Molecular Simulation of Carbon Dioxide, Brine, and Clay Mineral Interactions

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Sandia National Laboratories

U.S. DEPARTMENT OF ENERGY
Office of Science

TEXAS

NNSA National Nuclear Security Administration

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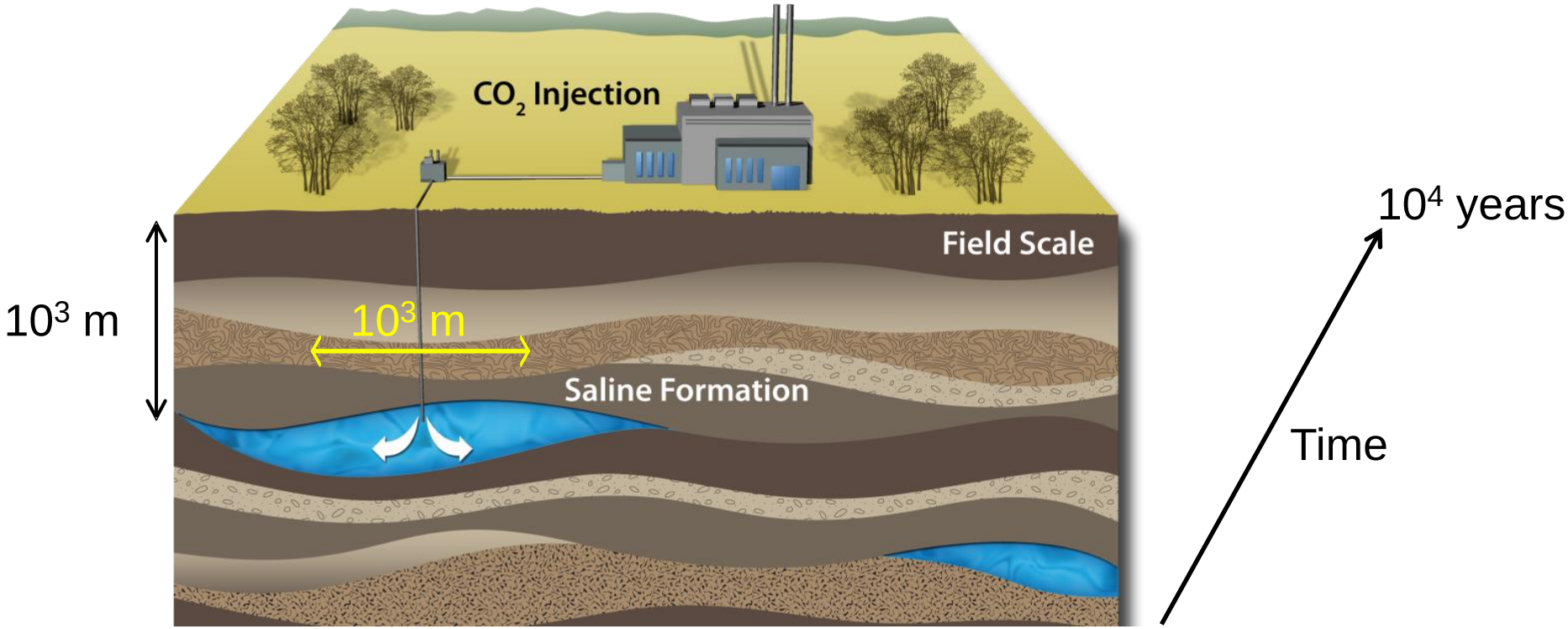
SAND 2013-6721P

Carbon capture and geological storage
Big Picture



1. Capture CO_2
2. Inject it deep underground
3. Hope it stays there

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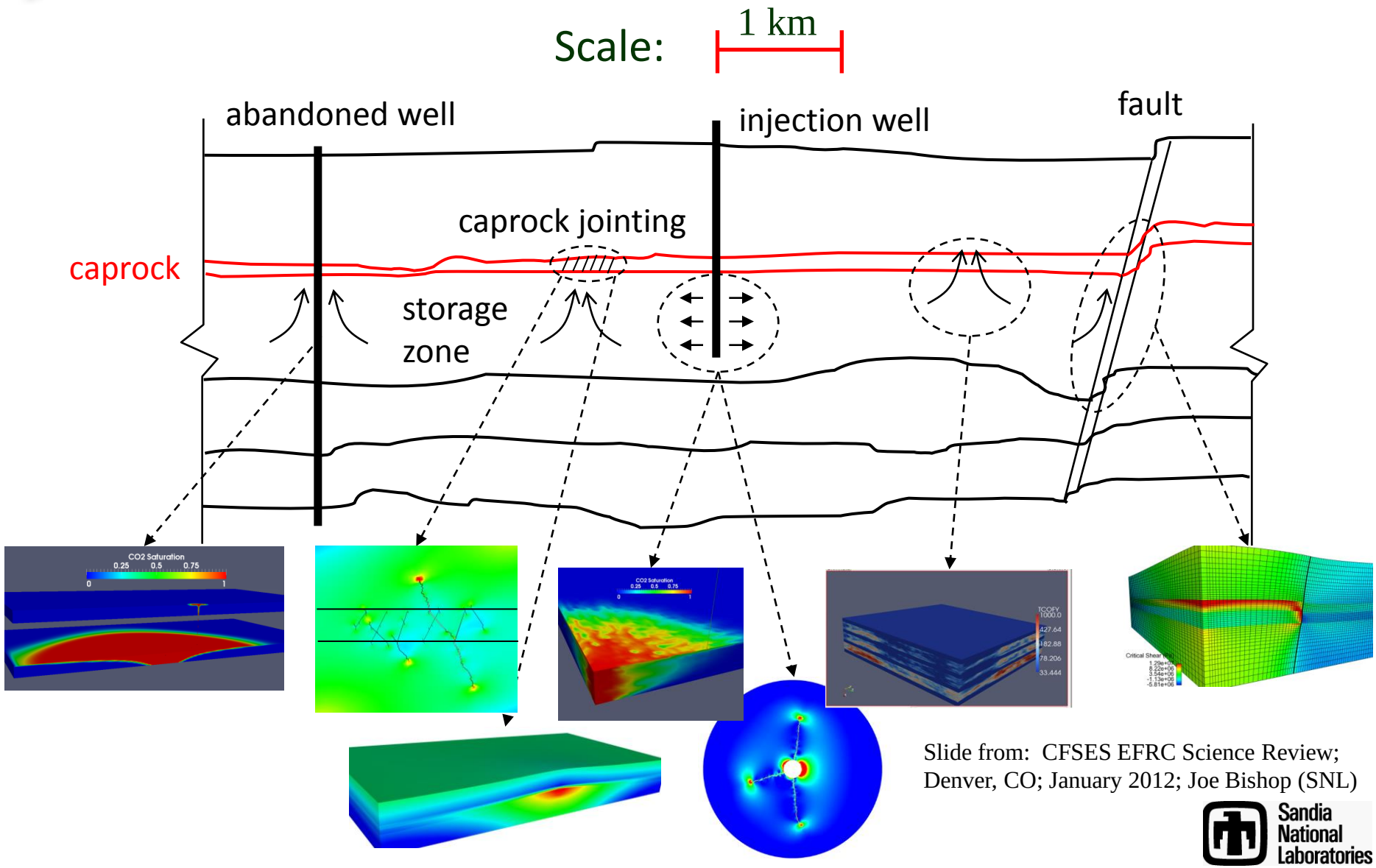
Field-scale modeling



Slide from: CFSES EFRC Science Review; Denver, CO; January 2012; Joe Bishop (SNL)

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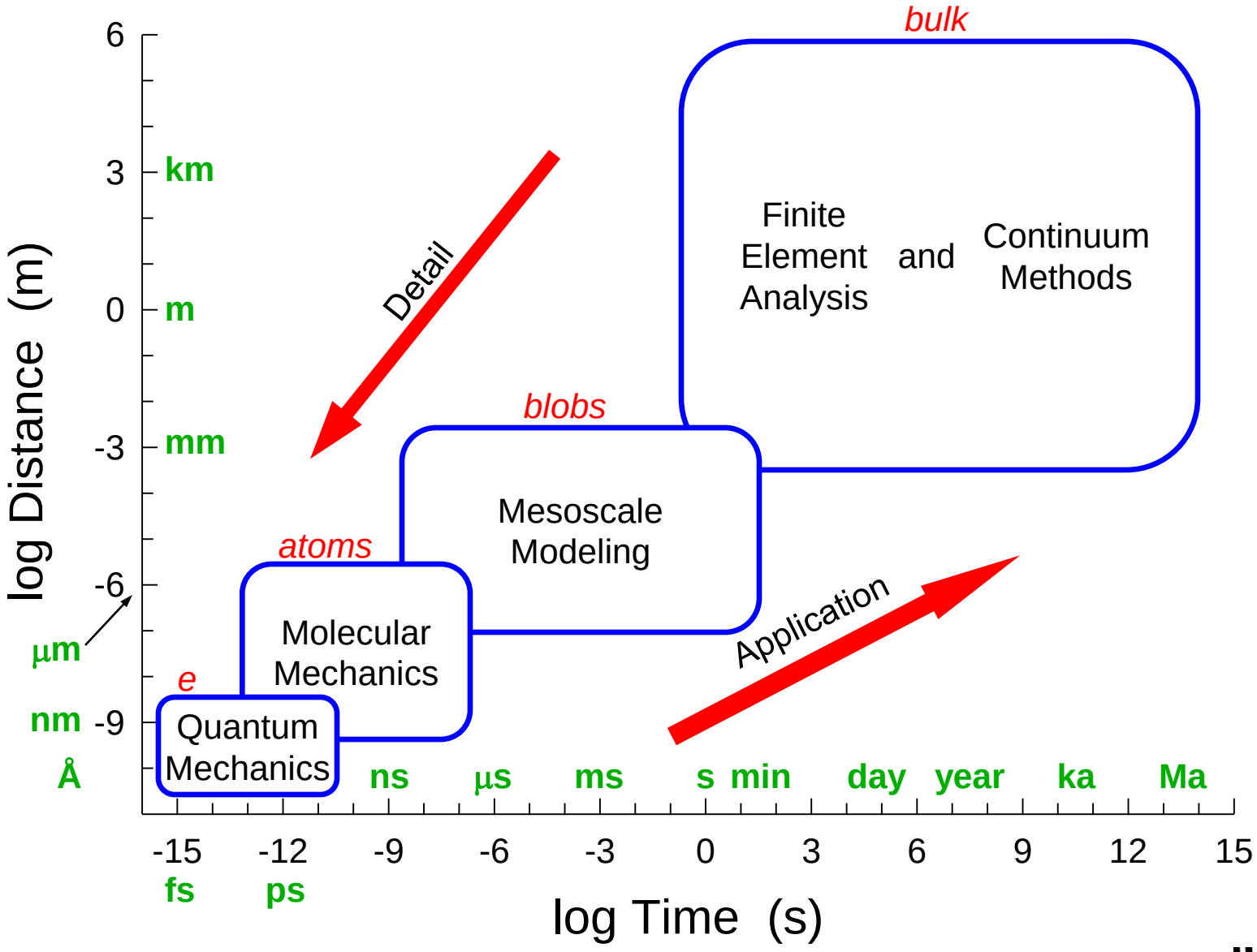
Multiscale simulation



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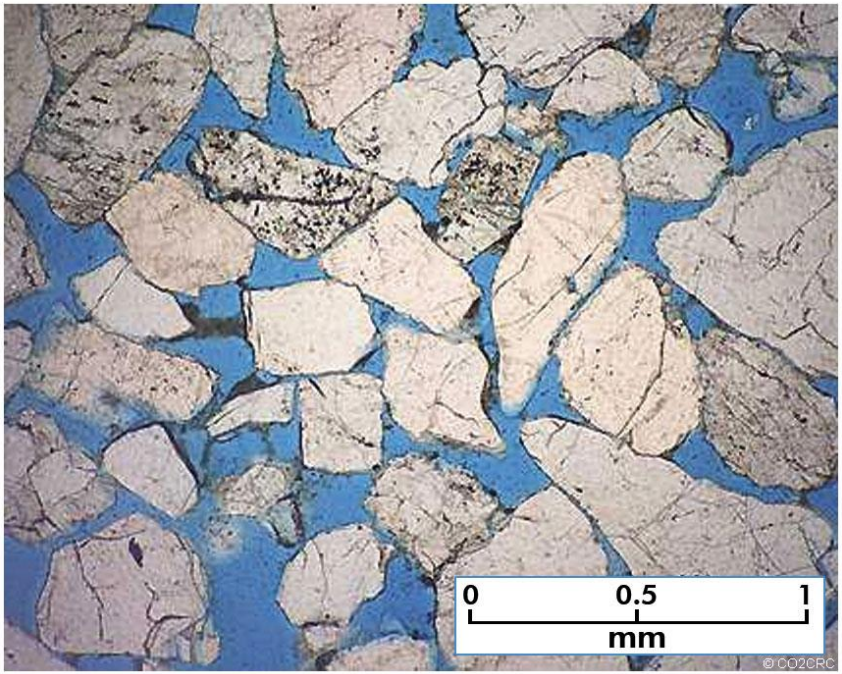
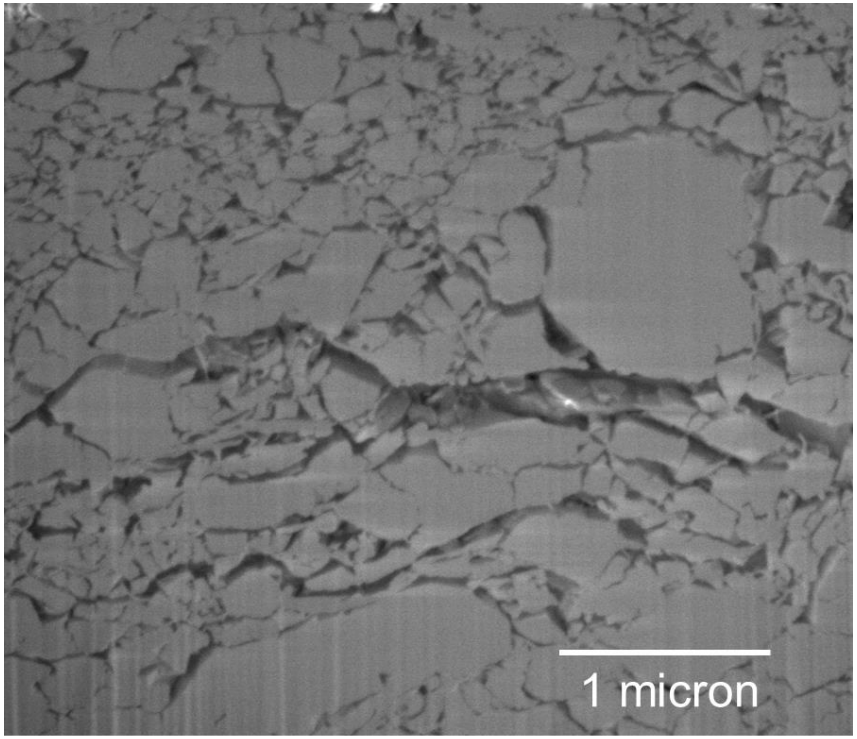
Geological carbon storage
Up Close

reservoir rock



Source: Cooperative Research Centre for Greenhouse Gas Technologies, http://www.co2crc.com.au/

cap rock



Source: Hongkyu Yoon (SNL)

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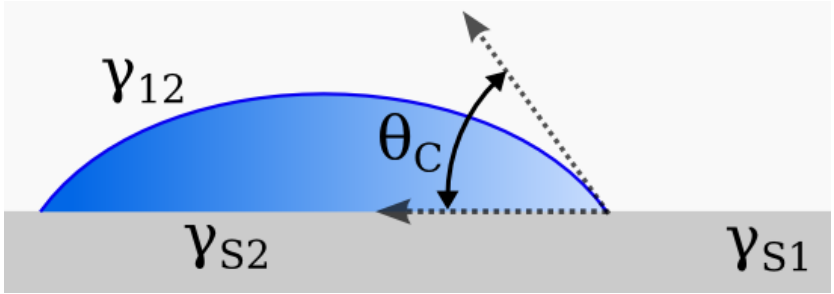
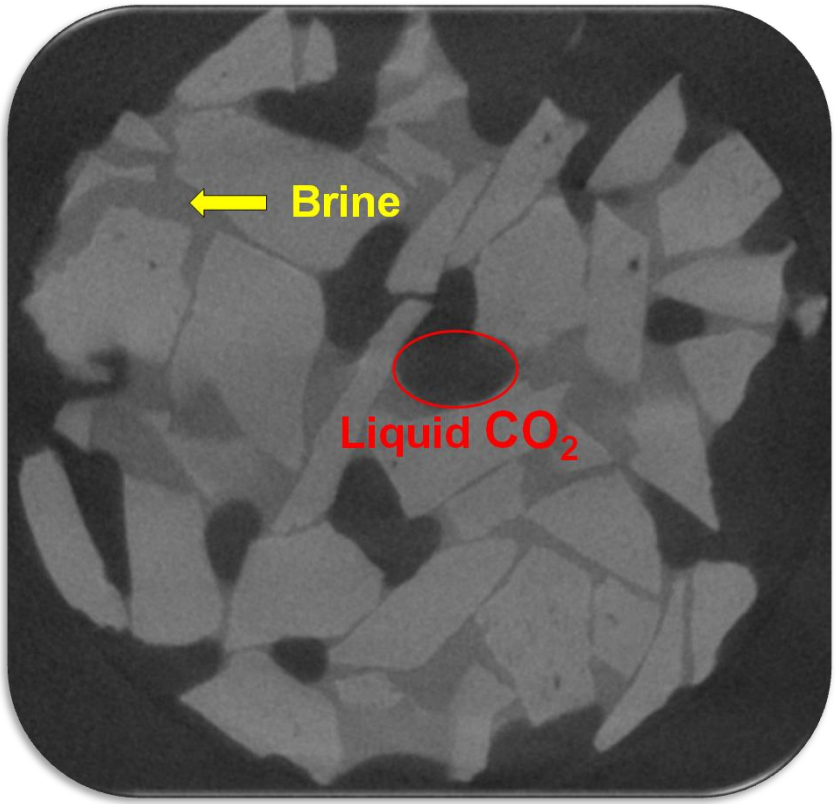
Sub-pore scale

- **interfacial tension γ_{12}**
  - **surface free energy**
- **contact angle θ_C**
  - **indication of wettability**



$$\gamma_{S1} - \gamma_{S2} = \gamma_{12} \cos\theta_C$$



Source: Kuldeep Chaudhary (UT Austin)

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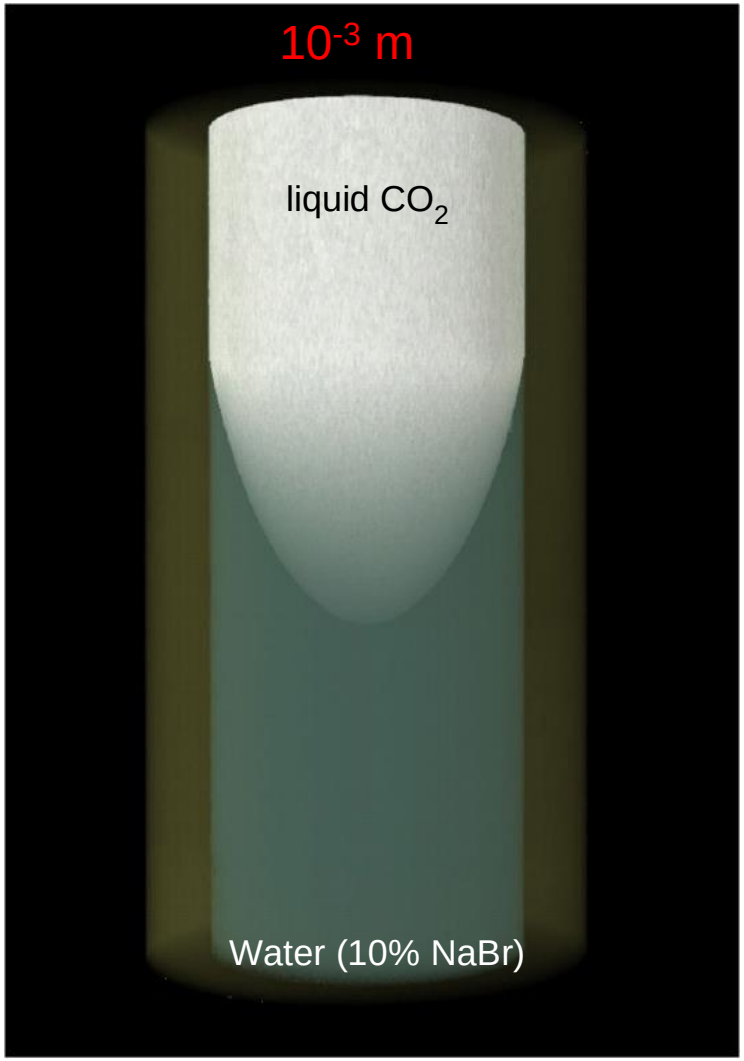
Pore- and field-scale

- **capillary pressure p_c**
  - **overpressure required to displace current fluid with new fluid**

$$p_c = \frac{2\gamma_{12}\cos\theta_C}{r}$$

- **relative permeability**
  - **fractional permeability of a fluid in the presence of other fluid(s)**



Source: Kuldeep Chaudhary (UT Austin)

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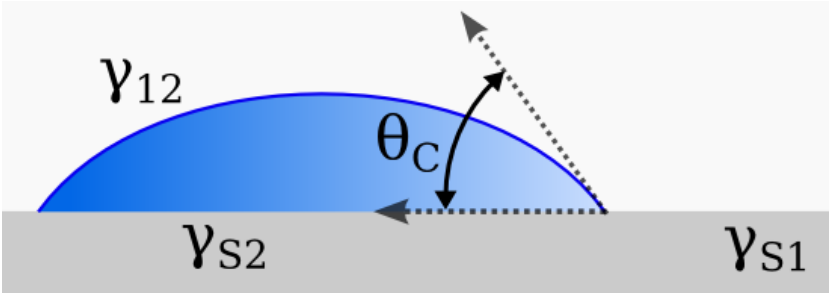
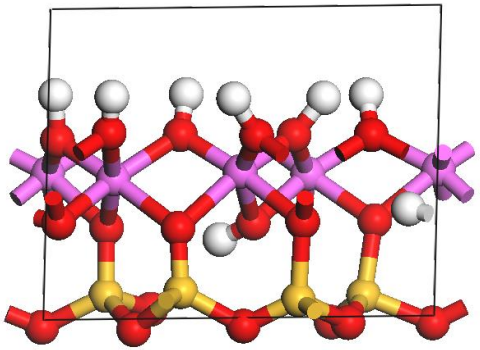
Molecular simulation of aqueous and supercritical CO_2 nanodroplets



kaolinite

gibbsite surface (hydrophilic) ⟶ **H**, **Al**

siloxane surface (hydrophobic) ⟶ **Si**, **O**



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Simulation details

- (20 x 20 x 17) nm
- 3 kaolinite layers
  (bottom two fixed)
- 330K, 20MPa
- 10-15 ns

- CO_2 in H_2O
  - 600k atoms
- H_2O in CO_2
  - 300k atoms
- brine systems
  - 0.7 M NaCl
  - 0.2 M $CaCl_2$

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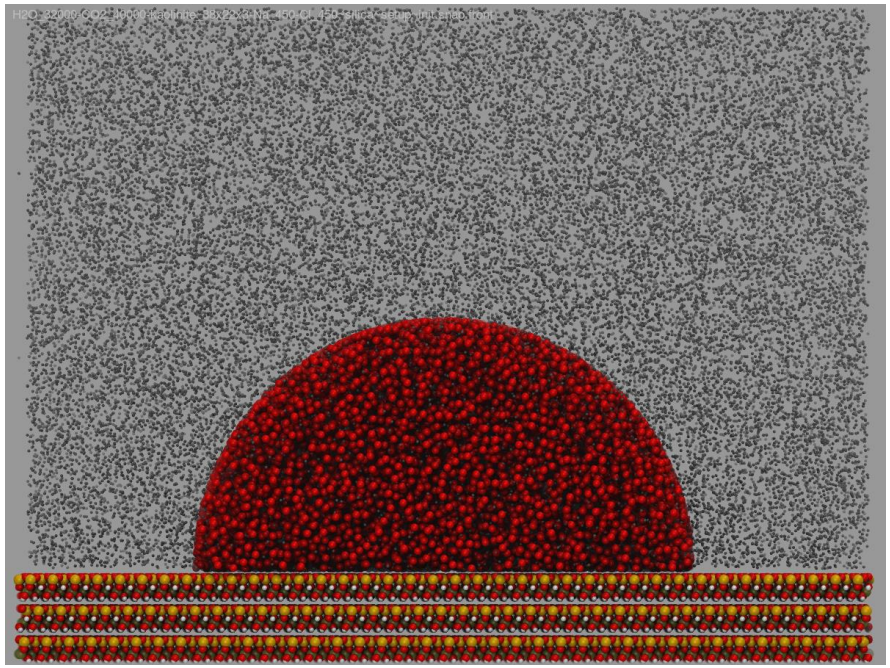
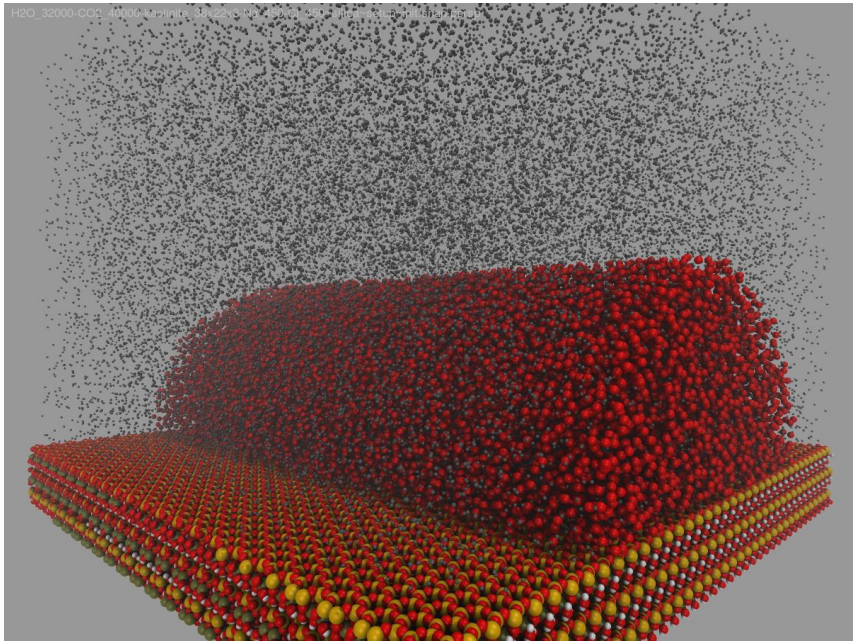
siloxane (hydrophobic) surface
0.7M NaCl in CO_2 – initial configuration

front view



oblique view



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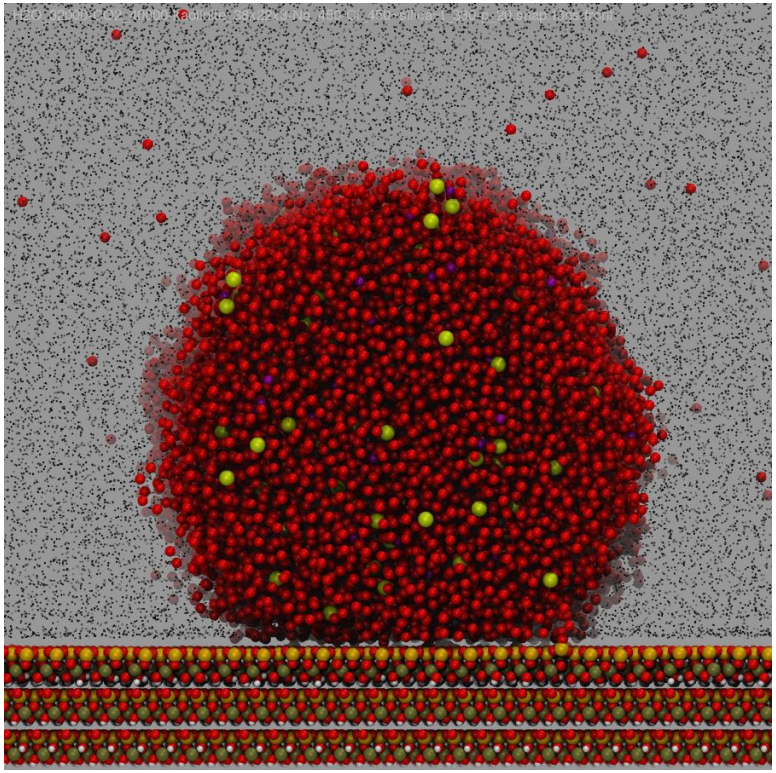
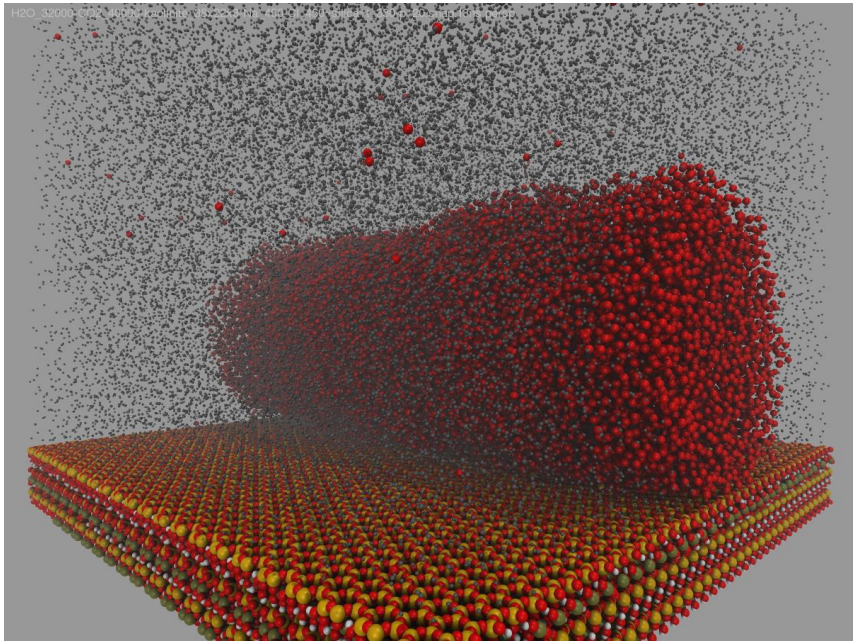
siloxane (hydrophobic) surface
0.7M NaCl in CO_2 – final configuration

front view



oblique view



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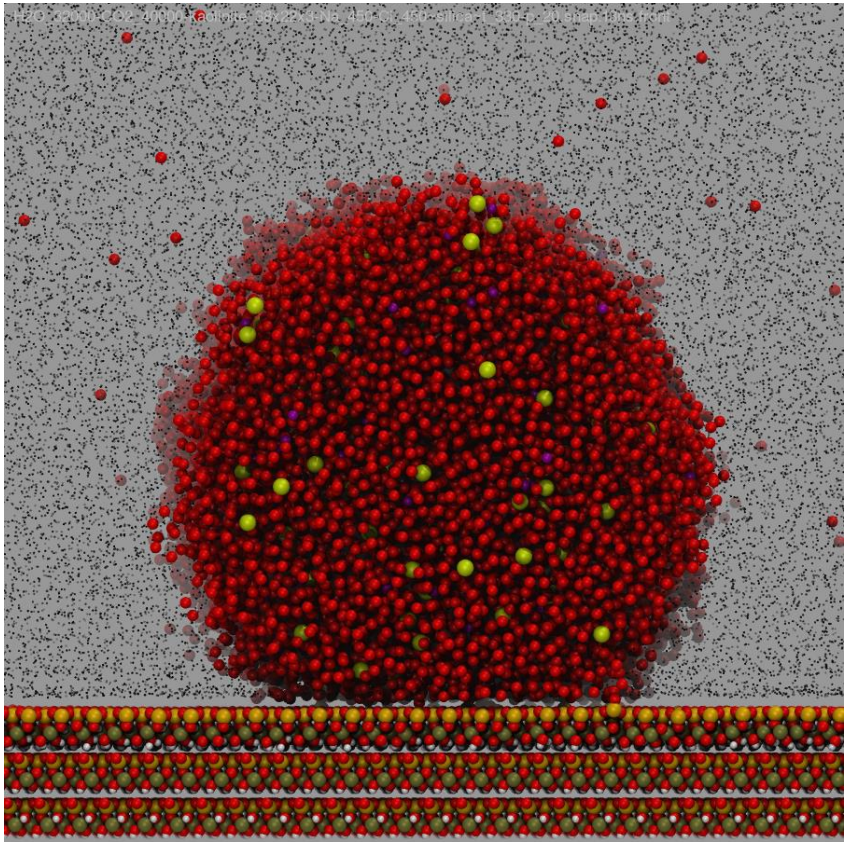

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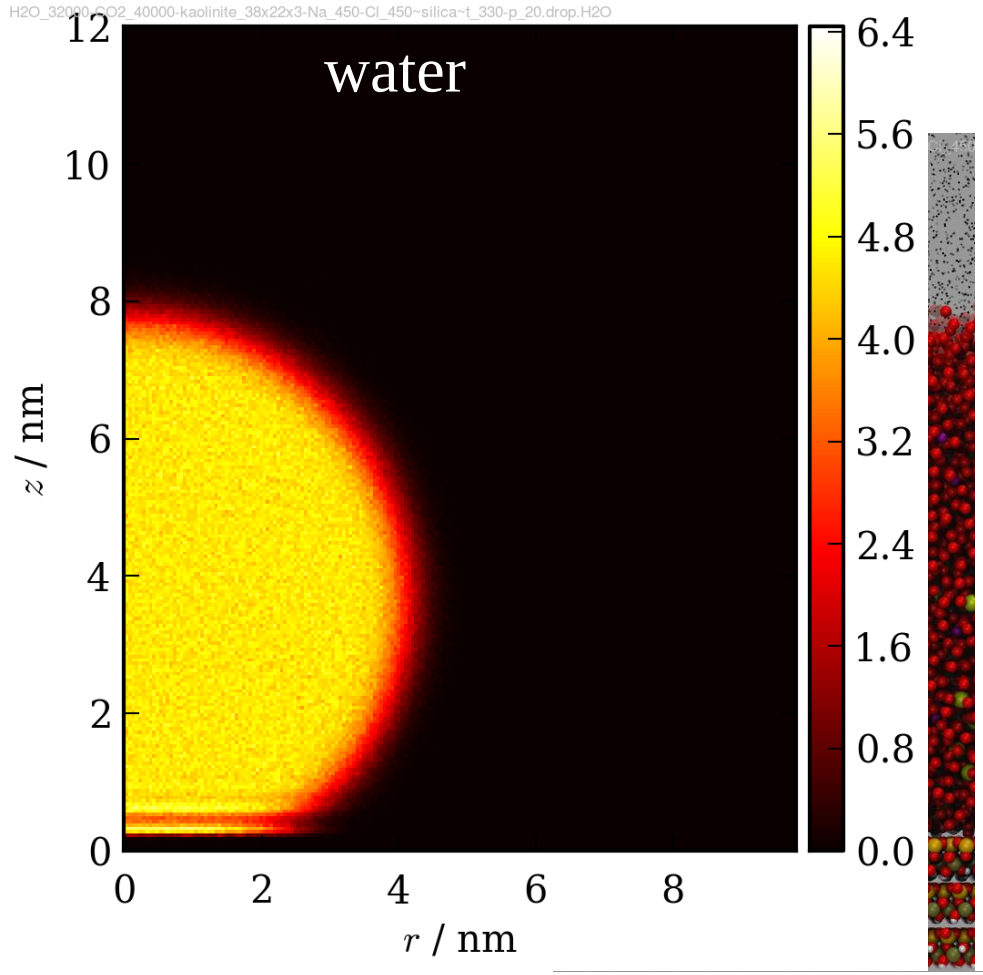

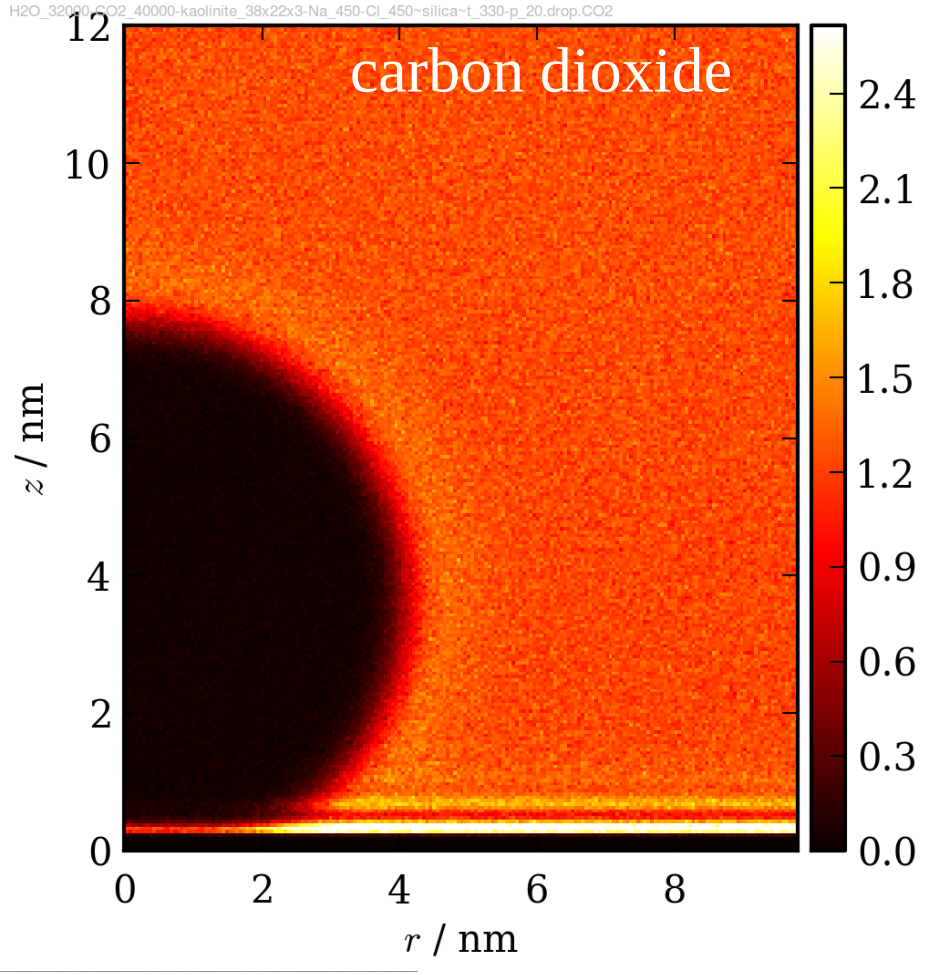

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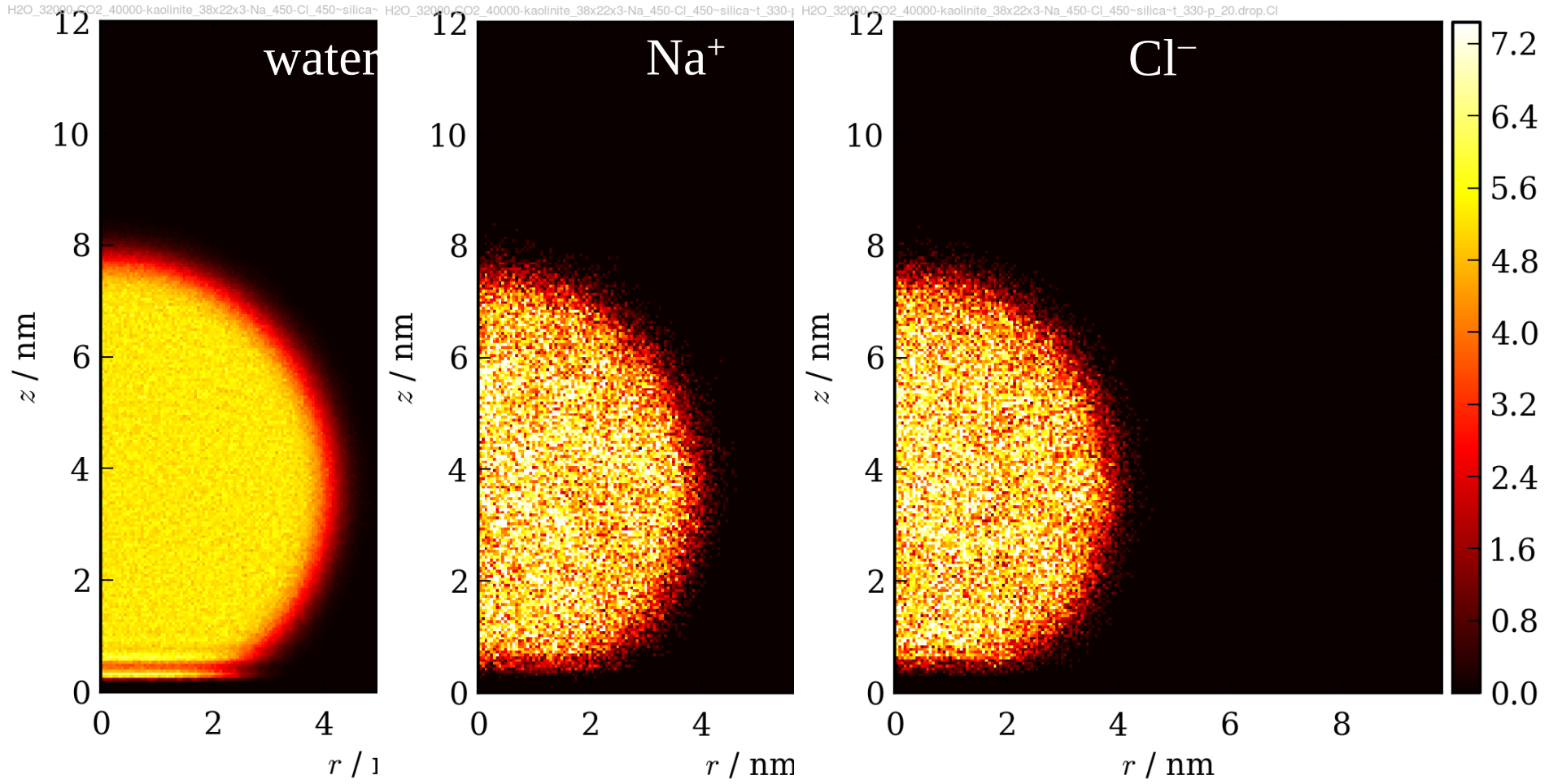

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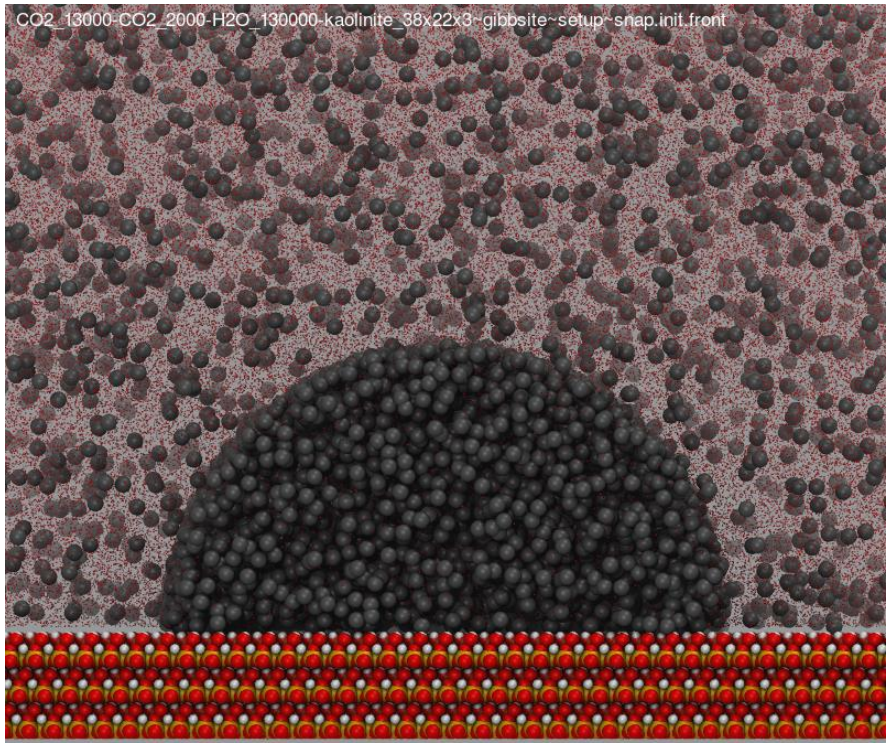
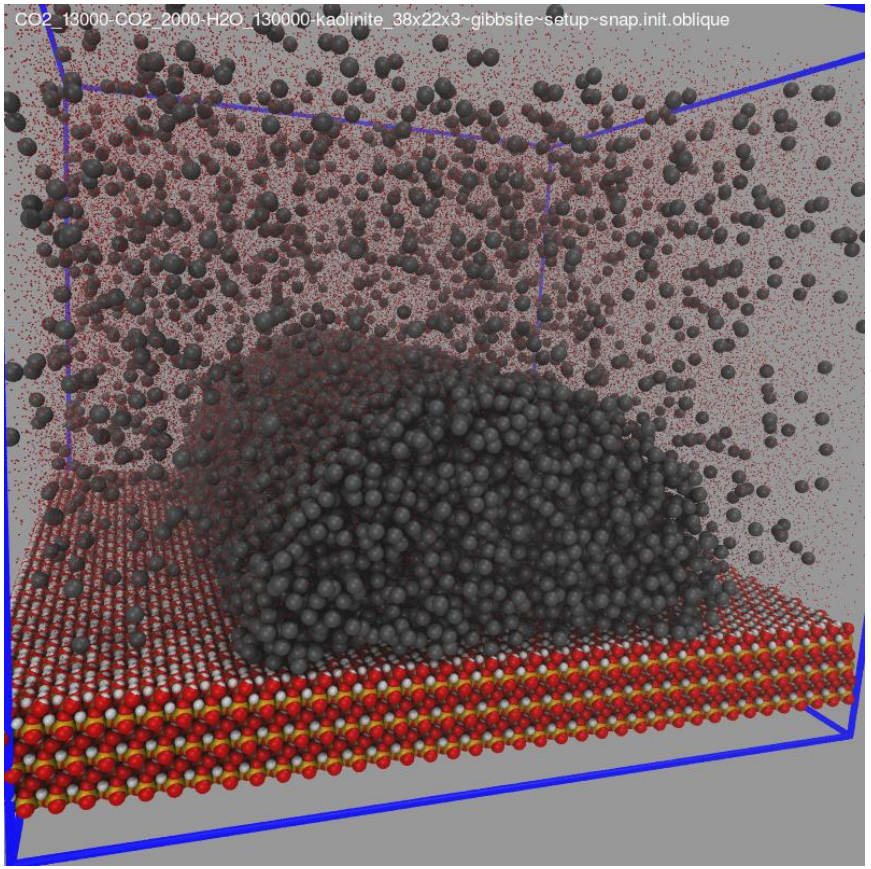
gibbsite (hydrophilic) surface
CO_2 in H_2O – initial configuration

front view



oblique view



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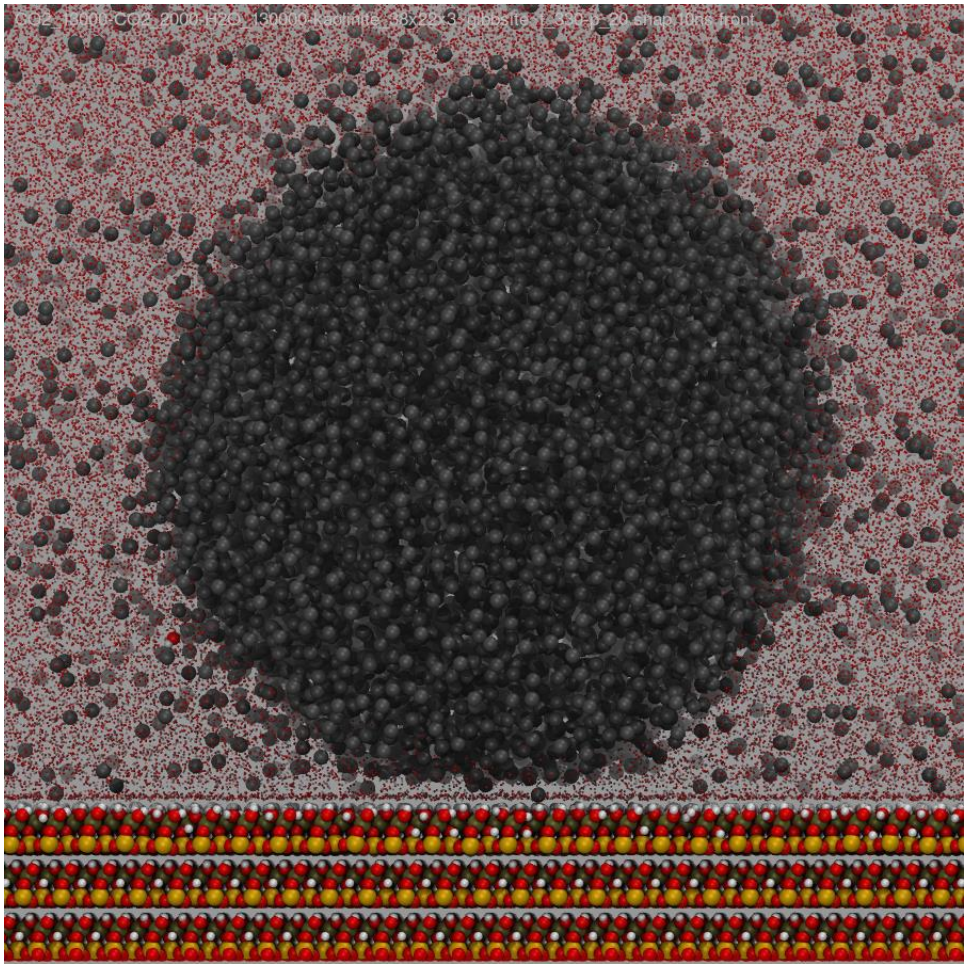
gibbsite (hydrophilic) surface CO_2 in H_2O – final configuration



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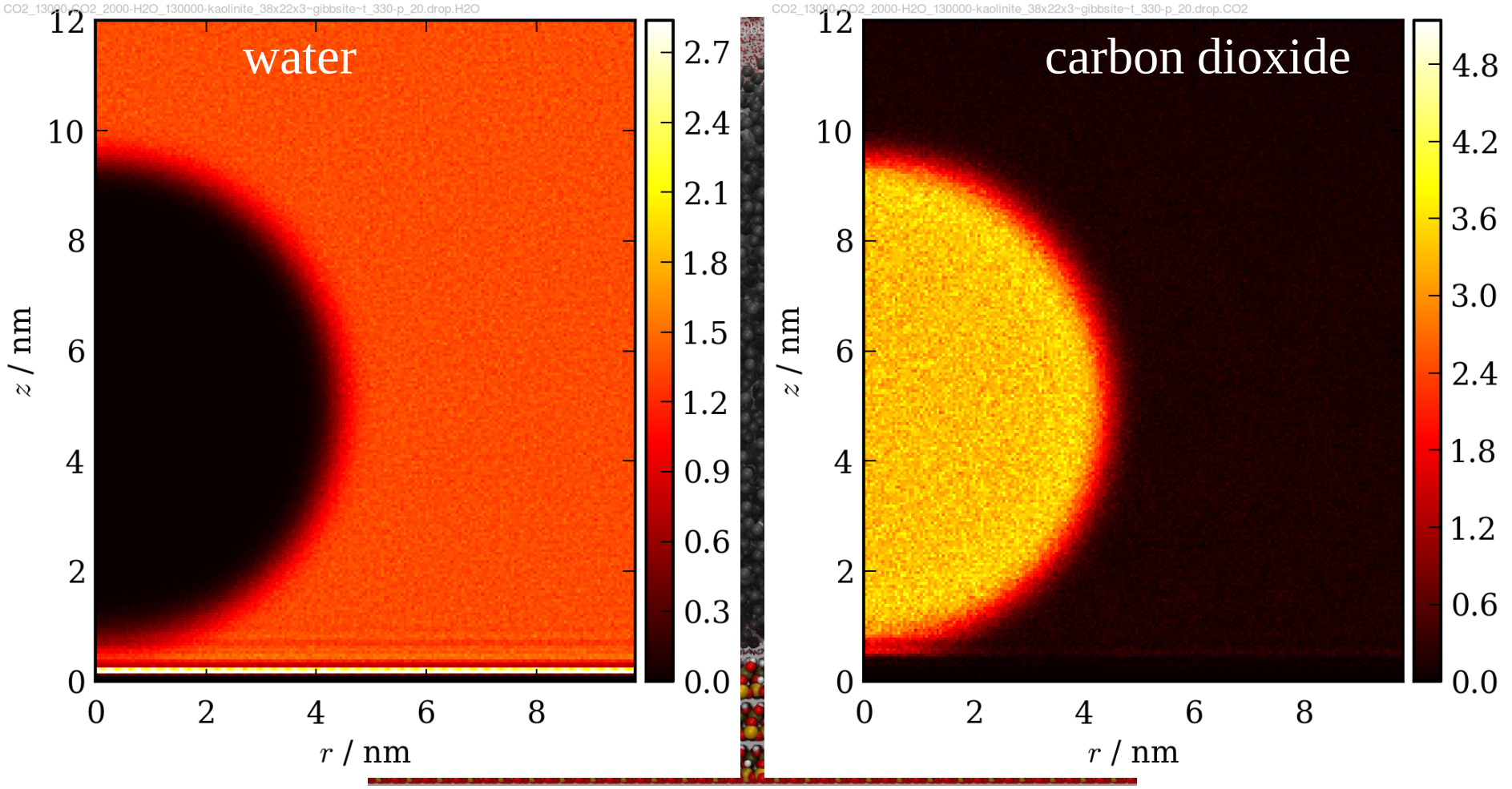
CO_2 in H_2O – final configuration



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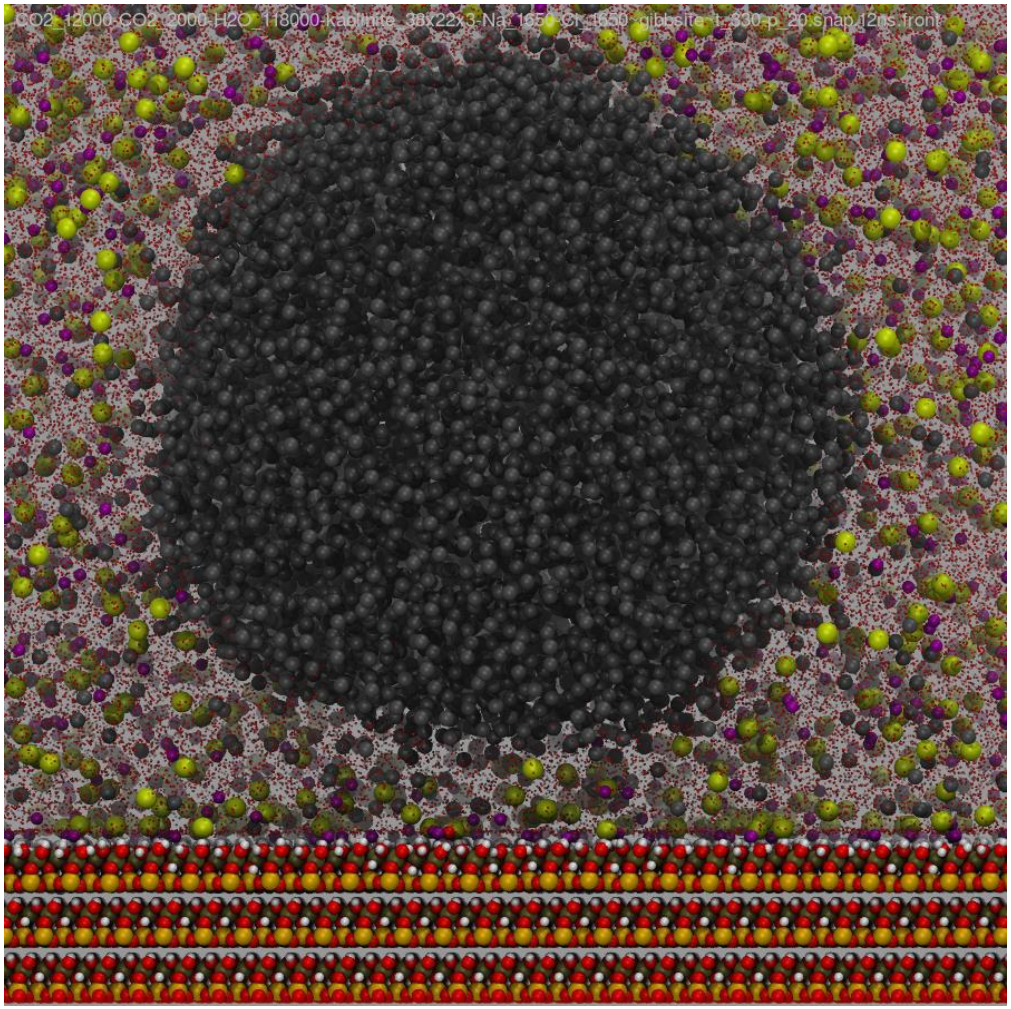
gibbsite (hydrophilic) surface
CO_2 in 0.7M NaCl – final configuration



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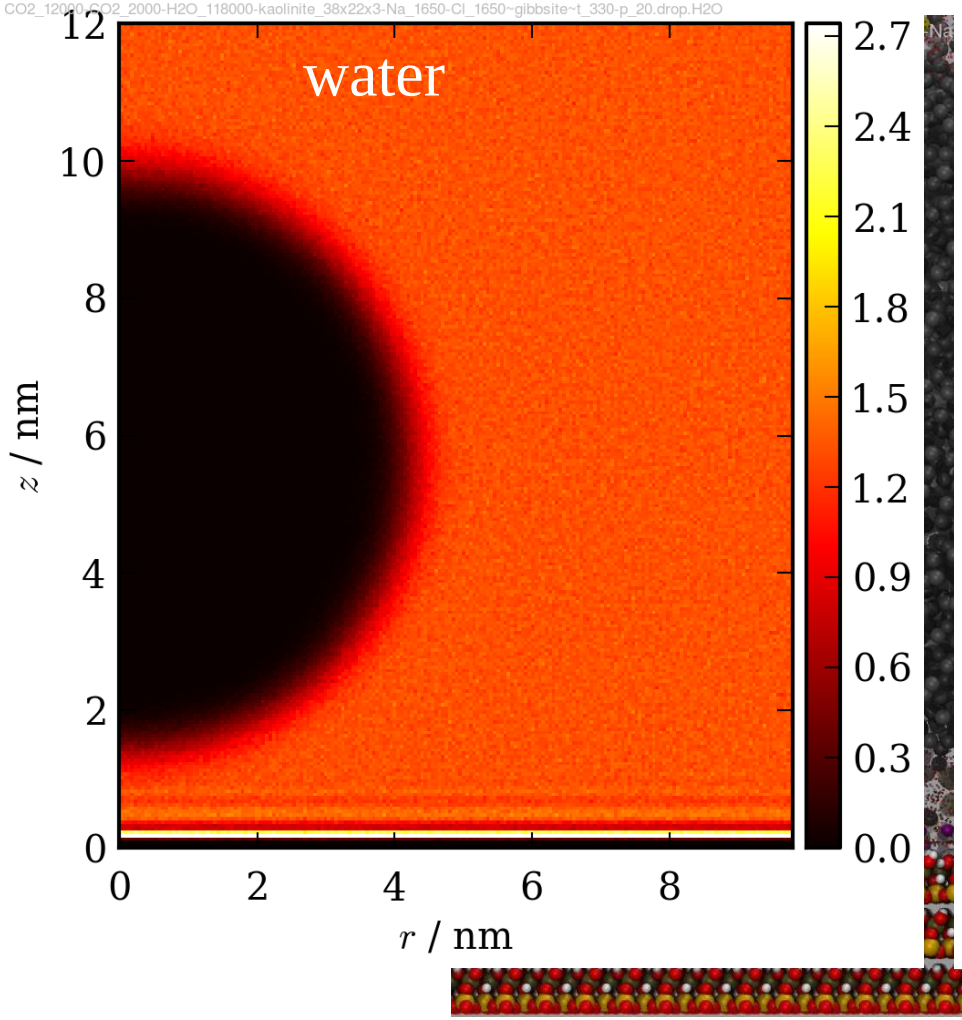

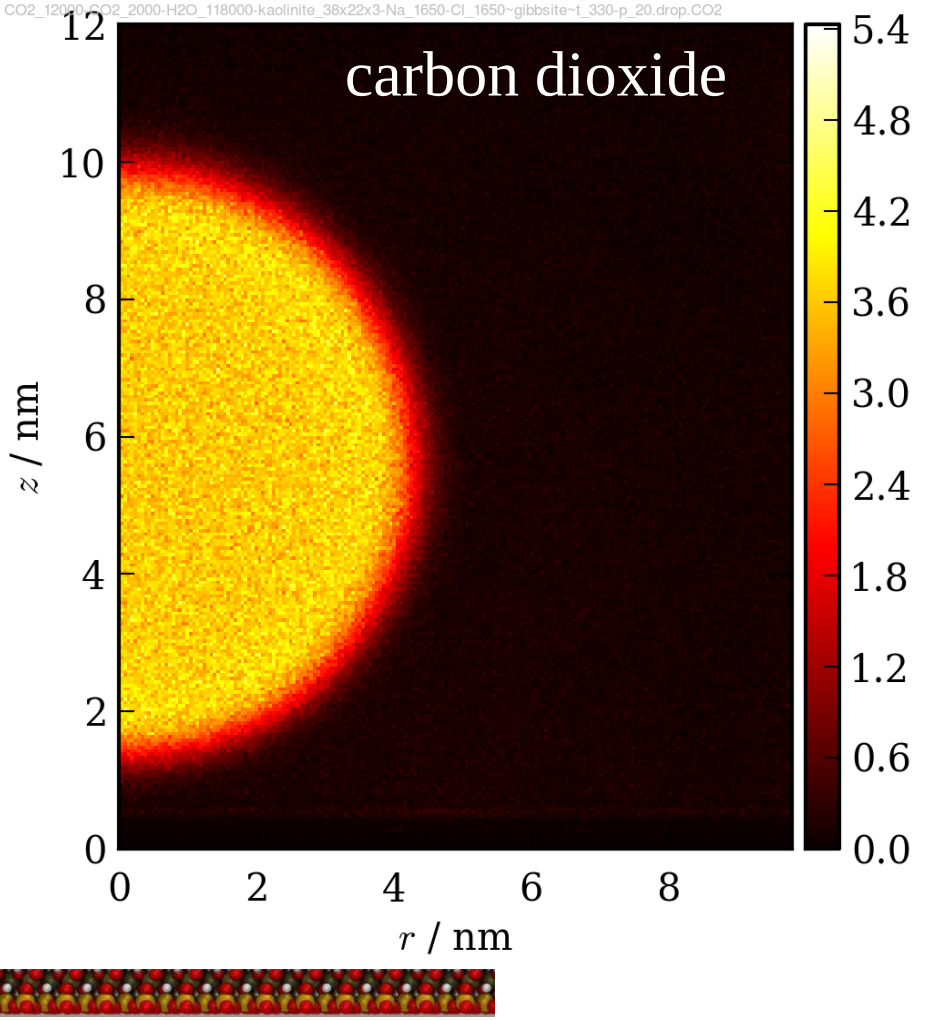

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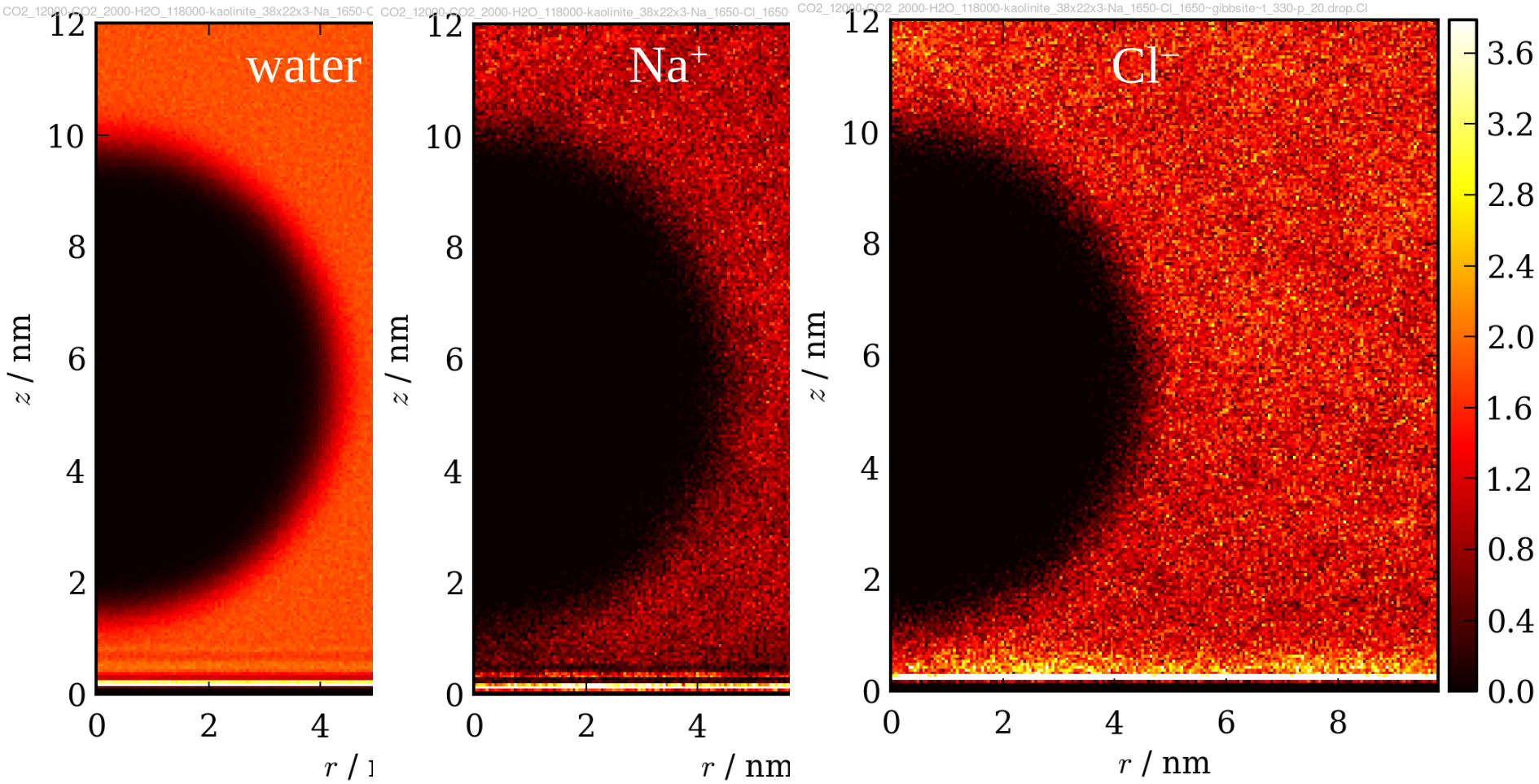
gibbsite (hydrophilic) surface
CO_2 in 0.7M NaCl – final configuration



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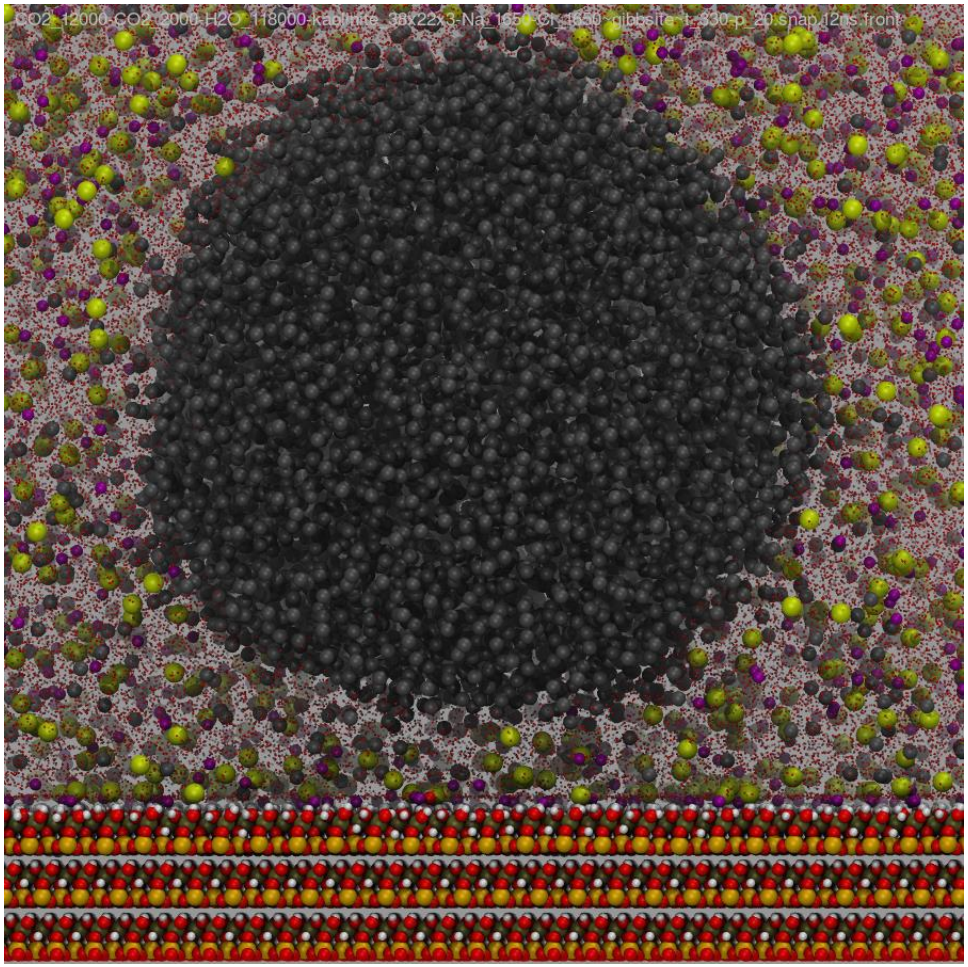
gibbsite (hydrophilic) surface
CO_2 in 0.2M $CaCl_2$ – final configuration



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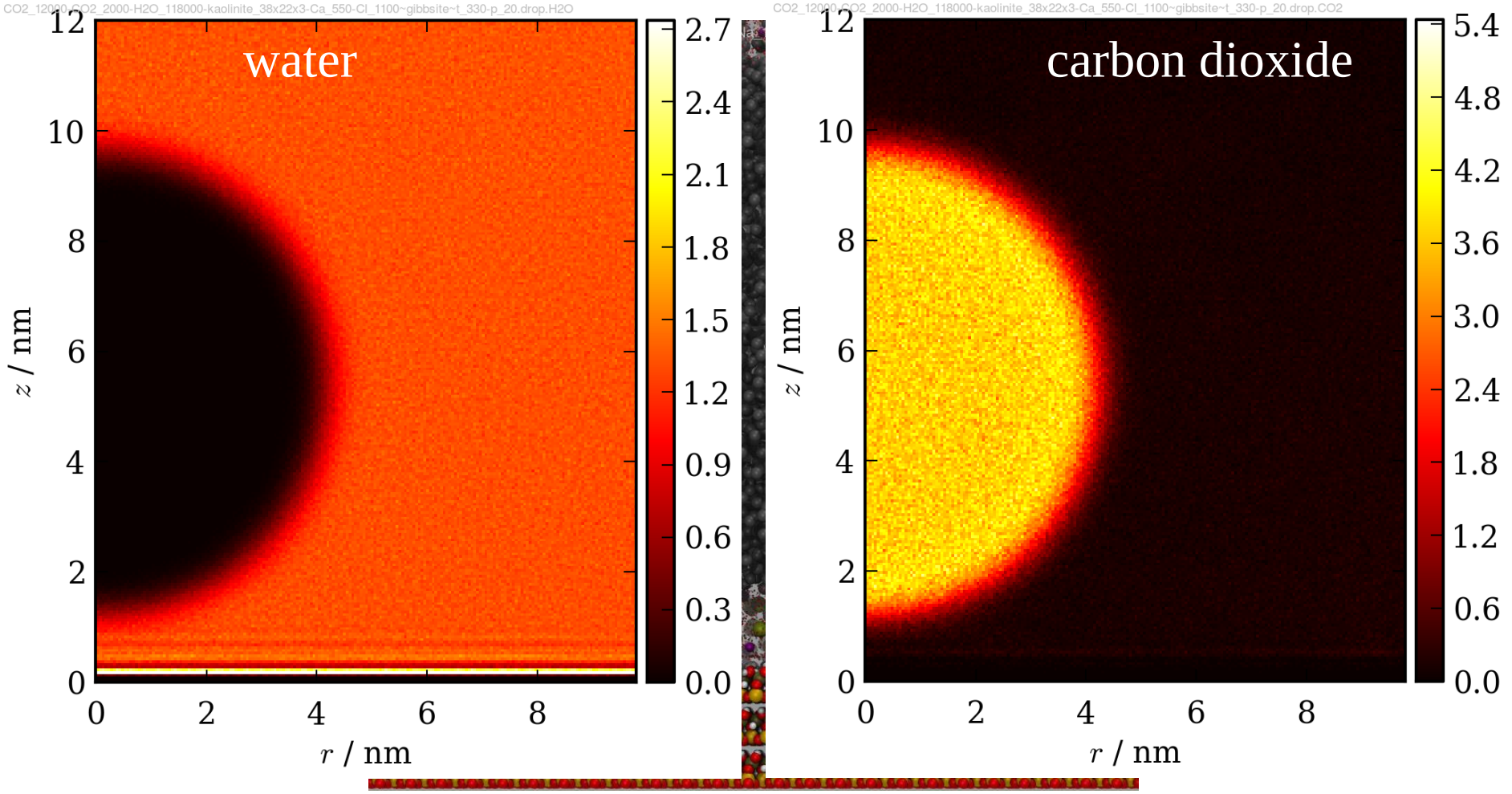
gibbsite (hydrophilic) surface
CO_2 in 0.2M $CaCl_2$ – final configuration



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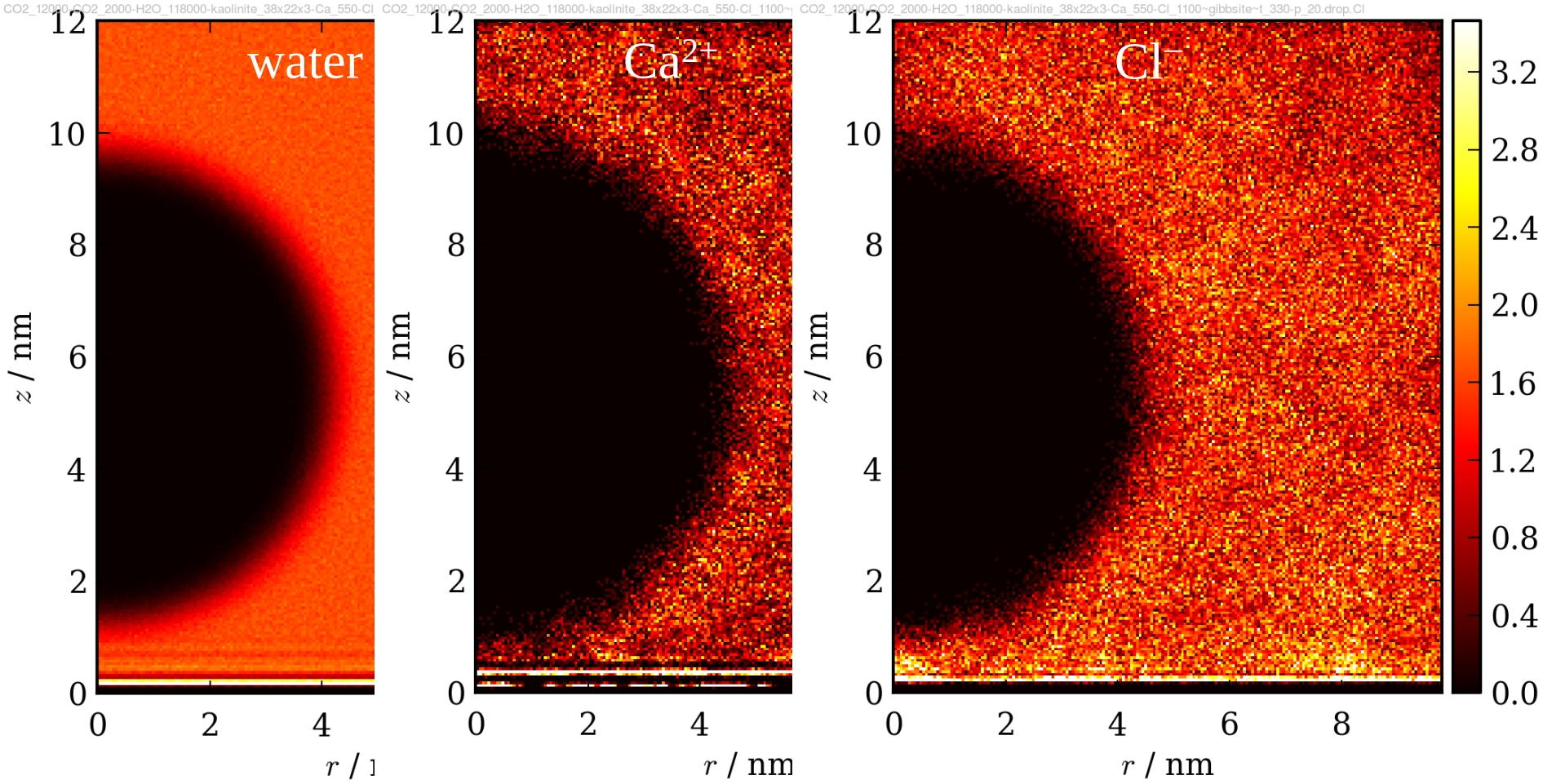
gibbsite (hydrophilic) surface
CO_2 in 0.2M $CaCl_2$ – final configuration



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Summary

- **Simulation shows realistic behavior for CO_2, H_2O, and ions on hydrophilic and hydrophobic basal surfaces of kaolinite.**

- **Possible future application: rational design of surfactants, coatings, enhanced oil or gas recovery**

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Acknowledgments

- Simulation development aided by discussions with
  - Louise Criscenti (SNL)
  - Ian Bourg (LBNL)

- This research is funded by the **Center for Frontiers of Subsurface Energy Security** (CFSES), a U.S. Department of Energy Office of Basic Energy Sciences Energy Frontier Research Center, under Contract No. DE-SC0001114.

TEXAS

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supplementary slides

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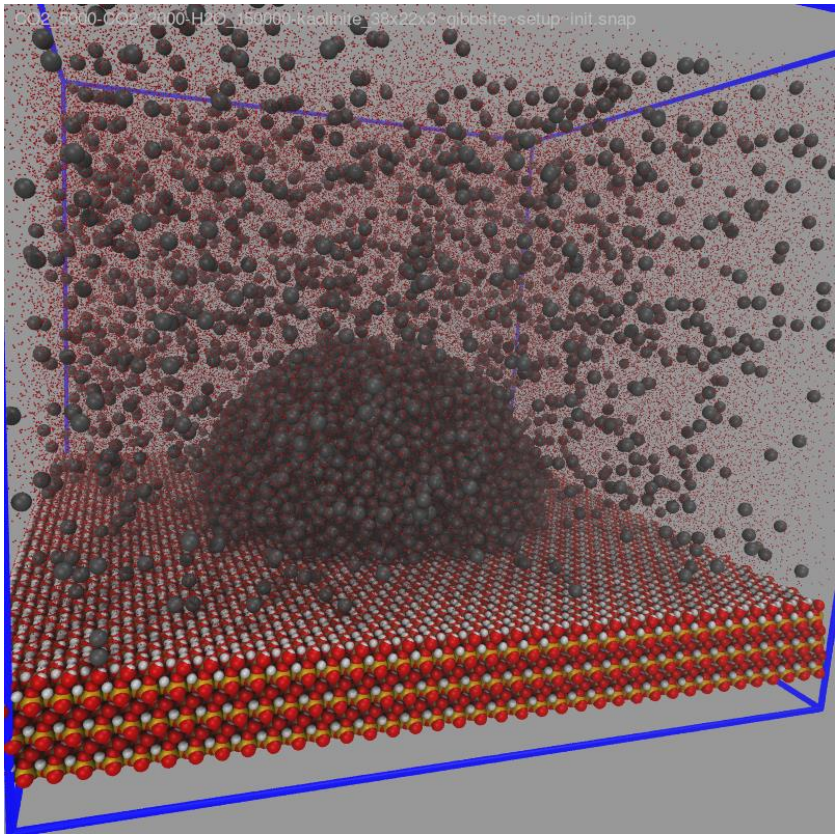
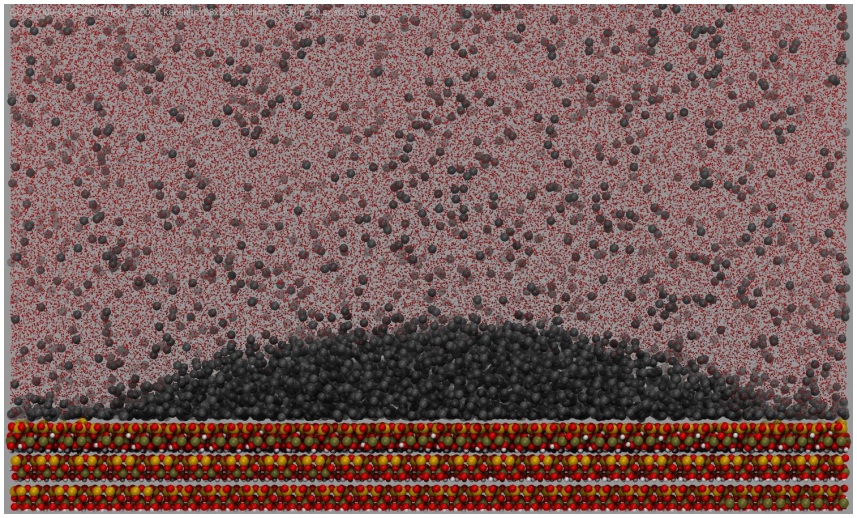
siloxane (hydrophobic) surface
CO_2 in H_2O

initial



final



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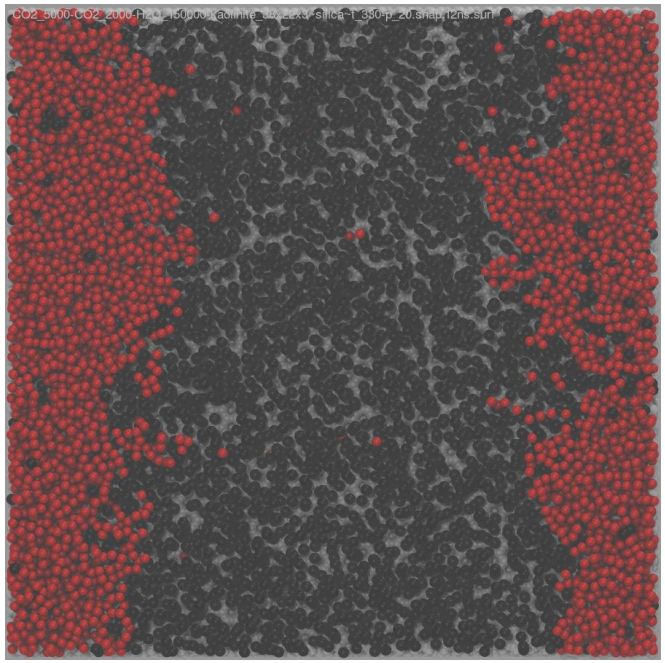
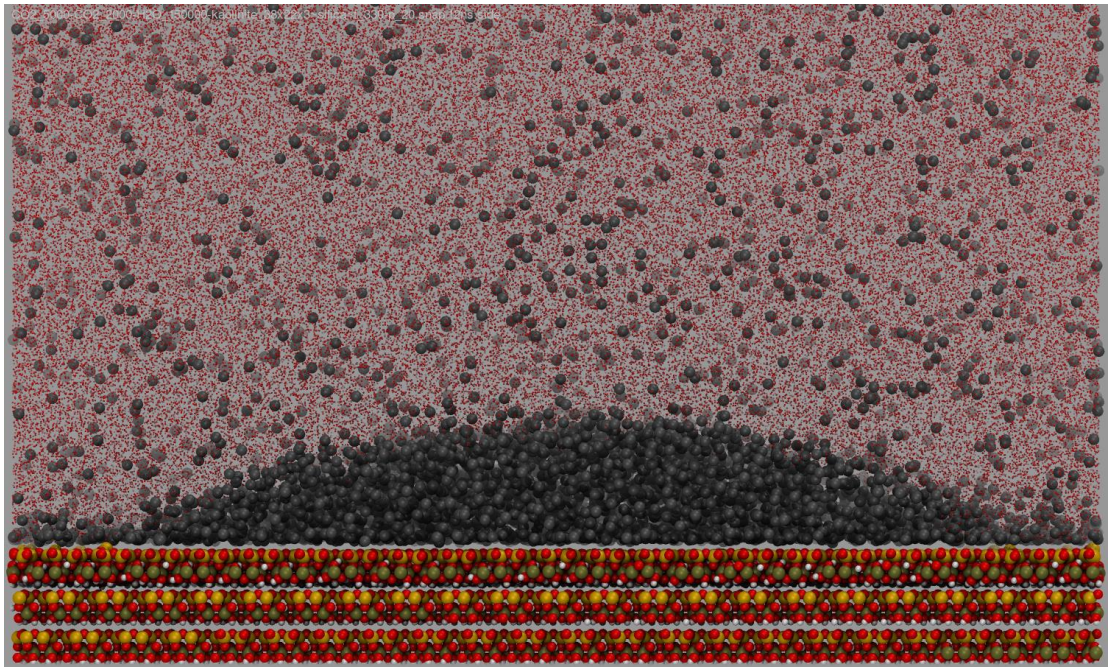
siloxane (hydrophobic) surface
CO_2 in H_2O – final configuration

top view



front view



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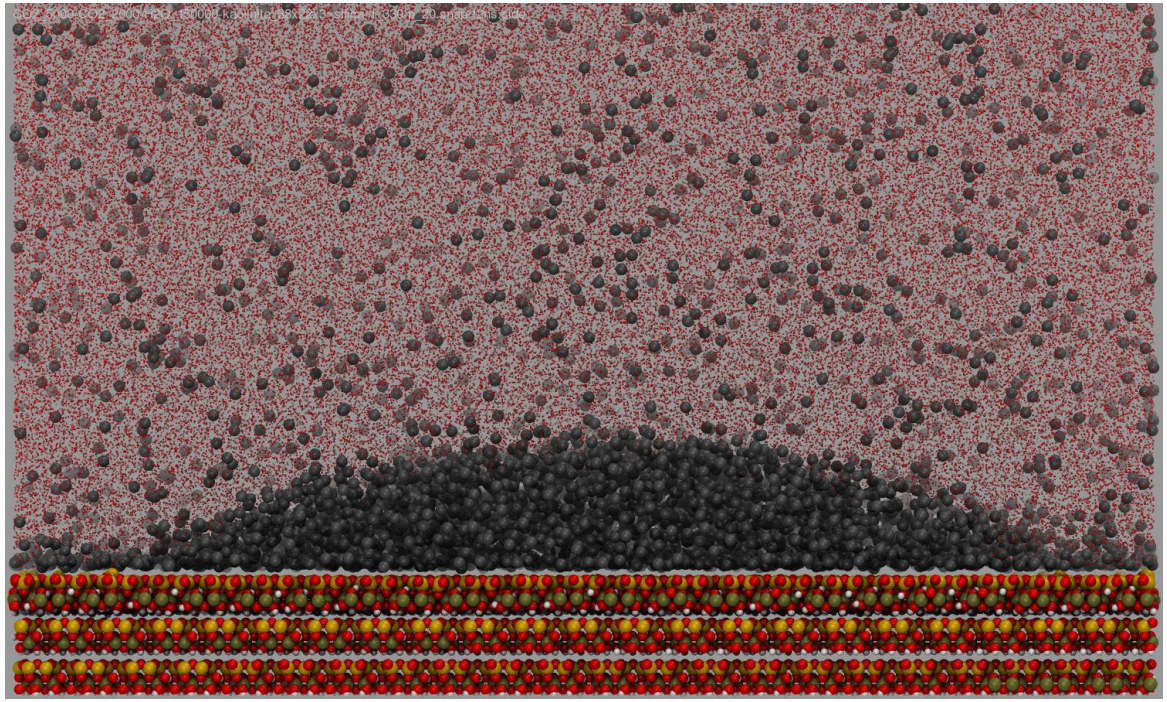
siloxane (hydrophobic) surface CO_2 in H_2O – final configuration



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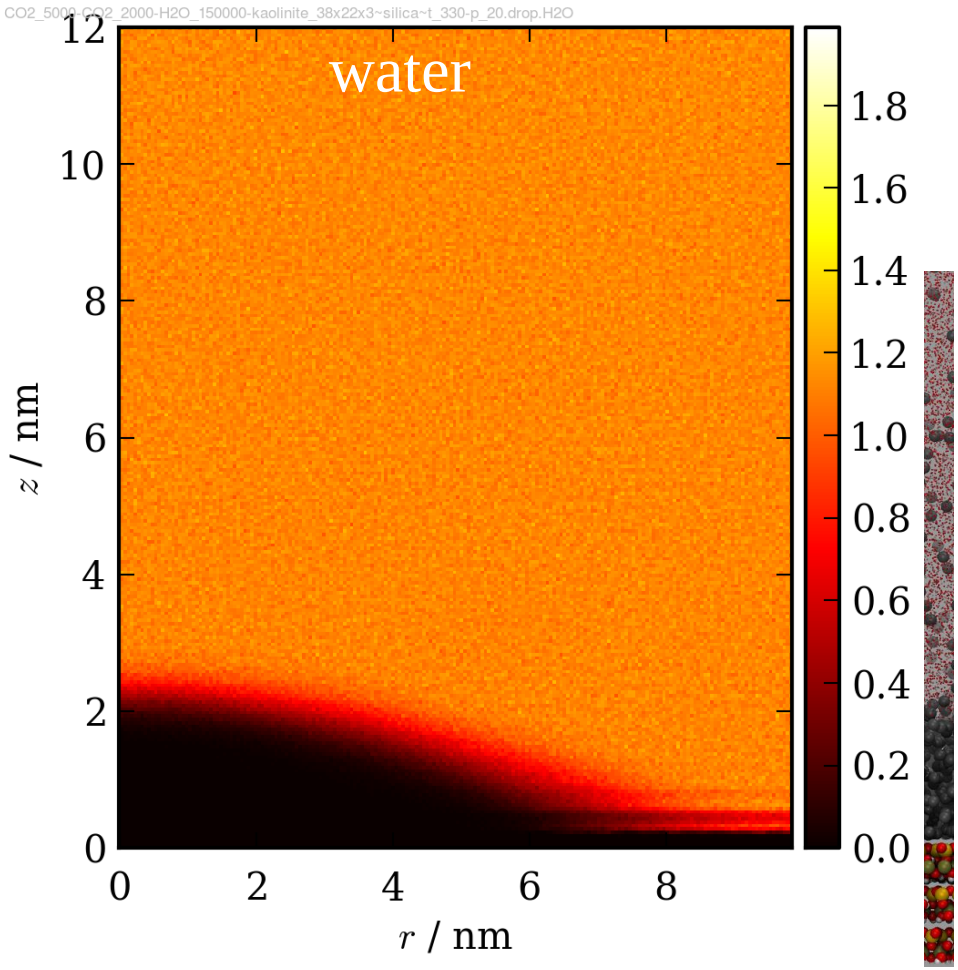
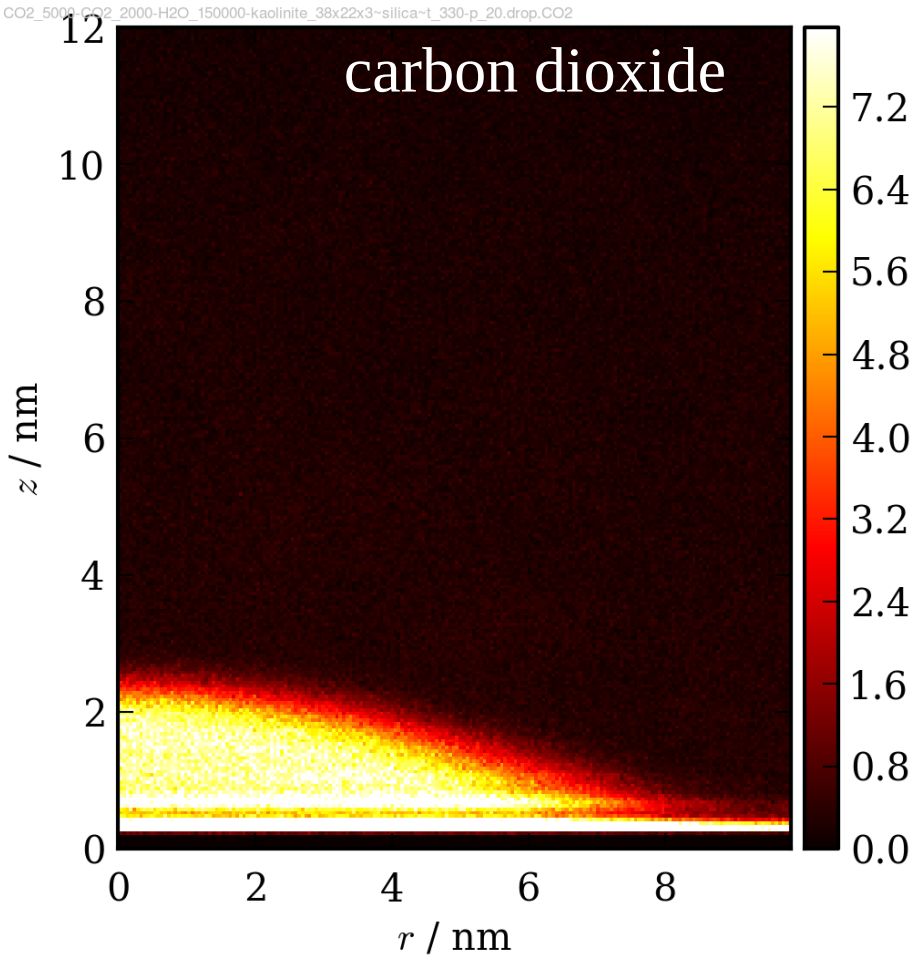
siloxane (hydrophobic) surface
CO_2 in H_2O – final configuration





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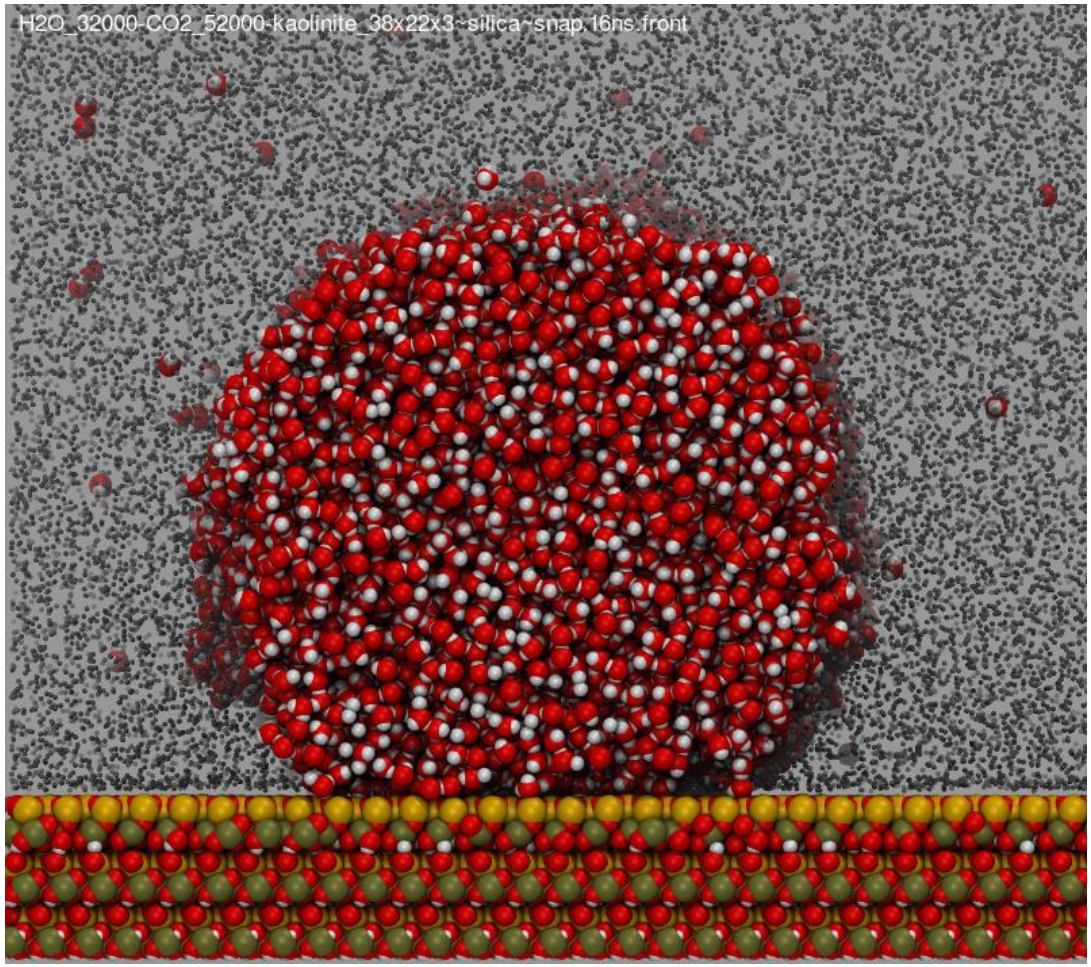
siloxane surface – H_2O in CO_2



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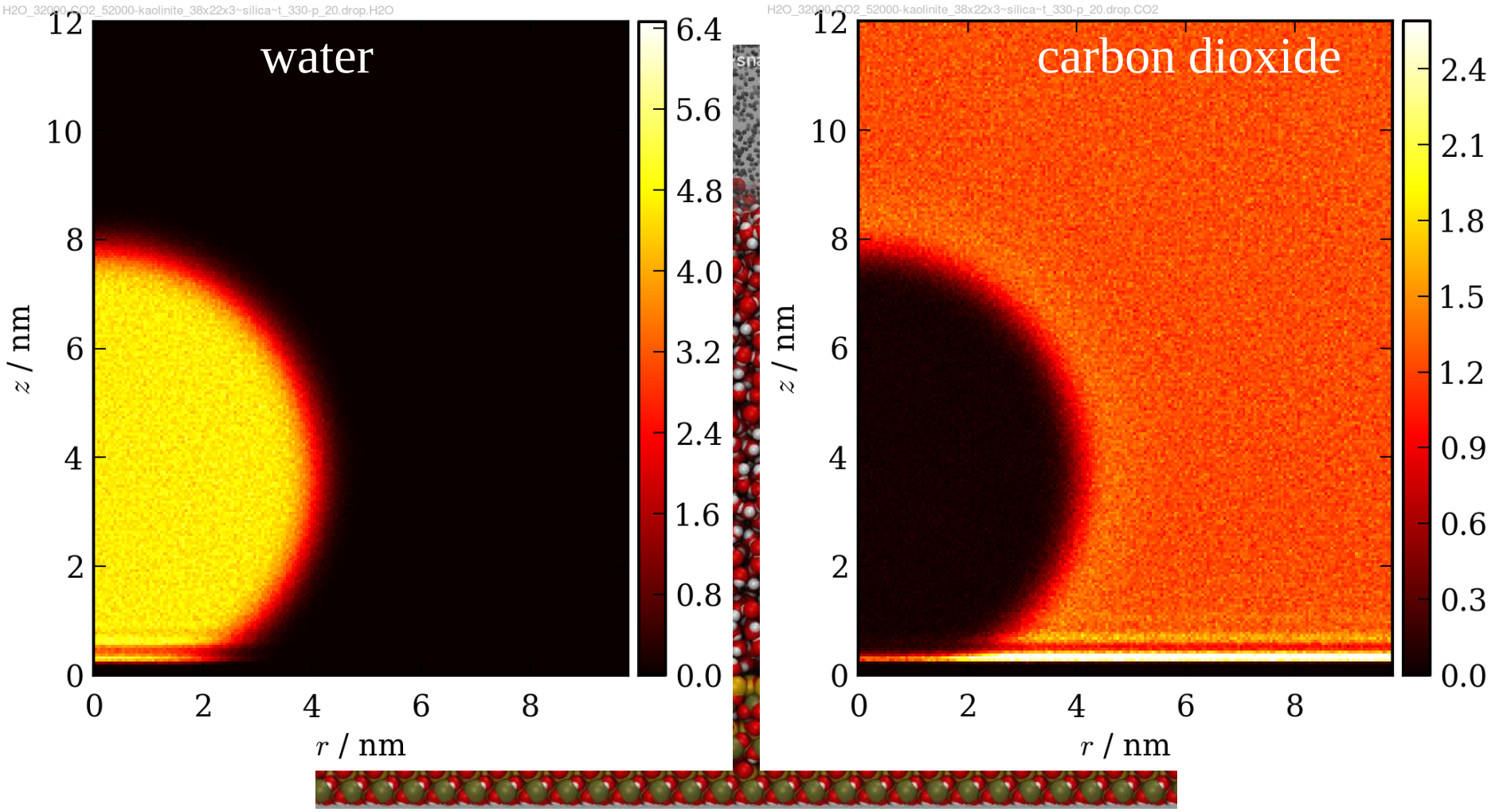
siloxane surface – H_2O in CO_2



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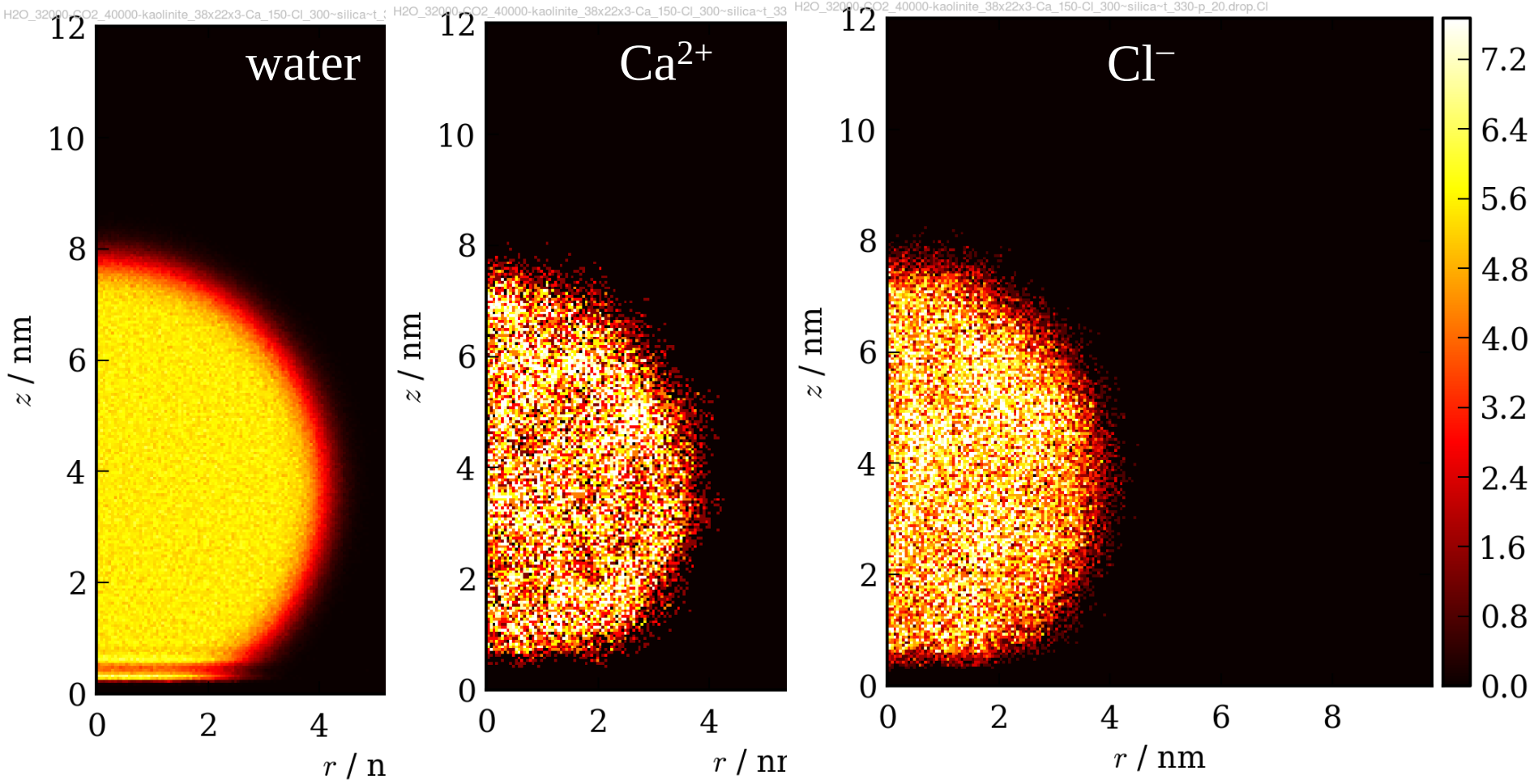
siloxane surface – 0.2M $CaCl_2$ in CO_2



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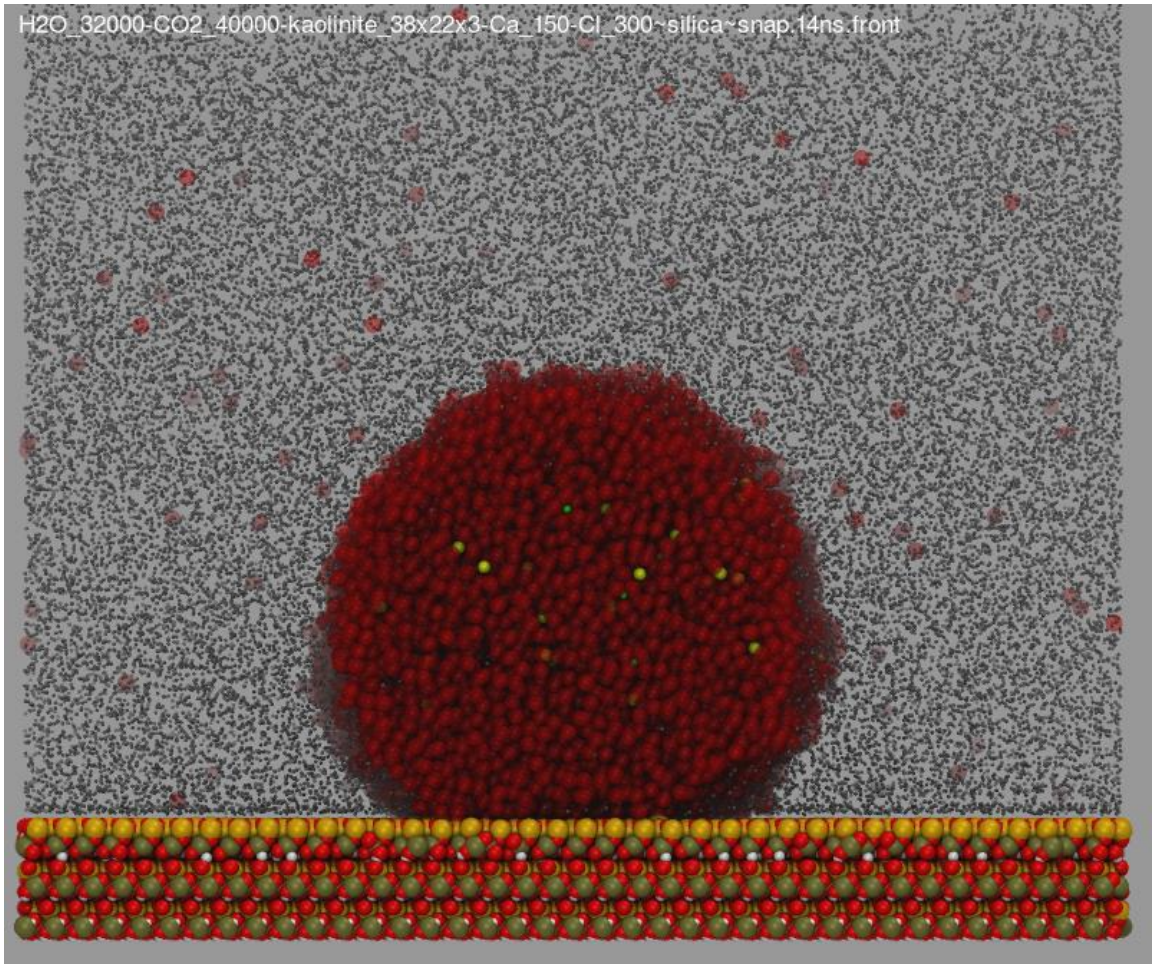
siloxane surface – 0.2M $CaCl_2$ in CO_2



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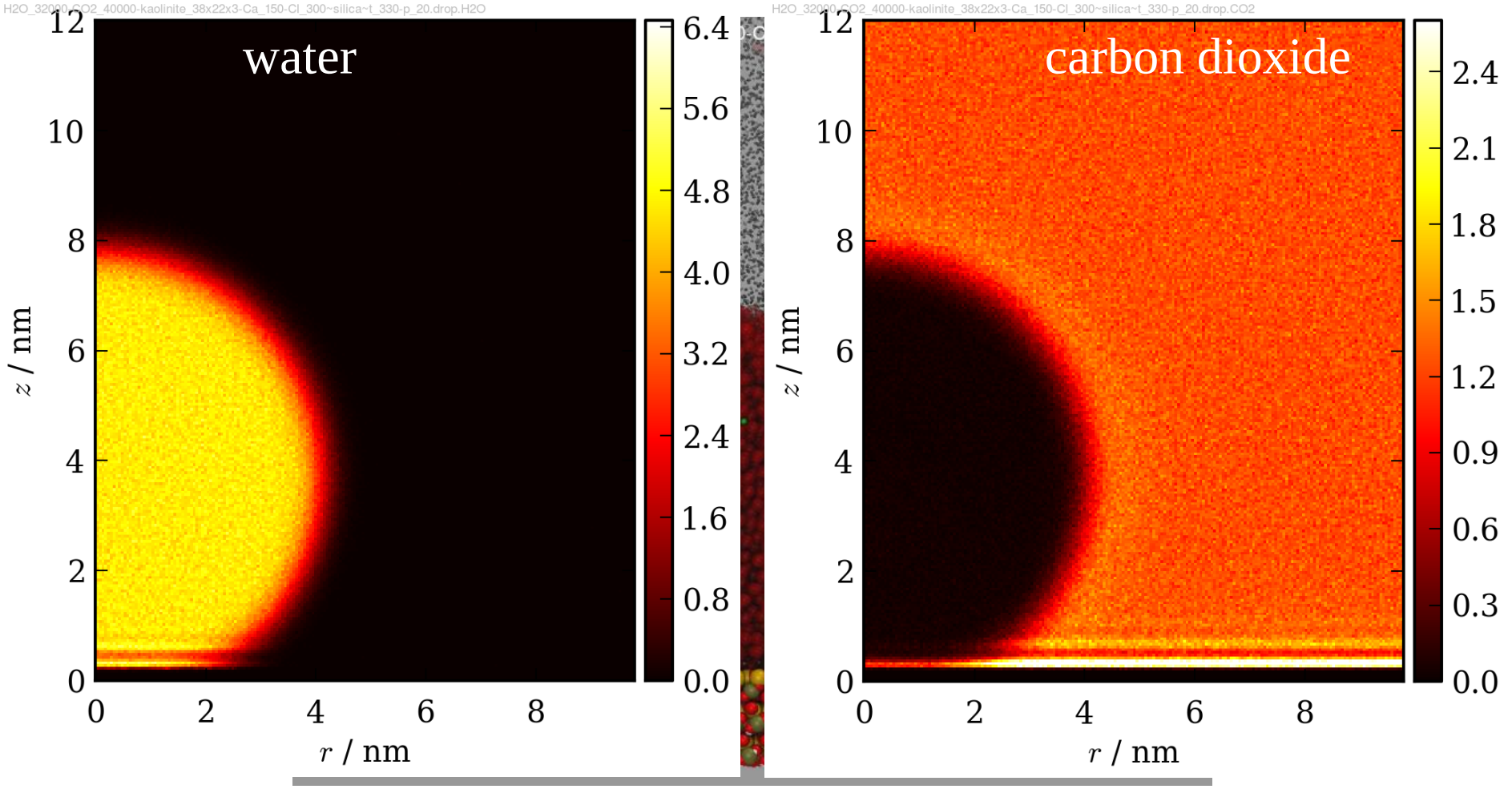

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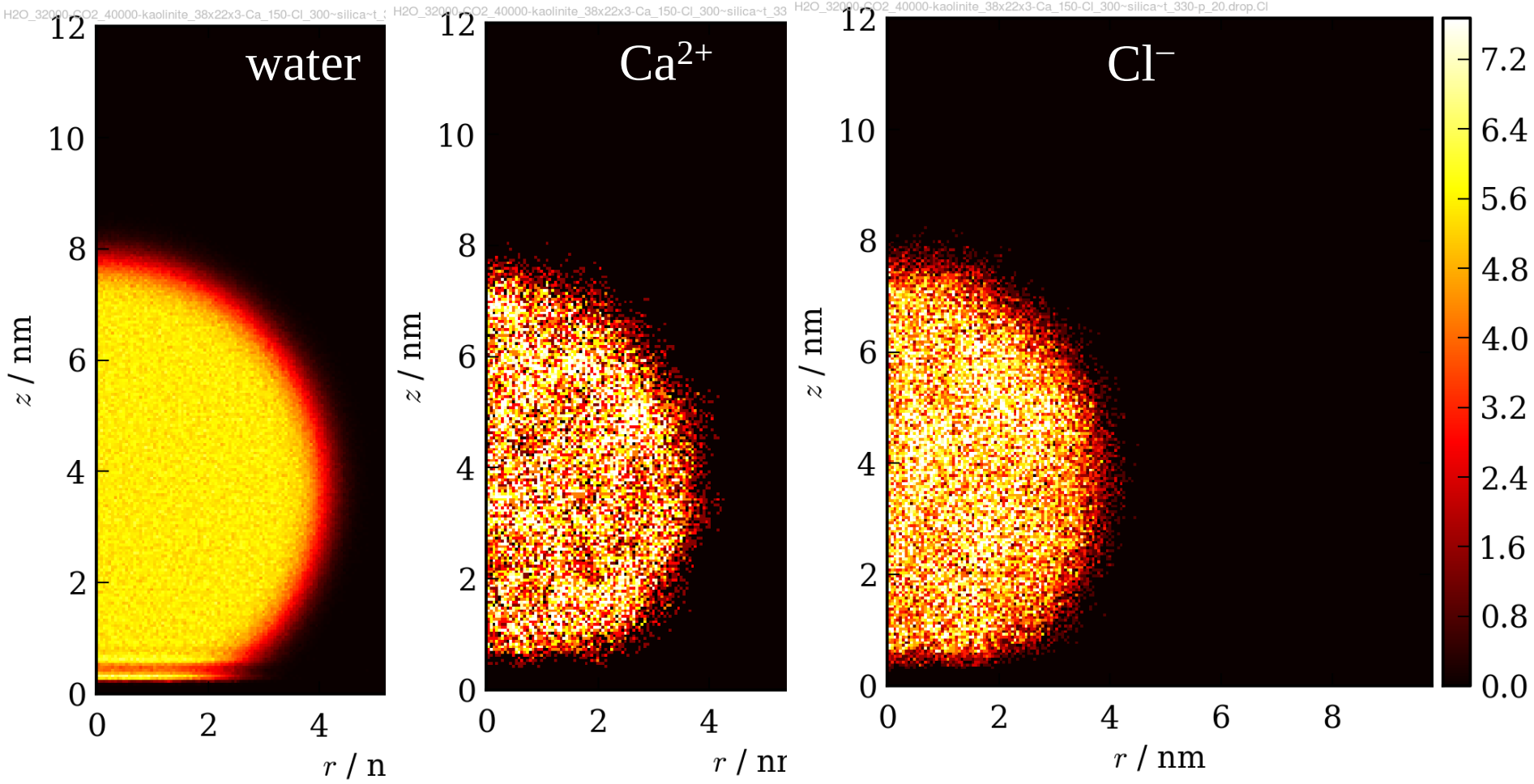

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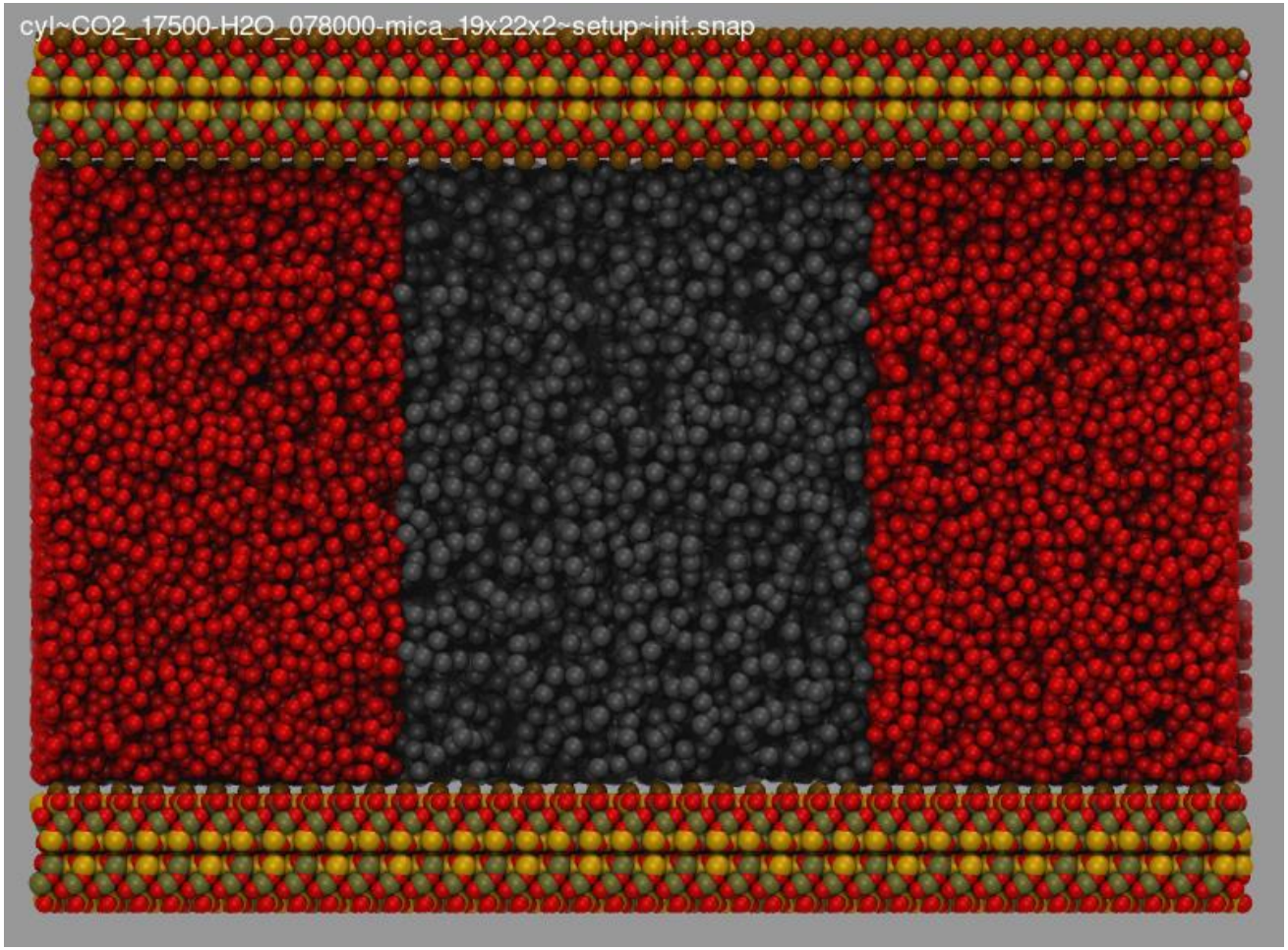
CO_2 and H_2O in mica slit pores initial configuration



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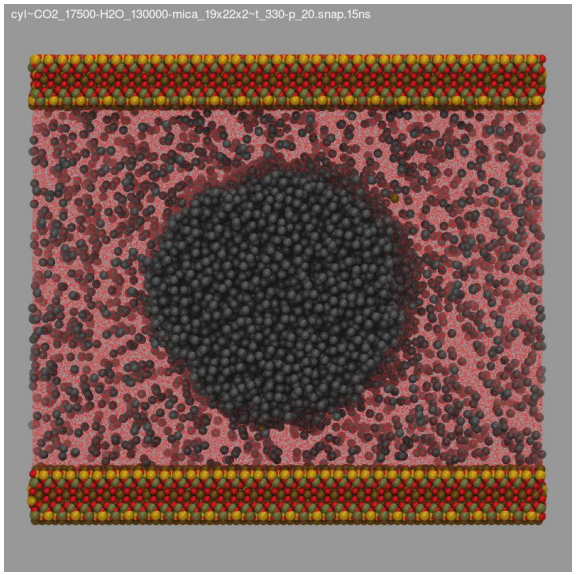
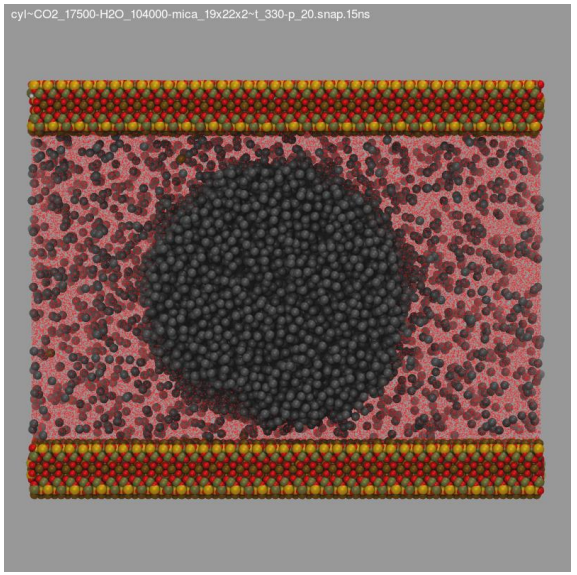
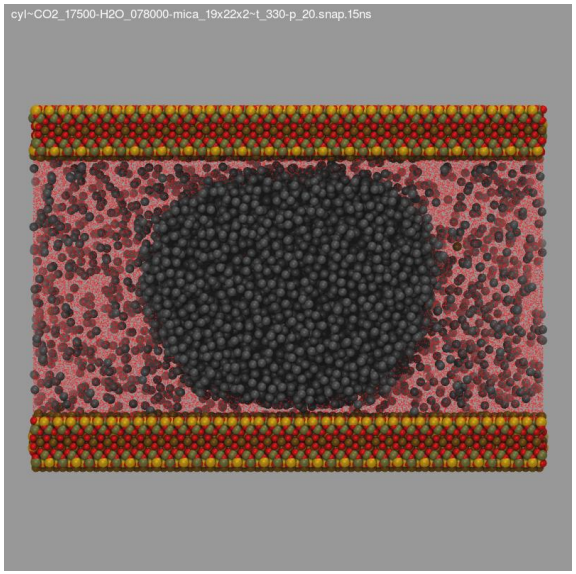
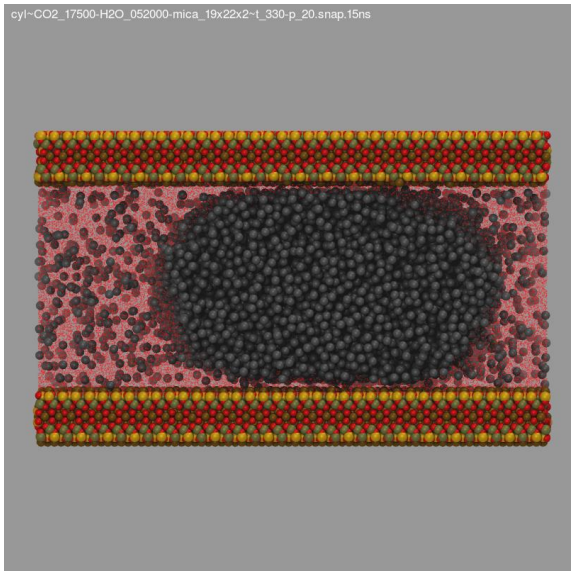
CO_2 and H_2O in mica slit pores final configurations









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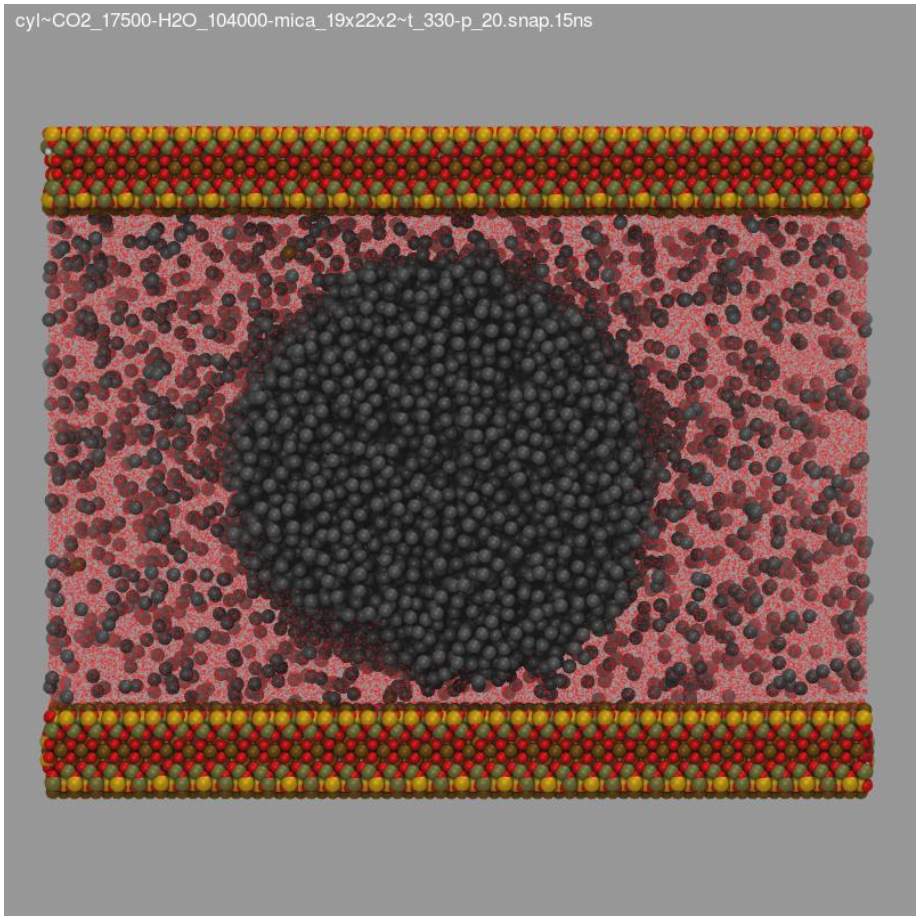
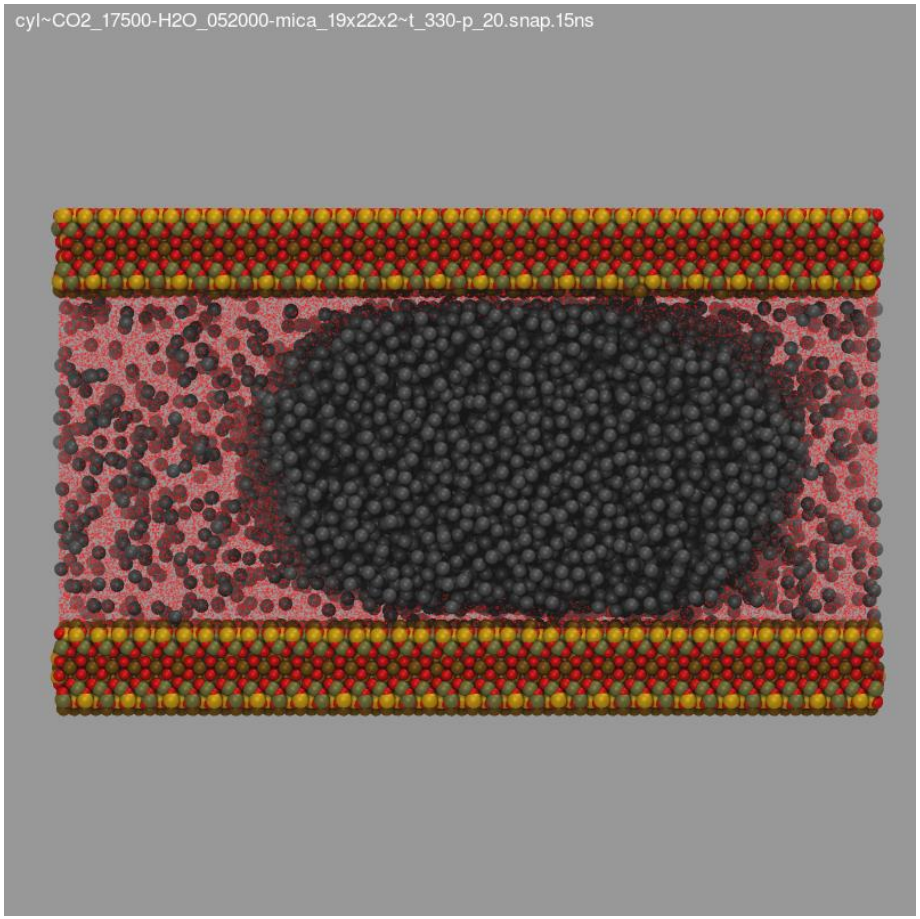
CO_2 and H_2O in mica slit pores final configurations





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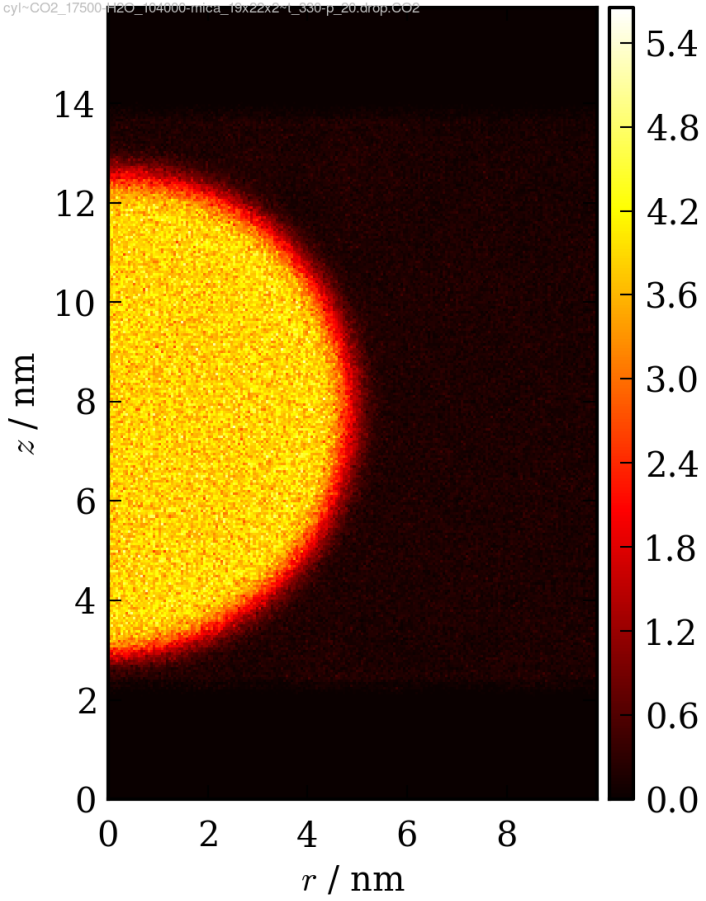
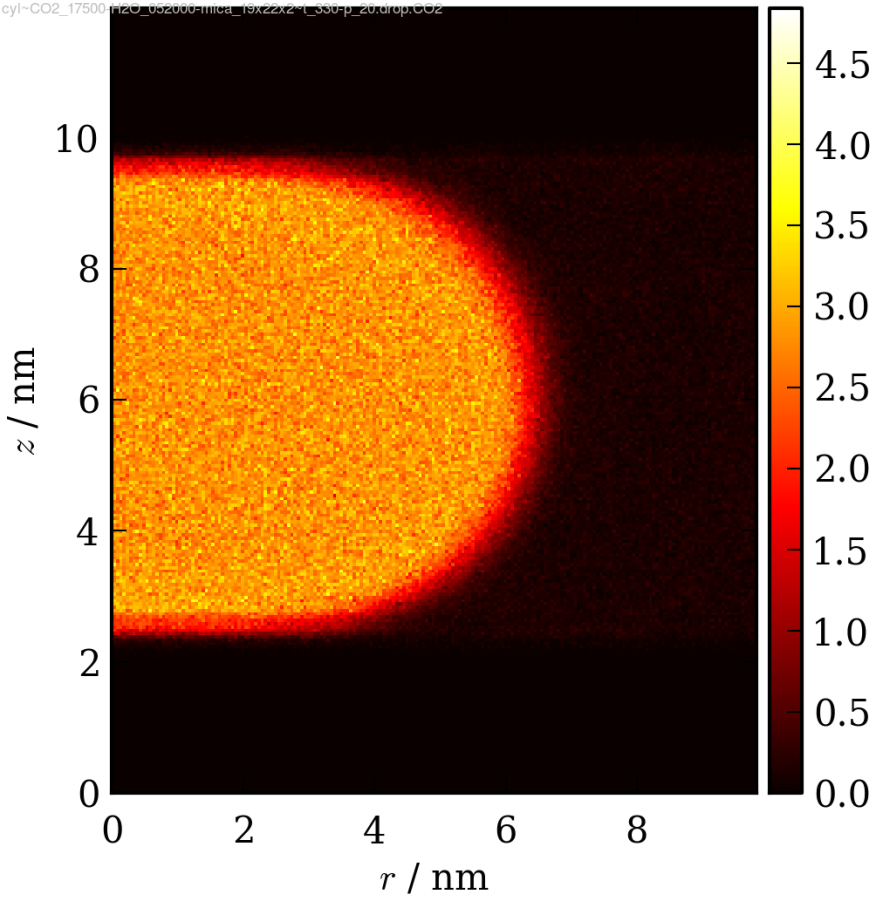
CO_2 and H_2O in mica slit pores
CO_2 density





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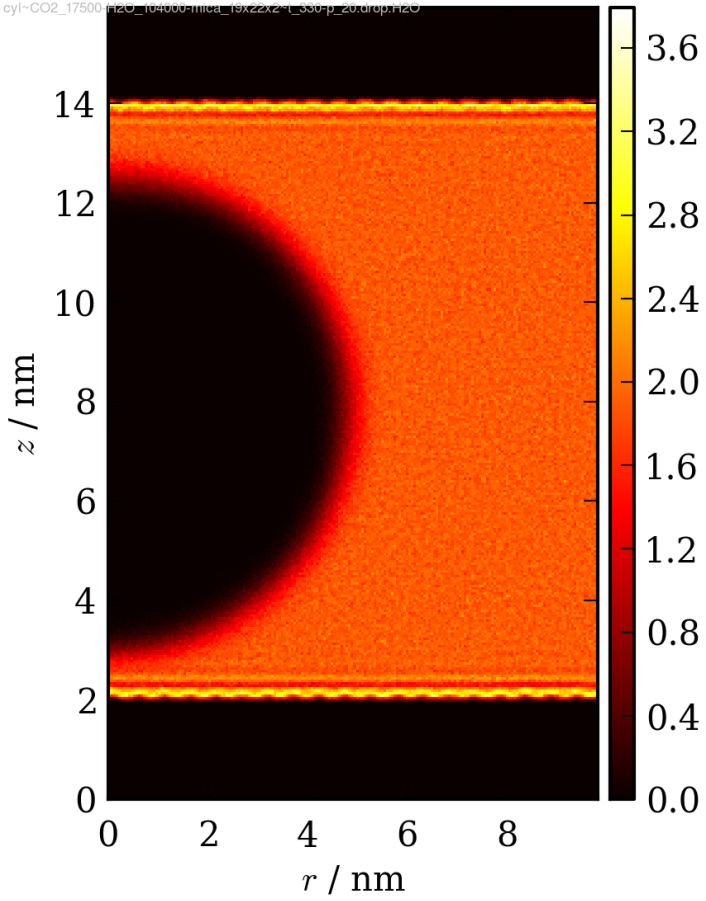
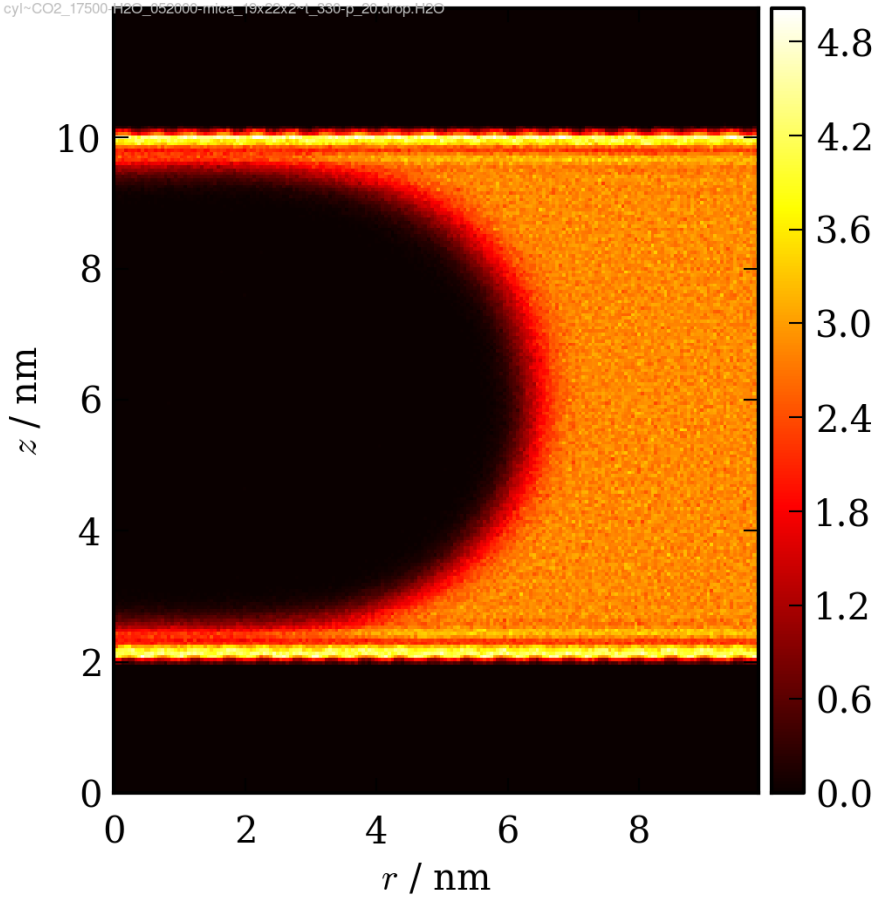
CO_2 and H_2O in mica slit pores
H_2O density





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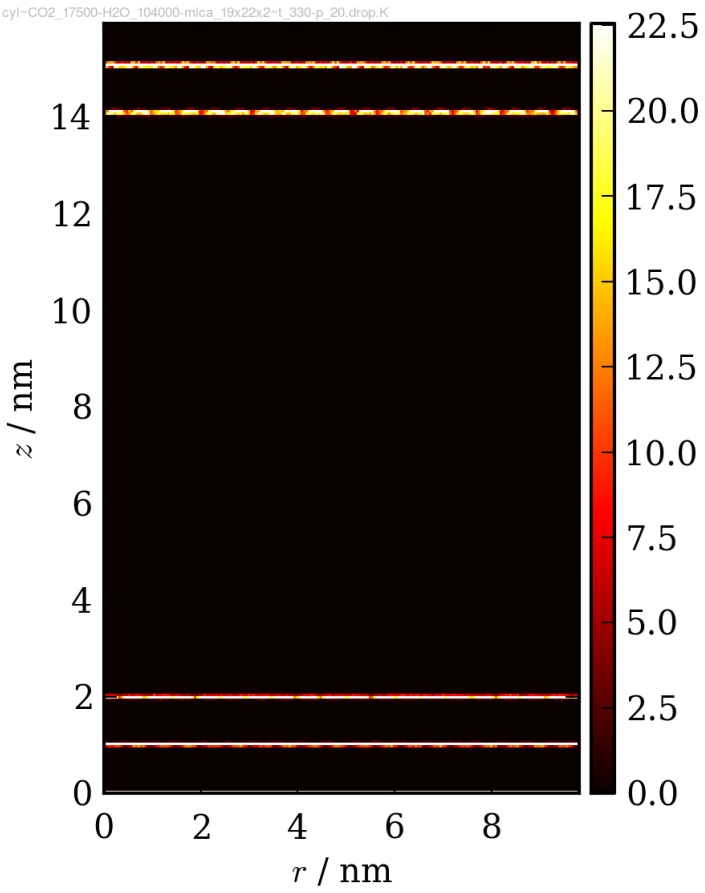
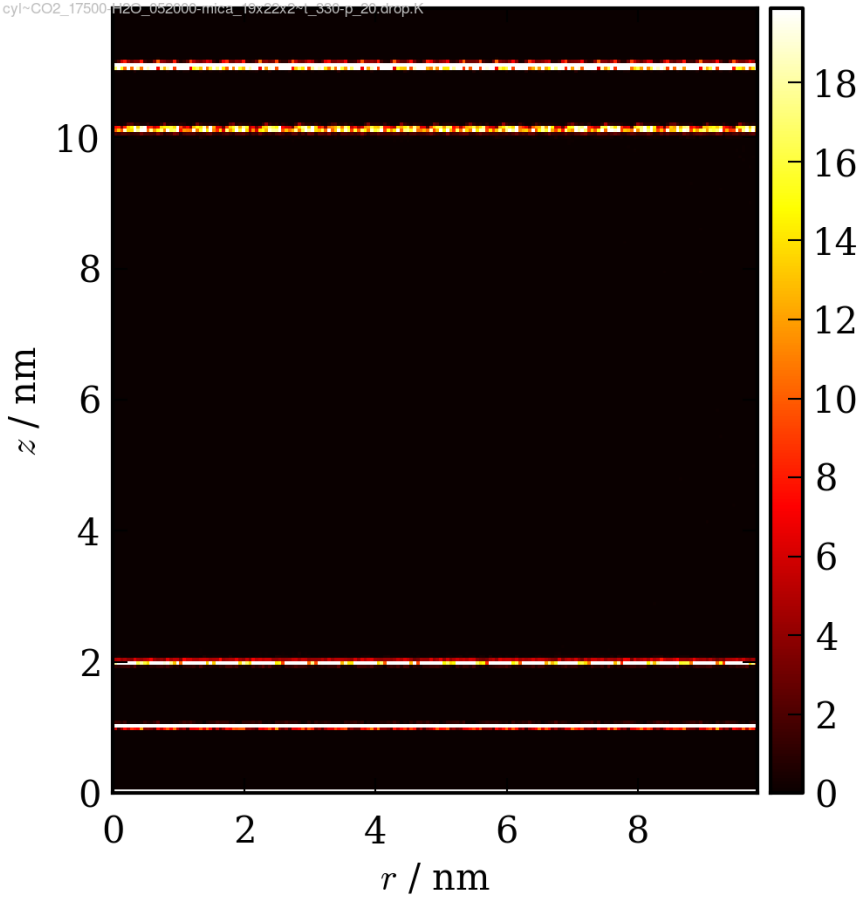
CO_2 and H_2O in mica slit pores
K^+ density





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Summary

- kaolinite hydrophilic surface
  - layers of water (and ions) completely displace CO_2
- kaolinite hydrophobic surface
  - strong CO_2 wetting in presence of water and brine
  - weak water and brine wetting in presence of CO_2
- mica slit pores
  - unconfined CO_2 is completely displaced by well-developed water layers
  - strongly confined CO_2 displaces diffuse water layers, slightly altering contact angle

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