

Understanding Water Controls on Shale Gas Mobilization into Fractures

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U.S. Department of Energy

National Energy Technology Laboratory

Mastering the Subsurface Through Technology Innovation, Partnerships and Collaboration:
Carbon Storage and Oil and Natural Gas Technologies Review Meeting

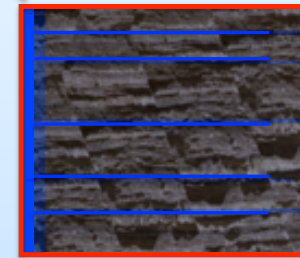
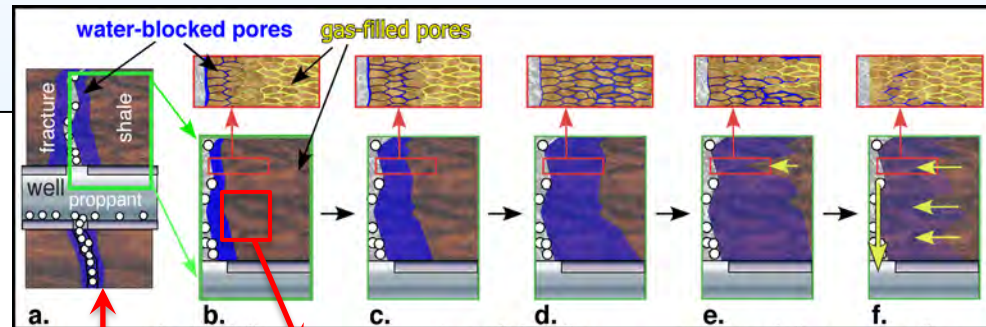
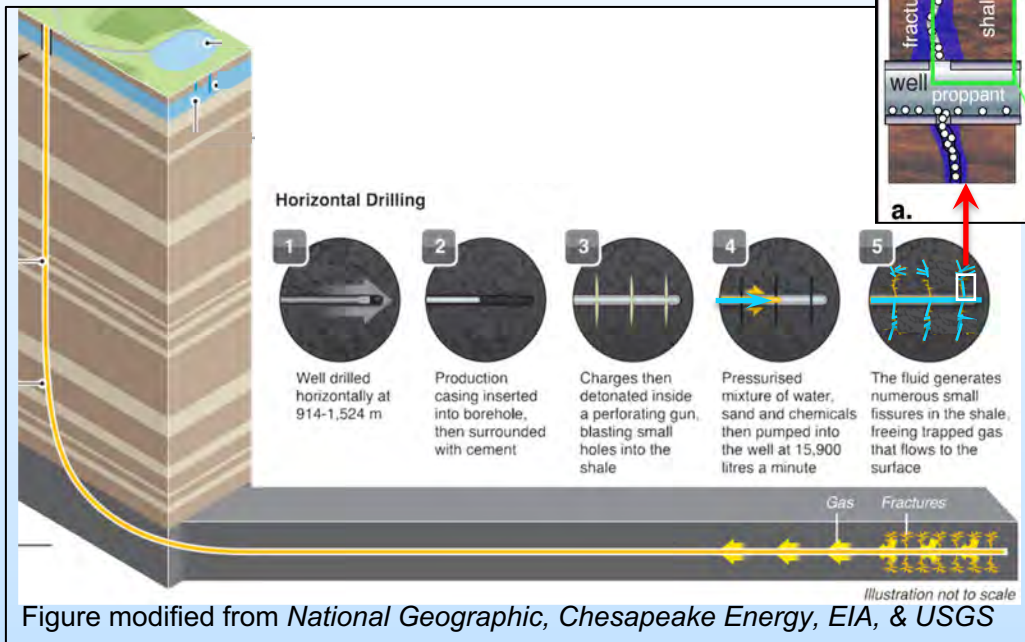
August 13-16, 2018

Presentation Outline

- General Background Information
 - Problem statement
 - Goals/Objectives
- Technical Status of current budget period (integrating accomplishments, lessons learned, and synergy opportunities within each task)
 - Task 2. Laboratory studies of shale-water interactions
 - Task 3. Laboratory studies of low-water fracturing fluids
 - Task 4. Modeling studies of shale-water interactions
- Summary of key findings
- Next Steps: Activities/tasks to be performed
- Appendix Materials

Problem Statement

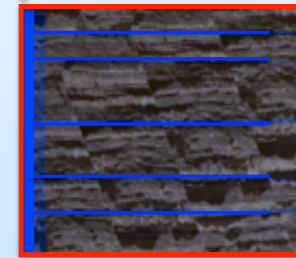
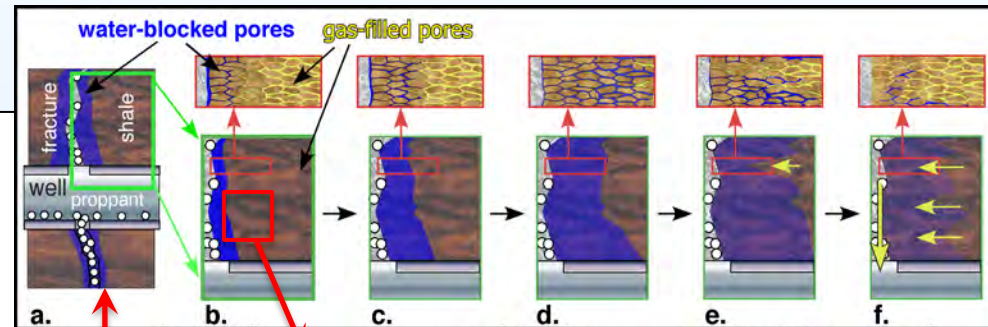
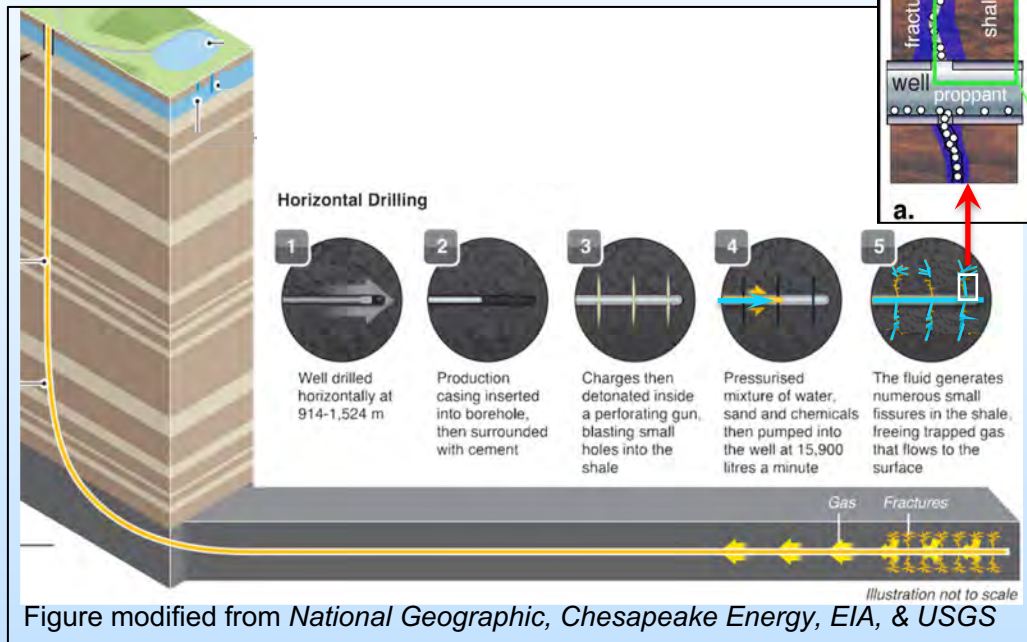
- $> 10^6$ gallons water/well is typically used to hydraulically fracture shale gas reservoirs.
- With typically $> 70\%$ of injected water (immiscible w. gas) usually remaining in the reservoir, why is gas production commonly high?
- Understanding is needed on how water distributes in shale and affects production, and how to improve gas/oil recovery, including via reducing water use.



Realistic representations of fluid displacement are needed to rationally improve extraction.

Goals and Objectives:

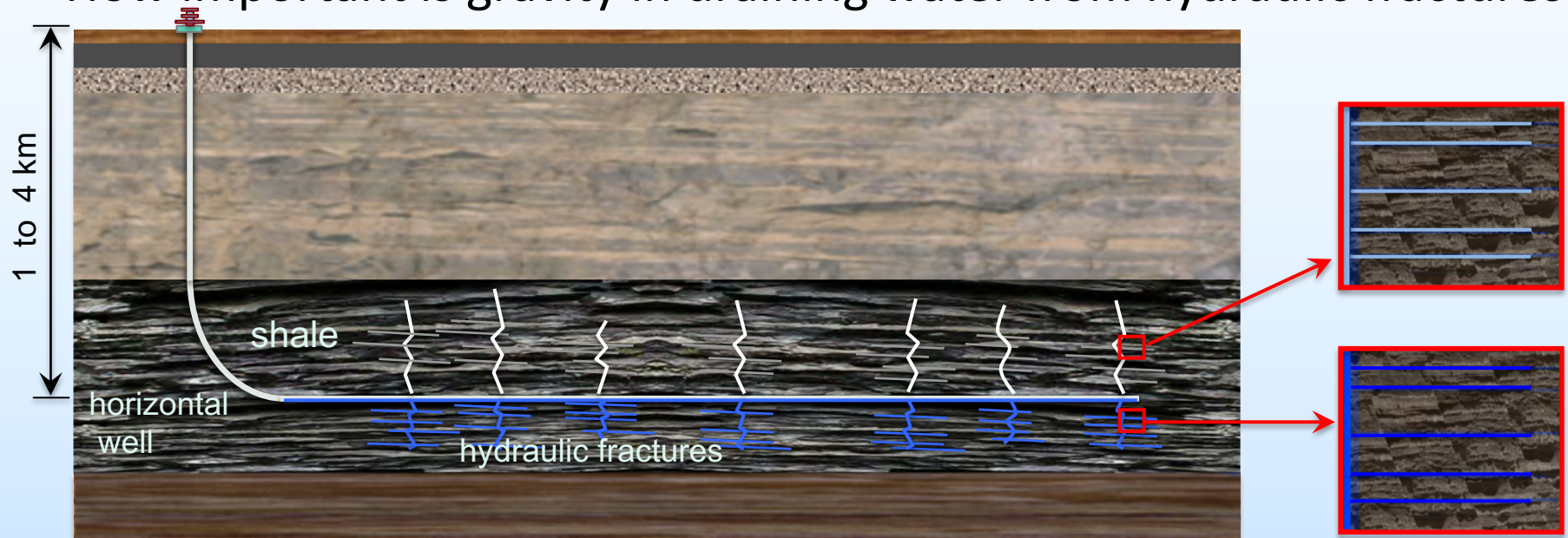
- Understand the coupling between water imbibition and gas counter-flow in shales in order to help identify approaches to improving production.
- Understand and improve effectiveness of decreased water injection, including low-water fracturing fluids for shale gas/oil production.



Realistic representations of fluid displacement are needed to rationally improve extraction.

Importance of orientation: anisotropy of transport properties and gravity influences in water-gas flow

- Nominally vertical hydraulic fractures connect with horizontally micro-fractured bedding planes of shale.
- Gas transport perpendicular to bedding planes is important.
- How important is gravity in draining water from hydraulic fractures?



Natural and stimulated secondary horizontal fractures along bedding planes are connected to primarily vertical hydraulic fractures. Above-well fractures drain more easily, but how important is this?

Task 2. Laboratory-based studies of shale-water interactions

2A. Physicochemical characterization of shales

2B. Measurements of anisotropic shale water uptake by vapor

diffusion/adsorption

2C. Measurements of shale water impacts on methane adsorption

Task 3. Laboratory-based studies of alternative fracturing fluids

3A. Natural biosurfactant extraction and foam properties measurements

3B. New foam generator system testing

Task 4. Modeling studies

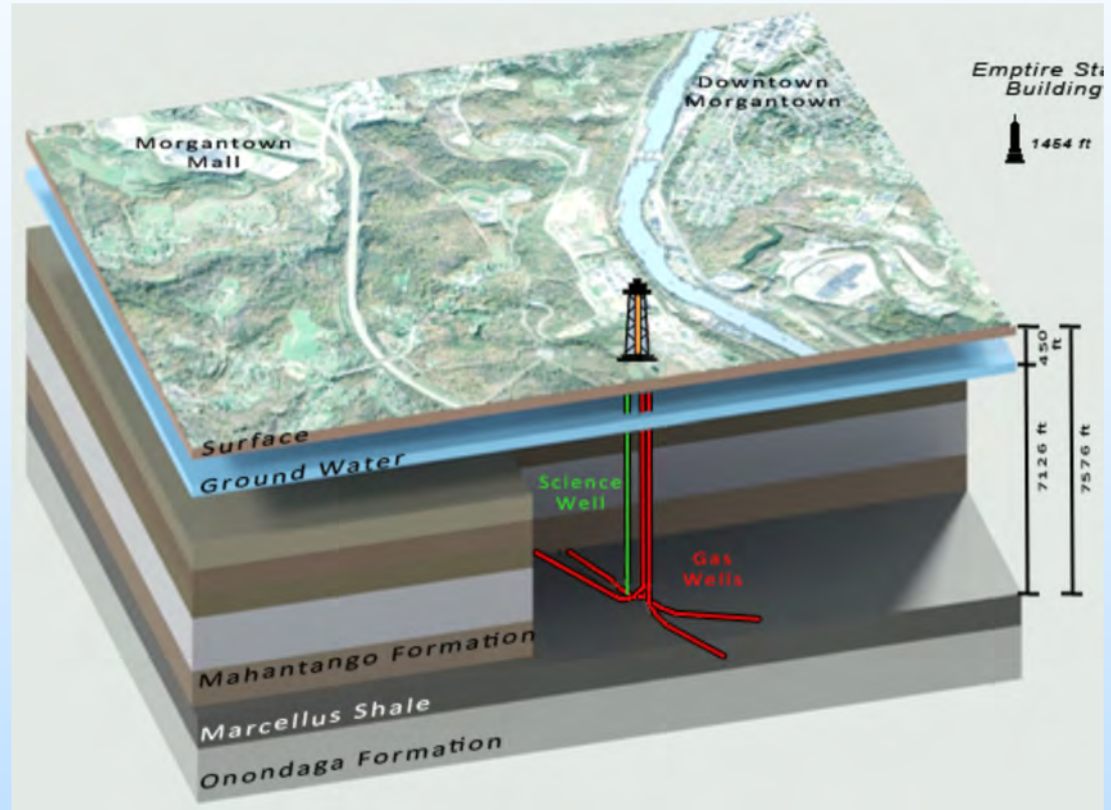
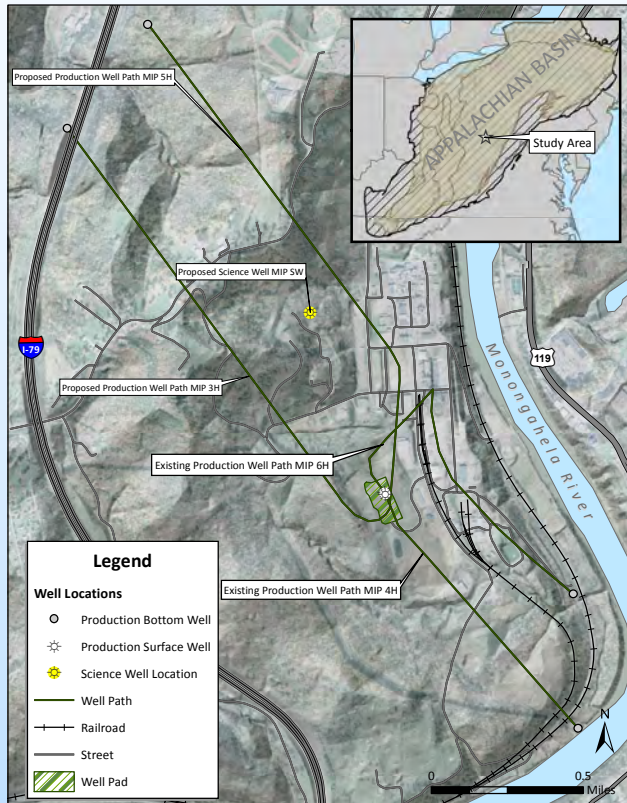
4A. Improve/apply multiphase flow models for shale

4B. Model water and non-water fluids at fracture-matrix interface

Marcellus and Mahantango
Shale samples for our recent
water uptake experiments

obtained from Marcellus Shale
Energy and Environment Lab
(MSEEL), Science Well

MARCELLUS SHALE PRODUCTION &
MSEEL SCIENCE WELLS



2B. Quantifying anisotropy in transport properties of Marcellus Shales: water vapor diffusion.



Vapor diffusion/adsorption in Mahantango and Marcellus Shale



7271'

7391'

7426'

7430'

7444'

7470'

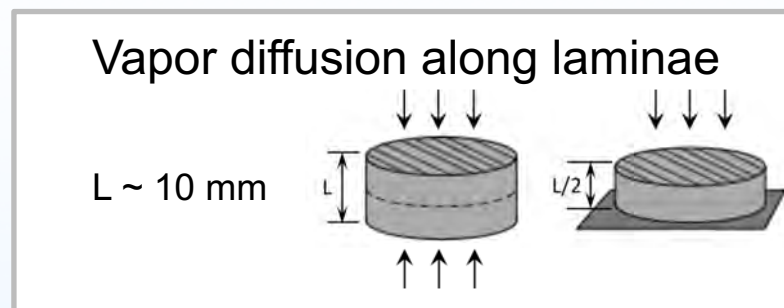
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110 °C (72 h) dry core plugs (sides sealed)

50 °C equilibrated in desiccator

transferred into 50 °C, 31% rh (MgCl_2 saturated salt solution)

periodically weighed



Elemental composition, X-ray fluorescence analyses

	Al	Si	Fe	K	Ca	Mg	Na	Ti	Ba	Sr	Mn	Cr	S	P	total C	organic C	inorganic C
	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%
Mahantango, 7271'	10.6	26.8	4.6	3.8	0.3	1.1	0.4	0.5	0.3	0.0	0.0	0.0	1.5	0.0	1.7	1.7	0.1
Mahantango, 7391'	8.6	27.4	5.2	3.1	0.9	0.9	0.3	0.4	0.1	0.0	0.0	0.0	2.3	0.0	2.5	2.4	0.1
<u>Marcellus, 7426'</u>	9.3	23.5	5.5	3.5	2.2	0.8	0.4	0.4	0.3	0.0	0.0	0.0	2.7	0.0	4.7	4.2	0.4
Marcellus, 7430'	10.0	26.6	4.3	3.7	0.4	0.8	0.4	0.5	0.1	0.0	0.0	0.0	2.2	0.0	2.8	2.7	0.1
Marcellus, 7444'	9.1	26.3	3.9	3.4	1.1	0.9	0.4	0.4	0.1	0.0	0.0	0.0	1.4	0.0	4.9	4.6	0.3
Marcellus, 7470'	8.4	26.6	3.4	3.1	1.1	0.9	0.5	0.4	0.1	0.0	0.0	0.0	1.3	0.0	7.0	6.6	0.4

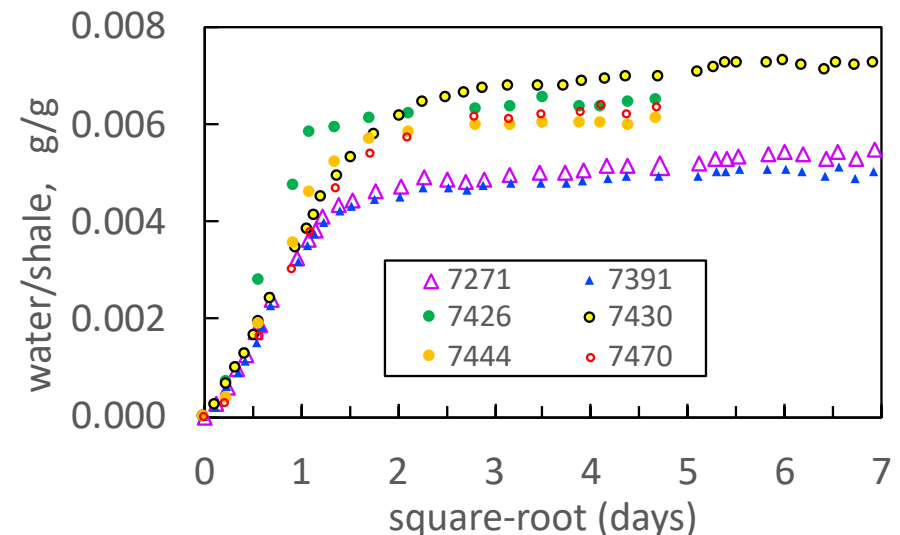
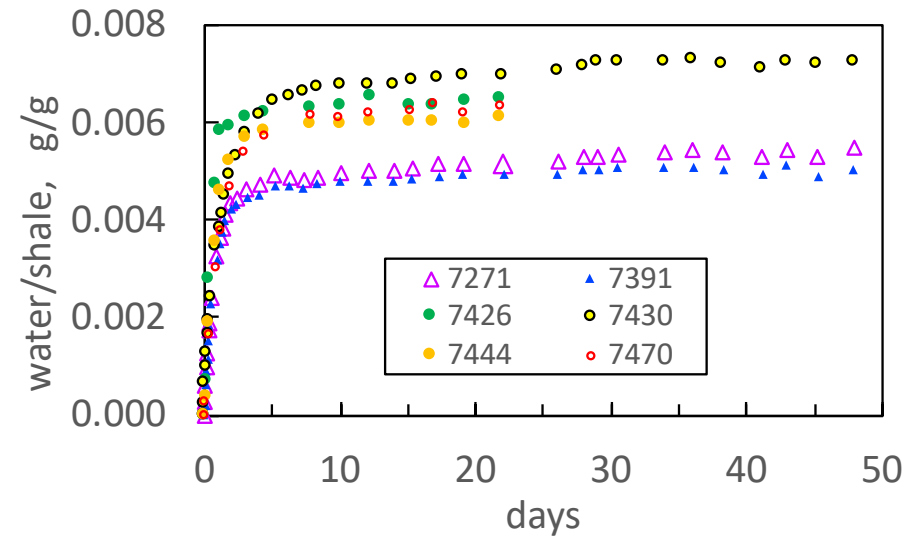
Water vapor diffusion parallel to laminae in Marcellus Shale:

Diffusion and adsorption at 31% rh and 50 °C in ~10 mm thick cores ($L/2 \sim 5$ mm) appeared to reach equilibria by ~30 days

110 °C (72 h) dry core plugs (sides sealed)
50 °C equilibrated in desiccator
31% rh condenses water in pores ≤ 1.6 nm

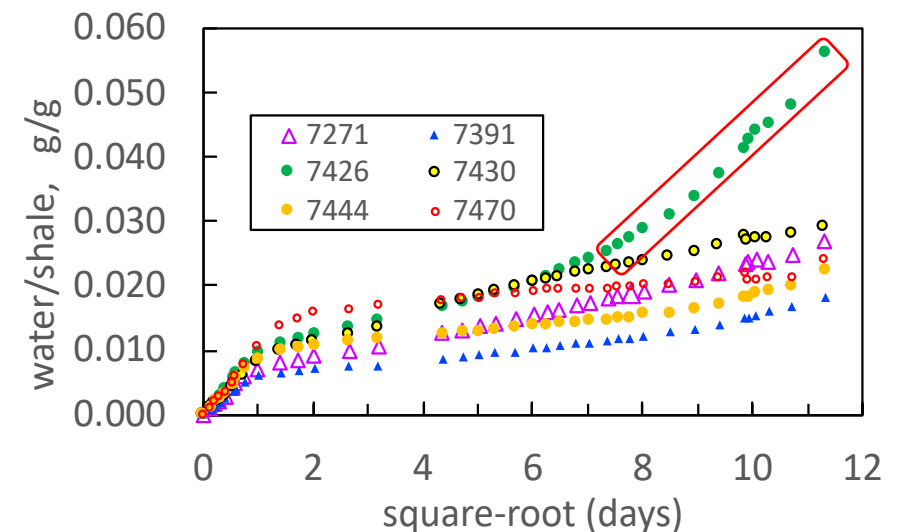
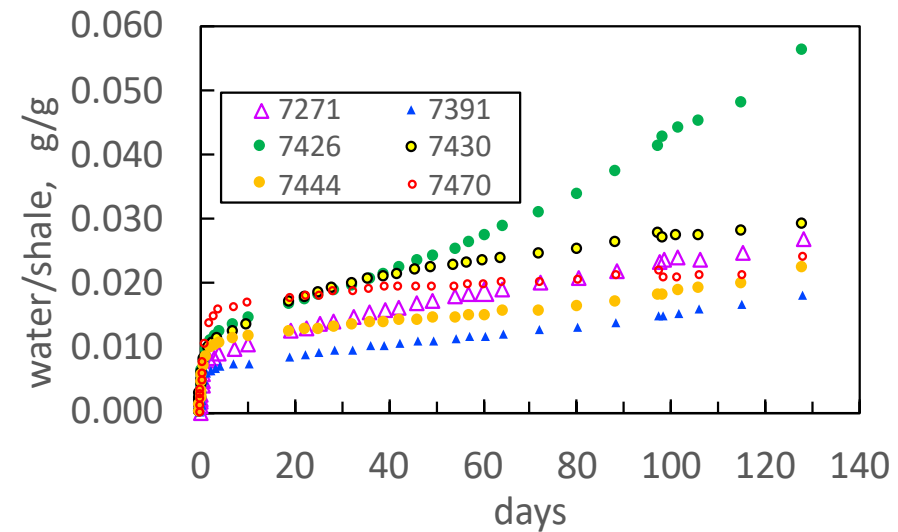


- Water vapor uptake is proportional to the square-root of time for the initial ~1.4 days, characteristic of diffusion-adsorption in semi-infinite systems.
- Later uptake appeared to level off to equilibrium adsorption/condensation.

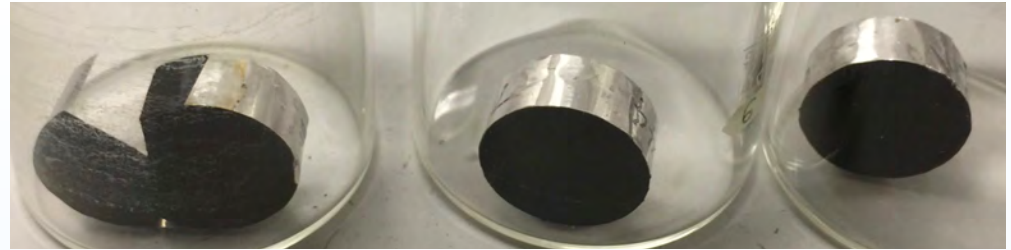
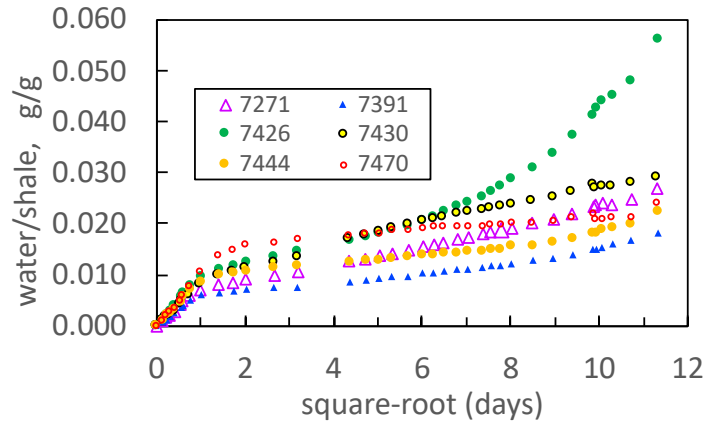


Water vapor diffusion parallel to laminae in Marcellus Shales: adsorption upon increasing rh from 31% to 51% (50 °C, on same samples)

- Initial water vapor uptake is again proportional to square-root of time.
- Later stage uptake lacked evidence of reaching equilibrium adsorption/condensation.
- 51% rh condenses water in pores ≤ 9 nm, slowing vapor diffusion.



Why do these shale samples continue to absorb water?

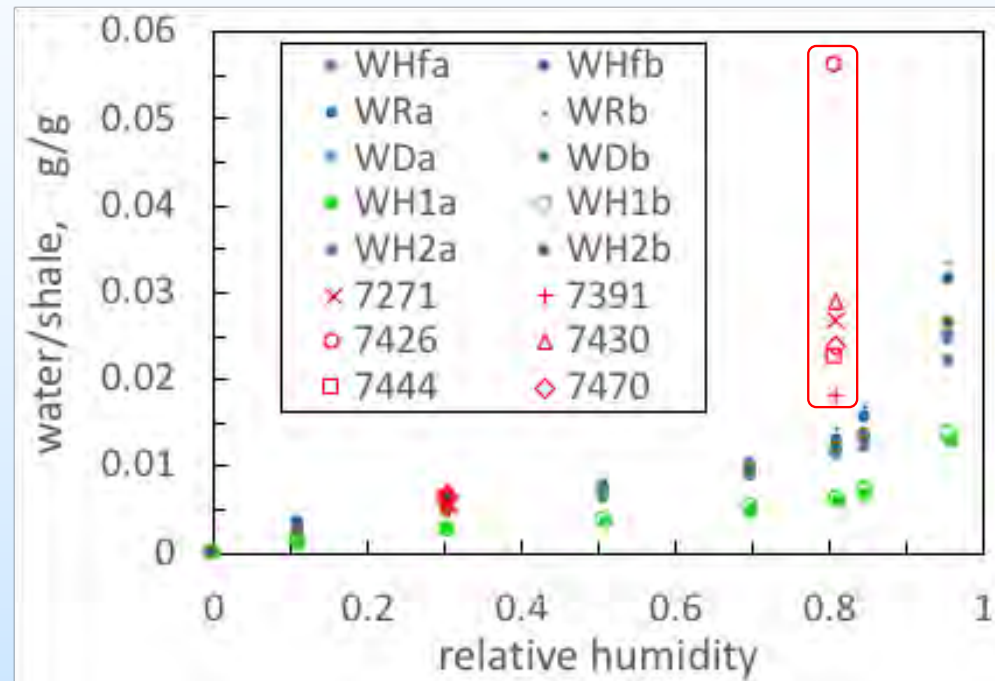


7426'

7444'

7470'

- Late stage water uptake did not level off
- Swelling of core 7426 even caused it to split open, breaking the epoxy-foil sleeve.
- Water uptake by day 120 in Marcellus Shale core plugs was much greater than levels obtained on crushed Woodford Shales (~5 day equilibrations).



Woodford Shale results are reported in Tokunaga et al., Water Resources Res., 2017.

Why do these shale samples continue to absorb water?

	Al	Si	Fe	K	Ca	Mg	Na	Ti	Ba	Sr	Mn	Cr	S	P	total C	organic C	inorganic C
	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%	%
Mahantango, 7271'	10.6	26.8	4.6	3.8	0.3	1.1	0.4	0.5	0.3	0.0	0.0	0.0	1.5	0.0	1.7	1.7	0.1
Mahantango, 7391'	8.6	27.4	5.2	3.1	0.9	0.9	0.3	0.4	0.1	0.0	0.0	0.0	2.3	0.0	2.5	2.4	0.1
<u>Marcellus, 7426'</u>	9.3	23.5	5.5	3.5	2.2	0.8	0.4	0.4	0.3	0.0	0.0	0.0	2.7	0.0	4.7	4.2	0.4
Marcellus, 7430'	10.0	26.6	4.3	3.7	0.4	0.8	0.4	0.5	0.1	0.0	0.0	0.0	2.2	0.0	2.8	2.7	0.1
Marcellus, 7444'	9.1	26.3	3.9	3.4	1.1	0.9	0.4	0.4	0.1	0.0	0.0	0.0	1.4	0.0	4.9	4.6	0.3
Marcellus, 7470'	8.4	26.6	3.4	3.1	1.1	0.9	0.5	0.4	0.1	0.0	0.0	0.0	1.3	0.0	7.0	6.6	0.4

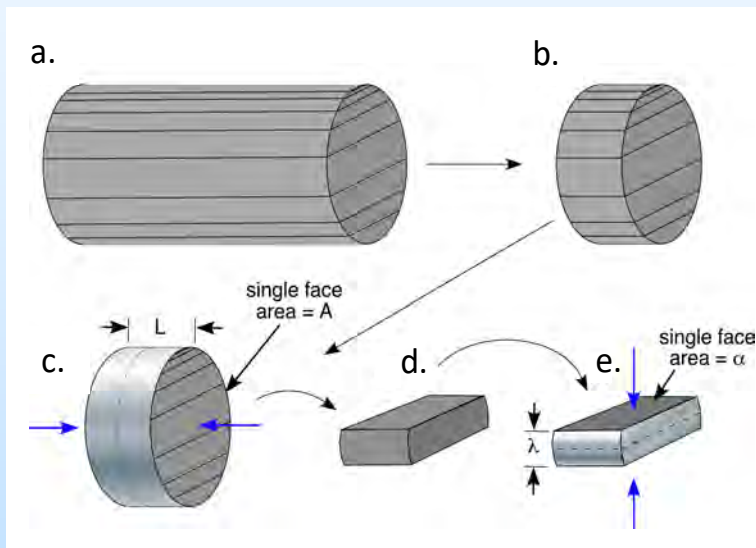
Based on elemental composition, with Ca and S both being elevated in the 7426' sample, we suspected anhydrite hydration and swelling to form gypsum

	Illite + Mica	Quartz	Pyrite	Chlorite	Plagiocl.	Calcite	Dolomite	Gypsum	K Feldspar	Anatase	Illite - Smectite	Kaolinite	Siderite
	%	%	%	%	%	%	%	%	%	%	%	%	%
Mahantango, 7271'	47.2	34.5	3.1	9.7	5.6	0.0	0.0	0.0	TR	TR	0.0	0.0	0.0
Mahantango, 7391'	37.8	40.6	8.2	7.7	4.5	1.1	TR	TR	TR	TR	0.0	0.0	0.0
<u>Marcellus, 7426'</u>	44.4	32.3	10.0	6.6	4.0	2.8	0.0	TR	0.0	TR	0.0	0.0	0.0
Marcellus, 7430'	45.7	36.1	6.6	7.3	4.3	0.0	0.0	TR	0.0	TR	0.0	0.0	0.0
Marcellus, 7444'	45.1	37.3	5.3	5.5	4.8	0.8	1.1	TR	TR	TR	0.0	0.0	0.0
Marcellus, 7470'	42.9	38.8	5.1	3.5	6.2	1.5	2.0	TR	TR	TR	0.0	0.0	0.0

- Based on XRD, neither anhydrite nor gypsum are at significant concentrations
- Pyrite is abundant in most samples, especially in the 7426' shale.
- Pyrite oxidation is enhanced under higher humidity, and causes swelling.

Next step: measuring water vapor diffusion/adsorption across shale laminae.

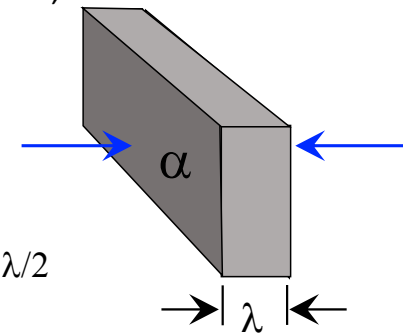
- Sliced core plug segments along bedding planes used in the previous along-laminae diffusion tests
- Sealed edges with epoxy and Al foil.
- Water uptake now occurs only by diffusion across laminae.



diffusion into opposite faces,
each with area = α ,
Total entry area = 2α ,

block thickness = λ ,
symmetry distance at = $\lambda/2$

f.



Water vapor diffusion across laminae in Marcellus Shale:

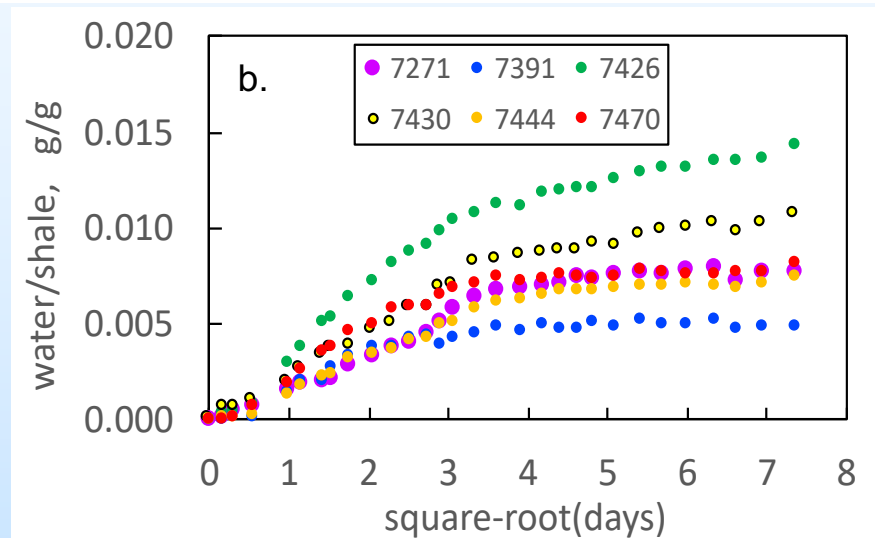
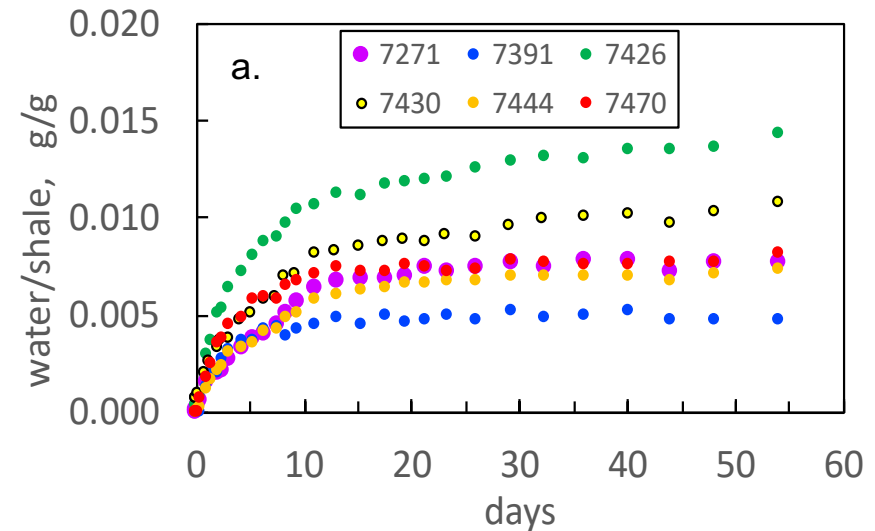
Diffusion and adsorption at 31% rh
and 50 °C

~5 mm thick slabs ($\lambda/2 \sim 2.4$ mm)

semi-infinite behavior for ~11 days

estimate $D(\text{parallel})/D(\text{perpendicular})$
from the ratios of depth/(sqrt time)

- $D(\text{parallel})/D(\text{perpendicular}) \sim 5.7$
- Values of $D(\text{parallel})$ and $D(\text{perpendicular})$ will be obtained from modeling of diffusion with nonlinear adsorption



2C. Influence of shale water on methane adsorption.

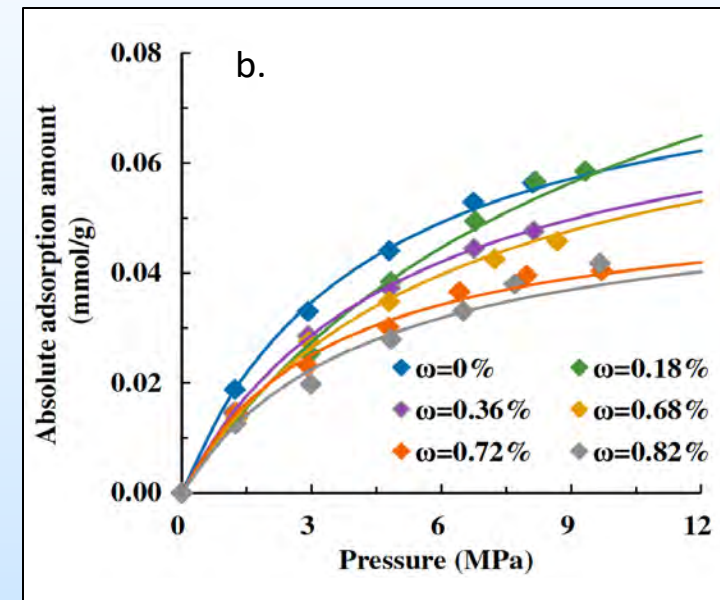
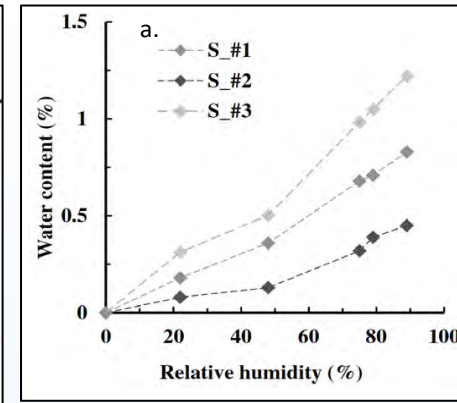
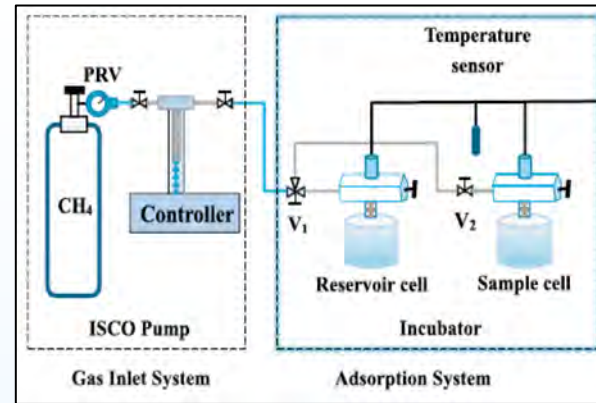
*Conducted by Lu Wang,
visiting Ph.D. student,
China University of Geosciences*

Shales from Qaidam Basin, China
Crushed to $< 74 \mu\text{m}$, dried.

Pre-equilibrated at 40°C , at fixed
relative humidities.

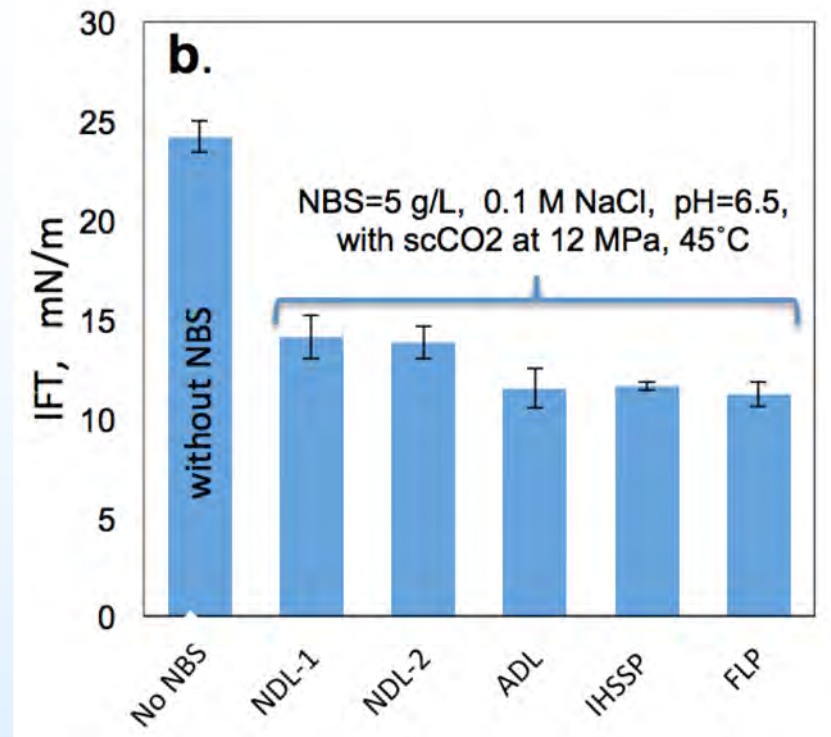
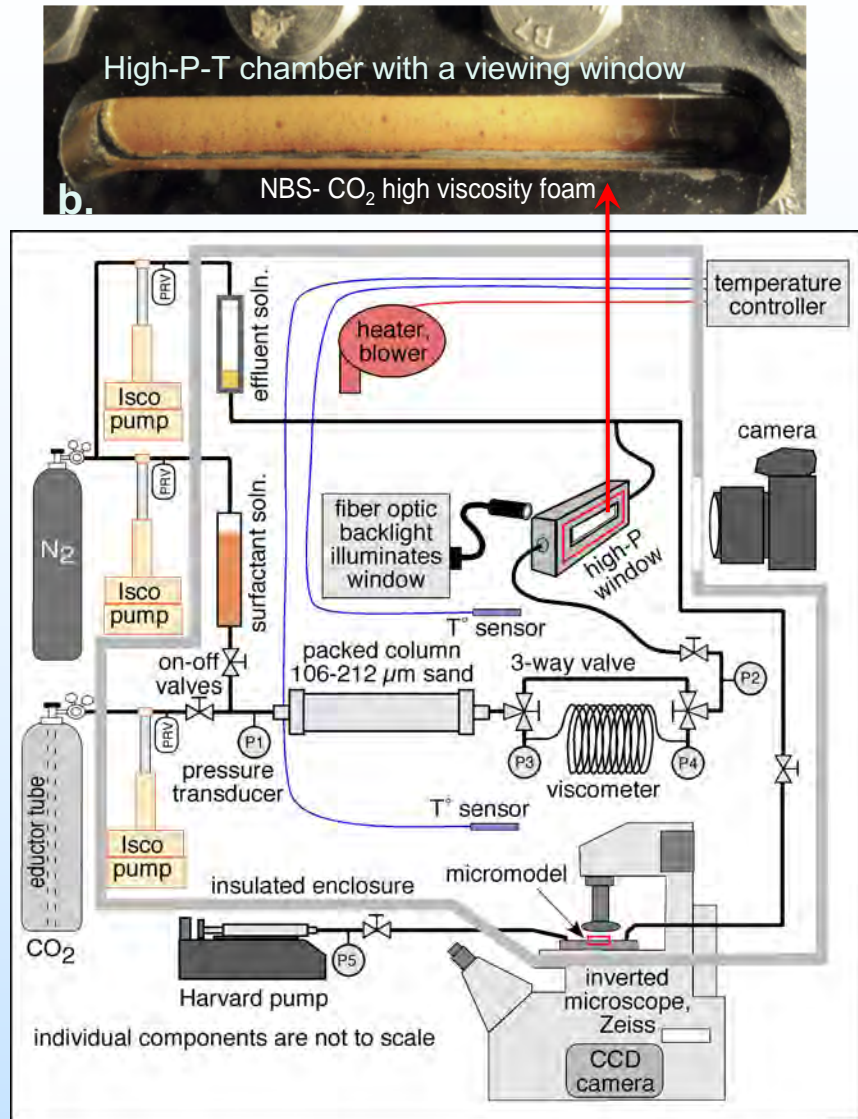
CH_4 equilibrated up to 10 MPa.

- CH_4 adsorption reduced by as much as 45% with water present.
- Further impacts of water on CH_4 adsorption become less significant at $\text{rh} > 72\%$.
- Publication. Wang et al., Water Resour. Res., 2018.



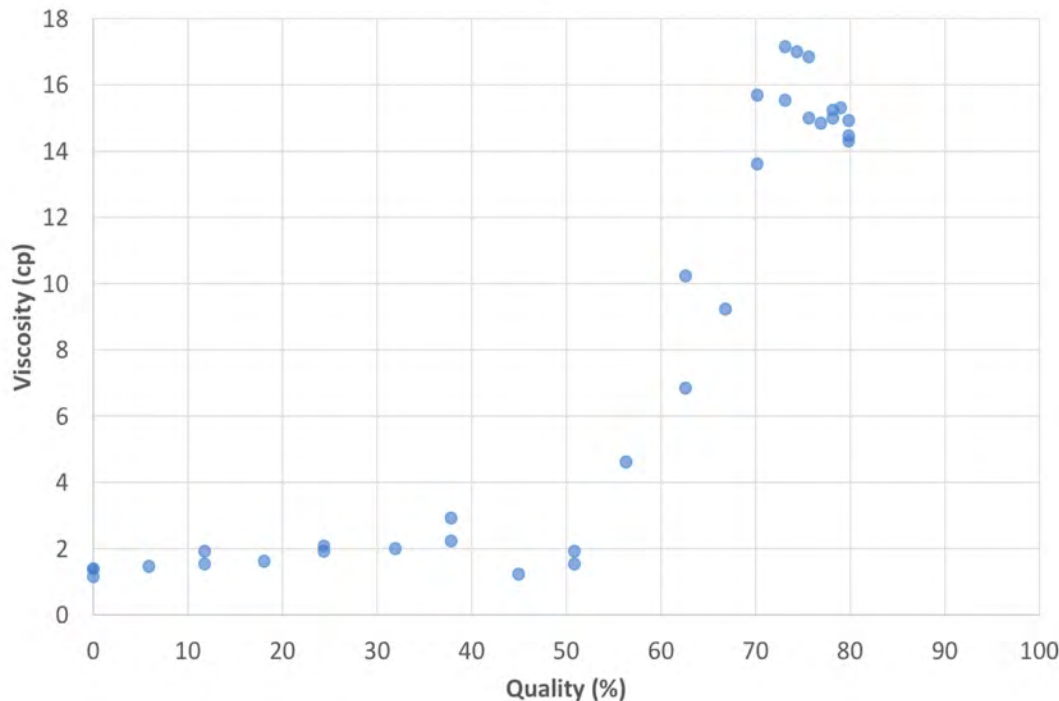
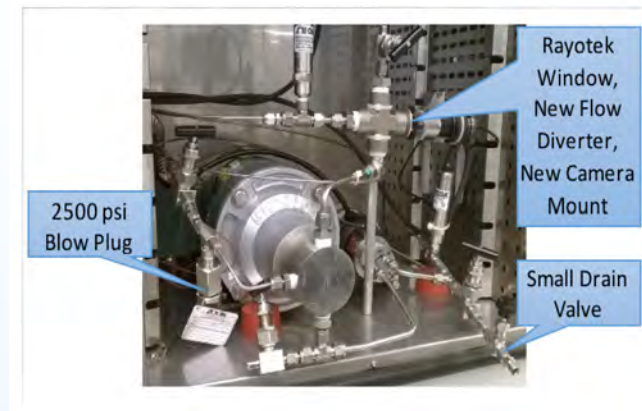
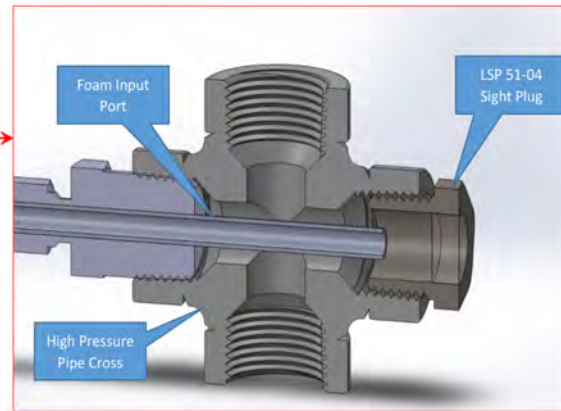
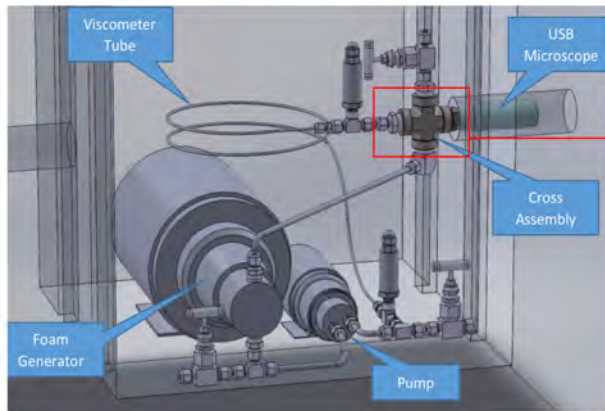
CH_4 adsorption isotherms for
different shale water contents.

Task 3A, Testing Natural Biosurfactants to Manipulate CO₂ Foam Viscosity



- Moderate decreases in CO₂-H₂O interfacial tension, down to ~10 mN/m.
- Moderate increases in effective viscosities, up to 20 cP.
- Publication. Wan et al., Energy & Fuel, 2017.

Task 3B, Developing a New CO₂ Foam Generator Test System



- Our previous bead-pack foam generator operates as a single pass.
- Desire for testing foams that are recirculated through porous and fractured media.
- Modifying a system developed by Hutchins and Miller, SPE 2005.

Task 4. Modeling studies for understanding fluid flow and transport processes in shale matrix and matrix-fracture interfaces

Task 4.A. Improve multiphase flow models for shale

Objectives:

- To understand mechanisms affecting multiphase fluid distribution in shale matrix surrounding hydraulic fractures
- To develop and improve predictive models for processes in shale.

Activities:

- Numerical and analytical models for diffusive transport with adsorption, to analyze the transient vapor adsorption measurements in shale laminae.
- Developed a new theoretical model for representing multiphase-multicomponent fluid flow in shale matrix and matrix-fracture interfaces.
- Testing and parametrization of the model parameters using pore-scale modeling tool (MDPD) and the experimental data developed in this project.

Modeling multiphase multicomponent flow phenomena using a new model

Motivation:

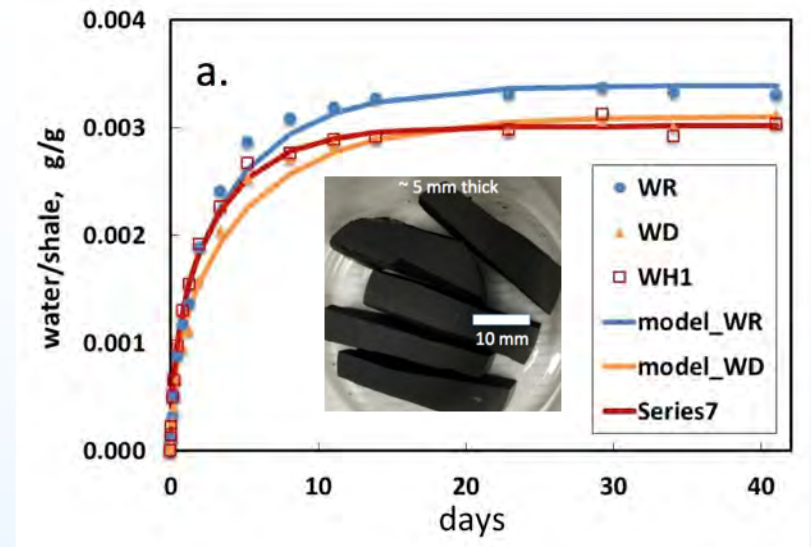
- Strong molecular interactions between fluid molecules and solid molecules exist in nanoscale pores of shale matrix.
- The fluid-solid interaction forces in shale pores can significantly influence the distribution of the fracturing fluids around the fractures and general continuum-scale behavior of multiphase fluids in shale matrix-fracture systems.
- Understanding the interaction forces on fluids in shale pores needed to find approaches to improve oil and gas recovery.

Approach:

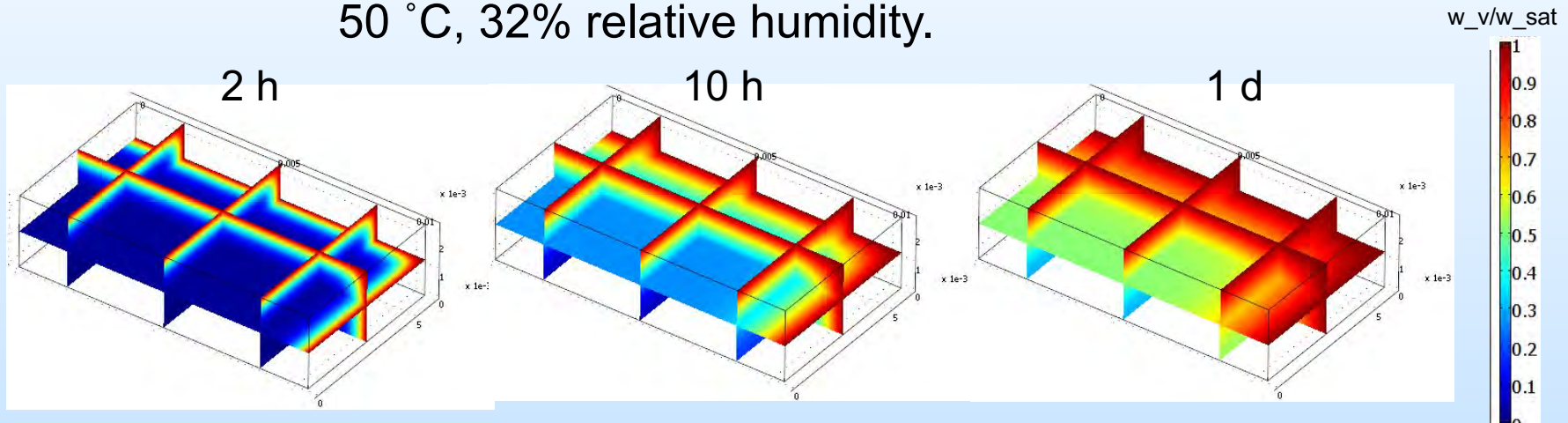
- Developed an initial version of a new multiphase multicomponent model based on the density functional approach, which led to development of new set of macroscopic equations. The equations explicitly include interaction forces between fluids and solids that influence multiphase processes at continuum scales of shale.
- Pore-scale modeling approach in conjunction with the experimental data will be used to characterize the fluid-fluid and fluid-solid interaction force terms in the new theory.
- Employ this new model to explore the sustained disequilibrium phenomenon and multiphase behavior with vapor diffusion and capillary condensation in shale rock samples in this project.

Diffusion-limited adsorption in shale

- Effective diffusion coefficients in the range of 9×10^{-9} to 3×10^{-8} m²/s were obtained through modeling of the diffusion process. These values are consistent with other measurements on low-porosity rocks.
- Simulations of diffusion in anisotropic shale are underway.



Time-dependence of water uptake in Woodford Shales,
50 °C, 32% relative humidity.

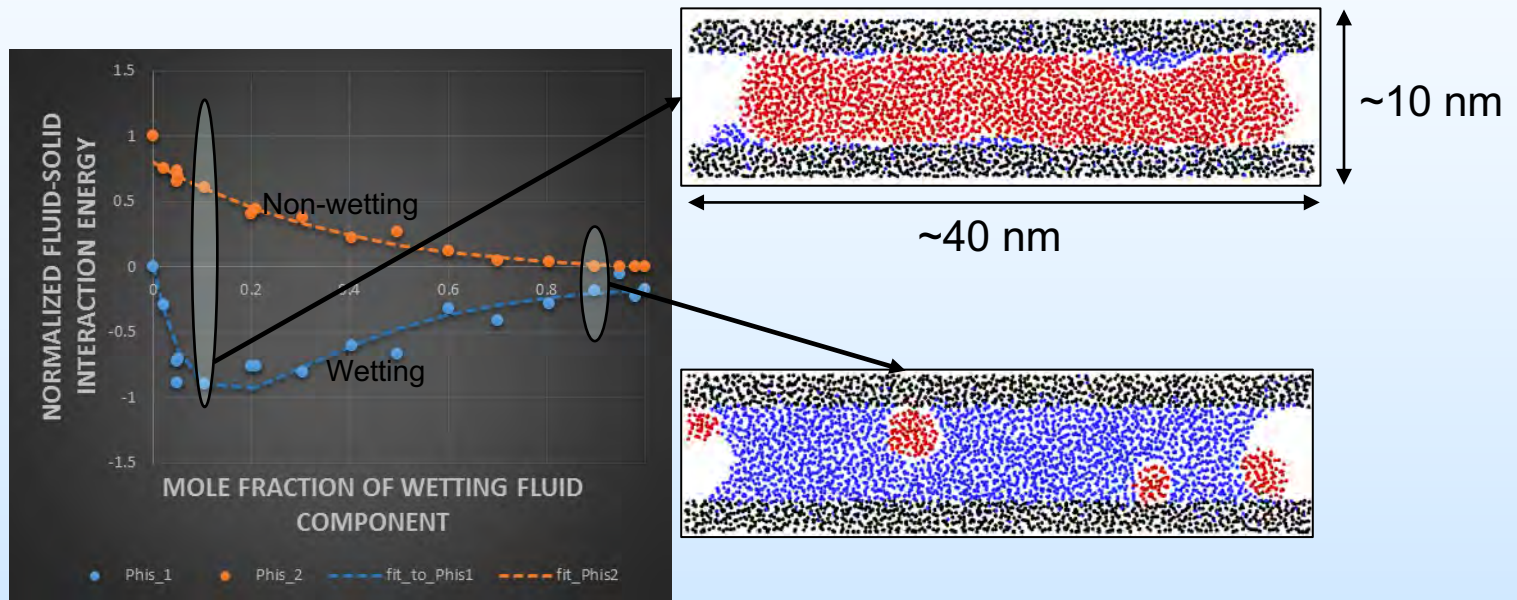


(Utilizing the symmetry, the numerical results are shown only for the quarter of the domain)

Modeling multiphase multicomponent flow phenomena using a new model

Preliminary application of MDPD-based pore-scale modeling approach to determine the functional relationships of the fluid-solid interaction forces in nanoscale pores

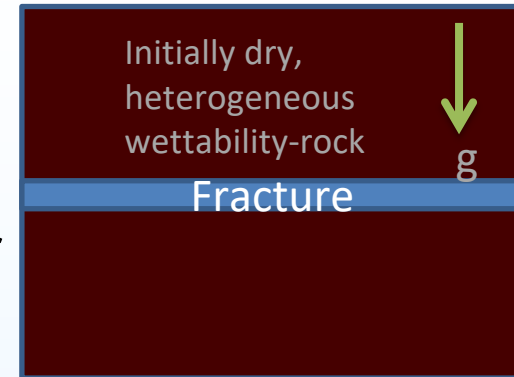
Strong attractive energy btw thin wetting fluid molecules (blue) and the wall molecules (mole fraction of blue=10%)



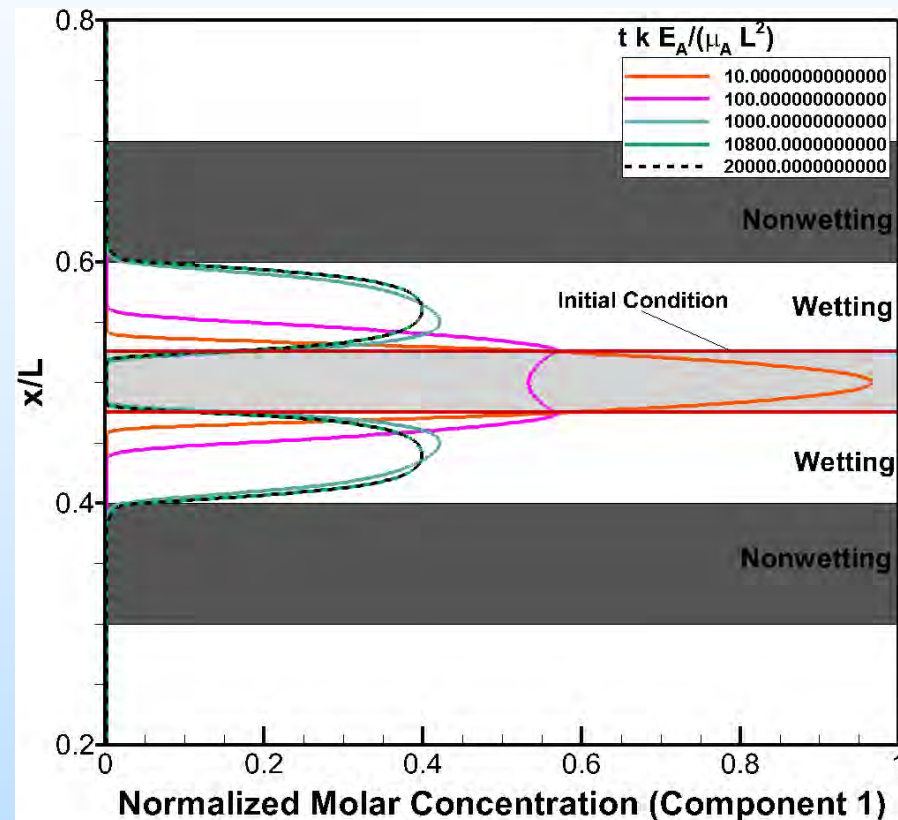
Attractive energy btw wetting fluid molecules (blue) and solid decreases with increasing moles of blue (mole fraction of blue =90%)

Modeling two-phase-two-component flow in shale: A preliminary simple demonstration

- Functional relationships of the fluid-solid interaction forces built based on the MDPD-based pore-scale modeling studies
- Example demonstration of the new model presented for simulating wettability effects on two-phase two-component fluid distribution in shale matrix around a fracture



- Water stays within the wetting zones (hydrophilic rock materials) around the fracture, potentially blocking oil/gas flow
- Gravity does not appear to play a significant role.
- Next, we will test the model with the experimental data generated from the vapor diffusion and capillary condensation experiments in shale

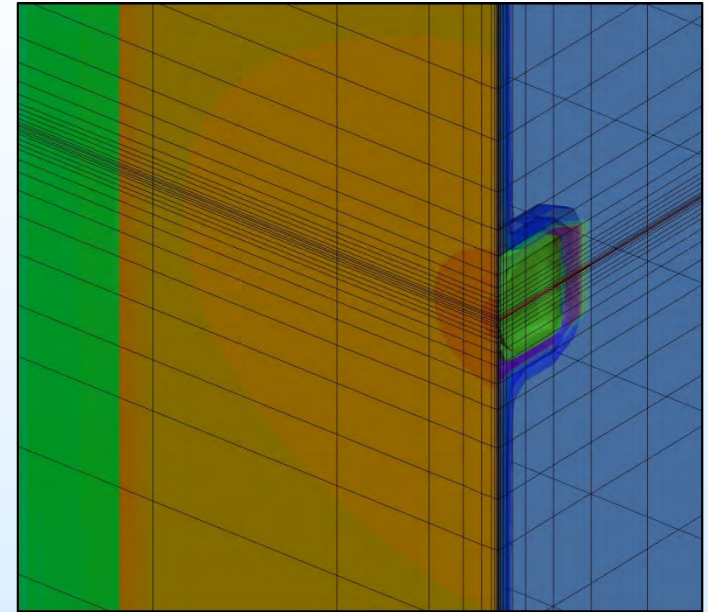


Task 4.B. Testing and development of macroscopic modeling approaches for flow processes of water and non-water fluids in shale matrix and across fracture-matrix interface

- Goal: to examine the potential influence of gravity segregation of fracturing fluid within the main hydrofracture and surrounding fracture network on gas productivity
- Activity: to simulate gravity drainage in fractured shale reservoir
 - Using iTOUGH2/EOS7c
 - A three stage scenario
 - Fracking for two hours
 - Soaking for seven days
 - Producing for five years

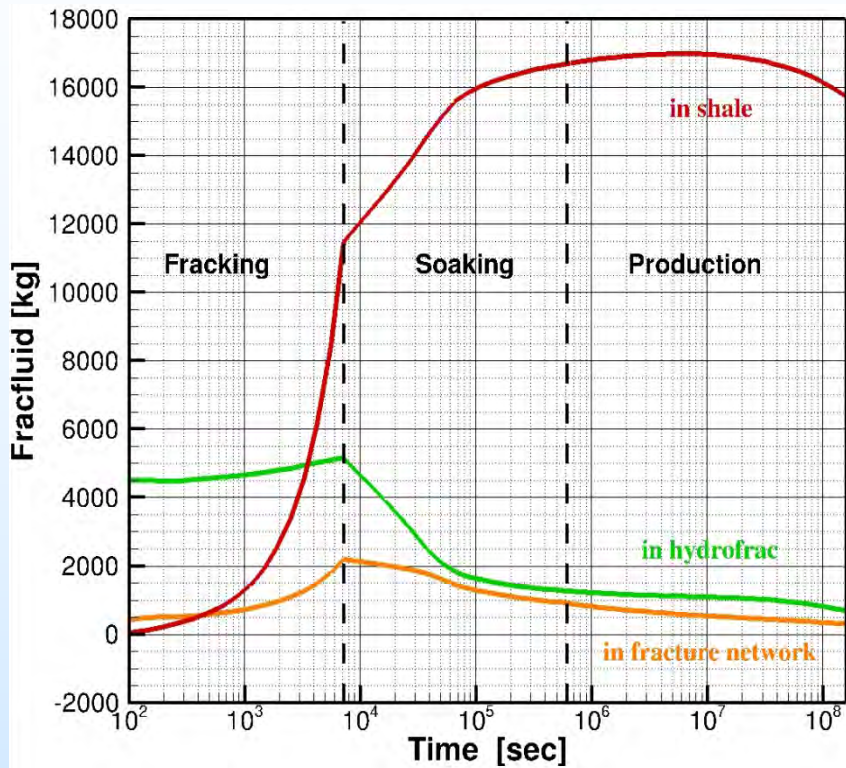
Model Setup

- Methane-bearing tight reservoir rock
- Horizontal stimulation/production borehole
- Vertical hydrofrack
- Stimulated small fracture around hydrofrack (five interacting continuum)

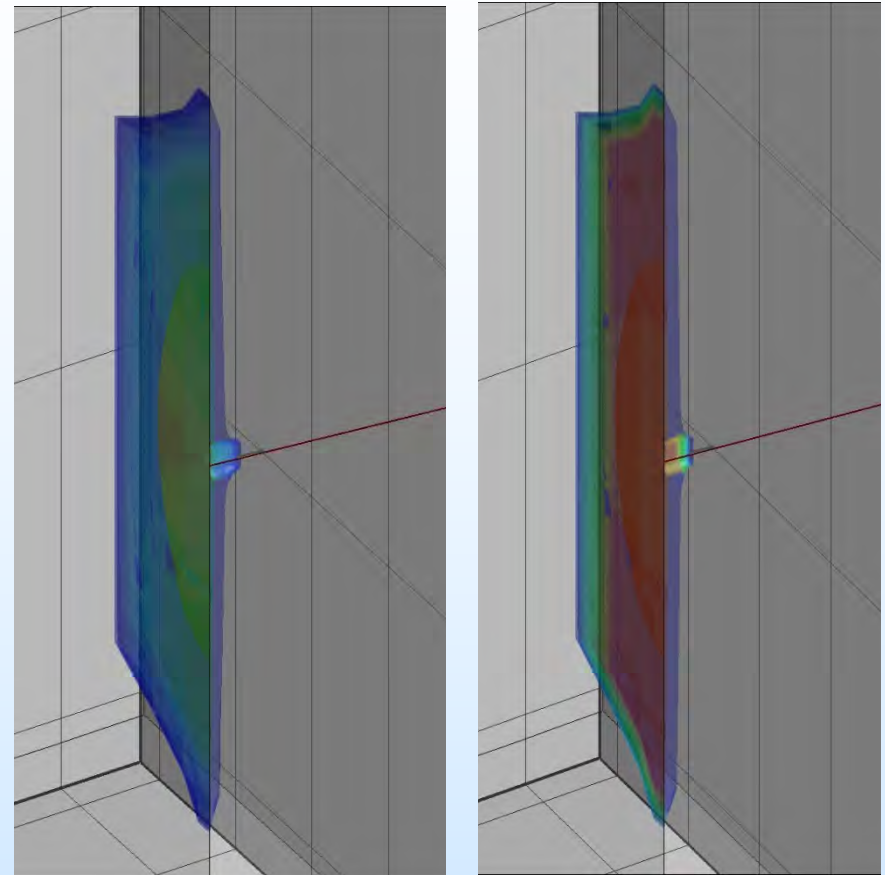


close-up of permeability structure near perforated well section

Fluid Balance



Fracking fluid distribution within stimulated tight gas reservoir



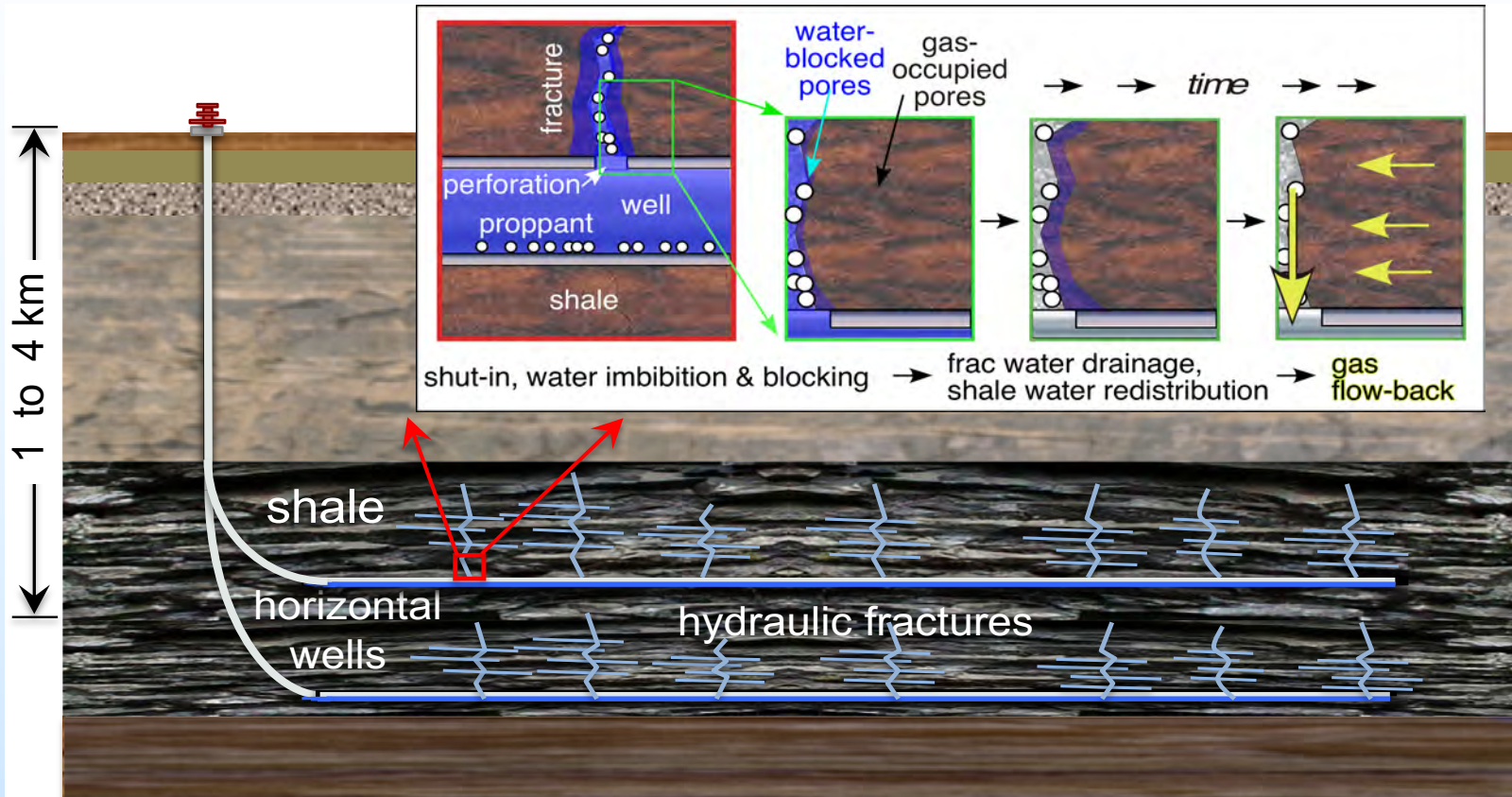
Concentration of fracking fluid in pore water after (a) one week of soaking, (b) five years of production

Observations

- Fracfluid imbibes into shale matrix, enabled by:
 - High fracking pressure
 - Dense network of secondary, small fractures around hydrofrac, providing large surface area for matrix imbibition
 - Assumption of strong capillary suction in gas-filled matrix
 - Limited storage volume in fracture
- Once in the low-permeable matrix, the fracfluid is difficult to remove, even during an extended production period.
- Imbibition of fracfluid into the matrix leaves hydrofrac essentially dry, reducing gravity drainage processes
- These conclusions are contingent on assumptions on matrix sorptivity properties

Implications for increased reservoir production per unit water use if gravity drainage is important

- Placement of horizontal wells deeper in pay zones.
- Directional, upward, perforating and hydraulic fracturing.

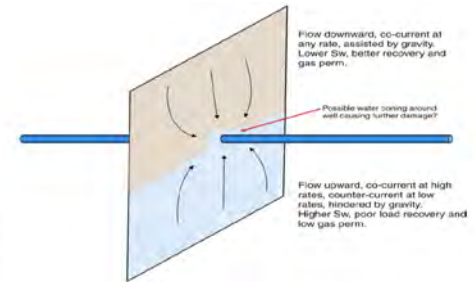


- Reduce water use, water-blocking, increase water recovery.
- Increase hydrocarbon production

Earlier models for gravity drainage effects in unconventional reservoirs

Taylor et al., Canadian Soc. Unconventional Gas, 2011

CSUG/SPE 148680



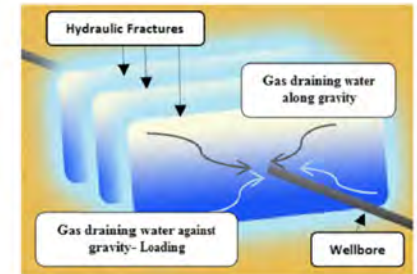
Why Not to Base Economic Evaluations on Initial Production Alone

Robert S. Taylor, SPE, Halliburton; Robert Barree, Barree and Associates; Roberto Aguilera, University of Calgary; Ottmar Hoch, Hoch & Associates Inc.; Ken Storozhenko, KJS & Associates Ltd.

Agrawal & Sharma, J. Unconventional Oil & Gas Res., 2015

Practical insights into liquid loading within hydraulic fractures and potential unconventional gas reservoir optimization strategies

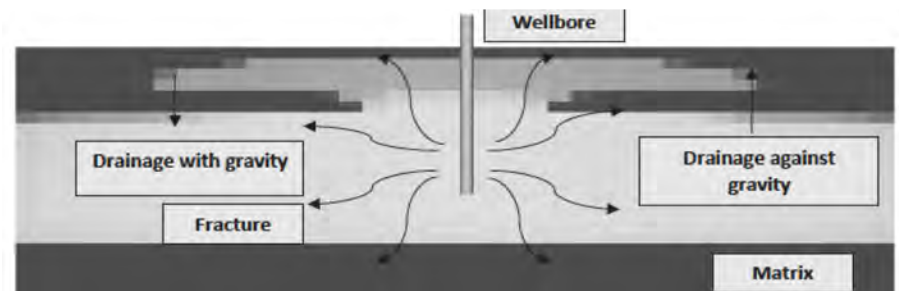
Samarth Agrawal*, Mukul M. Sharma



Sarkar et al., APPEA J., 2016, 369

A Cooper Basin simulation study of flow-back after hydraulic fracturing in tight gas wells

S. Sarkar, M. Haghighi, M. Sayyafzadeh, D. Cooke, K. Pokalai and F. Mohamed Ali Sahib



Accomplishments to Date

- Comprehensive analyses of shale-water retention relations, including hysteresis.
- Quantification of diffusion-limited equilibration in shale, including anisotropy.
- Working on improving conceptual models for immiscible fluids in reservoirs (gravity, hysteresis, wettability).
- Alternative, low-water, high-pressure foams being tested.
- 4 peer-reviewed journal papers have been published this past year, and others are in progress.

Lessons Learned

- Research gaps/challenges.
 - Experimental basis for reliably predicting immiscible fluid displacements over a wide range of matrix permeabilities and wettabilities.
- Unanticipated research difficulties.
 - Fragile core samples
 - Questionable ability to re-establish in-situ conditions for experiments on cores recovered from deep reservoirs
- Changes that should be made next time.
 - Experiments on other more competent porous media (geologic and synthetic) spanning the desired wide ranges in permeability and porosity needed to develop reliable scaling predictions for multiphase flow in tight rocks.

Synergy Opportunities

- Pursued novel low-water hydraulic fracturing foams in the context of enhanced geothermal systems through a collaboration proposal with Keith Johnson and Masa Prodanovic (UT Austin).
- Development of alternative low water content stimulation fluids were pursued through industry collaborations (Liang Xu, Multi-Chem, Halliburton).
- We are open to developing collaborations with other groups interested in multiphase flow in shales, particularly at complementary scales.

Project Summary

- Comprehensive analyses of shale-water retention relations.
- Quantification of diffusion-limited equilibration in shale.
- Improvement of conceptual models (gravity, hysteresis, wettability).
- Characterizing alternative, low-water, high-pressure foams.
- 4 publications in in the past year, and others in progress.

Next Steps

- Immiscible fluids equilibria and flow measurements in other geologic and synthetic media to develop the needed basis for more reliable hydraulic scaling.
- Experiments on fracture flow dynamics to develop predictive capabilities for gravity drainage of frac fluids.

Acknowledgments

- NETL: Stephen Henry, Jared Ciferno, Dustin Crandall, Robert Vagnetti, and Jonathan Moore
- Oklahoma Geological Survey, Brian Cardott: (Woodford Shale)
- MSEEL (Marcellus Shale)

Appendix

- These slides will not be discussed during the presentation, but are mandatory.

Benefit to the Program

- Gain understanding of water in unconventional reservoir stimulation through studies of water imbibition, redistribution, and gas counter-flow.
- Reduction in water use must be based on understanding of water dynamics in shale matrix pores and fractures.

Project benefits statement.

This research project is developing basic understanding of water partitioning in hydraulically fractured reservoirs, and of CO₂ foams in order to reduce water use and enhance hydrocarbon recovery.

Project Overview

Goals and Objectives:

- Understand the coupling between water imbibition and gas counter-current gas flow in shales in order to help identify approaches to improving production.
- Understand the impact of gravity drainage of water in hydraulic fractures on counter-current gas flow.
- Understand effectiveness of non-water fracturing fluids on shale gas/oil mobilization, and improve formulas of fracturing fluids.

Organization Chart

- Tetsu K. Tokunaga
 - Immiscible fluid phase equilibrium and flow.
- Jiamin Wan
 - Surface chemistry, wettability, low-water foams.
- Abdullah Cihan
 - Pore- to core-scale modeling of immiscible fluids.
- Yingqi Zhang, Stefan Finsterle
 - Continuum modeling of fracture-matrix systems
- Ken Tokunaga
 - Engineering
- Lu Wang
 - Graduate student assistant

Gantt Chart

Tasks	Year 1				Year 2			
	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4
start date	10/1/16	1/1/17	4/1/17	7/1/17	10/1/17	1/1/18	4/1/18	7/1/18
end date	12/31/16	3/31/17	6/30/17	9/30/17	12/31/17	3/31/18	6/30/18	9/30/18
1. Project Mgmt. Plan								M
2. Shale Properties, equilibrium and flow experiments								
2A. Shale water saturation vs capillary pressure	m	M						
2B. Water imbibition into shales				m		M		
2C. Gas permeabilities at specific saturations						M		
2D. Countercurrent water uptake and gas flow							m	M
3. Non-water fracking fluids experiments								
3A. Micromodel design & construction	m							
3B. Optimize experimental setup and procedures			m		M			
3C. Compare water-based vs CO2-based fluids						m		M
3D. Test other non-water fracturing fluids							m	M
4. Modeling								
4A.1. Multicomponent diffusion-adsorption model				M				
4A.1. Improved hysteretic two-phase flow models								M
4B. Model fluids at fracture-matrix interfaces								M

Bibliography

Peer-reviewed journal publications during this past year

- Tokunaga, T.K., Shen, W., Wan, J., Kim, Y., Cihan, A., Zhang, Y., and Finsterle, S., 2017, Water saturation relations and their diffusion-limited equilibration in gas shale: Implications for gas flow in unconventional reservoirs. *Water Resources Research*, v 53, p. 9757-9770, <https://doi.org/10.1002/2017WR021153>.
- Wan, J., Tokunaga, T.K., Dong, W., and Kim, Y., 2017, Extracting natural biosurfactants from humus deposits for subsurface engineering applications. *Energy & Fuels*, v. 31, p. 11902-11910, doi: 10.1021/acs.energyfuels.7b02203.
- Wang, L., Wan, J., Tokunaga, T.K., Kim, Y., Yu, Q., 2018, Experimental and modeling study of methane adsorption onto partially saturated shales. *Water Resources Research*, v 54, p. (in press), <https://doi.org/10.1029/2017WR020826>.
- Shen, W., Zheng, L., Oldenburg, C.M., Cihan, A., Wan, J., and Tokunaga, T.K., 2018, Methane diffusion and adsorption in shale rocks: A numerical study using the Dusty Gas Module in TOUGH2/EOS7C-ECBM. *Transport in Porous Media*, 123, p. 521-531.

Upcoming presentations at conferences

- Zhang, Y., Finsterle, S., Tokunaga, T.K., Wan, J., Cihan, A., Tokunaga, K., 2018, Impact of gravity segregation on gas production from tight shales. TOUGH Symposium 2018. October 8-10, 2018, Lawrence Berkeley National Laboratory, Berkeley, CA.
- Tokunaga, T.K., 2018, Gravitational and Matric/Capillary Potential Controls on Distributing Unsaturated Flow of Water in Fracture Rocks and Talus. American Geophysical Union Fall Meeting, December 2018, Washington D.C.