

Final Report: September 2011

Methane Recovery from Hydrate-bearing Sediments

Funding Number: DE-FC26-06NT42963

Submitted By:

J. Carlos Santamarina
Georgia Institute of Technology
School of Civil and Environmental Engineering
Atlanta, GA 30332-0355
Phone: (404)-894.7605
Fax: (404)-894.2278
E-mail: jcs@gatech.edu

Costas Tsouris
Oak Ridge National Laboratory
Georgia Institute of Technology
School of Civil and Environmental Engineering
Atlanta, GA 30332-0373
E-mail: costas.tsouris@ce.gatech.edu

Acknowledgment: This material is based upon work supported by the Department of Energy under Award Number DE-FC26-06NT42963

Disclaimer: This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

SCOPE AND CONTRIBUTIONS

Goals: Identifying, understanding and modeling processes involved in methane production from hydrate-bearing sediments.

Approach: Observation and interpretation of phenomena at multiple scales, ranging from pore-contact scale to the macro-reservoir scale, taking into consideration various possible driving forces (e.g., depressurization, thermal stimulation).

Salient contributions:

- *Hydrate formation and growth.* Different conditions (unsaturated from gas phase, from ice, from dissolved phase, in water-wet and oil-wet sediments, during gas exchange). Formation rates at gas-water interface. Transients. Spatial distribution (partial pore filling, cluster, segregated).
- *Relevance to marine and permafrost environments.*
- *Hydrate-mineral bonding and tensile strength.* Implications on the mechanical behavior of hydrate bearing sediments in view of production strategies.
- *Properties:* mechanical, conduction (hydraulic, thermal), mixed fluids: interfacial, mixed fluids: unsaturation.
- *Gas production by heating and depressurization.* Study in 5m long 1D cell. Experimental study and modeling.
- *Gas production by chemo-driven methods.* Fundamental understanding of CO₂-CH₄ exchange.
- *The role of effective stress.*
- *Gas invasion versus gas production – Evolution of degree of saturation and fluid conduction.* Fluid-driven fractures.
- *Fluid conductivity in spatially varying sediments.*
- *Thermodynamic formulation.*
- *Coupled Thermo- Hydro- Chemo-Mechanical analyses.*
- *Production strategies in different formations.*
- *Relevance to real systems.*
- *Emergent phenomena:* gas driven fractures, fines migration and clogging.
- *Gas recovery efficiency .*

TRAINING OF HIGHLY QUALIFIED PERSONNEL

- Dr. Tae Sup Yun (2005). Hydrate-Bearing Sediments (also obtained MSc). Sower's Award. Current Position: Assistant Professor, Yonsei University (formerly: *PC Rossin Assistant Prof.* at Lehigh U.).
- Dr. Patricia Taboada (Post Doc, 2007), Current Position: Assistant Professor, Rochester Institute of Technology.
- Dr. Changho Lee (Post-Doc, 2008 - 2010). Methane hydrates – natural formations. Current Position: Korea University.
- Dr. Hosung Shin (2009). Discontinuities (dissolution and polygonal faults, desiccation cracks, miscible and non-miscible fluid invasion). Sower's Award. Current Position: Assistant Professor, Ulsan University.
- Dr. Jong Won Jung (2010). Gas Production from Methane Hydrates. Current Position: Post Doctoral Fellow, Laurence Berkeley National Laboratory.

- Dr. Nicolas Espinoza (2011). CO2 sequestration – Fundamental Studies. Current Position: Research Fellow, Ecole Nationale des Ponts et Chaussées, Paris.
- Dr. Jaewon Jang (2011). Gas Production from Methane Hydrates. Current Position: Assistant Professor, Wayne State University, Detroit.
- Sheng Dai (PhD Starting Fall 2008 - Current). Theme: Hydrate Bearing Sediments – Characterization.
- Jun Bong Jang (PhD Started Fall 2008 - Current). Theme: Gas production from fine-grained sediments.
- Alessio Savioli (MSc 2011), Open Mode Discontinuities –Lenses.

COLLABORATIONS

During the implementation of this research, we developed close collaborations with researchers in the USA and abroad. The closest ties involved (shared related publications)

- Carolyn Ruppel (*USGS – no cost Co-PI*)
- William Waite (*USGS*)
- Tommy Phelps (*ORNL*)
- Claudia Rawn (*ORNL*)
- K. Soga (*Cambridge U.*)
- M. Sanchez (*Strathclyde U. in UK; moved to Texas A&M*)
- G.C. Cho (*KAIST, S. Korea*)
- C. Viggiani (*U. Joseph Fourier, Grenoble, France*)

SPECIAL ACTIVITIES

Special Events

Workshop on physical properties (2009)
 Hosted DOE Hydrates Program Review (2010)
 US-Japan Workshop (2010)
 International Laboratory Comparison (2010-2011)

Study of Real Systems

Gulf of Mexico
 Alaska North Slope
 South Korea
 India
 Japan (in progress)

PUBLICATIONS

Articles in Referred Archival Journals

- Espinoza, D.N. and Santamarina J.C. (2010), Ant Tunneling - A Granular Media Perspective, *Granular Matter*, vol. 12, pp. 607-616.
- Espinoza, D.N. and Santamarina J.C. (2010), Water-CO2-mineral systems: interfacial tension, contact angle and diffusion – Implications to CO2 geological storage, *Water Resources Research*, vol. 46, DOI: 10.1029/2009WR008634.

- Espinoza, D.N. and Santamarina, J.C., P-wave monitoring of hydrate-bearing sand during CH₄-CO₂ replacement (under review).
- Espinoza, D.N., Kim, S.H., Santamarina, J.C. (2011), Carbon Geological Storage, *KSCE Journal of Civil Engineering*, vol. 15, no. 4, pp. 707-719.
- Fragaszy, R.J., Santamarina, J.C., Amekudzi, A., Assimaki, D., Bachus, R., Burns, S.E., Cha, M., Cho, G.C., Cortes, D.D., Dai, S., Espinoza, D.N., Garrow, L., Huang, H., Jang, J., Jung, J.W., Kim, S.H., Kurtis, K., Lee, C., Pasten, C., Phadnis, H., Rix, G., Shin, H.S., Torres, M.C., and Tsouris, C. (2011). “Sustainable development and energy geotechnology – potential roles for geotechnical engineering”. *KSCE Journal of Civil Engineering, Special Issue on Energy Geotechnology*, vol. 15, no. 4, pp. 611-621.
- Gabitto, J., and C. Tsouris, “Physical Properties of Gas Hydrates: A Review,” *Journal of Thermodynamics*, Article ID 271291, [doi:10.1155/2010/271291](https://doi.org/10.1155/2010/271291), (2010).
- Jang, J. and Santamarina J.C. (2012), Gas Production from Hydrate Bearing Sediments: Pore-Network Model Simulation and Macro-Scale Analyses, *Journal of Geophysical Research - Solid Earth*, (in print).
- Jang, J., Narsilio, G. and Santamarina, J.C. (2011), Hydraulic Conductivity in Spatially Varying Media -A Pore-Scale Investigation, *Geophysical Journal International*, vol. 184, pp. 1167-1179
- Jung, J.W. and Santamarina, J.C. (2010), CH₄-CO₂ Replacement in Hydrate-Bearing Sediments: A Pore-Scale Study, *G-Cubed Geochemistry, Geophysics and Geosystems*, Vol. 11, Q0AA13, [doi:10.1029/2010GC003339](https://doi.org/10.1029/2010GC003339).
- Jung, J.W., Espinoza, D.N. and Santamarina, J.C. (2010), Hydrate Bearing Sediments: CH₄-CO₂ Replacement, *Journal of Geophysical Research*, vol. 115, B10102, [doi:10.1029/2009JB000812](https://doi.org/10.1029/2009JB000812)
- Jung, J.W., Jang, J., Santamarina, J.C., Tsouris, C., Phelps, T.J., and Rawn, C.J., Gas Production from Hydrate-Bearing Sediments – Emergent Phenomena (under review).
- Jung, J.W., Santamarina, J.C. (2012), Hydrate Adhesive and Tensile Strength, *G-Cubed Geochemistry, Geophysics and Geosystems* (in print).
- Jung, J.W., Santamarina, J.C., Hydrate Formation and Growth in Pores (under review).
- Jung, J.W., Santamarina, J.C., Soga, K., Stress-Strain Response of Hydrate Bearing Sediments – Numerical Study Using DEM Simulations, *Journal of Geophysical Research* (in print).
- Kwon, T., Cho, G.C., Santamarina, J.C., Kim, J., and Lee, J. (2009). Stability evaluation of hydrate-bearing sediments during thermally-driven hydrate dissociation, AGU Fall meeting, Abstract.
- Kwon, T.H., Cho, G.C. and Santamarina J.C. (2007). Hydrate Dissociation in Sediments: Pressure-Temperature Evolution, *Geochemistry, Geophysics, Geosystems*, vol. 9.
- Lucia, A., J. Lee, and C. Tsouris, “Advances in Gas Hydrate Thermodynamic and Transport Properties,” *Journal of Thermodynamics*, Article ID 523503, [doi:10.1155/2010/523503](https://doi.org/10.1155/2010/523503), (2010).
- Shin H. and Santamarina J.C. (2009). Mineral Dissolution and the Evolution of k_0 . *J. Geotechnical and Geoenvironmental Eng. ASCE*, vol. 135, no. 8, pp. 1141-1147.
- Shin H. and Santamarina J.C. (2011), Desiccation Cracks in Saturated Fine-Grained Soils: Particle Level Phenomena and Effective Stress Analysis, *Geotechnique* (in print).
- Shin, H. and Santamarina, J.C. (2010), Fluid-driven fractures in uncemented sediments: Underlying particle-level processes, *Earth and Planetary Science Letters*, vol. 299, pp 180-189.
- Shin, H. and Santamarina, J.C., Opening Mode Discontinuities in Soils (under review).
- Shin, H., Santamarina, J.C. and Cartwright, J. (2008). Contraction-Driven Shear Failure in Compacting Uncemented Sediments, *Geology*, vol. 36, no. 12, pp. 931-934.

- Shin, H., Santamarina, J.C. and Cartwright, J. (2010), Displacement Field In Contraction Driven Faults, *J. Geophysical Research*, 115, B07408, doi:10.1029/2009JB006572.
- Taboada-Serrano, P., S. Ulrich, P. Szymcek, S.D. McCallum, T.J. Phelps, A.V. Palumbo, and C. Tsouris, “A Multi-phase, Micro-dispersion Reactor for the Continuous Production of Methane Gas Hydrate,” *Industrial & Engineering Chemistry Research*, **48**, 6448–6452 (2009).
- Waite W.F., Santamarina J.C., Cortes DD, Dugan B, Espinoza DN., Germaine J., Jang J., Jung J.W., Kneafsey T., Shin HS, Soga K., Winters W., and Yun T-S. (2009), Physical Properties Of Hydrate-Bearing Sediments, *Review of Geophysics*, vol. 47, doi:10.1029/2008RG000279.
- Yun, T.S., Lee, C., Lee, J.S., Jang-Jun Bahk, and Santamarina, J.C., A Pressure Core Based Characterization of Hydrate Bearing Sediments in the Ulleung Basin, East Sea, *Journal of Geophysical Research*, *Journal Of Geophysical Research*, vol. 117, pp. 151-158. doi:10.1029/2010JB007468

Special Contribution

- Santamarina, J.C. and Jang, J. (2010), Energy Geotechnology: Implications of Mixed Fluid Conditions, Proc. 5th International Conference on Unsaturated Soils, Unsat 2010, Barcelona, Eds. A. Gens and E. Alonso (*see Keynote Lectures*).

Publications in Conference Proceedings

- Santamarina, J.C. and Jang, J. (2009), Gas Production from Hydrate Bearing Sediments: Geomechanical Implications, *DOE Fire in the Ice*, vol. 9, no.4, pp. 18-22.
- Santamarina, J.C. and Shin, H. (2009). Discontinuities in granular materials: Particle-level mechanisms, in *Symposium on the Mechanics of Natural Solids*, Horto, Greece, Eds. D. Kolymbas and C. Viggiani.
- Cartwright, J., Santamarina J.C., Shin, H. (2009). A diagenetic mechanism for the formation of shear fractures in shales hosting deep tight gas plays, AAAG meeting, June – Denver.
- Santamarina J.C. and Shin H. (2007). Particle Dissolution: Ko Effects, IUTAM, Brazil 2007 (Extended Abstract).

TEACHING - PRESENTATIONS

Knowledge gained during the execution of this project was also shared through classroom activities, lectures and both national and international events. A brief summary follows:

Teaching – New Graduate Courses

- *Geotechnical Aspects in Resource Recovery*
- *Explorations into Geo-Sustainability*

Keynote Lectures

- *Coussy Memorial, ENPC, France (April 2011).*
- *2010 Tewkesbury Lecturer, U. Melbourne, Australia (December 2010).*
- *Int. Conf. Unsaturated Soils, Barcelona, (September 2010).*
- *Int. Workshop on Multiscale & Multiphysics Processes in Geomechanics, Palo Alto (June 2010)*
- *Gordon Research Conference, Waterville, ME (June 2010)*
- *12th IACMAG Conference, Goa, India (October 2008)*
- *National Conference on the Advancement of Research, Savannah (April 2007)*

Invited Speaker- Universities

- *Kansas S. U (April 2011)*
- *U. North Carolina Charlotte (March 2011)*
- *ETH Zurich (March 2011)*
- *Shaw Lecture, North Carolina State University (February 2011)*
- *University of Pittsburg, January 21 2011.*
- *Texas A&M University (April 2010)*
- *Université Joseph Fourier, Grenoble, France (June 2009)*
- *Northwestern U., Evanston (March 2009)*
- *Colorado School of Mines, Golden (December 2008)*
- *Lafarge, Lyon, France (June 2008)*
- *Ecole de Ponts, France (June 2008)*
- *Korea Advanced Inst. of Science and Technology, Korea (February 2008)*
- *Arizona State U., Phoenix (October 2007)*
- *Cambridge U., UK (July 2007)*
- *Imperial College, UK (July 2007)*
- *Herriot-Watt U., Edinburg, UK (July 2007)*
- *U. Southern California, Los Angeles (April 2007)*
- *Tufts U., Boston (April 2007)*

PROJECT SUMMARY

This comprehensive project allowed us a wide range of activities and accomplishments. These can be summarized as follows:

Developed	unique <u>devices</u> and <u>laboratory facility</u>
Studied	fundamental pore and grain-scale phenomena
Analyzed	governing phenomena & variables
Identified	new emergent phenomena
Implemented	new algorithms for up-scaling
Programmed	formulation for macro-scale <u>modeling</u>
Published	results in leading journals
Educated	highly qualified <u>personnel</u>
Delivered	new courses
Presented	multiple lectures and seminars
Hosted	national and international events