U.S. Department of Energy
National Energy Technology Laboratory

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Fate of Methane Emitted from Dissociating Marine Hydrates: Modeling, Laboratory, and Field Constraints

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Ruben Juanes

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1 Executive summary

Work during this period focused on the following tasks:

- Task 1.0: Project Management, Planning and Reporting
- Subtask 2.1: Phase-field modeling of a static gas bubble
- Subtask 3.1: Laboratory experiments — flow-loop design, fabrication and construction
- Subtask 4.1: Quantitative analysis of newly-discovered US Atlantic margin methane plumes

We have finalized a revised Project Management Plan, which has been agreed upon with DOE. We have also participated, via Webex, in the kick-off meeting for the project. Juanes presented an overview of the administrative aspects of the project; and Juanes, Waite and Ruppel described the technical content of the various tasks of the projects (Tasks 2, 3 and 4, respectively).

Work on the project has now started in earnest. All PIs (Weber, Waite, Ruppel and Juanes) got together for a full-day meeting at the University of New Hampshire for an in-depth discussion of the laboratory experiments to be conducted. Waite visited Carolyn Koh's group at Colorado School of Mines for discussions on a related experimental facility — a summary of outcomes from that visit is included as part of this report. Ruppel and colleagues are finalizing the paper that summarizes the new East Coast seep database and its relationship to upper slope gas hydrate dissociation, and was on two Fall 2013 AGU Meeting posters about these seeps. The undergraduate student (Ms. Mali'o Kodis) collaborating with Ruppel, Weber, and Adam Skarke, the lead author on the seeps manuscript, won a best student paper award from the AGU Ocean Sciences section. The fully QA/QCed multibeam backscatter database, including times of observations, has been completed and will be published as a supplement to the forthcoming manuscript. Ruppel continues to make plans to visit some of the deepwater Nantucket seeps on the R/V Endeavor in July 2014 as part of a NSF cruise funded to J. Kessler (U. Rochester).

MIT and UNH have started staffing the project with graduate students, although this has been slightly delayed by the offset between the natural academic cycle and the beginning of project.

2 Accomplishments

2.1 Major goals and objectives of the project

The overall goals of this research are: (1) to determine the physical fate of single and multiple methane bubbles emitted to the water column by dissociating gas hydrates at seep sites deep within the hydrate stability zone or at the updip limit of gas hydrate stability, and (2) to quantitatively link theoretical and laboratory findings on methane transport to the analysis of real-world field-scale methane plume data placed within the context of the degrading methane hydrate province on the US Atlantic margin.

The project is arranged to advance on three interrelated fronts (numerical modeling, laboratory experiments, and analysis of field-based plume data) simultaneously. The fundamental objectives of each component are the following:

1. Numerical modeling: Constraining the conditions under which rising bubbles become armored with hydrate, the impact of hydrate armoring on the eventual fate of a bubbles methane, and the role of multiple bubble interactions in survival of methane plumes to very shallow depths in the water column.

2. Laboratory experiments: Exploring the parameter space (e.g., bubble size, gas saturation in the liquid phase, "proximity" to the stability boundary) for formation of a hydrate shell around a free bubble in water, the rise rate of such bubbles, and the bubbles acoustic characteristics using field-scale frequencies.

3. Field component: Extending the results of numerical modeling and laboratory experiments to the field-scale using brand new, existing, public-domain, state-of-the-art real world data on US Atlantic margin methane seeps, without acquiring new field data in the course of this particular project. This component will quantitatively analyze data on Atlantic margin methane plumes and place those new plumes and their corresponding seeps within the context of gas hydrate degradation processes on this margin.

2.2 Accomplishments in this reporting period

Work during this period focused on the following tasks:

- Task 1.0: Project Management, Planning and Reporting
- Subtask 2.1: Phase-field modeling of a static gas bubble
- Subtask 3.1: Laboratory experiments — flow-loop design, fabrication and construction
- Subtask 4.1: Quantitative analysis of newly-discovered US Atlantic margin methane plumes

Here, we report in some detail on our developments and accomplishments under Subtask 3.1. In subsequent progress reports we will describe our advances in Subtasks 2.1 and 4.1. A detailed Milestones Status Report is included as Appendix 1.

Task 2.0: Theoretical and computational models of coupled bubble rise and hydrate formation and dissociation

Subtask 2.1: Phase-field modeling of a static gas bubble

Introduction. We will develop a *phase-field modeling* framework to study hydrate formation at the gas-water interface of a buoyant bubble. Phase-field modeling is a mathematical approach to describe systems that are out of thermodynamic equilibrium [Anderson et al., 1998; Bray, 1994; Emmerich, 2008]. It was first introduced in the context of solidification processes and phase transitions [Boettinger et al., 2002; Cahn, 1961; Cahn and Hilliard, 1958], but has since been applied to other fields, including multiphase flows [Cueto-Felgueroso and Juanes, 2012, 2014; Cueto-Felgueroso and Peraire, 2008; Hou et al., 1994; Lee et al., 2002; Lowengrub and Truskinovsky, 1998; Sun and Beckermann, 2004, 2008].

The approach is particularly attractive in the context of bubble flows because it permits the incorporation of surface tension effects in a very natural way [Gomez et al., 2010]. Given a two-component, two-phase fluid system (e.g., a bubble in a liquid environment), the phase-field approach is based on a mathematical description of the energy of the system, which consists of 'bulk energy' (associated with the gas inside the bubble and the liquid outside the bubble) and also 'interfacial energy' (associated with the gas–liquid interface). The interface is then modeled as a diffuse, rather than sharp, interface. This choice has advantages from a computational point of view, as it does not require tracking a material surface through the computational domain.

References

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Y. Sun and C. Beckermann. A two-phase diffuse-interface model for Hele-Shaw flows with large property contrasts. *Physica D*, 237:3089–3098, 2008.

Task 3.0: Laboratory experiments on hydrate armoring, rise rate, and gas loss from ascending bubbles

Subtask 3.1: Flow-loop design, fabrication and construction

Introduction. The USGS is constructing a high-pressure flow loop designed to "capture" gas bubbles for subsequent visual and acoustic imaging studies as well as bubble evolution and rise-rate measurements. The apparatus must be able to operate at pressures high enough for the gas to form hydrate. Xenon was chosen for the hydrate-forming gas, meaning hydrate can be formed at ~1.3 MPa (190 psi) at room temperature (21°C, 70°F), and at lower pressures when the system is cooled [*Ohgaki et al.*, 2000].

Design Activities. The group's full-day meeting at the University of New Hampshire (UNH) included a demonstration and discussion of their low-pressure bubble-capture flow loop. Based on lessons learned at UNH, we settled on a design strategy we were then able to refine after visiting the high-pressure bubble-capture facility at the Colorado School of Mines (CSM). L. Chen and Prof. C. Koh provided a hands-on demonstration of the CSM methane bubble-capture flow loop, and additional discussions concerning their measured results were fruitful (Their paper, Chen et al. [2013], was released during Waite's visit to CSM).

Summary of outcomes, UNH:

- The decision was made to design the project's flow loop with a fully-transparent bubble-capture section, rather than using windows in a metal housing.
- The bubble-capture chamber's minimum geometrical specifications for housing an acoustic imaging transducer were established.

Summary of outcomes, CSM:

- Preliminary measurements of methane-bubble rise rates, performed during CSM's hands-on demonstration, verified the specifications needed for this project's flow-loop circulation pump.
- The hydrate former's concentration in the flow loop can be reasonably estimated based on the injected gas volume [*Chen et al.*, 2013], rather than requiring a dedicated concentration measurement. This project's system will be initially designed with that simplification in mind. To maintain the relevance of xenon-based hydrate dissolution studies to the methane hydrate system, we will be reporting xenon concentrations relative to xenon's solubility limit rather than reporting only concentration values. As recently reported by Lapham et al. [2014], dissolution phenomena depend upon the concentration of the hydrate former relative to that hydrate former's solubility limit, not upon the hydrate former species itself.

Fabrication Activities.

Bubble-capture chamber:

- Optically-clear pressure housing has been ordered. This housing material is suitable for operations at ~5 MPa (700 psi), an upper bound temperature for the xenon system because xenon reverts to a liquid state near this pressure at 10°C (50°F) [*Ohgaki et al.*, 2000].
- Drawings for an optically-clear bubble-capture cone have been made and verified with a fabrication company.
- Endcap drawings have been made for the stainless steel components required for

holding the pressure housing in place and providing fluid and electrical feed-through ports for water circulation, gas injection and acoustic measurements. Stock material for these components has been acquired.

Water circulation through the flow loop:

- A high-pressure pump capable of delivering the high flow rates necessary to counter the rise rate of a gas bubble has been identified. The ordering process is underway.
- Building modifications necessary to supply the power required by the water-circulation pump have been discussed with the facility electricians. Plans for modifying the electrical service are underway.
- A high-pressure flow meter suitable for our expected flow rates has been identified, and the ordering process is underway.

References.

Chen, L., E. D. Sloan, C. A. Koh, and A. K. Sum (2013), Methane Hydrate Formation and Dissociation on Suspended Gas Bubbles in Water, Journal of Chemical & Engineering Data, doi: 10.1021/je400765a, 7p.

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Ohgaki, K., T. Sugahara, M. Suzuki, and H. Jindai (2000), Phase behavior of xenon hydrate system, Fluid Phase Equilibria, 175(1-2), 1-6.

Task 4.0: Field data analysis to link models and laboratory data to real world gas hydrate dynamics

Subtask 4.1: Quantitative analysis of newly-discovered US Atlantic margin methane plumes

Ruppel and colleagues are finalizing the paper that summarizes the new East Coast seep database and its relationship to upper slope gas hydrate dissociation, and was on two Fall 2013 AGU Meeting posters about these seeps. The undergraduate student (Ms. Mali'o Kodis) collaborating with Ruppel, Weber, and Adam Skarke, the lead author on the seeps manuscript, won a best student paper award from the AGU Ocean Sciences section. The fully QA/QCed multibeam backscatter database, including times of observations, has been completed and will be published as a supplement to the forthcoming manuscript. Ruppel continues to make plans to visit some of the deepwater Nantucket seeps on the R/V Endeavor in July 2014 as part of a NSF cruise funded to J. Kessler (U. Rochester).

We are anticipating receiving the raw split-beam and multibeam echosounder (SBES and MBES) data corresponding to the East Coast seep database, which are archived on tape at the National Geophysical Database Center, in early 2014. We will then begin the process of determining which subset of seeps were observed with the SBES, and proceed to extract vertical target strength profiles for that subset of seeps. This will build upon techniques used by Weber et al. (submitted) as well as the UNH master's thesis work of Kevin Jerram (manuscript in preparation). Ultimately, we will analyze these data to help constrain the fate and evolution of methane bubbles escaping the seabed.

2.3 Opportunities for training and professional development

The project has offered opportunities for training of our graduate students Christos Nicolaides (MIT) and Xiaojing Fu (MIT), who just recently started to work on the project (see section 4 of this report). A graduate student at UNH will also join the project shortly.

2.4 Dissemination of results to communities of interest

As the project has just started, we do not yet have results to communicate. However, discussions about the experimental work took place during Waite's visit to Colorado School of Mines. The objectives and goals of the project were also presented at the kick-off meeting.

2.5 Plans for the next reporting period

The project is progressing according to the anticipated plan. In the next reporting period we will continue to work on the following tasks:

- Subtask 2.1: Phase-field modeling of a static gas bubble
- Subtask 3.1: Laboratory experiments — flow-loop design, fabrication and construction
- Subtask 4.1: Quantitative analysis of newly-discovered US Atlantic margin methane plumes

We also anticipate that several PIs and graduate students will participate in the Gordon Research Conference on Natural Gas Hydrate Systems, to be held in March 23-28, 2014, in Galveston, TX. PI Ruppel will be giving one of the keynote talks, titled "Investigating climate-sensitive marine gas hydrates to evaluate late Pleistocene to contemporary climate change".

3 Products

3.1 Journal publications, conference papers, and presentations

3.1.1 Journal publications

- L. Cueto-Felgueroso and R. Juanes. A phase-field model of two-phase Hele-Shaw flow. *J. Fluid Mech.*, 2014. Submitted for publication.
- Weber, T., Mayer, L., Jerram, K., Beaudoin, J., Rzhanov, Y. and Lovalvo, D., 2014. Acoustic estimates of methane gas flux from the seabed in a 6000 km2 region in the Northern Gulf of Mexico. *G-cubed*, 2014. Submitted for publication.

3.1.2 Conference papers

Nothing to report.

3.1.3 Presentations

- Skarke, A., Ruppel, C.D., Kodis, M., Lobecker, E., and Malik, M., 2013, Geologic significance of newly-discovered methane seeps on the Northern US Atlantic Margin, EOS Trans. Amer. Geophys. Union, Fall Meeting, OS21A-1613.
- Kodis, M., Skarke, A.D., Ruppel, C.D., Weber, T., Lobecker, E., and Malik, M., 2013, US Atlantic Margin methane plumes identified from water column backscatter data acquired by NOAA ship Okeanos Explorer, EOS Trans. Amer. Geophys. Union, Fall Meeting, OS21A-1612

3.2 Website(s) or other Internet site(s)

Nothing to report.

3.3 Technologies or techniques

Nothing to report.

3.4 Inventions, patent applications, and/or licenses

Nothing to report.

3.5 Other products

(such as data or databases, physical collections, audio or video products, software or NetWare, models, educational aids or curricula, instruments, or equipment)

Nothing to report.

4 Participants and collaborating organizations

4.1 Individuals working on the Project

- Name: Ruben Juanes
  Project Role: Principal Investigator / Project Director
  Nearest person month worked: 1
  Contribution to Project: Ruben Juanes, as project director, is responsible for overall coordination of the effort and for the technology transfer activities, including progress and topical reports, and project review presentations. He takes the lead in the modeling and simulation of hydrate formation and dissociation in rising methane bubbles (Task 2.0), and advises the MIT graduate student responsible for doing the modeling. He also serves as primary advisor to the MIT student who conducts the laboratory experiments of bubble rise and hydrate formation with analogue multiphase fluids (Task 3.0), in collaboration with Waite (USGS).
  Funding Support: MIT academic-year salary / DOE summer salary
  Collaborated with individual in foreign country: No
  Country(ies) of foreign collaborator: Not applicable
  Travelled to foreign country: Not applicable
  Duration of stay in foreign country(ies): Not applicable

- Name: Thomas Weber
  Project Role: Co-Principal Investigator
  Nearest person month worked: 1
  Contribution to Project: Thomas Weber leads the field component of the project (Task 4.0), particularly the quantitative analysis of existing public domain data for northeast Atlantic margin bubble plumes. He also advises a graduate student at UNH. Weber also assists with the acoustics aspects of the laboratory experiments (Task 3.0), both in design of the acoustic component and the interpretation of the resulting data.
  Funding Support: MIT academic-year salary / DOE summer salary
  Collaborated with individual in foreign country: No
  Country(ies) of foreign collaborator: Not applicable
  Travelled to foreign country: Not applicable
  Duration of stay in foreign country(ies): Not applicable

- Name: Carolyn Ruppel
  Project Role: Co-Principal Investigator
  Nearest person month worked: 1
  Contribution to Project: Carolyn Ruppel has responsibility for keeping the project grounded in natural gas hydrates systems and in the issues of greatest relevance for the US gas hydrates research community, particularly the part of the community focused on the environmental impact of methane emissions from gas hydrate deposits. She is also responsible for ensuring that appropriate resources (salary support) are allocated to herself, Waite, and the USGS engineers supporting this project and interacts frequently with Juanes and his students at MIT, where she maintains a second office. She is also responsible for regional analysis and integration of observational data related to

hydrate-derived seeps and plumes on the U.S. Atlantic margin and for linking the newly emerging observational data to other existing data sets (e.g., BOEMs gas hydrates assessment of the Atlantic margin) in this area and in other areas worldwide (Task 4.0).
Funding Support: USGS salary
Collaborated with individual in foreign country: No
Country(ies) of foreign collaborator: Not applicable
Travelled to foreign country: Not applicable
Duration of stay in foreign country(ies): Not applicable

- Name: William Waite
  Project Role: Co-Principal Investigator
  Nearest person month worked: 1
  Contribution to Project: William Waite leads the lab component of the project (Task 3.0) and has primary responsibility for design and construction oversight of the xenon hydrate lab apparatus. He interacts with the USGS engineers, visits UNH to see existing devices at Webers lab, and meets with MIT staff to understand the parameters for the cell installation at MIT. After completion of the testing phase of the laboratory work at the USGS, Waite is responsible for moving the apparatus to MIT. Waite takes on primary responsibility for developing the collaboration among MIT, UNH, and the USGS for the multifaceted lab experiments and working directly with the MIT graduate student on the experiments at MIT.
  Funding Support: USGS salary
  Collaborated with individual in foreign country: No
  Country(ies) of foreign collaborator: Not applicable
  Travelled to foreign country: Not applicable
  Duration of stay in foreign country(ies): Not applicable

- Name: Christos Nicolaides
  Project Role: Graduate Student at MIT
  Nearest person month worked: 3
  Contribution to Project: Christos Nicolaides works on Task 2.0: Theoretical and computational models of coupled bubble rise and hydrate formation and dissociation.
  Funding Support: Vergottis Fellowship (MIT cost-share)
  Collaborated with individual in foreign country: No
  Country(ies) of foreign collaborator: Not applicable
  Travelled to foreign country: Not applicable
  Duration of stay in foreign country(ies): Not applicable

- Name: Xiaojing Fu
  Project Role: Graduate Student at MIT
  Nearest person month worked: 0
  Contribution to Project: Xiaojing Fu works on Task 2.0: Theoretical and computational models of coupled bubble rise and hydrate formation and dissociation.
  Funding Support: DOE
  Collaborated with individual in foreign country: No

Country(ies) of foreign collaborator: Not applicable
Travelled to foreign country: Not applicable
Duration of stay in foreign country(ies): Not applicable

4.2 Other organizations involved as partners

Nothing to report.

4.3 Other collaborators or contacts

We have established a collaboration with Luis Cueto-Felgueroso, formerly a research scientists in Juanes's group and currently a researcher at the Technical University of Madrid, and with Hector Gomez, a professor at the University of La Coruña and who has visited MIT on several occasions and has published joint papers with Juanes. Both researchers are experts in phase-field modeling, and the collaboration will bring new perspectives on the mathematical aspects of multiphase–multicomponent flows.

We have also established contact with Carolyn Koh's group at Colorado School of Mines, where they have built an experimental system that is related to the one proposed in our project. William Waite has already visited their group and we anticipate that this contact will be very beneficial for the experimental aspects of the project.

Ruppel continues to make plans to visit some of the deepwater Nantucket seeps on the R/V Endeavor in July 2014 as part of a NSF cruise funded to J. Kessler (U. Rochester).

5 Impact

5.1 Impact on the principal discipline of the Project

No impact to report yet.

5.2 Impact on other disciplines

No impact to report yet.

5.3 Impact on the development of human resources

The project is supporting the training of graduate students.

5.4 Impact on physical, institutional, and information resources that form infrastructure

Nothing to report yet.

5.5 Impact on technology transfer

Nothing to report yet.

5.6 Impact on society beyond science and technology

No impact to report yet.

5.7 Dollar amount of the awards budget spent in foreign country(ies)

Zero.

6 Changes and problems

Nothing to report.

7 Special reporting requirements

Nothing to report.

8 Budgetary information

The Cost Plan is included as Appendix 2.

MILESTONE STATUS REPORT

Milestone	Task/	Project Milestone	Year 1				Year 2				Year 3				Planned	Planned	Actual	Actual	Comments (notes, explanation
	Subtask	Description	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Start date	End date	Start date	End date	of deviation from plan)
1	1.0	Revise PMP	X												1-Oct-13	31-Dec-13	1-Oct-13	3-Dec-13	Revised PMP sent by email on Dec 3, 2013
2	1.0	Kick-off meeting	X												1-Oct-13	31-Dec-13	1-Oct-13	14-Nov-13	Webex meeting on Nov 14, 2013
3	2.1	Model of static gas bubble in 3D				X									1-Oct-13	30-Sep-14	1-Oct-13		
4	3.1	Verify flow-loop				X									1-Oct-13	30-Sep-14	1-Oct-13		
5	4.1	Extract MBES/SBES seep parameters				X									1-Oct-13	30-Sep-14	1-Oct-13		
6	3.2	Acoustic signature due to hydrate formation							X						1-Oct-14	31-Jul-15			
7	4.2	Estimate of methane flux from Atlantic							X						1-Oct-14	31-Jul-15			
8	2.2	Model of buoyant hydrate-coated gas bubble								X					1-Oct-14	30-Sep-15			
9	3.3	Measure gas-loss rate at low initial pressures								X					1-Jul-15	30-Sep-15			
10	4.1	Analyze plume data acquired by NOAA OE								X					1-Oct-14	30-Sep-15			
11	2.3	Model of bubble-bubble interactions										X			1-Apr-15	31-Mar-16			
12	3.3	Measure gas-loss rate at high initial pressures										X			1-Oct-15	31-Mar-16			
13	4.2	Extend bottom water temperature database										X			1-Jun-15	31-Mar-16			
14	2.4	Model formulation and comparison with field observations												X	1-Oct-15	30-Sep-16			
15	all	Manuscripts submitted / Final project synthesis and report												X	1-Oct-14	30-Sep-16			

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