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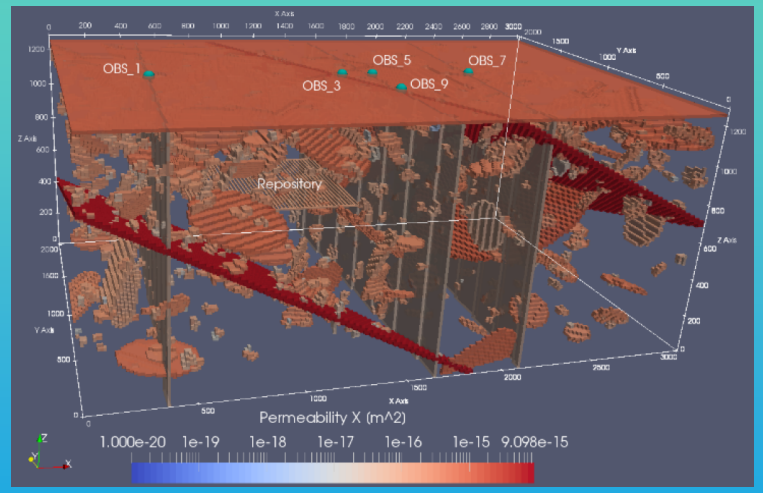

Molecular dynamics simulation of water transport phenomena in smectite

Spent Fuel and Waste Disposition
Annual Program Meeting

Tuan A. Ho, Eric Coker, Carlos F. Jove-colon, and Yifeng Wang
Sandia National Laboratories

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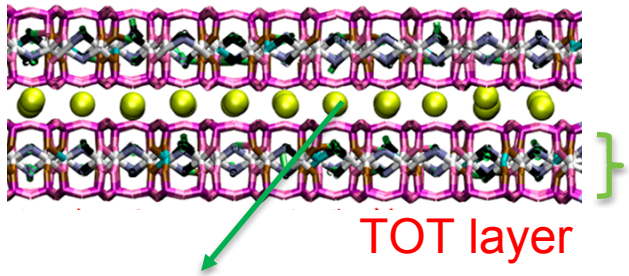
Introduction

Swelling/shrinking of montmorillonite (MMT) with increasing/decreasing relative humidity

Increasing relative humidity (RH)

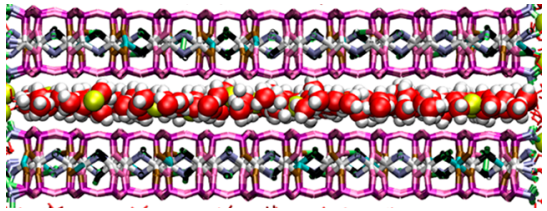
Dry-MMT (0W)



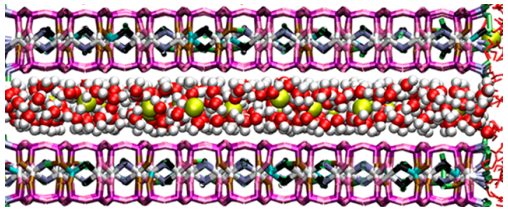
TOT layer

Interlayer cation (e.g. Na^+)

(1W)

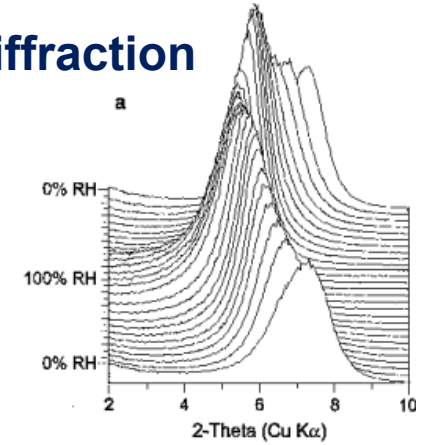


(2W)



Decreasing relative humidity (RH)

in situ X-ray diffraction



Chipera et al, *Advances in X-ray Analysis* 2019, *39*, 713-722.

Our research questions:

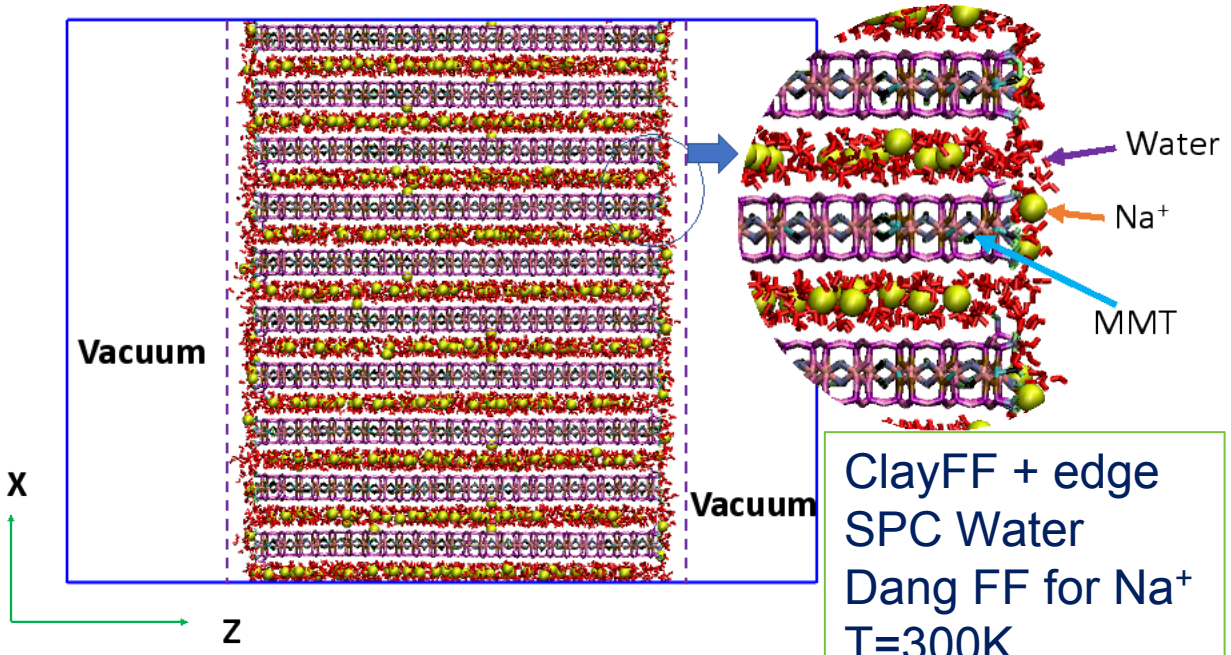
Microscopically, how does the hydration/dehydration process occur?

How does the system transform from 0W to 1W, 2W and vice-versa?

Using combined molecular dynamics (MD) simulation and TGA/DSC/in situ XRD techniques to study the dehydration process

Method

Dehydration MD simulation



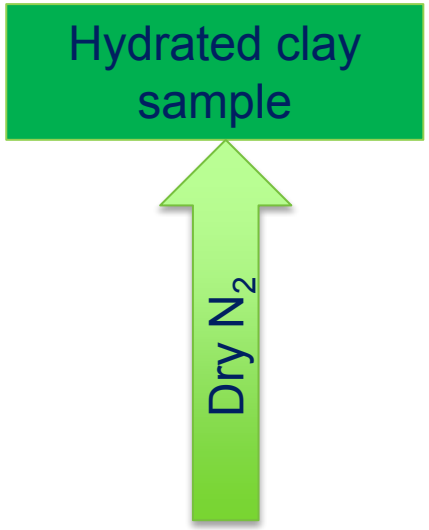
ClayFF + edge
SPC Water
Dang FF for Na^+
T=300K
P=1atm

Unique features:

Remove water molecules that move to the vacuum region
Deform of materials in the X direction

Simulate the transport, chemical, mechanical coupling effects during the dehydration process

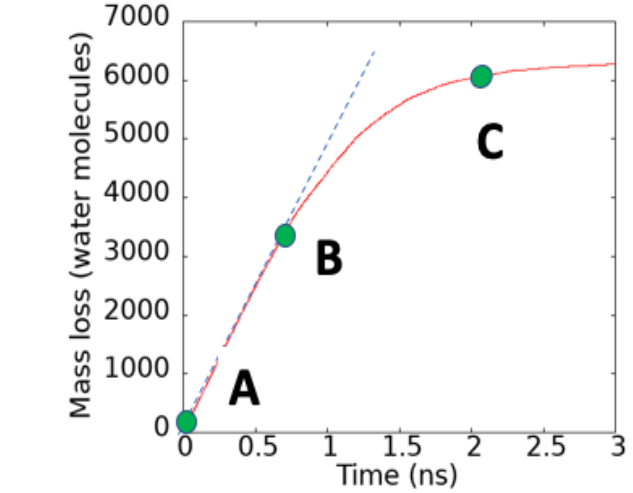
TGA/DSC/in situ XRD experiment on FEBEX bentonite

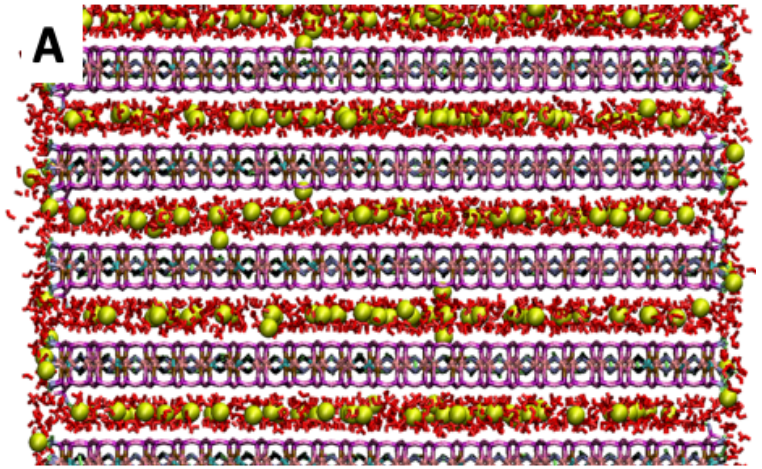


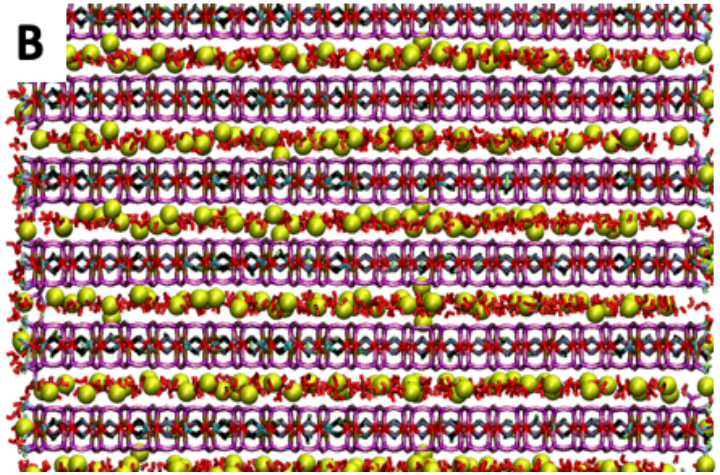
Measurement: Mass loss, heat flow, and the 001 d-spacing

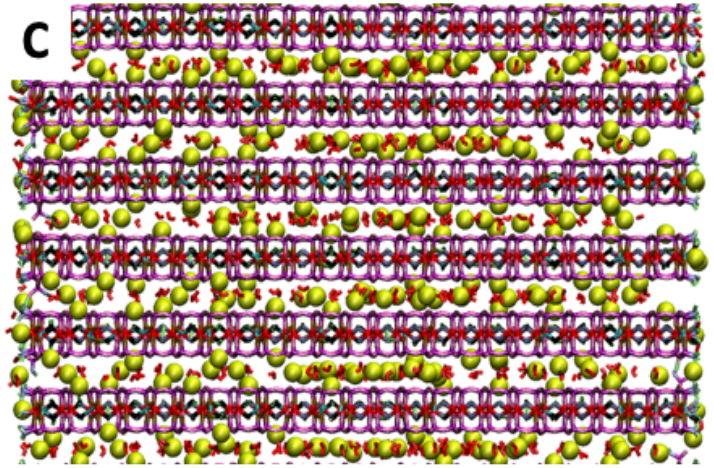
Result: Mass loss

MD dehydration simulation for 2W state



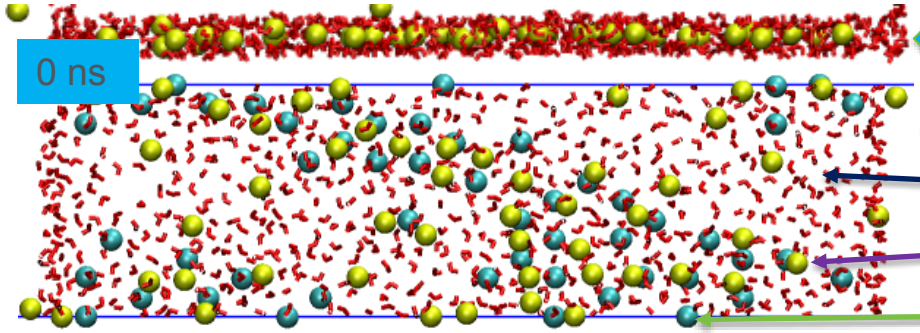






Water transport early in the dehydration process can be by advection in the interlayer

Result: Transport pathway



Side view

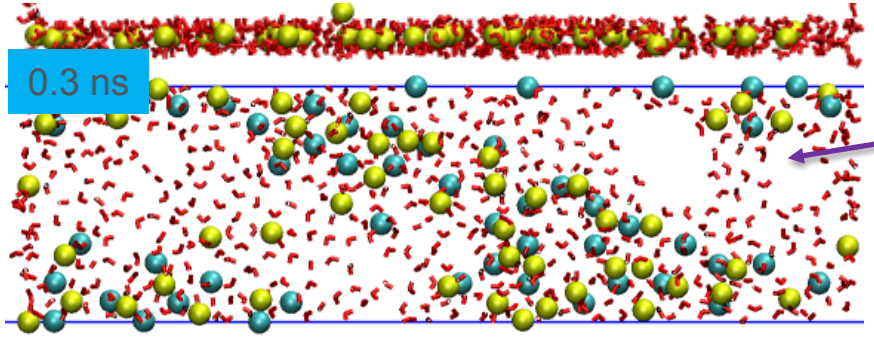
Top view

Water (red)

Na^+ (yellow)

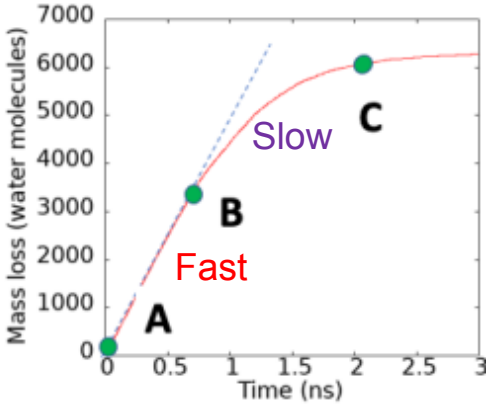
Mg (cyan): substitute for Al in MMT layer

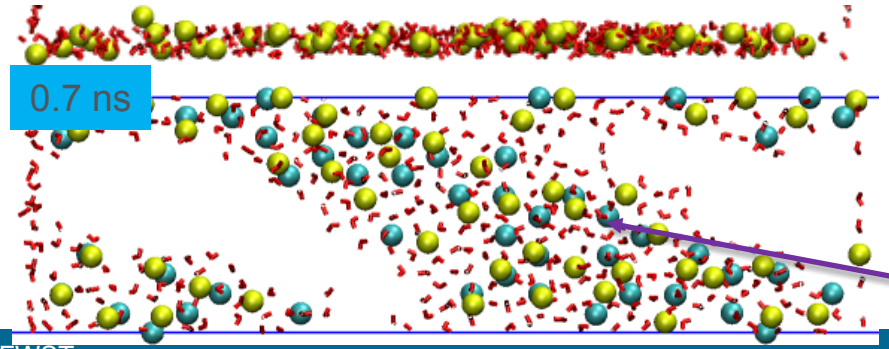
Mg:Al ratio of 0.75:3.25



Hydrophobic/siloxane sites

Water transport is fast near these sites

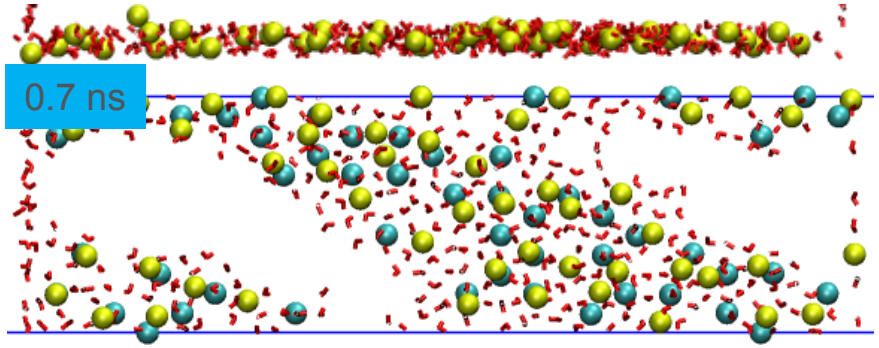


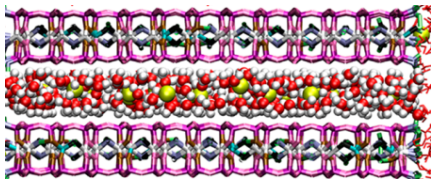


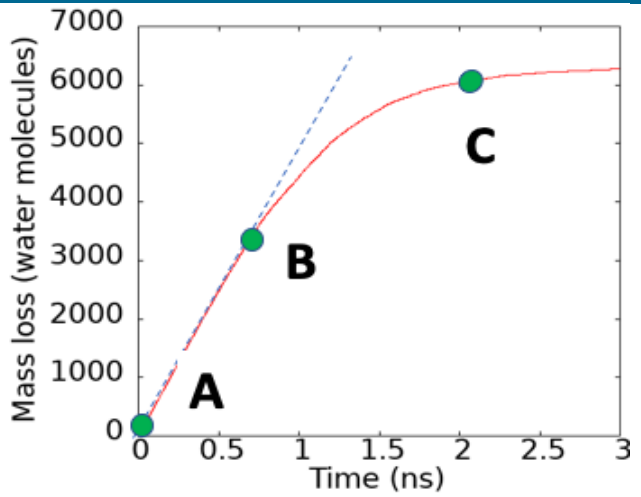
Hydrophilic/Na^+/metal substitution sites

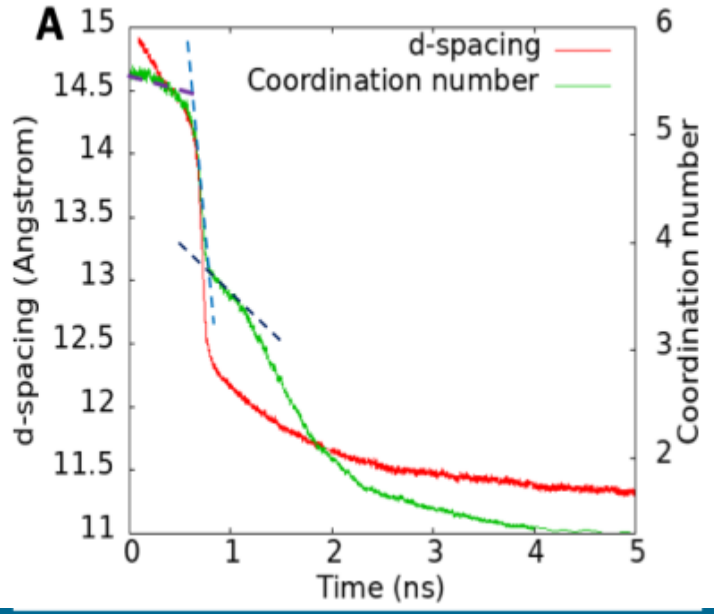
Water transport is slow near these sites

Result: Transport-chemo-mechanical coupling effect



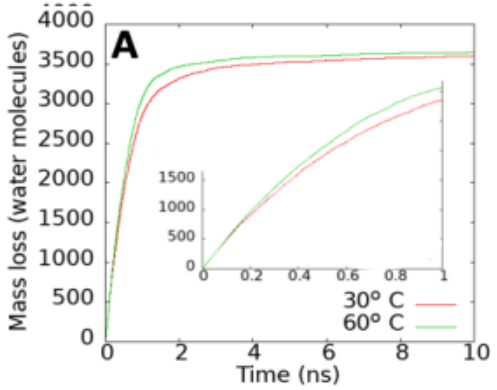


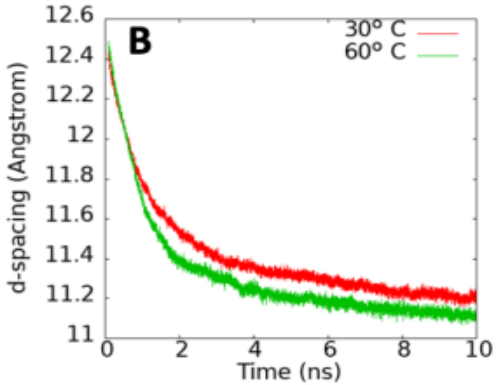


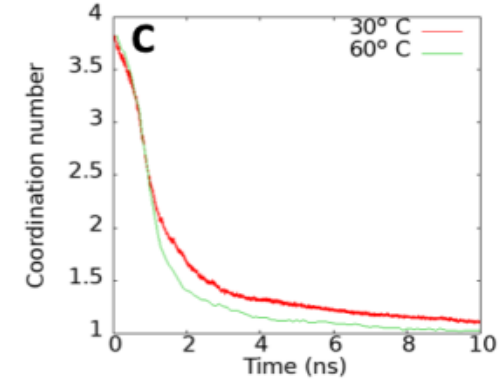


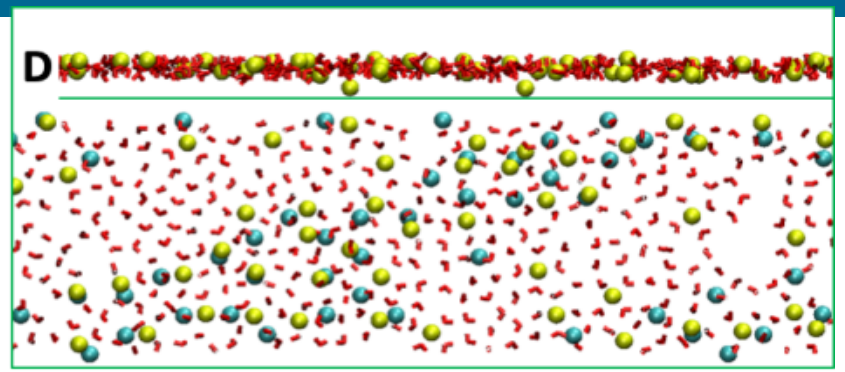
Great correlation between Na^+ coordination number and d-spacing

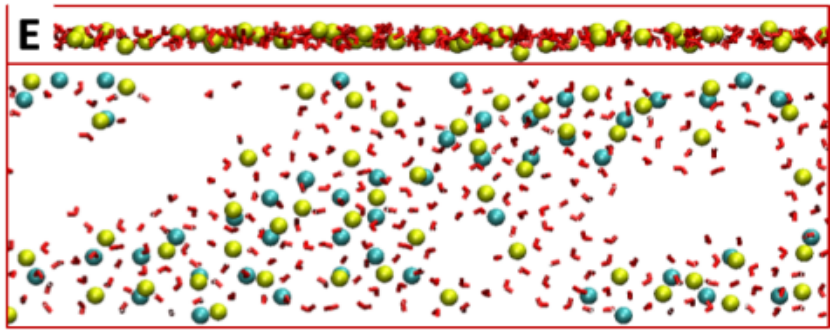
Result: Dehydration process for 1W state

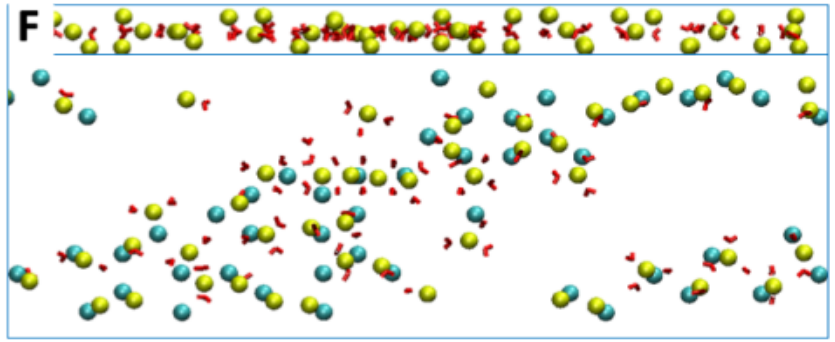




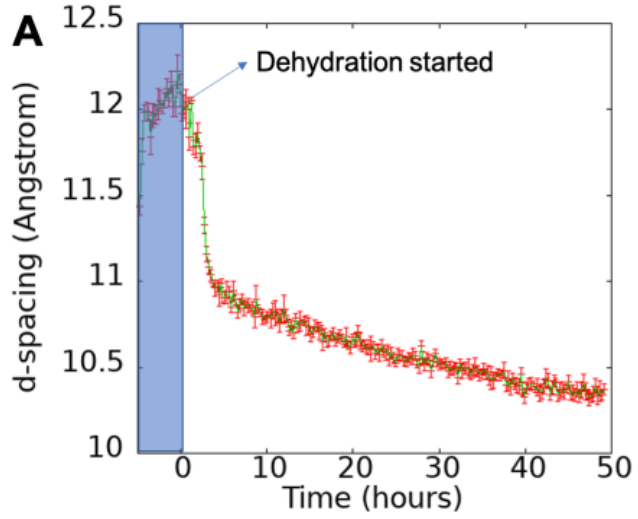


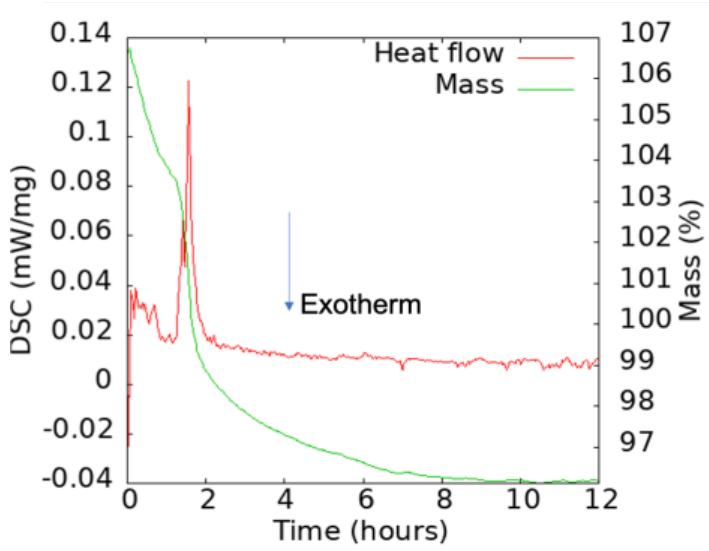




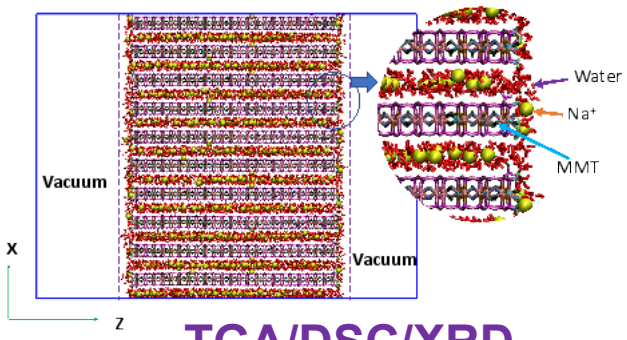


Experimental results

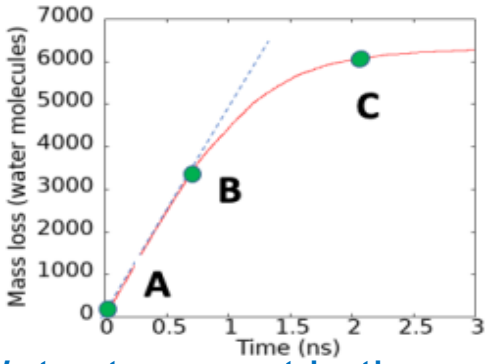


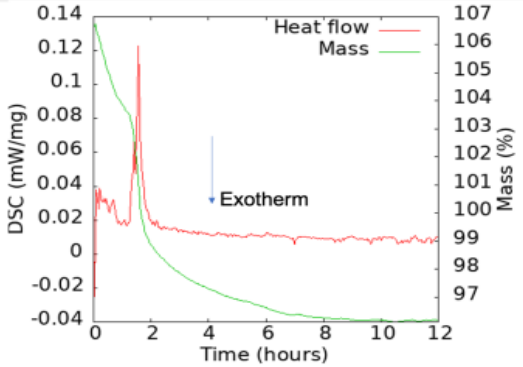


Conclusions

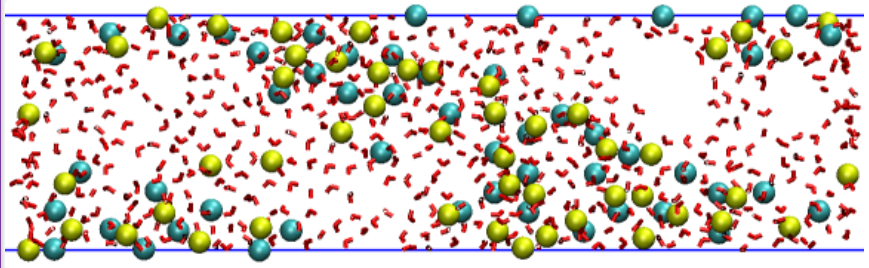


TGA/DSC/XRD





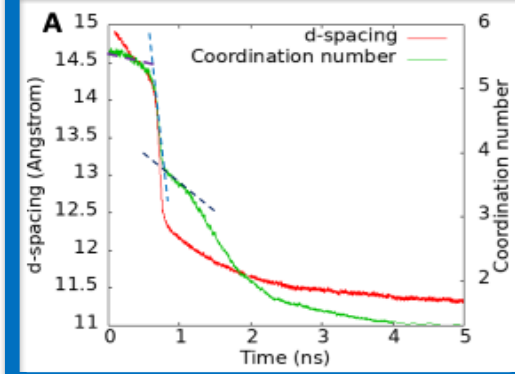
Water transport in the early dehydration process can be an advection



Water transport is:

fast near hydrophobic/siloxane sites

slow near hydrophilic/metal substitutions sites



Chemo-mechanical coupling effect

Future research

- Effect of cation type (Na^+, K^+, Ca^{2+}) on the dehydration process
- Effects of charge location (in the octahedral/tetrahedral sheet) and surface charge density on the dehydration process
- H_2 gas adsorption and transport in the clay interlayers

Thank you!

Questions?