



Gas hydrate research advances steadily on multiple fronts: A collection of published research (2009–2015)



1. Introduction

This article serves as the introduction and overview to the *Journal of Natural Gas Science and Engineering's* virtual special issue (VSI) on gas hydrates compiled in April 2015. The compilation includes forty-three articles addressing diverse topics related to gas hydrates that were published by *Journal of Natural Gas Science and Engineering* during the period 2009 to April 2015. Each article included in the compilation represents a stand-alone research topic that has passed the journal's rigorous peer-review editorial system. Collectively these articles provide key insight to the progress being made towards better understanding of gas hydrates, and their potential to be exploited as a resource, harnessed for other commercial applications, and inhibited from causing problems in other industrial processes.

The published articles compiled in this virtual special issue provide a useful synthesis of progress made in recent years in several diverse research areas pertaining to gas hydrates. These articles build upon a substantial existing body of research published in recent decades, of which that relating to the history of gas hydrate research, methane hydrates as a potential energy source and their risks with respect to global climate is recently presented in some detail by the United Nations Environment Program "Global Outlook on Gas Hydrates" publication volume 1 (Beaudoin et al., 2014a) and volume 2 (Beaudoin et al., 2014b). However, as this virtual special issue compilation demonstrates, there are many other areas of gas hydrate research worthy of attention that go beyond the exploitation of methane hydrates as a resource and the environmental risks associated with naturally occurring hydrate deposits. In order to make this synthesis more digestible than just an assemblage and listing of research articles, the articles are organized into related categories, and, in this introductory article, a brief summary is provided of the work conducted, and the conclusions drawn, by each article included in the compilation.

The articles compiled in this JNGSE virtual special issue are organized into eleven distinct categories:

1. Introduction to the JNGSE virtual special issue and summary of compiled articles (i.e., this article)
2. Reviews of gas hydrates as an energy source
3. Description, characterization and production performance of naturally occurring hydrate deposits
4. Seismic analysis to detect and characterize natural gas hydrate reservoirs
5. Tests and models to produce gas from gas hydrates
6. Formation of hydrocarbon hydrates and phase stability studies
7. Formation and phase stability of hydrates in pipelines and flowlines
8. Correlation studies providing insight to hydrate formation and inhibition
9. Inhibiting the formation of hydrates
10. Hydrates of CO₂ and THF: their stability and potential to aid the capture and sequestration of carbon dioxide
11. Potential exploitation of hydrates for gas storage, transportation and gas separation

JNGSE is also planning a special issue with the title "Gas Hydrates and Applications" with an expected publication date in 2016. That special issue will be in honour of Professor P. R. (Raj) Bishnoi for his outstanding contributions to gas hydrate research, and is being coordinated with contribution to a symposium of that title to be held at the 65th Canadian Chemical Engineering Conference. We hope that this virtual special issue compilation will contribute to the useful foundation of recent gas hydrate research for JNGSE 2016 special issue to build upon. This virtual special issue should also highlight to researchers JNGSE's keen interest in gas hydrates and our willingness to peer-review research articles on gas hydrates and related topics on an ongoing basis.

The brief summaries of the articles included in this virtual special issue now follow, organized into the categories described above.

2. Reviews of gas hydrates as an energy source

Makogon (2010) reviewed gas hydrates as a promising future source of energy. This commenced with a history of the discovery of natural gas hydrates, including a description of the characteristics and morphology of known gas deposits. Pressure–temperature equilibrium curves were presented for methane–water system delineating zones of hydrate formation and for other gases (i.e. C₃H₈, CO₂, H₂S and a natural gas mixture). The location of naturally-occurring, gas hydrate zones were identified and primary and secondary gas hydrates were distinguished: A primary deposit is one which does not melt after its formation (i.e. which is typical of deep water hydrates); A secondary deposit may be formed and melt repeatedly over its existence and require impermeable cap rocks in structural or stratigraphic traps in order to be preserved (i.e. such deposits are usually located in the Arctic onshore associated with natural gas reservoirs). Potential reserves of gas in hydrated deposits distributed were estimated at greater than $1.5 \times 10^{16} \text{ m}^3$. More than 230 gas hydrate deposits had been discovered up to 2010 with about 97% of those known natural gas hydrates located offshore, and only 3% located on land. The

characteristics of the commercially-producing Messoyakha gas hydrate field (Russia) were described, and compared on a P-T graph with other known gas hydrate deposits of different gas compositions. Experience in Messoyakha field showed that the cost required to produce gas from that hydrate deposit was about 15–20% higher than for a conventional gas field in the same area.

Koh et al. (2012) reviewed the available worldwide published knowledge base on natural gas hydrates as a resource. From their analysis they highlight the following points (1) there is substantial methane in hydrate reservoirs around the world; at least twice that available in conventional gas resources (2) the most accessible hydrates are in sandy sediments, with lithological controls, (3) laboratory characterization tools are available, (4) field detection tools are acceptable, (5) many of the national hydrate research programs are in the phase of resource identification and characterization, with two exceptions (U.S.A and Japan), and (6) the first long-term production tests of methane hydrates were expected to start imminently on the Alaska North Slope permafrost, and in deep water sediments offshore Japan.

3. Description, characterization and production performance of naturally occurring hydrate deposits

Makogon and Omelchenko (2013) described the history of commercial gas production from Messoyakha gas hydrate field (located in Siberia), 43 years after this, the only successfully-developed gas hydrate field was brought into production. Initial gas-in-place was estimated at up to $36 \times 10^9 \text{ m}^3$ (1271 BSCF) of which $24 \times 10^9 \text{ m}^3$ (847 BSCF) was likely to be present as free gas, and between 9 and $12 \times 10^9 \text{ m}^3$ (440 BSCF) present as hydrate. Cumulative gas produced from this reservoir was $12.9 \times 10^9 \text{ m}^3$ (455 BSCF) at the end of 2011 of which $5.4 \times 10^9 \text{ m}^3$ (190 BSCF) was obtained through hydrate decomposition. Cumulative water produced to 2011 was $48 \times 10^3 \text{ m}^3$ (1700×10^3 SCF) of which $45 \times 10^6 \text{ m}^3$ (1060×10^6 SCF) was obtained through hydrate decomposition. Reservoir pressure has remained constant for more than 25 years. The low water production rates have led to the formation of a water saturated zone between free gas and hydrate zone. An expected decrease in the reservoir pressure due to gas production was offset/balanced by additional the gas formed in the reservoir from hydrates decomposition. The position of the gas hydrate and free gas interface has continued to change during field development.

Uddin et al. (2014b) compared simulations of the expected hydrate dissociation response of the three hydrate bearing zones (lower, middle and upper) of the Mallik well 2L-38 to a single vertical well depressurization test. The Mallik gas hydrate deposit is located in the Mackenzie Delta of northern Canada, and in this study its characteristics and test results are described in detail, particularly the effects of salinity and hydration number in hydrate dissociation. A previously-developed model was extended, to better-history-match and represent, the physical and thermodynamic mechanisms involved with hydrate dissociation in the Mallik field test. The simulation model supports potential commercial gas production from the middle hydrate zone is feasible. On the other, hand the low pressure and low temperature conditions limit the commercial potential of the upper zone. The analysis suggest that a bottom aquifer determines water and gas flows in all three hydrate zones with the existing hydrate zones acting as an upward gas migration block. This latter characteristic has implications for well designs to might effectively exploit the deposit.

Sun et al. (2014) compared the production trial results and numerical simulations of gas production from the multilayer hydrate deposits in the Qilian Mountain permafrost (China). Both depressurisation and thermal stimulation methods were used in the 2011 production tests, which produced a total of 101 m^3 of natural

gas for 95 h. The focus of subsequent tests was to determine the hydrate saturation and to analyse the performance of different production methods using TOUGH β HYDRATE (T + H) simulator with a view to optimizing gas recovery. Detailed test results were reported. The simulation of depressurisation-induced gas production under different initial hydrate saturations showed that the simulated gas production was close to the actual value observed in the test. However, the hot steam injection simulation produced more than double the gas produced in the actual test, suggesting that the simulation model was over-simplified. The conclusions drawn suggest that depressurisation-induced production was relatively more effective than thermal stimulation for the low saturation hydrate deposits in the Qilian Mountain permafrost.

Marinakakis et al. (2015) reported results of laboratory tests with multi-component gas hydrates formed in the pore space of the clay-rich-treated-sediment samples retrieved from the Amsterdam subsea mud volcano located in deep water (i.e., 2000 m) in Eastern Mediterranean Sea. The gas hydrates discovered in this area occur just 40 cm below the sea floor, with relatively high seabed temperatures (i.e. ranging between 12°C and 14°C). The test results on the samples, focussing on permeability and consolidation behaviour, led the authors to conclude that gradual dissociation of the hydrates (e.g. due to minor changes in seabed temperature) could significantly change the integrity of the host deep-sea sediments. This work implies that in situ hydrate dissociation could be a possible cause of subsea landslides.

4. Seismic analysis to detect and characterize natural gas hydrate reservoirs

Luo et al. (2015) revealed the influence of heterogeneous hydrate distribution (i.e. high- and low-hydrate saturation; from 40% to 15%) on the compressional wave velocity of hydrate-bearing sediment. Simulation analysis revealed that the compressional wave velocity increases as the length of the high-hydrate saturation segment increases. This occurrence introduces uncertainty to models used to correct gas hydrate saturation estimates based on sonic wireline log data and seismic data recorded from sub-surface hydrate deposits.

Uddin et al. (2014a) explored the use of seismic data, and available well log data, to quantify the areally-heterogeneous distribution of the Mallik gas hydrate deposit located in the Mackenzie Delta of northern Canada. Two scenarios of variable/heterogeneous gas hydrate distributions within the Lower hydrate zone were simulated, with a single vertical well, comparing peak gas production with a homogeneous distribution case. The simulations indicate similar production profiles for each scenario, but with delays in peak gas production, and those delays being further increased when geomechanical factors are included in the simulation model, compared to a homogeneous reservoir scenario. The geomechanical effects contrast with gas production increases that would be expected for conventional gas reservoirs.

5. Tests and models to produce gas from gas hydrates

Tabatabaie and Pooladi-Darvish (2009) analysed gas production from hydrate reservoirs with zones of free gas underlying the gas hydrate zone. They developed a time-dependent-pressure-reduction model to predict constant rate gas production from flat hydrate reservoirs with hydrate dissociation behaving as a moving boundary problem with a sharp decomposition interface. Their model involves coupling the energy balance equation of 1D hydrate decomposition ahead of a moving interface with the material balance equation in a volumetric gas reservoir, and includes the thermodynamic relationships of hydrate decomposition. The

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