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OIL & GAS

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Research Performance Progress Report

(Period Ending 09/30/2017)

Impact of clays on the compressibility and permeability of sands during methane extraction from gas hydrate

Project Period (10/1/2016 to 9/30/2019)

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EXECUTIVE SUMMARY

Background: The quantity of methane potentially recoverable from gas hydrate is large enough to motivate federally-supported production tests in several countries, which in turn motivates studies of reservoir production efficiency. Evaluating long-term production well viability involves modeling permeability evolution in the reservoir sediments around the production well because processes reducing the flow of gas into the production well also reduce the long-term economic viability of the well. Fine particles, such as clays, exist nearly ubiquitously in the permafrost and marine settings that typically host gas hydrate, and fines reacting to fluid flow by migrating and clogging pore throats can reduce flow toward the production well. Many fines are sensitive to variations in pore-fluid chemistry, swelling in reaction to in situ pore brine being displaced by fresh water liberated from hydrates during dissociation. Additionally, fine particles tend to collect at gas/water interfaces created by the multiphase flow of gas and water. Thus, as methane and fresh water flow from the hydrate-dissociation front toward the production well, fine particles in the reservoir sands, interbedded fine-grained layers and seal layers can be swelled, migrated (or both), potentially clogging pathways and limiting flow to the production well.

Objective: This project seeks to provide a quantitative basis for reservoir models to account for the impact of clays and other fine-grained material ("fines") on reservoir compressibility and permeability, two key factors controlling the flow of gas and fluids toward a production well. This overall objective is addressed through a combination of site-specific and more generalized, fundamental science goals:

Site-specific measurement goals: quantify the change in compressibility and permeability due to the reaction of fines to pore-water freshening in sediment from the 2015 NGHP-02 gas hydrates research cruise offshore India.

Fundamental measurements on pure fines goal: distinguish between, and quantify, mechanisms for sediment compressibility and permeability change due to physical and chemical responses of fines to the flow of freshened pore water and gas:

- Chemical response: quantify and catalog the sensitivity of pure fines (fines with only a single component, or "endmember" fines) to pore-water chemistry.
- Physical response: quantify the link between fines migration and clogging during single and multiphase flow.

ACCOMPLISHMENTS

The overall project timeline is shown in Figure 1. This report details activities in the fourth quarter of Year 1. A full list of milestones and Success Criteria is provided in the Appendix.

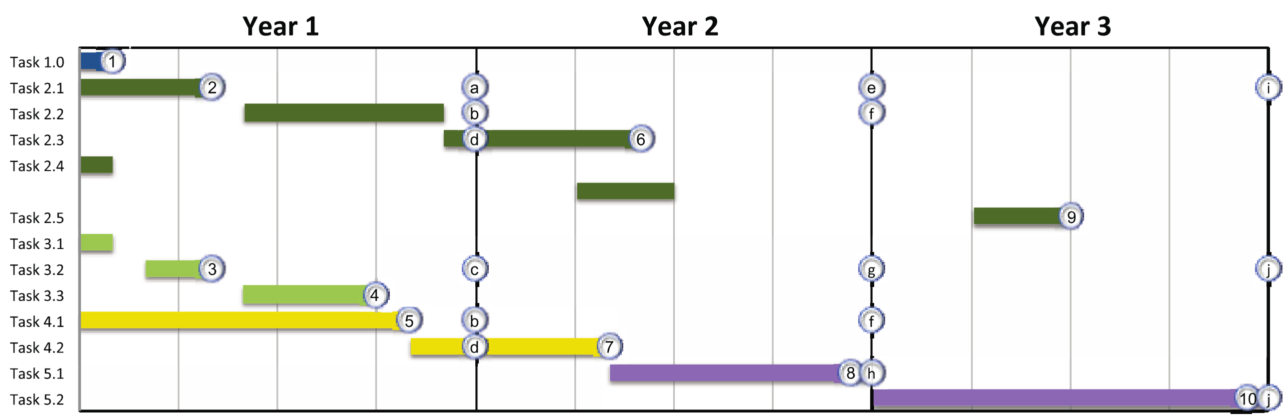

Figure 1: Project timeline, including times of activity (color bars), Milestones (numbered circles) and Success Criteria (lettered circles). A complete list of Milestones and Success Criteria are given in the Appendix.

Active Tasks this quarter included **Tasks 2.2** (2D microfluidic model visualization of fines migration), **2.3** (Microfluidic model visualization of NGHP-02 fines migration and clogging in a 2D pore network), **4.1** (Dependence of fines migration and clogging on physical conditions in porous media containing pure, endmember fines), and **4.2** (Dependence of fines migration and clogging on pore-fluid chemistry in porous media containing pure, endmember fines). A summary of accomplishments for each Task is provided below.

Task 2.2: 2D microfluidic model visualization of fines migration

This subtask is an opportunity to compare results with 2D micromodeling between LSU (Task 4), and the USGS (Task 2.2). Observations were made on endmember fines as a function of fines concentration and pore fluid chemistry to examine fines migration and permeability shutdown (clogging). In this quarter, the focus was on the measurement and interpretation of micromodel tests run at LSU. Plans to build micromodels at LSU and ship them to the USGS have been rescheduled for Fall 2017.

Results from the LSU studies completed thus far are provided in the sections covering Tasks 4.1 and 4.2.

Task 2.3: Microfluidic model visualization of NGHP-02 fines migration and clogging in a 2D pore network

Natural sediments generally contain mixtures of the pure, endmember fines selected for this study. Mapping out the response of each endmember provides a guide for estimating the response of real sediment. Measurements on sediment from the 2015 NGHP-02 coring program offshore eastern India offer opportunities to examine how effectively the endmember results can be used to predict the response of natural material. In this quarter, the USGS selected material from two high-value targets within the NGHP-02 program and made specimen splits with which to run 2D micromodel studies at LSU, but also run X-ray diffraction (XRD) studies at the USGS to determine the content of the fines fraction for each target. Reservoir modeling studies are ongoing at DOE/NETL for both targets. Preliminary results from the x-ray diffraction studies are pending follow-up measurements using techniques keyed to resolving the presence of phases that are challenging to capture in XRD spectra, such as montmorillonite (the primary component in the bentonite used as an endmember fine in this study).

Task 4.1: Dependence of fines migration and clogging on physical conditions in porous media containing pure, endmember fines

Fines existing in coarse-grained material can migrate due to pore-fluid flow, collect at pore-throats and clog flow pathways, reducing the overall permeability required for efficient methane extraction from hydrate-bearing sands. In this task, endmember fines were assessed in terms of their mechanical capacity to migrate and clog pores of various sizes relative to the grain size of the fines themselves. Accomplishments this quarter include:

- LSU micromodel tests for all endmember fines in water and brine are complete. For each fine-type and fluid, tests have been run on micromodels with pore throat widths of 10, 20, 40, 60, and 100 μm, plus a micromodel containing a random distribution of those pore-throat widths. For each combination, increasing concentrations of fines are used to establish a matrix describing the particle concentration and particle size/pore throat size geometry that result in apparent pore-throat clogging. For each endmember fine, a "clogging map" has been generated to visualize the specific combinations of mechanical process parameters that lead to clogging (Figures 4 and 6).

- Follow-up experiments were developed for clarifying the impact of a moving fluid/gas interface on clogging via enhanced fines concentrations at the fluid/gas interface. These simplified experiments using a flow tube provide an edge-on view of the moving meniscus, which helps refine conclusions drawn from the map-view perspective provided by the 2D micromodel imagery.

<u>Task 4.2: Dependence of fines migration and clogging on pore-fluid chemistry in porous media containing pure, endmember fines</u>

In addition to the mechanical drivers for pore-throat clogging by fines, there are chemical stimuli as well. Fine-grained particles are generally smaller than the pore throats they end up clogging, so the clogging behaviors described in Task 4.1 are caused when clusters or clumps of fine-grained particles form, growing large enough to span the pore throats. The tendency for fine-grain clusters to form depends strongly on interactions between the fines and the surrounding pore fluid. As discussed in the March 31, 2017 quarterly report, Task 3.2 (Electrical sensitivity of pure, endmember fines) was able to observe a fundamental difference in how pore fluid chemistry impacts clustering in particles such as silica silt and bentonite compared to that in illite and kaolinite. That distinction is apparent in the clustering observed in the 2D micromodel clogging studies as well:

- Particle/fluid interactions governed by the overall excess surface charge of the particle: Because of their small size, interactions between fines and their surroundings (fluid or other fines) can be strongly influenced by electrical interactions. Some fines, such as silica silt and bentonite, have interactions that stem primarily from the overall excess surface charge each particle possesses. The smaller the particle and the larger the excess surface charge density, the larger the effect can be, hence the larger electrical sensitivity of bentonite than silica silt. Because these particles all possess an overall negative electrical charge, they tend to repel each other and in fluids such as deionized water with very low ionic concentrations, this interparticle repulsion can hinder clustering. As shown in Figure 3a, silica particles in deionized water (DW) are evenly distributed throughout the micromodel, and show no signs of clogging pore throats. In contrast, Figure 3b shows the same concentration of silica particles in brine, which has a significantly higher concentration of ions than DW. The positively charged cations in the brine are attracted to the spaces between negatively charged particles, and their presence between particles generates attractive electrical interactions that cause silica particles to cluster.

In certain locations, these clusters clog pore throats. Figure 4 shows

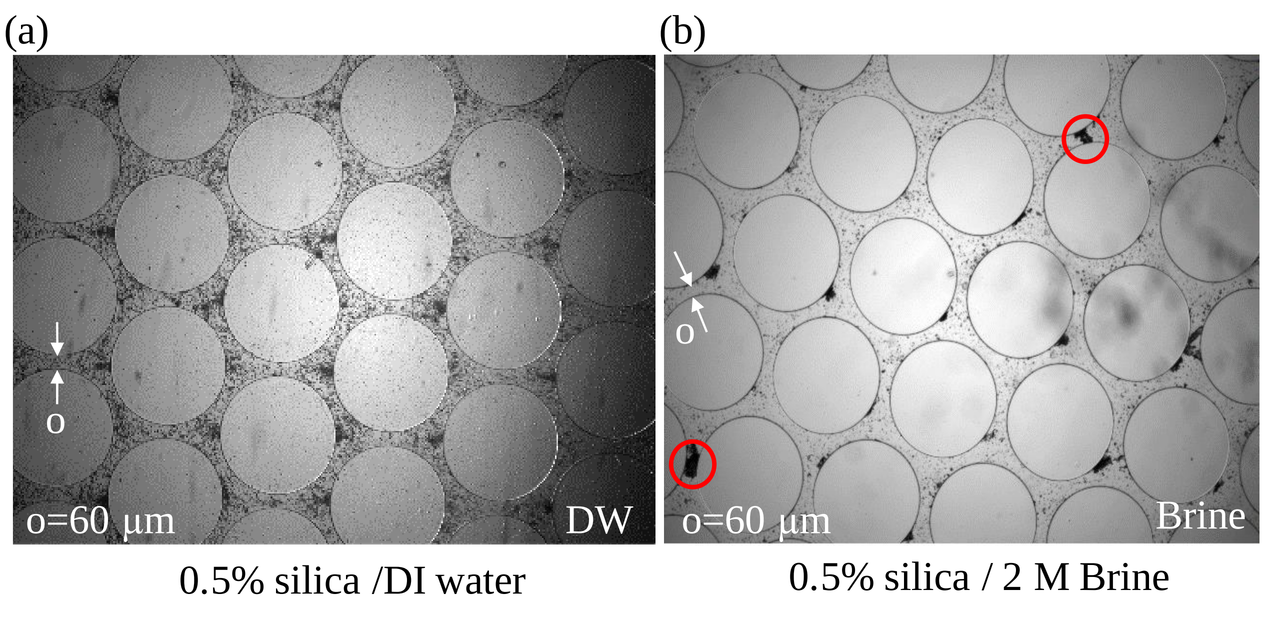

Figure 3: 2D micromodel images for silica. Pore throat width (distance between circular "particles") is 60μm, fines concentration is 0.5% by weight. (**a**) Silica particles suspended in deionized water (DW) are injected into the micromodel, saturating the micromodel and dispersing the silica particles (black flecks in the pore space) relatively homogenously. (**b**) Silica particles suspended in brine tend to cluster (small dark regions adjacent to circular "particles"), and these clusters can cause pore-throat clogging (highlighted by red circles).

the "clogging map" for these two silica silt cases. Relevant to Task 4.1, the map indicates clogging will occur only above a certain size ratio of fine particle size to pore throat size, and only above a certain concentration of fines in the pore fluid. As anticipated, for a constant silica silt size, clogging becomes more prevalent when either the pore throat size decreases (increased size ratio) or the concentration of fines increases. A second clogging boundary shows clogs form more easily in brine than in deionized water (clogging occurs at lower size ratios and lower fines concentrations).

- Particle/fluid interactions governed by the unbalanced distribution

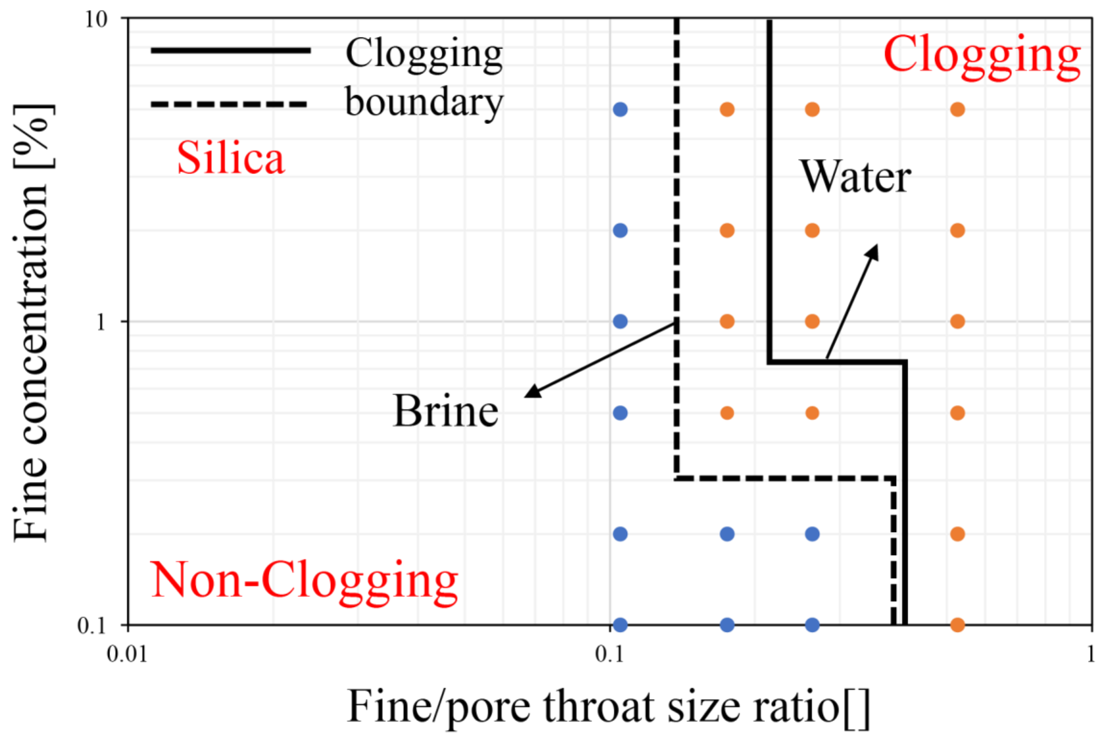

Figure 4: Clogging map for silica in deionized water ("water") and brine. Clogging occurs for conditions above and to the right of each boundary. The boundary is solid for the water case, and dashed for the brine case. Measured data points are shown as filled dots, with blue signifying no clogging in either case, and orange signifying clogging in brine or water. The shifting boundary shows that in brines, clogs form at lower silica concentrations, and for larger pore throats (smaller fine/pore throat size ratio), than for identical conditions in water.

of excess surface charge on the particle: Some particles, particularly platy particles such as illite or kaolinite, have unbalanced charge distributions. In the case of these platy particles, their flat surfaces tend to be negatively charged, while their edges tend to be positively charged. In the absence of ions in the surrounding fluid, these platy particles prefer to cluster in "edge-to-face" configurations which are fairly open and tend to occupy a large volume relative to the volume of the platy particles themselves. Because these clusters can grow large without requiring many particles, clusters can clog pore throats at relatively low concentrations of fines. In fluids with higher ionic concentrations, such as brines, the pore-fluid cations are attracted to the spaces between the negatively-charge platy particle faces, and in a similar fashion to the silica case described above, the presence of the cations generates an electrical attraction that allows the platy particles to cluster in more of a face-to-face configuration that makes much more efficient use of space. Consequently, for a cluster to grow large enough to clog a pore throat, a great many more particles have to join the cluster. For a given concentration of fines, clogging is therefore much more apparent in deionized water (Figure 5a) than in brine (Figure 5b). A "clogging map" for illite (Figure 6) shows this clogging boundary reversal relative to the silica case shown in Figure 4. These results demonstrate the importance of understanding the response of each endmember fine to a given pore-fluid chemistry change, such as the pore fluid freshening that occurs when gas hydrate is dissociated in situ to extract the methane as an energy resource. These 2D micromodel results are part of an abstract submitted to the 2017 Fall American Geophysical Union Conferences to be held December 11-15, 2017.

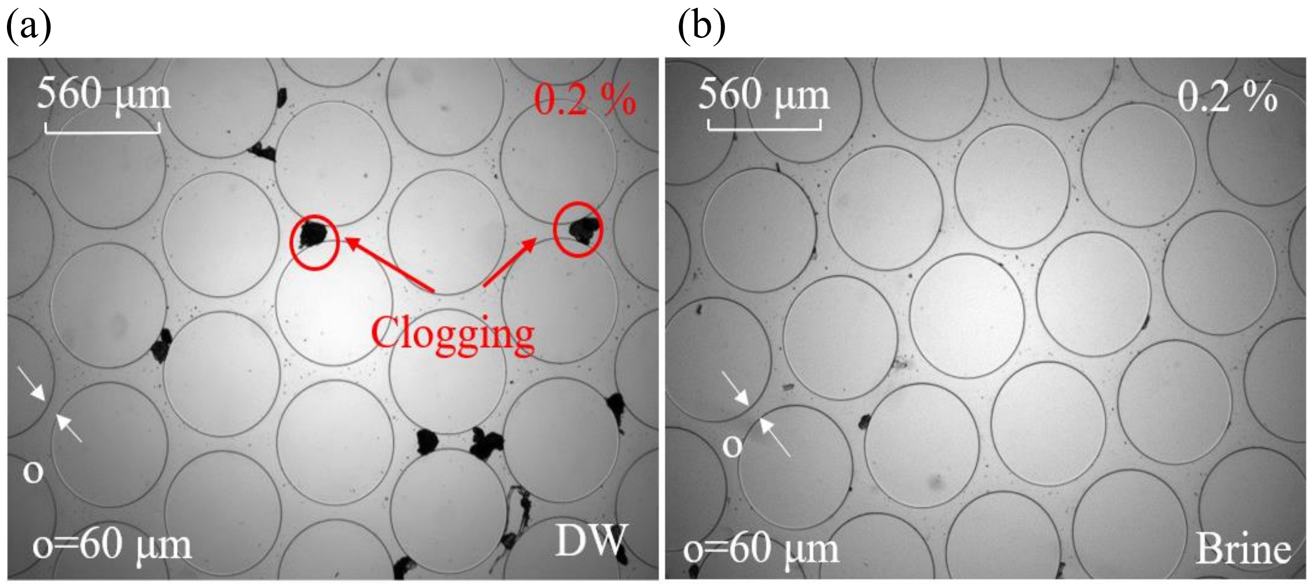

Figure 5: 2D micromodel images for illite. Pore throat width (distance between circular "particles") is 60µm, fines concentration is 0.2% by weight. (**a**) Illite particles suspended in deionized water (DW) are injected into the micromodel, clustering in large-volume, edge-to-face configurations (small dark regions adjacent to circular "particles") that are big enough to clog pore throats (highlighted by red circles). (**b**) Illite particles suspended in brine do not cluster as quickly, and clusters that do form (small dark regions) contain efficient face-to-face particle packings that do not occupy enough space to clog pore throats.

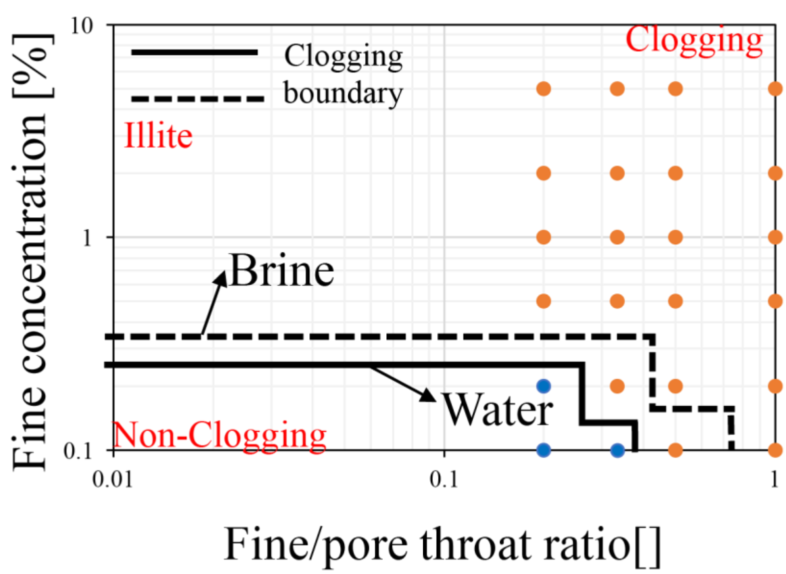

Figure 6: Clogging map for illite in deionized water ("water") and brine. Clogging occurs for conditions above and to the right of each boundary. The boundary is solid for the water case, and dashed for the brine case. Measured data points are shown as filled dots, with blue signifying no clogging in either case, and orange signifying clogging in brine or water. The shifting boundary shows that, opposite to the silica case, clogs form at lower illite concentrations, and for larger pore throats (smaller fine/pore throat size ratio) in water than for identical conditions in brine.

PRODUCTS

Cao, S.C., Jang, J., Waite, W.F., Jafari, M., Jung, J., A 2D micromodel study of fines migration and clogging behavior in porous media: Implications of fines on methane extraction from hydrate-bearing sediments [Abstract]. Submitted to the 2017 Fall American Geophysical Union Conference, New Orleans, LA, December 11-15, 2017.

Jang, J., Waite, W.F., Jung, J., Pore-fluid sensitivity of clays and its impacts on gas production from hydrate-bearing sediments [Abstract]. Accepted for the 9th International Conference on Gas Hydrates, June 25-30, 2017, Denver, Colorado.

Jang, J., Cao, S., Waite, W.F., Jung, J., Impact of pore-water freshening on clays and the compressibility of hydrate-bearing reservoirs during production. Conference paper submitted for the 9th International Conference on Gas Hydrates, June 25-30, 2017, Denver, Colorado.

APPENDIX: PROJECT TIMELINE & MILESTONE TRACKING

Figure A1 is the original complete Project timeline. Milestones and Success Criteria are listed thereafter, with updates given for elements in the current reporting period.

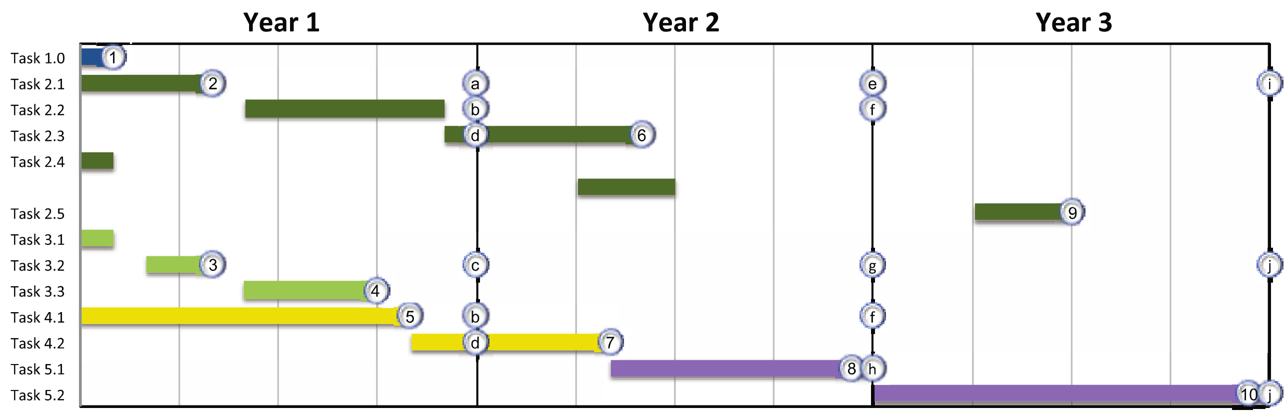

Figure A1: Original project timeline, including times of activity (color bars), Milestones (numbered circles) and Success Criteria (lettered circles). A complete list of Milestones and Success Criteria are given below.

<u>Milestones (listed according to the numbers given in Figure A1)</u>

Budget Period 1

1. Task 1, Project Management (LSU/USGS). This task will be completed October 31, 2016 and verified through DOE acceptance of the project SOPO, annual budget forecasts and Project Management Plan.

   <u>Status</u>: Completed. SOPO and PMP accepted by DOE. Kickoff meeting presentation complete.

2. Task 2, Site-specific pore fluid sensitivity study (USGS). This data acquisition component of Task 2 will be completed January 31, 2017 and verified through comparison of NGHP-02 data obtained with available shipboard data from the NGHP-02 cruise offshore India.

   <u>Status</u>: Initial phase of milestone completed. NGHP data has been collected on shipboard depressurized core material, but project will take the opportunity to collect additional data as pressure core material becomes available. Data will be integrated into a set of NGHP-02 special science volume papers currently with a February 2018 submission deadline.

3. Task 3, Endmember fines – electrical sensitivity index (USGS). This data acquisition component of Task 3 will be completed January 31, 2017. Results will be verified

through duplicate measurements of targeted specimens using LSU equipment, literature comparison where available.

> *<u>Status:</u> Completed. Data from this milestone have been incorporated into a conference paper and poster presented at the Ninth International Conference on Gas Hydrates (June 25- 30, 2017 in Denver, CO).*

4. Task 3, Endmember fines – dependence of compressibility and permeability on pore fluid chemistry (LSU). This data acquisition component of Task 3 will be completed June 30, 2017. Results will be verified through duplicate measurements of targeted specimens using USGS equipment.

> *<u>Status:</u> Completed. Data from this task is partly included in the conference paper and poster presented at the Ninth International Conference on Gas Hydrates (June 25-30, 2017 in Denver, CO). Remaining data are being incorporated into a manuscript for peer-reviewed journal publication.*

5. Task 4, 2D micromodel studies – mechanical contribution of endmember fines to clogging (LSU). This data acquisition component of Task 4 will be completed July 31, 2017. Results will be verified through duplicate measurements of targeted specimens using USGS equipment.

> *<u>Status:</u> LSU contribution completed. Data from this task is partly included in the conference abstract submitted to the Fall American Geophysical Union Conference (December 11-15, 2017 in New Orleans, LA). Remaining data are being incorporated into a manuscript for peer-reviewed journal publication. Micromodels to be used at the USGS will be constructed at LSU in the first quarter of BP 2 and shipped to the USGS.*

Budget Period 2

6. Task 2, 2D micromodel studies – mechanical contribution of NGHP-02 fines to clogging (USGS). This data acquisition component of Task 2 will be completed March 1, 2018. Results will be verified through linkages between imaged clogs and measured evolution of pressure and flow parameters.
7. Task 4, 2D micromodel studies – clogging dependence of endmember fines on pore fluid chemistry (LSU). This data acquisition component of Task 4 will be completed January 31, 2018. Results will be verified through duplicate measurements of targeted specimens using USGS equipment.
8. Task 5, 3D visualization of clogging and clog fracturing – dependence on endmember fines (LSU). This task has been removed from the project, which will now end upon completion of BP2.

Budget Period 3

9. Task 2, Site-specific dependence of compressibility and permeability on pore fluid chemistry (USGS). This data acquisition component of Task 2 will be completed March 31, 2019. Results will be verified for brines and freshened pore water by comparisons with pressure core data obtained elsewhere in the NGHP-02 project. This task will be shifted into BP2 and incorporated into an NGHP-02 special science volume paper with a February 2018 submission deadline.
10. Task 5, 3D visualization of clogging and clog fracturing – dependence on pore water chemistry (LSU). This task has been removed from the project, which will now end upon completion of BP2.

<u>Success Criteria (listed according to the letters given in Figure A1)</u>

End of Budget Period 1

a. Subtasks 2.1, 2.4: NGHP-02 fines properties (Offshore India). Index property measurements and liquid limit tests should have begun on NGHP-02 conventional core sediment. Additional index property and liquid limit tests can be run on NGHP-02 material as the material becomes available from pressure cores that were previously dedicated for USGS study during NGHP-02.

   <u>Status</u>: Initial phase of criteria completed. NGHP data has been collected on shipboard depressurized core material, but project will take the opportunity to collect additional data as pressure core material becomes available. Data will be integrated into a set of NGHP-02 special science volume papers currently with a February 2018 submission deadline.

b. Subtasks 2.2 and 4.1 (linked): 2D microfluid models – clogging via physical processes. Measurements of clogging by endmember fines should have been run separately by both participants. Results should be quantified in terms of clogging potential due to mechanical activity (fines migration) and geometry (pore throat size relative to grain size of the fines). Results should demonstrate similar behavior within the subset of LSU and USGS tests that are paired for interlaboratory verification purposes.

   <u>Status:</u> LSU contribution completed. Data from this task is partly included in the conference abstract submitted to the Fall American Geophysical Union Conference (December 11-15, 2017 in New Orleans, LA). Remaining data are being incorporated into a manuscript for the NGHP-02 special science volume, with a February 2018 submission deadline. Micromodels to be used at the USGS will be constructed at LSU in the first quarter of BP 2 and shipped to the USGS.

c. Task 3: Endmember fines assessment of pore fluid chemistry impact on compressibility and permeability. All data for a manuscript detailing the implications of the electrical sensitivity (pore fluid sensitivity) of fines on compressibility and permeability should be in hand, and a conference abstract prepared.

<u>Status</u>: Criteria complete. Conference paper and poster have been presented on this material at the Ninth International Conference on Gas Hydrates (June 25-June 30, 2017 in Denver, CO).

d. Subtasks 2.3 and 4.2 (linked): 2D microfluid models – clogging dependence on pore fluid chemistry. 2D micromodel experiments should have been started by both participants to assess the dependence of clogging by fines in relation to fluid chemistry. Initial comparisons between participants should guide subsequent efforts and dictate any additional tests that may need to be run.

   <u>Status:</u> LSU contribution completed. Data from this task is partly included in the conference abstract submitted to the Fall American Geophysical Union Conference (December 11-15, 2017 in New Orleans, LA). Remaining data are being incorporated into a manuscript for the NGHP-02 special science volume, with a February 2018 submission deadline. Micromodels to be used at the USGS will be constructed at LSU in the first quarter of BP 2 and shipped to the USGS.

End of Budget Period 2

e. Subtasks 2.1, 2.4: NGHP-02 fines properties (Offshore India). Index property measurements and liquid limit tests should continue on NGHP-02 pressure core sediment as the material becomes available from pressure cores that were previously dedicated for USGS study during NGHP-02. The publication moratorium should have expired in time to allow a conference abstract submission covering the NGHP-02 fines study to date.
f. Subtasks 2.2, 2.3 and Task 4: 2D Micromodel studies of clogging by endmember fines. All data for a manuscript detailing the implications of mechanical and chemical controls on clogging by endmember fines should be in hand. A joint manuscript should be submitted for peer reviewed journal publication, though the review process will likely be ongoing at the end of Budget Period 2.
g. Task 3: Endmember fines assessment of pore fluid chemistry impact on compressibility and permeability. Based on feedback from presenting this material at a conference, a peer-reviewed journal manuscript should have been written and submitted during this budget period, though the review process will likely be ongoing at the end of Budget Period 2.
h. Subtask 5.1: 3D micromodel imagery of the role of endmember fines in clogging, clog fracturing, and relative permeability. This Subtask is a 3D extension of the Subtasks 2.2 and 4.1 2D micromodel tests. By the end of Budget Period 2, comparisons between 2D and 3D observations of fines clogging based on mechanical and geometric factors should be providing insight into how the 2D micromodel results scale up to 3D, and these insights should be captured in a submitted conference abstract.

End of Budget Period 3

i. Subtasks 2.1, 2.4: NGHP-02 fines properties (Offshore India). Index property measurements and liquid limit tests should be complete on NGHP-02 pressure core sediment as the material. Based on feedback from presenting this material at a conference, a peer-reviewed journal manuscript should have been written and submitted during this budget period, though the review process for an NGHP-02 special volume may be ongoing even by the end of Budget Period 3.

j. Tasks 3 and 5: Interaction of fines with pore water – effect of pore water chemistry on index properties and flow behavior of endmember fines. Tying the macroscopic property insights from Task 3 with the 3D pore-scale behaviors observed in Task 5 provides the scientific content for the capstone publication in this project. Based on reviewer feedback from Task 3 and conference feedback from Subtask 5.1, a manuscript covering the interaction between fines and pore water and the subsequent impact on index and flow properties will be submitted.

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