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Hydrate-Bearing Clayey Sediments: Morphology, Physical Properties, Production and Engineering/Geological Implications Project Period (10/1/2012 to 9/30/2017)

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ENERGY NATIONAL ENERGY TECHNOLOGY LABORATORY

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ACCOMPLISHMENTS

Context – **Goals**. Fine grained sediments host more than 90% of the global gas hydrate accumulations. Yet, hydrate formation in clayey sediments is least understood and characterized. This research focuses on <u>hydrate bearing clayey sediments</u>. The goals of this research are (1) to gain a fundamental understanding of hydrate formation and ensuing morphology, (2) to develop laboratory techniques to emulate "natural" formations, (3) to assess and develop analytical tools to predict physical properties, (4) to evaluate engineering and geological implications, and (5) to advance gas production alternatives to recover methane from these sediments.

Accomplished

The main accomplishments for this period include:

- Physical properties numerical simulation
 - o Thermal field with ice/hydrate lens inclusion
 - o Bulk modulus with lens inclusion
- Impacts of supercooling on hydrate morphology in clays
 - o X-ray CT studies

Plan - Next reporting period

- 1. Elastic properties of THF hydrate bearing clays
- 2. Gas production from hydrate-bearing clayey sediments

RESEARCH IN PROGRESS

Fundamental physical properties – numerical simulation

Figure 1 illustrates the influence of two types of segregated hydrate distribution on the thermal conduction process in hydrate-bearing sediments. Gas hydrate has significantly lower thermal conductivity in comparison to the sediment matrix. Therefore, the heat flow line in Figure 1 shifts to reduce the length of the conduction path through the hydrate.



Figure 1: Lenses distribution and the influence on thermal conduction fields. Arrows denote the heat flow direction. Horizontal lines represent the temperature contour fields.

Figure 2 compiles the simulation results of the thermal conductivities K_T as a function of hydrate mass orientation θ . The effective thermal conductivity of the sediments with a single ellipse lens follows the equation below,

$$K_T = K_0 \cdot \cos^2 \theta + K_{90} \cdot \sin^2 \theta$$

where K_T is the effective thermal conductivity, K_0 and K_{90} are the effective thermal conductivities when the lens is perpendicular and parallel to the thermal gradient respectively, and θ is the orientation of the hydrate mass. By contrast, the effective thermal conductivity of the sediments that contain crossed hydrate lenses is not sensitive to the lens orientation.

Results for thermal conductivity apply to electrical conductivity, magnetic permeability, and dielectric permittivity as well. The effect of hydrate lenses on effective media properties re-

flects the corresponding physical properties. For example, the hydraulic and electrical conductivities of hydrate approach zero. Therefore, the cutoff effect of hydrate on the water flow and electric current is more significant than the effect on heat flow.



Figure 2: Effective thermal conductivity of hydrate-bearing fine-grained sediments as a function of hydrate mass orientation θ . Numerical simulation results. Note: lines represent estimations, dots are numerical simulation results.

Figure 3 presents the negligible influence of hydrate mass orientation θ on the bulk modulus of hydrate-bearing fine-grained sediments. This is primarily due to the inherent isotropic stress boundary condition associated with bulk modulus. The influences of the hydrate fraction and the type of geometrical distribution are much more significant.



Figure 3: Effective bulk modulus of hydrate-bearing fine-grained sediments as a function of hydrate mass orientation θ . Numerical simulation results.

THF hydrate morphology in clays

To avoid experimental difficulties, THF is used as a proxy of hydrate formed in clayey sediments (i.e., Kaolinite in this case) to investigate their mechanical properties. Specimens are made by mixing kaolinite with a certain mass fraction of 100% stoichiometric THF solution (i.e., H_2O :THF = 81:19). Since hydrate nucleation in such fine-grained sediments is in segregated form, the definition of hydrate saturation is defined as the ratio of the hydrate volume over the total specimen volume, instead of over the pore volume as defined in coarse-grained sediments. We here show CT results of kaolinite mixed with such solution with mass ratios of 100:50, 100:60, and 100:70 (Figure 4). Note that a specimen made of Kaolinite:solution = 100:50 inherently has the mass ratios of Kaolinite:H₂O:THF = 100: 40.5: 0.95. Even with identical mixture to prepare the specimen, the morphology and saturation of formed hydrate in kaolinite vary depending on the supercooling temperature, i.e., preferred nucleation at core boundaries under low supercooling temperature in compared with random nucleation under high supercooling temperature.



Figure 4: CT results of hydrate morphology in kaolinite: impacts of mass fracton and surpercooling temperature.

MILESTONE LOG

| Milestone | Planed completion date | Actual completion date | Verification method | Comments |
|--|------------------------------|------------------------------|---|--------------------------------|
| Literature review | 5/2013 | 5/2013 | Report | |
| Preliminary laboratory proto- col | 8/2013 | 8/2013 | Report (with preliminary val- idation data) | |
| Cells for Micro-CT | 8/2013 | 8/2013 | Report (with first images) | |
| Compilation of CT images: segregated hydrate in clayey sediments | 8/2014 | 8/2014 | Report (with images) | Additional studies in progress |
| Preliminary experimental studies on gas production | 12/2014 | 12/2014 | Report (with images) | |
| Analytical/numerical study of 2-media physical properties | 5/2015 | 5/2015 | Report (with analytical and numerical data) | Additional studies in progress |
| Experimental studies on gas production | 12/2015 | 12/2015 | Report (with data) | Additional studies in progress |
| Early numerical results related to gas production | 5/2016 | 2/2016 | Report | Additional studies in progress |
| Comprehensive results (in- cludes Implications) | 9/2016 | 9/2016 | Comprehensive Report | |

PRODUCTS

- Publications & Presentations:
 - Jang, J., Sun, Z. and Santamarina, J.C., (2017) Capillary pressure across a pore throat in the presence of surfactants. *Water Resources Research*. (Published online).
 - Dai, S., and Santamarina, J.C., (2017) Stiffness evolution in frozen sands subjected to stress changes. *Journal of Geotechnical and Geoenvironmental Engineering* (Published online).
 - Park, J., & Santamarina, J. C. (2017). Revised Soil Classification System for Coarse-Fine Mixtures. Journal of Geotechnical and Geoenvironmental Engineering, (Published online).
 - Jang, J. and Santamarina, J.C., (2016). Hydrate bearing clayey sediments: Formation and gas production concepts. *Marine and Petroleum Geology*, 77, pp.235-246.
 - Shin, H. and Santamarina, J.C., (2016). Sediment-well interaction during depressurization. *Acta Geotechnica*, pp.1-13.
 - Dai, S., Shin, H. and Santamarina, J.C., (2016). Formation and development of salt crusts on soil surfaces. *Acta Geotechnica*, 11(5), pp.1103-1109.
 - Jang, J., & Carlos Santamarina, J. (2015). Fines Classification Based on Sensitivity to Pore-Fluid Chemistry. *Journal of Geotechnical and Geoenvironmental Engineering*, 142(4), 06015018.

- Website: Publications and key presentations are included in http://pmrl.ce.gatech.edu/ (for academic purposes only)
- Technologies or techniques: X-ray tomographer and X-ray transparent pressure vessel
- Inventions, patent applications, and/or licenses: None at this point.
- Other products:

Lei, L (2017). Gas Hydrate in Fine-grained Sediments - Laboratory Studies and Coupled Processes Analyses. PhD Thesis, Georgia Institute of Technology.

PARTICIPANTS & OTHER COLLABORATING ORGANIZATIONS

Research Team: The current team involves:

- Carlos Santamarina (Professor)
- Sheng Dai (Assistant Professor)
- Zhonghao Sun (PhD student)
- Jongchan Kim (PhD student)



IMPACT

Understanding of fine grained hydrate-bearing sediments.

CHANGES/PROBLEMS:

None at this point.

SPECIAL REPORTING REQUIREMENTS:

We are progressing towards all goals for this project.

BUDGETARY INFORMATION:

As of the end of this research period, expenditures are summarized in the following table.

| | _ | | | - | - | | | _ | | - |
|---|----------|---------------------|----------|---------------------|----------|---------------------|-----------|---------------------|----------|---------------------|
| | | Budget P | eriod 4 | | | | | Budget Pe | riod 5 | |
| | σ | 2 | a | 3 | ð | _ | a | 1 | ð | 2 |
| Baseline Reporting Quarter DE-FE009897 | 1/1/16 - | 3/31/16 | 4/1/16 - | 6/30/16 | 7/1/16-9 | 9/30/16 | 10/1/16 - | 12/31/16 | 1/1/17-3 | 3/31/17 |
| | 02 | Cumulative Total | 8 | Cumulative Total | Q4 | Cumulative Total | 01 | Cumulative Total | 07 | Cumulative Total |
| Baseline Cost Plan | | | | | | | | | | |
| Federal Share | 41,547 | 544,299 | 41,547 | 585,846 | 41,547 | 627,393 | 0 | 627,393 | 0 | 627,393 |
| Non-Federal Share | 11,935 | 158,904 | 11,935 | 170,839 | 11,935 | 182,774 | 0 | 182,774 | 0 | 182,774 |
| Total Planned | 53,482 | 703,203 | 53,482 | 756,685 | 53,482 | 810,167 | 0 | 810,167 | 0 | 810,167 |
| Actual Incurred Cost | | | | | | | | | | |
| Federal Share | 32,381 | 494,341 | 45,285 | 539,627 | 17,607 | 557,234 | 11,416 | 568,650 | 16,598 | 585,248 |
| Non-Federal Share | 10,111 | 162,556 | 5,056 | 167,612 | 2,505 | 170,116 | 5,009 | 175,126 | 5,009 | 180,135 |
| Total Incurred Costs | 42,492 | 656,897 | 50,341 | 707,238 | 20,112 | 727,350 | 16,425 | 743,775 | 21,607 | 765,382 |
| Variance | | | | | | | | | | |
| Federal Share | -9,166 | -49,957 | 3,738 | -46,219 | -23,940 | -70,159 | 11,416 | -58,743 | 16,598 | -42,145 |
| Non-Federal Share | -1,824 | 3,652 | -6,879 | -3,227 | -9,430 | -12,658 | 5,009 | -7,648 | 5,009 | -2,639 |
| Total Variance | -10,990 | -46,305 | -3,141 | -49,447 | -33,371 | -82,817 | 16,425 | -66,392 | 21,607 | -44,785 |
| | | | | | | | | | | |

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