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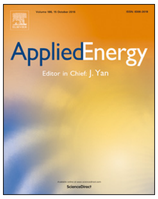

# Numerical analysis of experimental studies of methane hydrate dissociation induced by depressurization in a sandy porous medium

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## HIGHLIGHTS

- Numerical analysis of MH dissociation and fluids production by depressurization.
- A function describing the change in MH surface area is incorporated in the T + H code.
- Flow, thermal, kinetic parameters are optimized using a history-matching technique.
- Simulation validated by experiment elucidates the issue of spatial heterogeneity.

## ARTICLE INFO

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## ABSTRACT

Methane Hydrates (MHs) are a promising energy source abundantly available in nature. Understanding the complex processes of MH formation and dissociation is critical for the development of safe and efficient technologies for energy recovery. Many laboratory and numerical studies have investigated these processes using synthesized MH-bearing sediments. A near-universal issue encountered in these studies is the spatial heterogeneous hydrate distribution in the testing apparatus. In the absence of direct observations (e.g. using X-ray computed tomography) coupled with real time production data, the common assumption made in almost all numerical studies is a homogeneous distribution of the various phases. In an earlier study (Yin et al., 2018) that involved the numerical description of a set of experiments on MH-formation in sandy medium using the excess water method, we showed that spatially heterogeneous phase distribution is inevitable and significant. In the present study, we use as a starting point the results and observations at the end of the MH formation and seek to numerically reproduce the laboratory experiments of depressurization-induced dissociation of the spatially-heterogeneous MH distribution. This numerical study faithfully reproduces the geometry of the laboratory apparatus, the initial and boundary conditions of the system, and the parameters of the dissociation stimulus, capturing accurately all stages of the experimental process. Using inverse modelling (history-matching) that minimized deviations between the experimental observations and numerical predictions, we determined the values of all the important flow, thermal, and kinetic parameters that control the system behaviour, which yielded simulation results that were in excellent agreement with the measurements of key monitored variables, i.e. pressure, temperature, cumulative production of gas and water over time. We determined that at the onset of depressurization (when the pressure drop – the driving force of dissociation – is at its maximum), the rate of MH dissociation approaches that of an equilibrium reaction and is limited by the heat transfer from the system surroundings. As the effect of depressurization declines over time, the dissociation reaction becomes kinetically limited despite significant heat inflows from the boundaries, which lead to localized temperature increases in the reactor.

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# 1. Introduction

## 1.1. Background

Gas hydrates are solid crystalline compounds, which consist of gas molecules (called “guests”) that are trapped in a crystal lattice formed by water molecules. Typical guest gases are hydrocarbons with low molecular weight (e.g. $CH_4$, $C_2H_6$, $C_3H_8$), as well as other gases like $CO_2$, $N_2$ and $H_2S$ [1]. Depending on the attributes of the gas molecules, three primary hydrate structures have been identified, namely structure I (sI), structure II (sII) and structure H (sH) [2]. If the hydrate-forming gas is pure $CH_4$, the resulting methane-hydrate (MH) crystals have a sI structure, with two $5^{12}$ small cages and six $5^{12}6^2$ large cages. MH is thermodynamically stable under favourable low temperature (*T*) and high pressure (*P*) conditions.

MH occur abundantly (~20,000 TCM [3,4] in nature at (a) appropriate permafrost locations and (b) in oceanic deposits at and below the seafloor near the continental shelf [5]. The amount of $CH_4$ trapped in MH is vast, possibly twice as large as the equivalent of the combined world reserves of oil and gas [6]. An important feature of MH is that it has a high energy storage capacity: upon dissociation, 1 $m^3$ of MH releases 164 $m^3$ of $CH_4$ under STP condition [7]. Because of these reasons, MH is being considered as a potential future energy resource, and as such it has become the focus of considerable attention in several recent studies conducted by several countries (USA, Canada, Japan, S. Korea, China, India, Singapore, etc.).

Four different methods have been proposed so far to produce $CH_4$ gas from hydrates: thermal stimulation, depressurization, the use of inhibitors and gas exchange [6]. The working principle of these methods is based on (a) altering the reservoir $P/T$ condition to a region outside the MH stability zone, or (b) by shifting the hydrate equilibrium curve away from the original MH stability region [8]. A number of field tests have been carried out to investigate the technical feasibility of recovering $CH_4$ gas from hydrate reservoir at both permafrost [9–11] and marine locations [12]. A summary of these field tests, of the associated production methods and of the corresponding production rates are summarized in Table 1. Under the conditions of the limited-duration field tests conducted thus far, the gas production rate from hydrate deposits has not yet reached the level of economic viability of conventional gas reservoirs (which is considered to be on the scale of 100,000 $m^3$/day [13], but the production of gas from hydrates have been exhibiting a consistent (and promising) upward trend. Although considerable progress has been attained on the subject of gas production from natural hydrate deposits, technical challenges still remain in the areas of well design and completion, sand production and control, and optimization of fluids production [14].

Lack of knowledge on significant parameters that define key hydrate properties and behaviour, coupled with the limited number of field tests (which could provide some answers) and the significant challenges encountered in these field explorations [15], necessitate the study of MH-bearing sediments (an analogue of natural hydrate-bearing geologic media) under controlled laboratory conditions. Of particular interest is the investigation on the kinetic (dynamic) behaviour of MH formation and dissociation, as well as the associated behaviour of the reservoir fluids under a variety of conditions and flow regimes [16–21]. A common limitation of these studies is that hydrate saturation is estimated based on the measurement of *P* and *T* in the reactor on a bulk basis, without consideration of its spatial variability.

Recently, a variety of techniques have been applied in laboratory studies of MH formation and dissociation experiments in order to quantify the spatial distributions of in-situ phase saturations, mainly those of the hydrate phase ($S_H$) and of the aqueous phase ($S_A$). In-situ observations of $S_A$ during MH-dissociation under varying back-pressures were carried out using Magnetic Resonance Imaging (MRI) techniques [22]. This study determined that MH dissociation propagates radially inside a cylindrical testing apparatus, and that the resulting $S_A$ is spatially heterogeneous during dissociation. The same MRI technique was applied to investigate gas production under different initial $S_A$ conditions [23,24], and revealed that (a) higher $S_A$ occurs at the centre of the reactor after hydrate formation, and (b) that the heterogeneous water saturation (with high $S_A$ at the vessel bottom) impedes gas production during depressurization-induced dissociation. A more direct measurement on the spatial distribution of $S_H$ using MRI was carried out by Birkedal et al. [25]. This study indicates that higher saturation of $S_H$ is present in the vicinity of the end-piece of the vessel, while higher concentrations of $S_G$ is located at outer radial positions with significant excess water in the middle segment of the core. Similar MRI findings of spatially heterogeneous hydrate distribution was also reported in a study of MH formation in a silica sand bed inside a millimetre-scale reactor [26].

Besides MRI, X-ray Computed Tomography (CT) also demonstrated its ability to track $S_\mathrm{H}$ both spatially and temporally in a number of formation and dissociation studies on (a) hydrate cores extracted from natural field deposits [27–29] and (b) hydrate-bearing sediments synthesized in the laboratory [30–34]. These studies have provided visual evidence that the $S_H$ distribution in the samples is not uniform because of the uneven distribution and migration of water under gravity and capillary effect during hydrate formation [32]. Electrical Resistivity Tomography (ERT) is another experimental technique that has been used to describe the $S_\mathrm{H}$ spatial distribution in reactors. In their study of MH formation and dissociation behaviour in a meter-scale reactor, Priegnitz et al. [35,36] indicated that localized $S_H$ can deviate significantly from the bulk saturation estimate (computed for the entire reactor). These earlier experimental studies have consistently provided strong evidence on the nature of spatially heterogeneous $S_\mathrm{H}$ in laboratory apparatus.

In addition to experimental studies, a number of simulation studies have been carried out to model the behaviour of hydrate dissociation

**Table 1**
Summary of gas hydrate filed production tests.

| Site (Location) | Time | Method | Production duration (day) | Cumulative gas production volume ($m^3$) | Average gas production rate ($m^3$/day) |
|---|---|---|---|---|---|
| Mallik (Permafrost) | Mar 2002 | Thermal Stimulation [9] | 5 | 515 | 103 |
| | Apr 2007 | Depressurization [74] | 1 | 830 | 830 |
| | Mar 2008 | Depressurization [74] | 5.5 | 13,200 | 2400 |
| Ignik Sikumi (Permafrost) | May 2012 | Injection of $CO_2$ and $CO_2 + N_2$ [11] | 38 | 24,210 | 637 |
| Nankai Trough (Marine) | Jan 2013 | Depressurization [12] | 6 | 120,000 | 20,000 |
| | April 2017 | Depressurization [75] | 12 | 35,000 | 2917 |
| | June 2017 | Depressurization [75] | 24 | 200,000 | 8333 |
| Shenhu Area (Marine) | May 2017 | Depressurization [76] | 60 | 300,000 | 5000 |