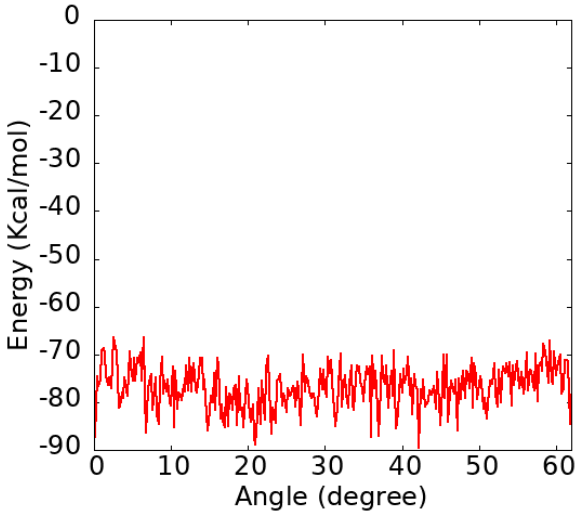
Sliding motion

Rotating motion

Interaction in vacuum



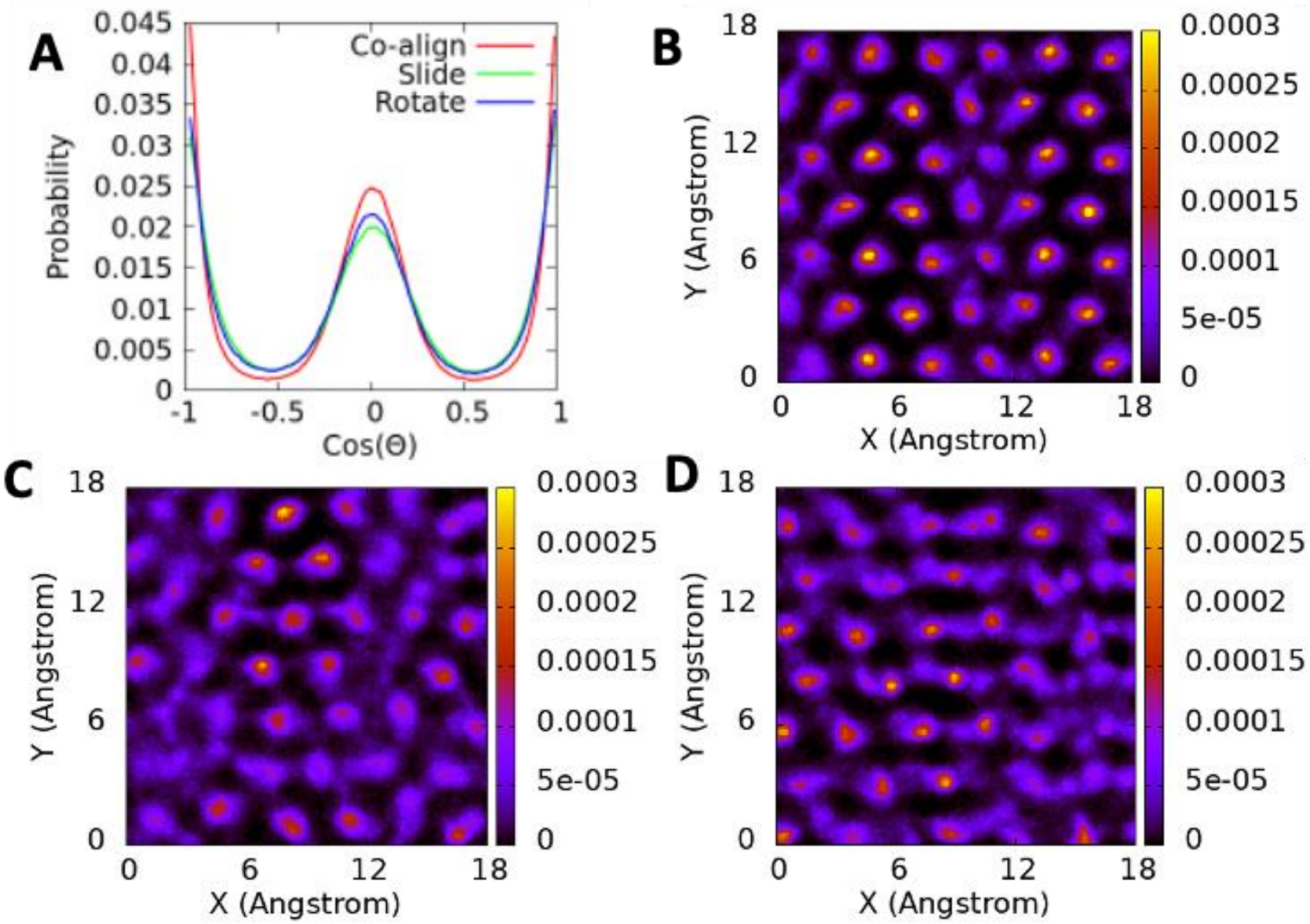




- Water controls the fluctuations in the PMF profiles for all three motions studied
- Water reduces the interaction between two particles
- However, particles still “feel” each other in water.

Water structure

Perfect alignment



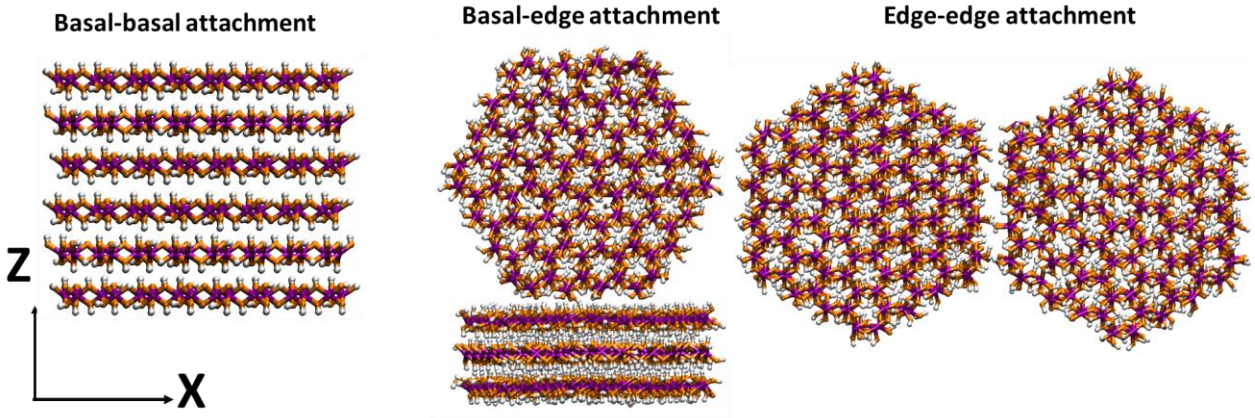
Misalignment (rotating)

Misalignment (sliding)

Water is less structured for the mis-aligned configurations, compared to the co-aligned configuration

Conclusions

- Energy-structure relationships during particle motions: approaching, sliding, and rotating



Per surface area: edge-edge attachment is more favorable
Large particle: basal-basal attachment is more favorable

- Approaching motion encounters highest energy barrier
- Water properties and atom-by-atom mismatch control the energy-structure relationship during the motions

Thank you!

taho@sandia.gov