A Multi-Scale Experimental Investigation of Flow Properties in Coarse-Grained Hydrate Reservoirs During Production

DE-FE0028967

Steve Phillips

University of Texas at Austin

PI: Peter Flemings Co-PIs: Hugh Daigle, David DiCarlo, Nicolas Espinoza, Nicola Tisato, Afu Lin

U.S. Department of Energy

National Energy Technology Laboratory

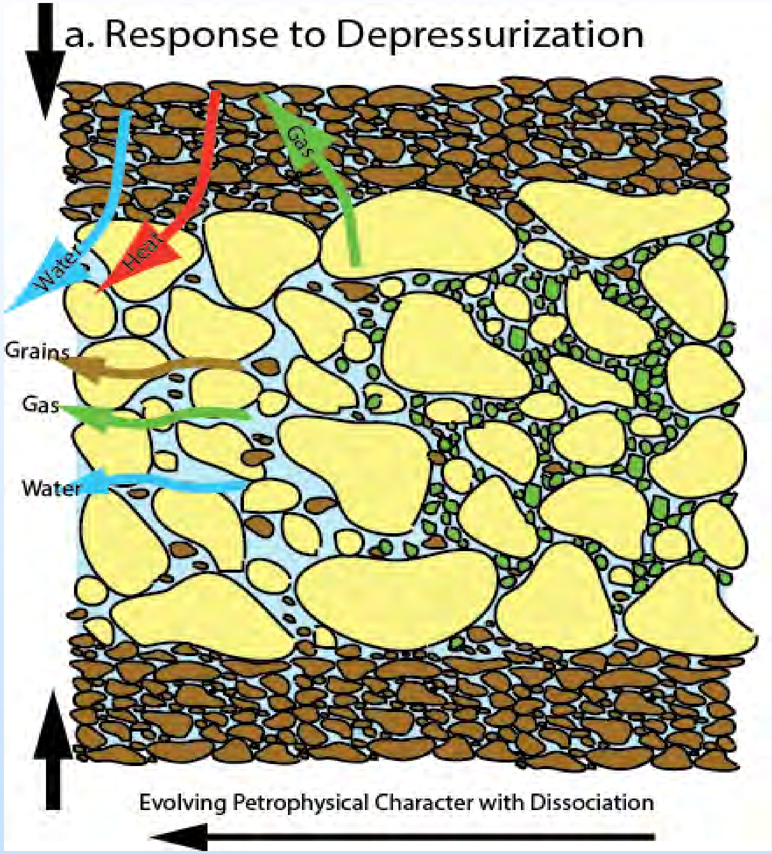
Mastering the Subsurface Through Technology Innovation, Partnerships and Collaboration:
Carbon Storage and Oil and Natural Gas Technologies Review Meeting

August 13-16, 2018

Presentation Outline

- Project Goals and Summary of Approach
- Technical Status
  - Relative permeability
  - Depressurization
  - Micro-CT
  - Micro-Raman
- Accomplishments to Date and Lessons Learned
- Synergy Opportunities
- Summary

Project Goals



Boswell and Collett, 2016

- Controls on relative permeability and transport processes of coarse-grained hydrate reservoirs
- The response of these reservoirs to depressurization at the macro-(1 m) and micro-(1×10^{-6} m) scale.

Summary of Approach

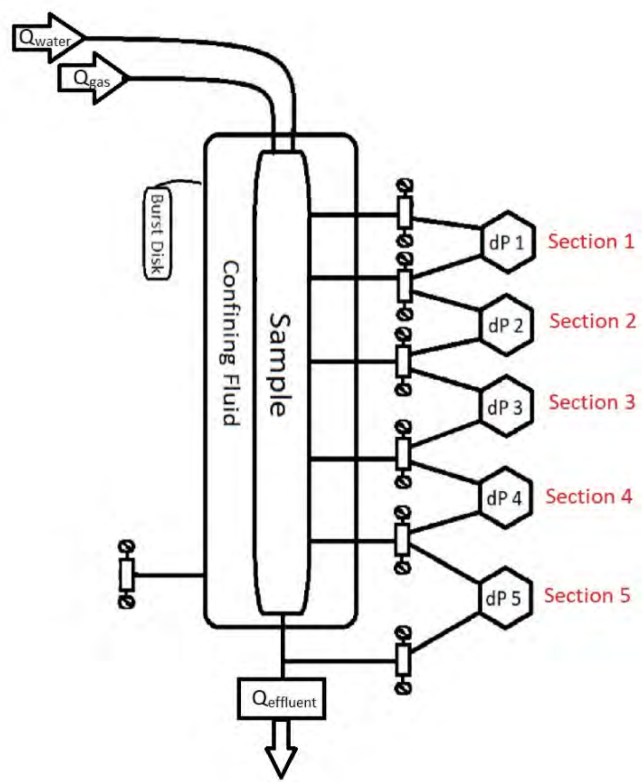
Macro-Scale	Micro-Scale
<u>Relative Permeability</u> Measurement of gas/brine relative permeabilities in the presence of hydrate	**<u>Micro-CT experiments</u>** Image hydrate habit and evolution
<u>Depressurization Tests</u> Performed controlled depressurization at range of hydrate saturations and monitor pressure response to shut-in periods	**<u>Micro-Raman experiments</u>** Micro-scale phase distribution, and local diffusion associated with perturbation

Analysis of both synthetic and natural hydrate samples

Technical Status

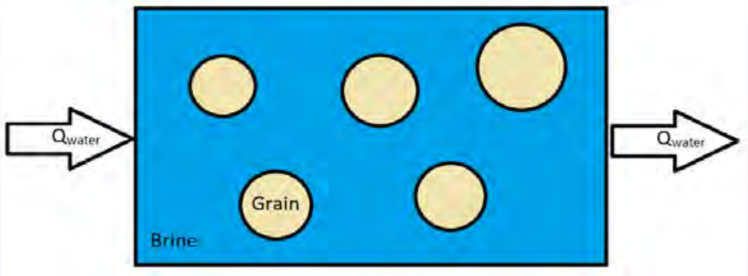
Relative permeability in hydrate-bearing sand

PIs: Hugh Daigle, David DiCarlo
Graduate students: Zach Murphy, David Fukuyama



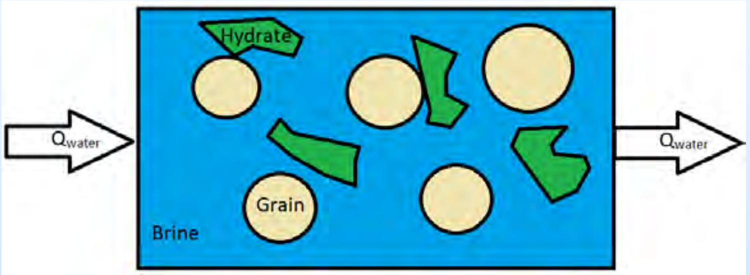
- Hydrate formation using the excess gas method in sandpack and sandstone samples
- First relative three-phase permeability measurements using the steady state method

Relative permeability in hydrate-bearing sand



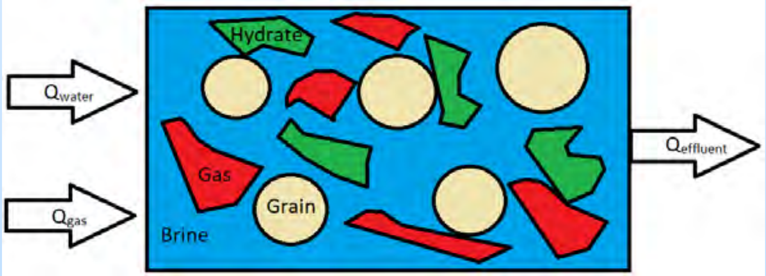
- Φ=0.20
- S_{water}=1.00
- $k_{absolute}$=6 darcy

- Successfully measured absolute permeability with no hydrate present



- S_{water}= 0.75
- $S_{hydrate}$=0.25
- $K_{effective}$=2.5 darcy

- Successfully measured two phase relative permeability (hydrate+brine)



- S_{water}=0.0-0.75
- S_{gas}=0.0-0.75
- $S_{hydrate}$=0.25

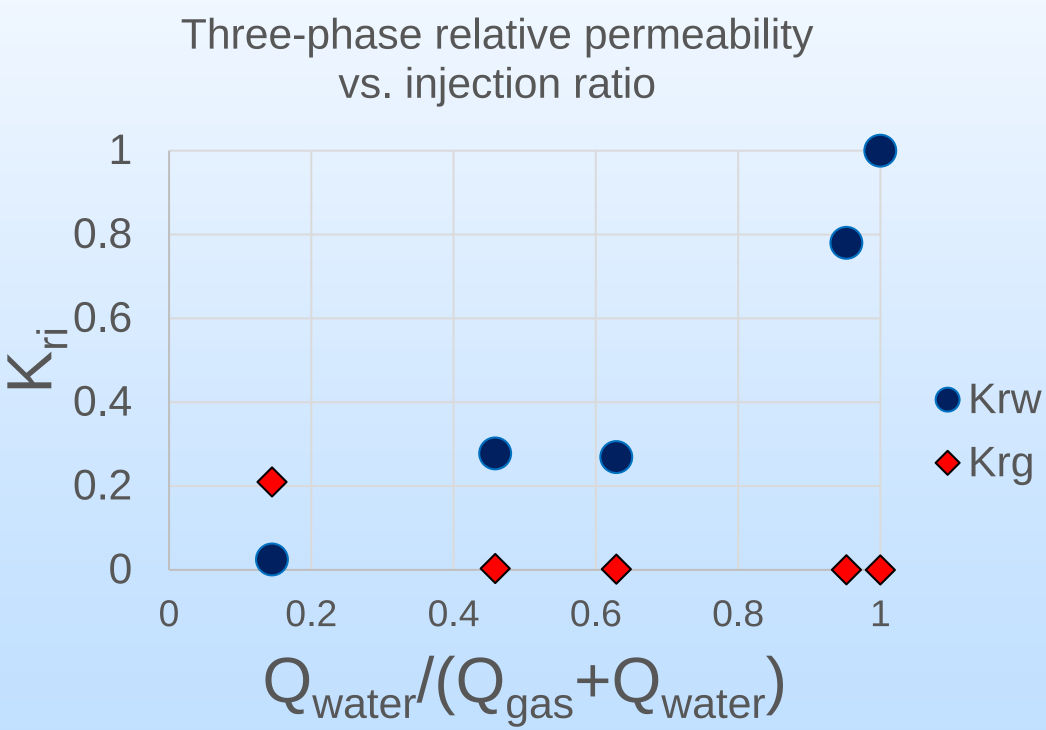
- Currently measuring three phase relative permeability (hydrate+brine+gas)

Hydrate-Brine Permeability

Section	Absolute Permeability, Darcy (S_h=0) (S_w=1.00)	Effective Permeability, Darcy (S_h=0.25) (S_w=0.75)	Relative Permeability (S_h=0.25) (S_w=0.75) (S_g=0.00)
1	4.7	3.0	0.64
2	3.9	2.3	0.58
3	5.5	2.4	0.43
4	4.8	2.2	0.46
5	4.5	0.53	0.12

- Endpoint measurements of the water relative permeability values for S_h=0.25
- No gas present in sample

Three-phase relative permeability

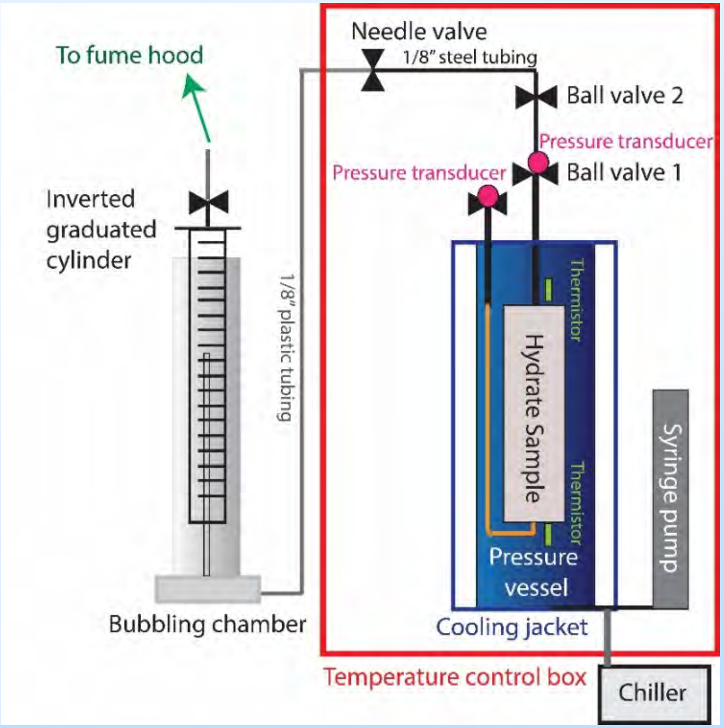


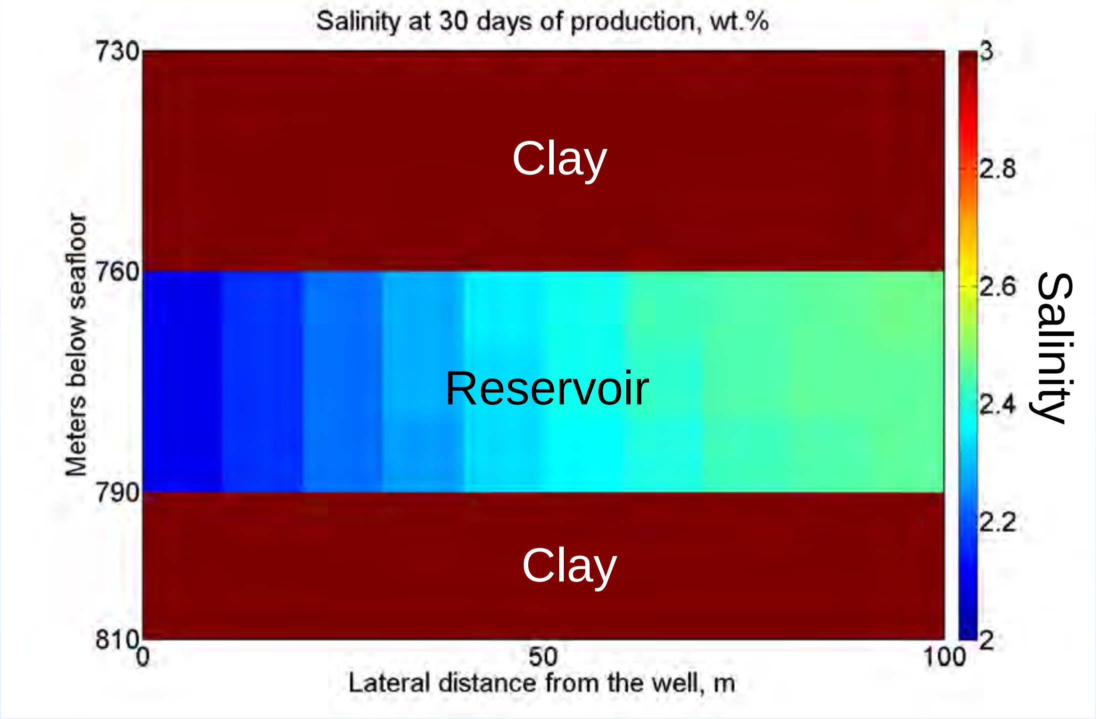
- We have successfully co-injected methane and brine
- Relative permeabilities for multiple injection ratios (Q_w/Q_{gas}) are presented
- We cannot yet measure phase saturations
  - We are working to CT scan experiments

Technical Status

Core-scale depressurization

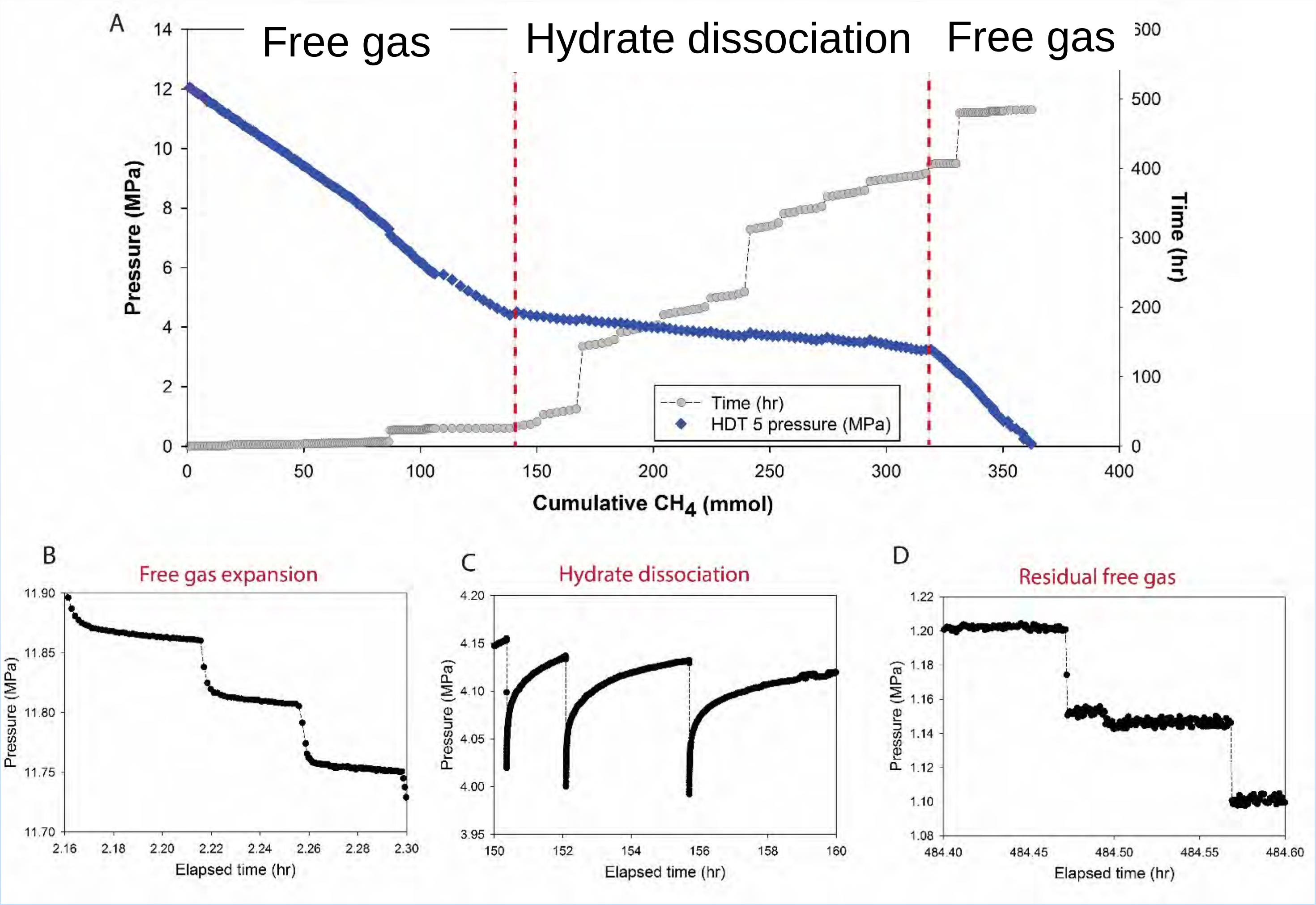
PI: Peter Flemings
Postdoc: Steve Phillips





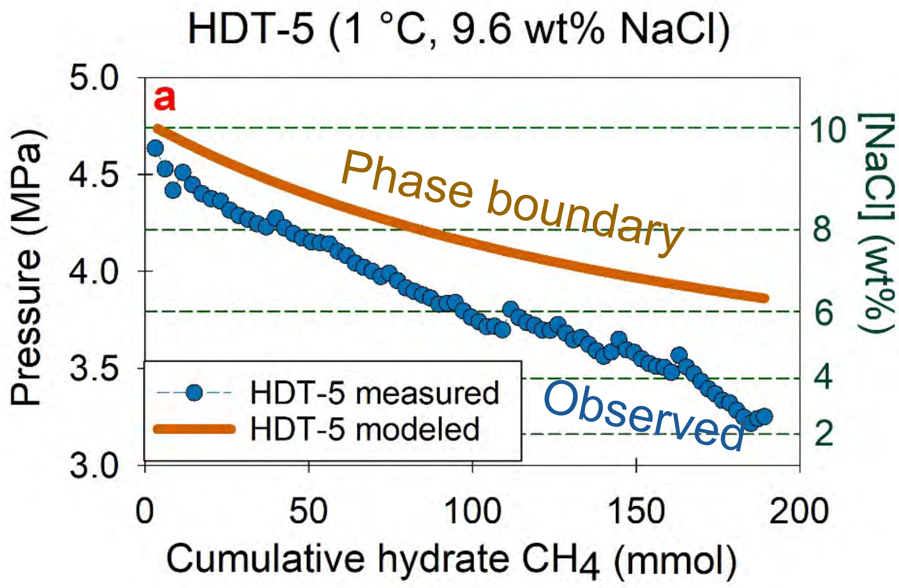
How does the salinity of a sample evolve during production by depressurization?

3 stages of depressurization



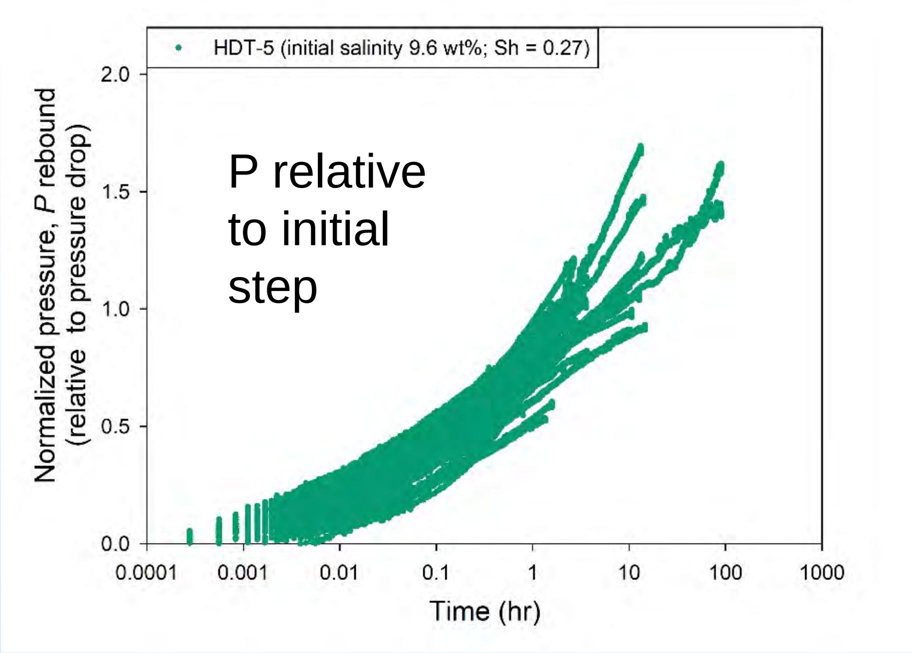
Synthetic hydrate in sand

Effect of local pore freshening and cooling

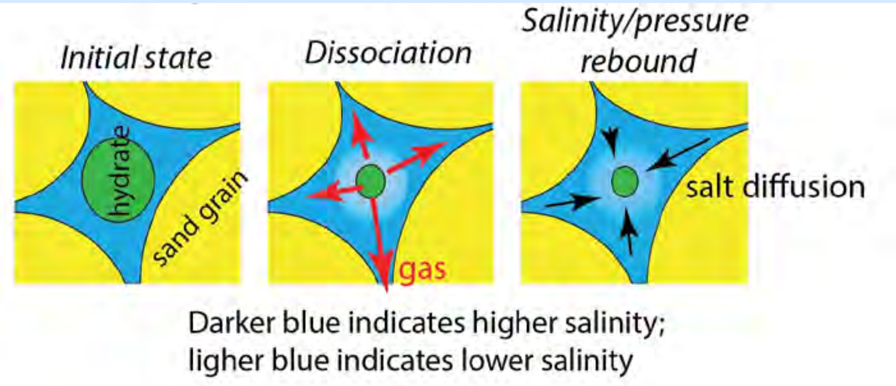


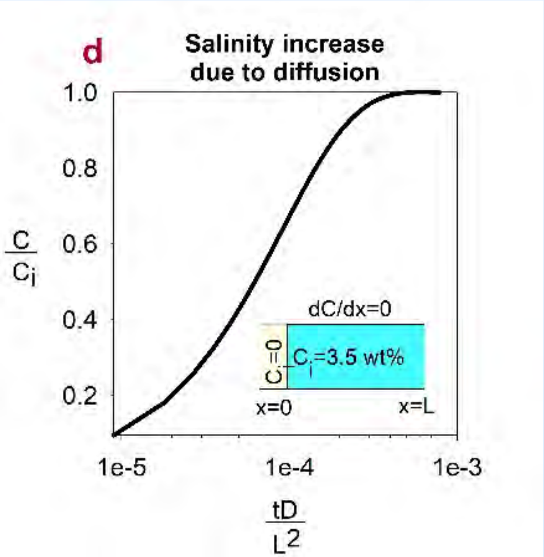
- Pressure decline more rapid than if salinity and temperature were homogenous in sample
- Heat transfer occurring more rapidly than salt diffusion
- Fast depressurization moves sample to freshwater phase boundary, but slow recovery towards bulk equilibrium

Pressure rebound behavior



- P rebounds more rapid in lower salinity
- Rebound shape evolves to more concave up over shut-in periods
- Suggests salt diffusion limits recovery
- Pressure rebound evolves over dissociation





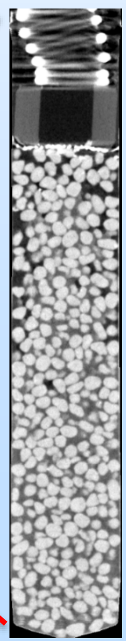
Ongoing work: interpretation of CT scans during dissociation, P rebounds on natural GoM samples

Technical Status

Micro X-ray CT analysis

Micro Consolidation Device
(X-ray axial CT slice)

Cooled Pressure Vessel



Stainless steel spring

Teflon spacer

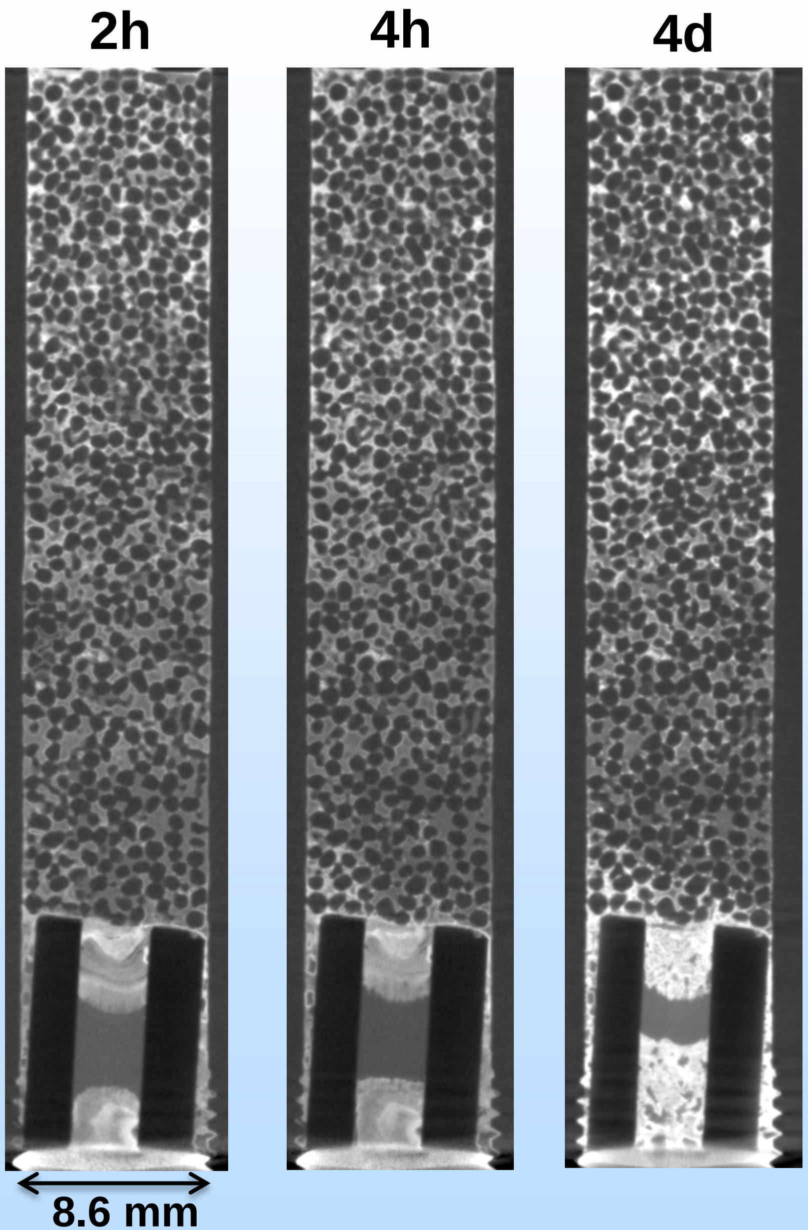
Sand Partially Saturated With 0.6 wt% NaBr Brine

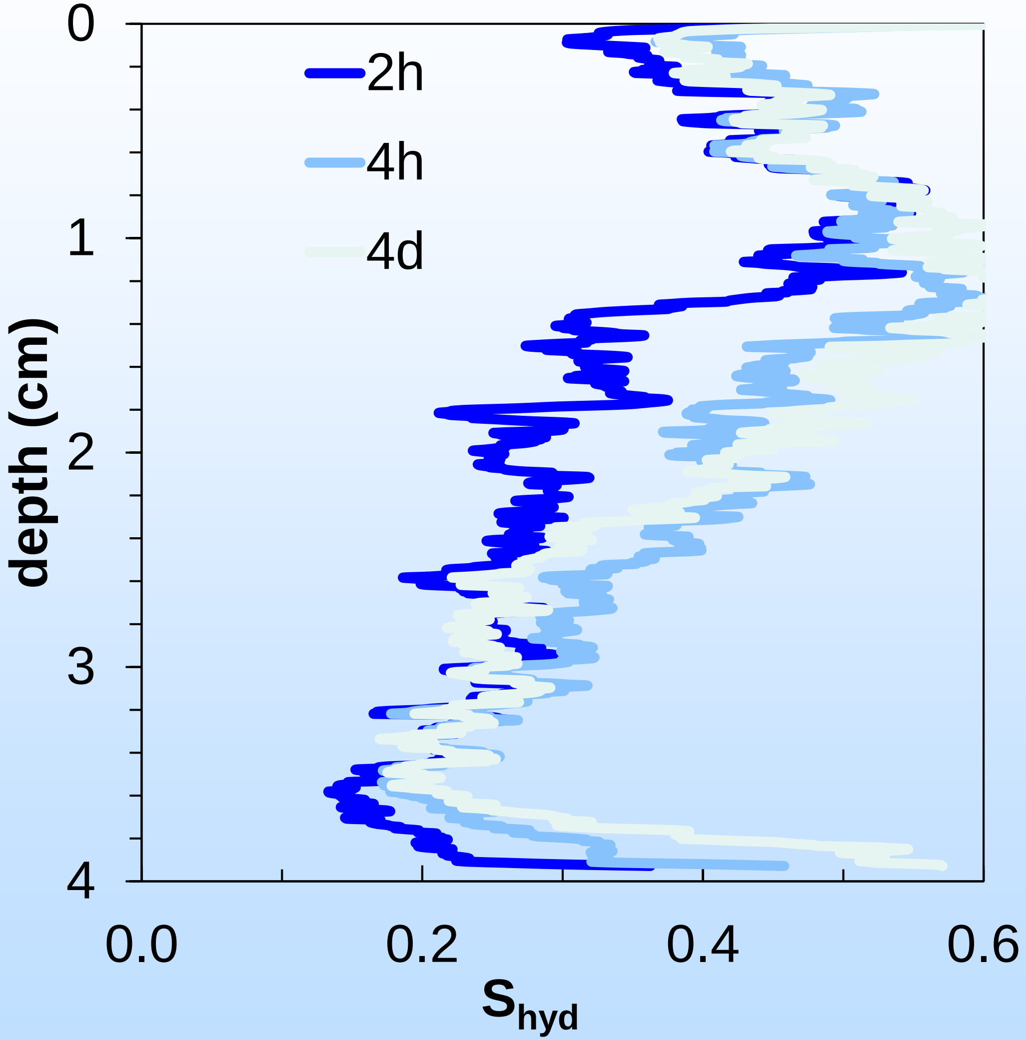
4 cm

PIs: Nicolas Espinoza, Nicola Tisato

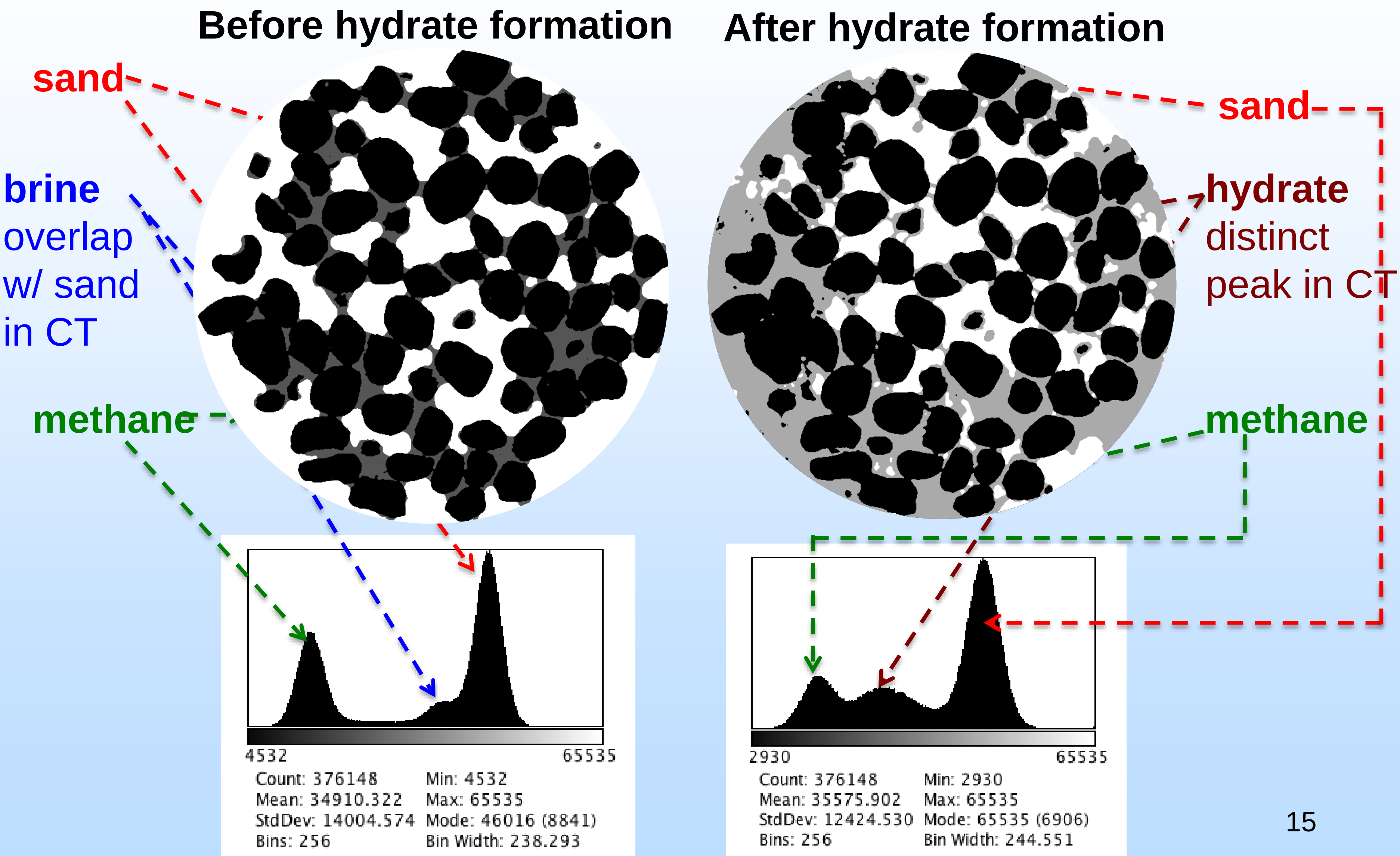
Postdoc: Xiongyu Chen

Hydrate formation w/ Xe: Patchy Hydrate Saturation



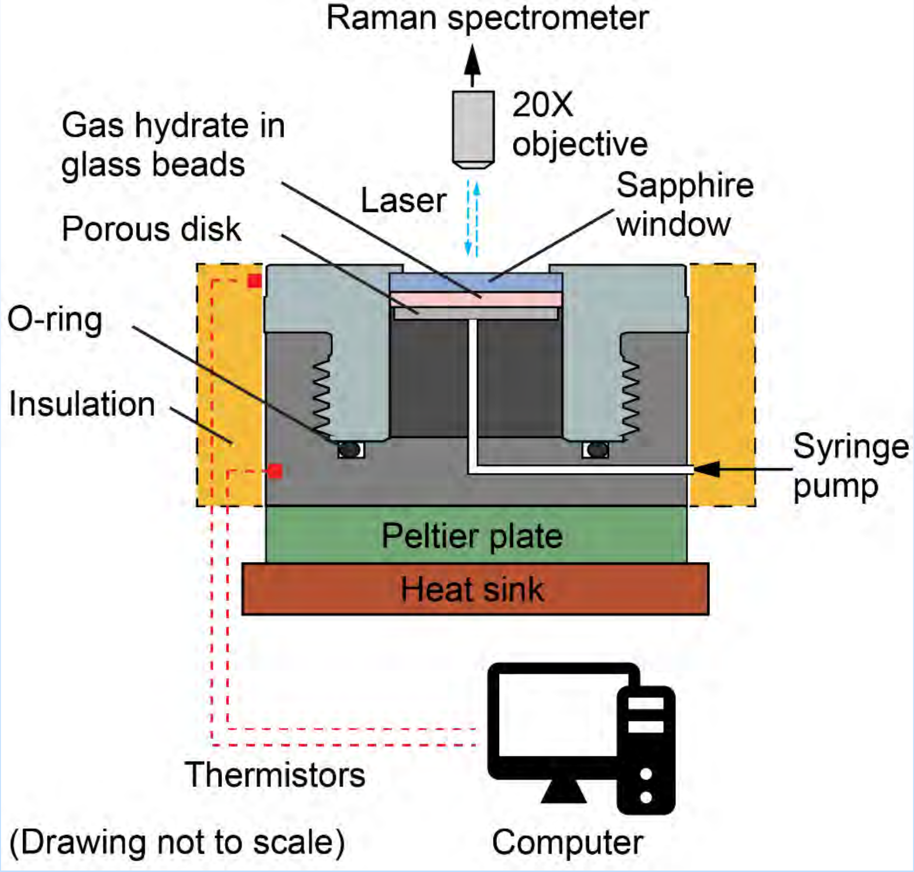


Methane Hydrate Experiment
-- Sharp μCT contrast for brine (KI) vs. hydrate



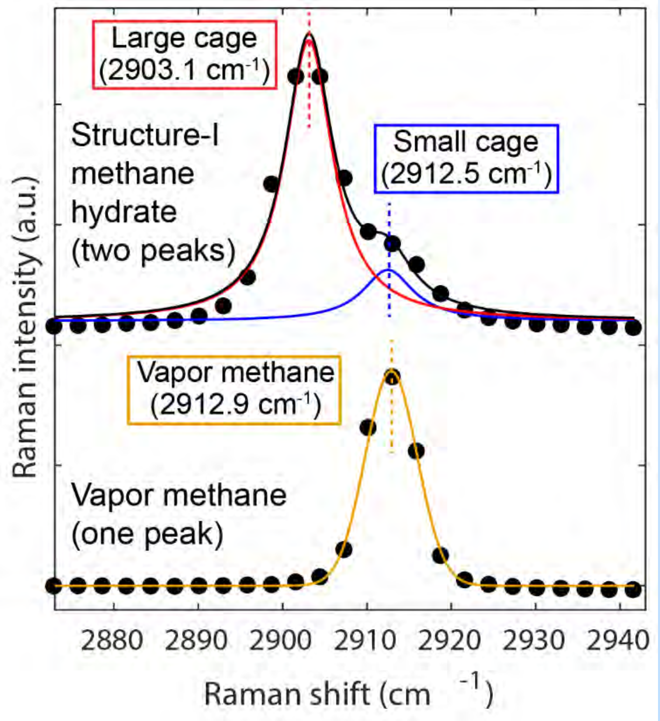
Technical Status

Micro Raman analysis



PI: Afu Lin

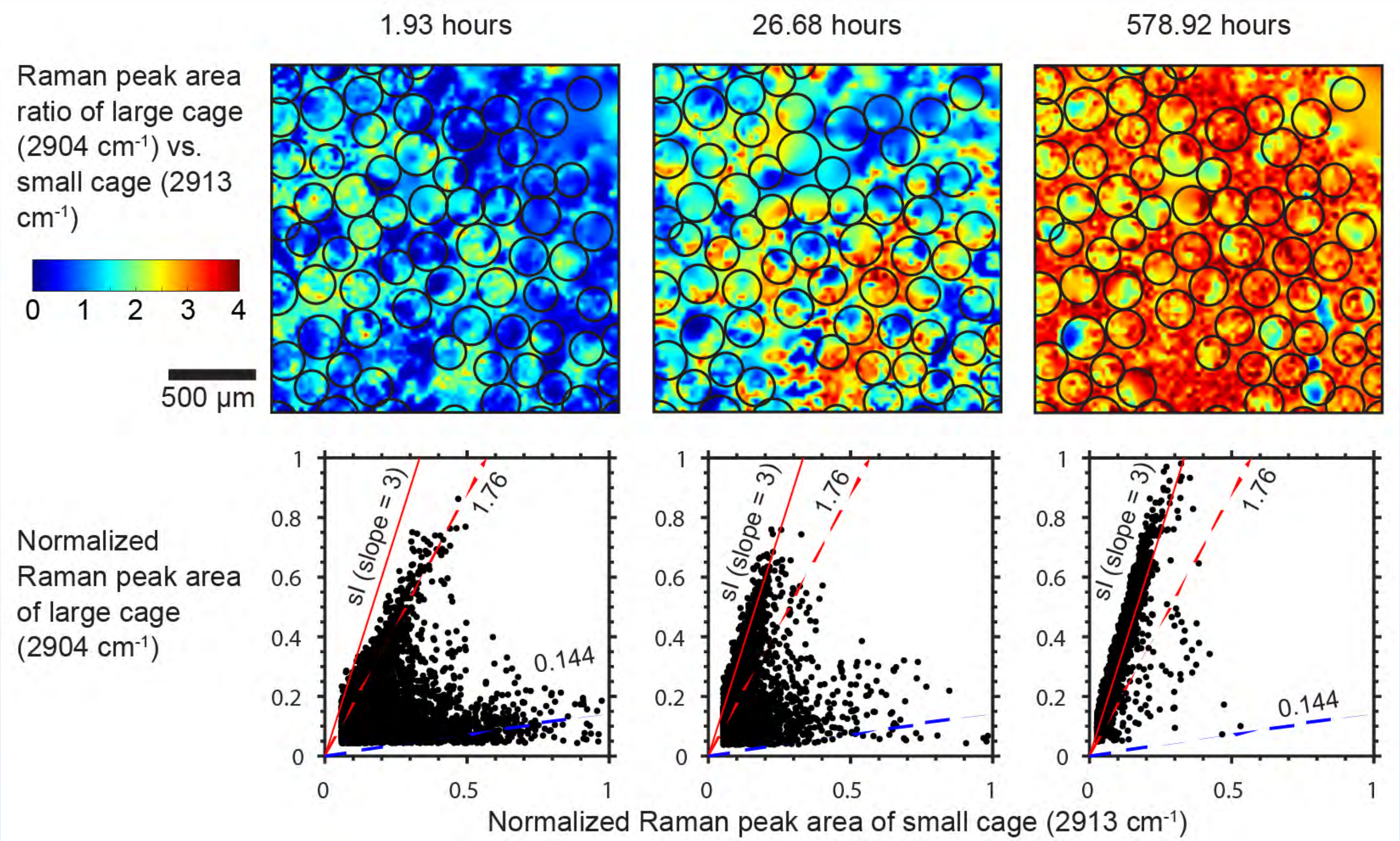
Graduate student: Tiannong Dong



Gas-hydrate-bearing sample dimension:
1.52 mm in thickness, 21.36 mm in diameter

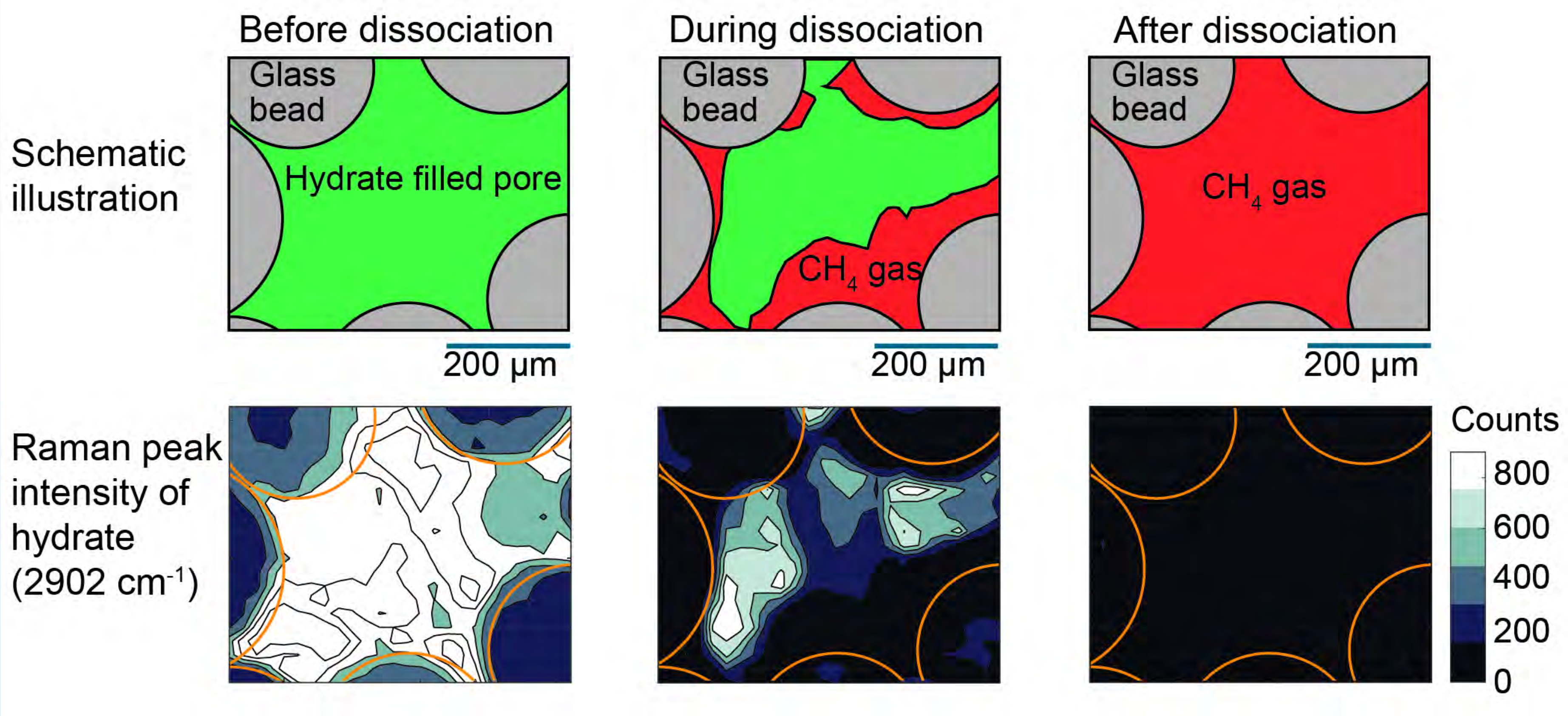
Methane Hydrates Formation: Spatial and Temporal Evolution

15 MPa
6 °C



- Black circles are glass beads (porous media)
- Structure-I hydrate (large cage/small cage = 3) is the thermodynamically stable hydrate phase
- Orange/red color (ratio of 3) indicates structure-I hydrate

Pore-scale hydrate dissociation using Raman spectroscopy



Hydrate dissociates from the porous media surface first, and then progresses into the pore center

Accomplishments to Date

- Design and building of micro-Raman and micro-CT cells
- Design and building of relative permeability core holder
- Achieved hydrate formation up to 45% saturation and maintain mass balance during depressurization
- Measurement of water permeability in hydrate sample
- Imaging of methane hydrate in KI solution
- Spatial and temporal mapping of Raman spectra of hydrate, water and gas
- Measurement of evolving pressure rebounds during dissociation at varying salinity

Lessons Learned

- Hydrate formation/blockage in core and tubing
- Heterogeneous distribution of hydrate in core
- Difficulty in measuring methane hydrate via CT

Synergy Opportunities

- Collaboration with the UT GOM^2 project to work with pressurized cores and run experiments with depressurized GoM sediments
- Collaboration with Yongkoo Seol and Liang Lei at NETL by sharing data and expertise in methane hydrate formation and imaging with micro-CT.
- Discussions with Tim Kneafsey from LBNL

Project Summary

Macro scale key findings

- Two phase (brine+hydrate) effective permeability has been successfully measured for hydrate bearing cores
- We document pressure rebound behavior and deviation from bulk equilibrium conditions during dissociation indicating influence of salinity on the phase boundary

Accomplishments to Date

Micro scale key findings

- Patchy hydrate distribution during Xe hydrate formation in sandy sediments due to Ostwald ripening
- Methane hydrate displays complex pore-scale morphology in sand and can be well segmented from KI brine in μCT images
- Raman peak ratios indicate that during hydrate formation the ratio of large to small cage hydrae increases until it is nearly all Structure I hydrate after several weeks
- Hydrate dissociation begins near the grain surface and then progresses into the pore interior

Project Summary

– **Next Steps:**
- Dissociation behavior of methane hydrate in CT
- Raman measurement of natural hydrate samples from GoM before and during dissociation
- Relative permeability of gas and water in methane hydrate-bearing sand
- Interpretation of pressure rebound behavior across range of hydrate saturations, formation methods, and in natural samples
- Integration of pore scale and core scale observations- how does pore habit influence relative permeability and pressure recovery behavior?

Appendix

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

Benefit to the Program

- Identify the program goals being addressed.

- Benefits statement
  - Methane hydrates within sand-rich marine reservoirs represent a potentially enormous reservoir for methane. Previous drilling/logging in marine sand reservoirs within the Gulf of Mexico has verified that methane hydrate filled sand reservoirs are present and that sand reservoirs can be identified from seismic analysis .DOE is now focusing on acquiring intact samples through its project 'Genesis of Methane Hydrate in Coarse-Grained Systems: Northern Gulf of Mexico Slope, DOE Award No.: DE-FE0023919'. We hope that the first conventional and pressurized cores of these reservoirs will be collected in spring 2017.
  - As pointed out by (Moridis et al., 2009), laboratory studies to determine the effect of solid phases (hydrate) on relative permeability are of the highest importance because this behavior has a large impact on gas recovery in hydrate bearing systems. Current modeling approaches are limited to relying on theoretical extensions of conventional multi-phase flow models. It is vital now to go beyond these limitations and pursue an experimental program that will illuminate at the core and the pore scale the effect of methane hydrate on gas flow behavior and the process of hydrate dissociation due to perturbation. A successful testing program grounded in sand-pack experiments and then extended to the analysis of intact cores is the appropriate way to gain this understanding. The learnings that result will provide a significant step forward in our ability to simulate hydrate production and make realistic estimates of the ability of the methane hydrate resource to be a viable energy source.

Project Overview

Goals and Objectives

The goals of this project are to provide a systematic understanding of permeability, relative permeability and dissipation behavior in coarse-grained methane hydrate - sediment reservoirs. The results will inform reservoir simulation efforts, which will be critical to determining the viability of the coarse-grained hydrate reservoir as an energy resource. We will perform our investigation at the macro- (core) and micro- (pore) scale.

At the macro- (core) scale, we will: 1) measure the relative permeability of the hydrate reservoir to gas and water flow in the presence of hydrate at various pore saturations; and 2) depressurize the hydrate reservoir at a range of initial saturations to observe mass transport and at what time scale local equilibrium describes disassociation behavior. Simultaneously, at the micro (pore) scale, we will 1) use micro-CT to observe the habit of the hydrate, gas, and water phases within the pore space at a range of initial saturations and then image the evolution of these habits during dissociation, and 2) use optical micro-Raman Spectroscopy to images phases and molecules/salinity present both at initial saturations and at stages of dissociation. We will use our micro-scale observations to inform our macro-scale observations of relative permeability and dissipation behavior.

In Phase 1, we first demonstrated our ability to systematically manufacture sand-pack hydrate samples at a range of hydrate saturations. We then measured the permeability of the hydrate-saturated sand pack to flow a single brine phase and depressurized the hydrate-saturated sand packs and observed the kinetic (time-dependent) behavior. Simultaneously we built a micro-CT pressure container and a micro-Raman Spectroscopy chamber and imaged the pore-scale habit, phases, and pore fluid chemistry of sand-pack hydrate samples. We then made observations on our hydrate-saturated sand-packs.

In Phase 2, we will measure relative permeability to water and gas in the presence of hydrate in sand-packs using co-injection of water and gas. We will also extend our measurements from sand-pack models of hydrate to observations of actual Gulf of Mexico material. We will also measure relative permeability in intact samples to be recovered from the upcoming Gulf of Mexico 2017 hydrate coring expedition. We will also perform dissipation experiments on intact Gulf of Mexico pressure cores. At the micro-scale we will perform micro-Raman and micro-Ct imaging on hydrate samples composed from Gulf of Mexico sediment.

Project Overview

Goals and Objectives – Success Criteria

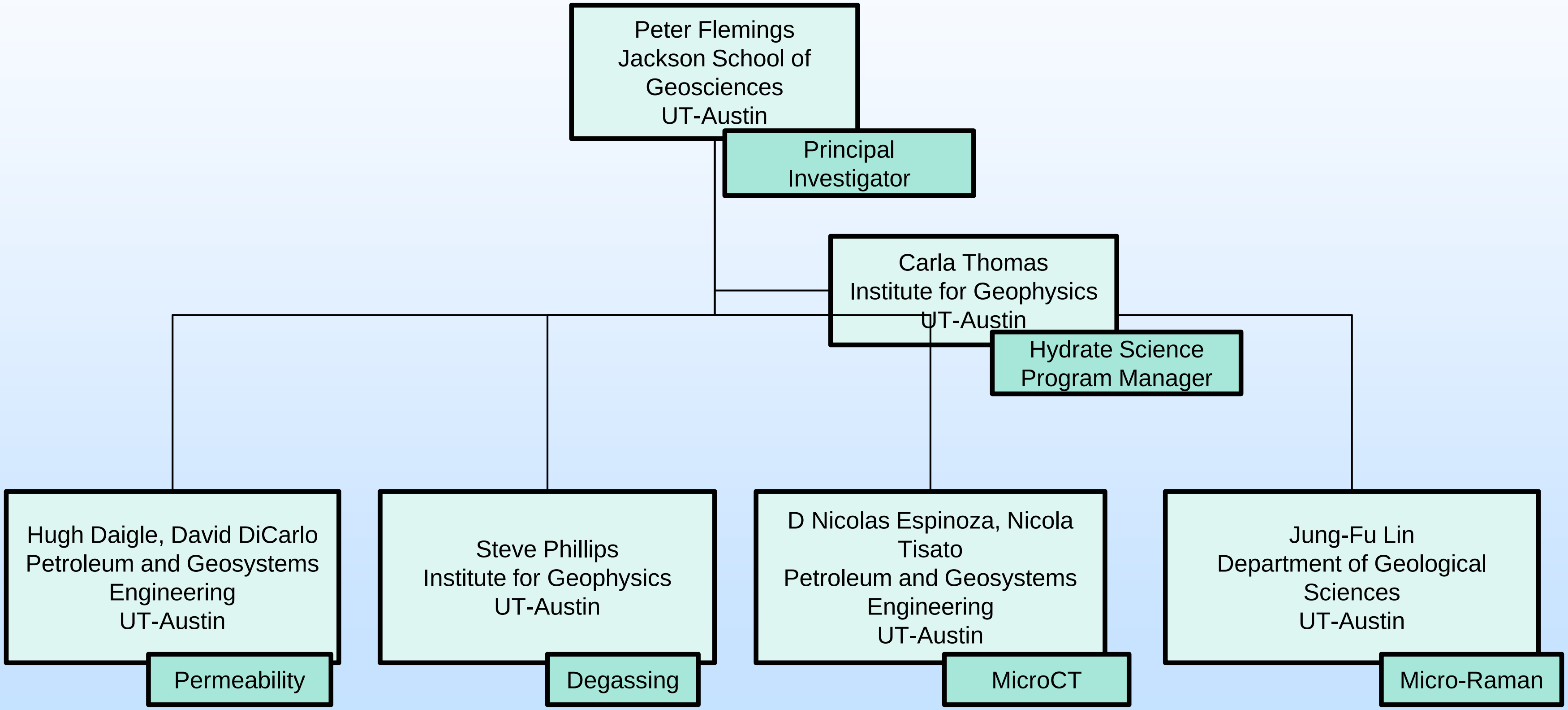
Milestone Description	Planned Completion	Actual Completion	Verification Method	Comments
Milestone 1.A: Project Kick-off Meeting	11/22/2016 (Y1Q1)	11/22/16	Presentation	Complete
Milestone 1.B: Achieve hydrate formation in sand-pack	6/27/2017 (Y1Q3)	8/11/17	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 2.1)	Complete, *Documentation in the Y1Q3 quarterly and Phase 1 report*
Milestone 1.C: Controlled and measured hydrate saturation using different methods	3/27/2018 (Y2Q2)	3/27/18	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 2.1)	Complete, *Documentation in Y2Q2 quarterly and Phase 1 report*
3 Milestone 1.D: Achieved depressurization and demonstrated mass balance	3/27/2018 (Y2Q2)	12/18/2017	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 3.1)	Complete, *Documentation in the Y2Q1 quarterly and Phase 1 report*
Milestone 1.E: Built and tested micro-consolidation device	6/27/2017 (Y1Q3)	6/27/2017	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 4.1)	Complete, *Documentation in Y1Q3 quarterly and Phase 1 report*
Milestone 1.F: Achieved Hydrate formation and measurements in Micro-CT consolidation device	3/27/2018 (Y2Q2)	2/15/18	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 4.1)	Complete, *Documentation in Y2Q2 quarterly and Phase 1 report*
Milestone 1.G: Built and integrated high-pressure gas mixing chamber	3/27/2018 (Y2Q2)	6/27/17	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 5.1)	Complete, *Documentation in Y1Q3 quarterly and Phase 1 report*
Milestone 1.H: Micro-Raman analysis of synthetic complex methane hydrate	3/28/2018 (Y2Q2)	3/27/18	Documentation of milestone achievement within required project reporting / deliverables (Deliverable 5.1)	Complete, *Documentation in Y2Q2 quarterly and Phase 1 report*

Project Overview

Goals and Objectives – Success Criteria, conintued

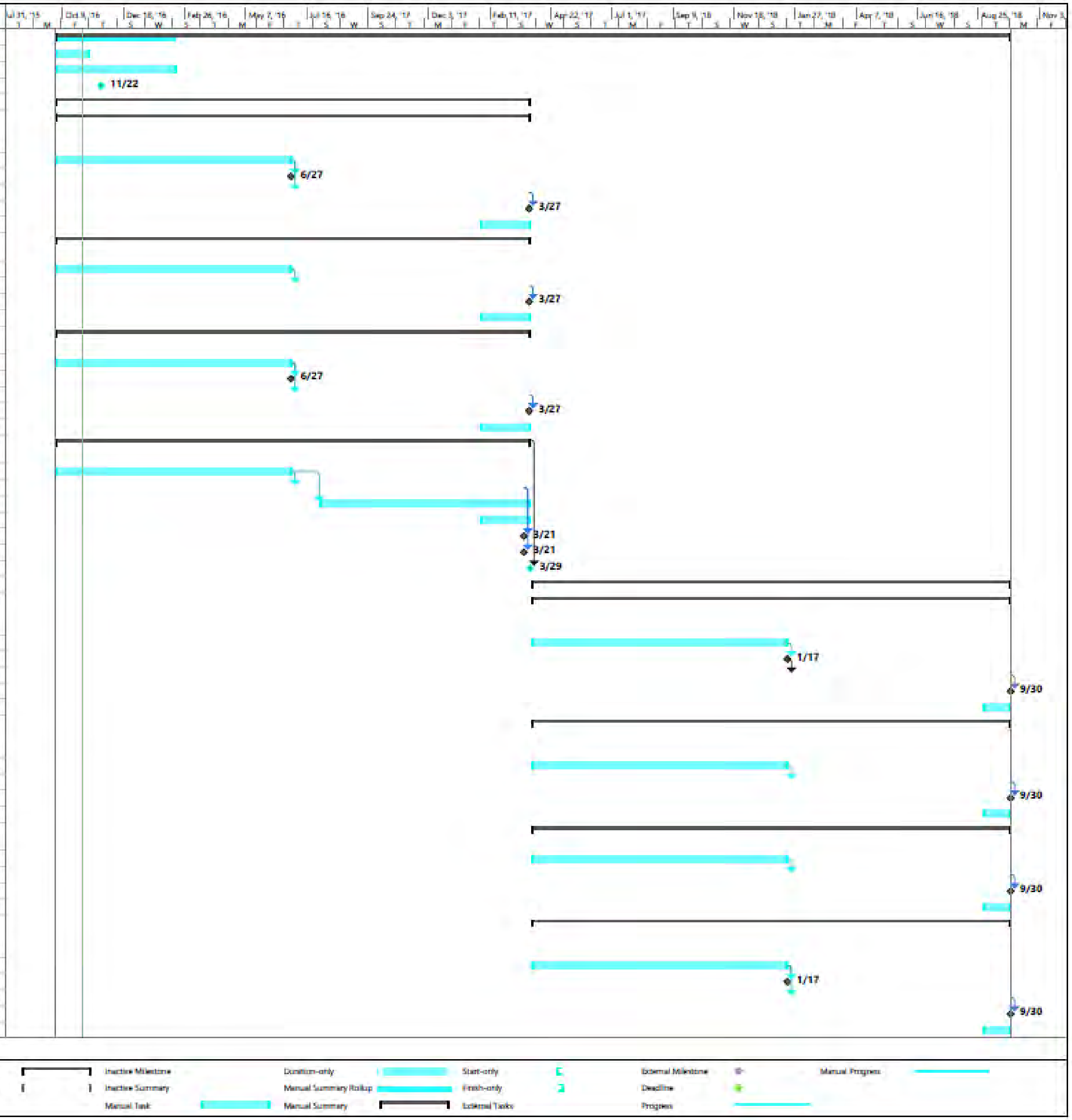
Milestone Description	Planned Completion	Actual Completion	Verification Method	Comments
Milestone 2.A - Measurement of relative permeability in sand-pack cores. (See Subtask 6.1)	1/17/2019 (Y3Q2)		Documentation of milestone achievement within required project reporting / deliverables (Deliverable 6.1)	In progress
Milestone 2.B - Measurement of relative permeability in intact pressure cores. (See Subtask 6.2)	9/30/2019 (Y3Q4)		Documentation of milestone achievement within required project reporting / deliverables (Deliverable 6.1)	
Milestone 2.C -Depressurization of intact hydrate samples and documentation of thermodynamic behavior. (See Subtask 7.1 and 7.2)	9/30/2019 (Y3Q4)		Documentation of milestone achievement within required project reporting / deliverables (Deliverable 7.1)	In progress
Milestone 2.D - Achieved gas production from GOM^2 samples monitored by micro-CT. (See Subtask 8.1 and 8.2)	9/30/2019 (Y3Q4)		Documentation of milestone achievement within required project reporting / deliverables Report (Deliverable 8.1)	In progress
Milestone 2.E - Building a chamber to prepare natural samples for 2D-3D micro-Raman analysis; (See Subtask 9.1 and 9.2)	1/17/2019 (Y3Q2)		Documentation of milestone achievement within required project reporting / deliverables (Deliverable 9.1)	In progress
Milestone 2.F - 2D micro-Raman analysis of natural methane hydrate samples at depressurization; (See Subtask 9.1 and 9.2)	9/30/2019 (Y3Q4)		Documentation of milestone achievement within required project reporting / deliverables (Deliverable 9.1)	In progress

Organization Chart



Gantt Chart

ID	Task Name	Start	Finish
1	**Task 1.0 - Project Management and Planning**	**Sat 10/1/16**	**Mon 9/30/19**
2	*Deliverable 1.1: Project Management Plan*	Sat 10/1/16	Mon 11/7/16
3	Deliverable 1.2: Data Management Plan	Sat 10/1/16	Tue 2/14/17
4	*Milestone 1.A: Project Kick-off Meeting*	Tue 11/22/16	Tue 11/22/16
5	**PHASE 1**	**Sat 10/1/16**	**Thu 3/29/18**
6	**Task 2.0 – Macro Scale: Relative Permeability of Methane Hydrate Sand Packs**	**Sat 10/1/16**	**Wed 3/28/18**
7	Sub-Task 2.1	Sat 10/1/16	Tue 6/27/17
8	*Milestone 1.B*	Tue 6/27/17	Tue 6/27/17
9	Sub-Task 2.2	Wed 6/28/17	Tue 3/27/18
10	*Milestone 1.C*	Tue 3/27/18	Tue 3/27/18
11	*Deliverable 2.1*	Thu 2/1/18	Wed 3/28/18
12	**Task 3.0 – Macro Scale: Depressurization of Methane Hydrate Sand Packs**	**Sat 10/1/16**	**Wed 3/28/18**
13	Subtask 3.1	Sat 10/1/16	Tue 6/27/17
14	Subtask 3.2	Wed 6/28/17	Tue 3/27/18
15	*Milestone 1.D*	Tue 3/27/18	Tue 3/27/18
16	*Deliverable 3.1*	Thu 2/1/18	Wed 3/28/18
17	**Task 4.0 – Micro Scale: CT Observation of Methane Hydrate Sand Packs**	**Sat 10/1/16**	**Wed 3/28/18**
18	Sub-Task 4.1	Sat 10/1/16	Tue 6/27/17
19	*Milestone 1.E*	Tue 6/27/17	Tue 6/27/17
20	Sub-Task 4.2	Wed 6/28/17	Tue 3/27/18
21	*Milestone 1.F*	Tue 3/27/18	Tue 3/27/18
22	*Deliverable 4.1*	Thu 2/1/18	Wed 3/28/18
23	**Task 5.0 – Micro Scale: Raman Observation of Methane-Gas-Water Systems**	**Sat 10/1/16**	**Wed 3/28/18**
24	Sub-Task 5.1	Sat 10/1/16	**Tue 6/27/17**
25	Sub-Task 5.2	Wed 6/28/17	Wed 3/21/18
26	Sub-Task 5.3	Tue 8/1/17	Tue 3/27/18
27	*Deliverable 5.1*	Thu 2/1/18	Wed 3/28/18
28	*Milestone 1.G*	Wed 3/21/18	Wed 3/21/18
29	*Milestone 1.H*	Wed 3/21/18	Wed 3/21/18
30	**TECHNICAL GO/NO GO DECISION POINT 1**	Thu 3/29/18	Thu 3/29/18
31	**PHASE 2**	**Sun 4/1/18**	**Mon 9/30/19**
32	**Task 6.0 – Macro Scale: Relative Permeability of Methane Hydrate Sand Packs and Intact Pressure Core Samples**	**Sun 4/1/18**	**Mon 9/30/19**
33	Sub-Task 6.1	Sun 4/1/18	**Thu 1/17/19**
34	*Milestone 2.A*	Thu 1/17/19	Thu 1/17/19
35	Sub-Task 6.2	Wed 1/23/19	Mon 9/30/19
36	*Milestone 2.B*	Mon 9/30/19	Mon 9/30/19
37	*Deliverable 6.1*	Sun 9/1/19	Mon 9/30/19
38	**Task 7.0 – Macro Scale: Depressurization of Methane Hydrate Sand Packs and Intact Pressure Core Samples**	**Sun 4/1/18**	**Mon 9/30/19**
39	Sub-Task 7.1	Sun 4/1/18	**Thu 1/17/19**
40	Sub-Task 7.2	Mon 1/21/19	Mon 9/30/19
41	*Milestone 2.C*	Mon 9/30/19	Mon 9/30/19
42	*Deliverable 7.1*	Sun 9/1/19	Mon 9/30/19
43	**Task 8.0 – Micro Scale: CT experiments on Gulf of Mexico Sand Packs**	**Sun 4/1/18**	**Mon 9/30/19**
44	Sub-Task 8.1	Sun 4/1/18	**Thu 1/17/19**
45	Sub-Task 8.2	Mon 1/21/19	Mon 9/30/19
46	*Milestone 2.D*	Mon 9/30/19	Mon 9/30/19
47	*Deliverable 8.1*	Sun 9/1/19	Mon 9/30/19
48	**Task 9.0 – Micro Scale: Raman Observation on hydrate-bearing sand packs**	**Sun 4/1/18**	**Mon 9/30/19**
49	Sub-Task 9.1	Sun 4/1/18	**Thu 1/17/19**
50	*Milestone 2.E*	Thu 1/17/19	Thu 1/17/19
51	Sub-Task 9.2	Mon 1/21/19	Mon 9/30/19
52	*Milestone 2.F*	Mon 9/30/19	Mon 9/30/19
53	*Deliverable 9.1*	Sun 9/1/19	Mon 9/30/19



Bibliography

Publications

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Chen, X., Verma, R., Nicolas Espinoza, D., & Prodanović, M. (2018). Pore-scale determination of gas relative permeability in hydrate-bearing sediments using X-ray computed micro-tomography and Lattice Boltzmann method. Water Resources Research, 54(1), 600-608. https://doi.org/10.1002/2017WR021851

Chen, X and Espinoza, DN (2018), Surface area controls gas hydrate dissociation kinetics in porous media, Fuel, 234, 358-363. https://doi.org/10.1016/j.fuel.2018.07.030

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