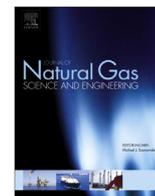




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Review of gas hydrate dissociation kinetic models for energy recovery

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ABSTRACT

In this work, a comprehensive review of gas hydrate dissociation kinetic models and hydrate reservoir simulators are provided. The main features of the dissociation kinetic models are summarized, with model formulation, assumptions, governing equations, solution method and limitations highlighted. A clear relationship between different kinetic models is identified according to the driving mechanisms and the progression of each hydrate dissociation kinetic model is presented to capture major improvements. Besides, key intrinsic kinetic parameters used in the kinetic models are extracted, which could be applied directly in future hydrate dissociation modelling study. Various aspects of hydrate reservoir simulator, including its theoretical basis, computational algorithms and applications have also been reviewed to provide a holistic view on the development of reservoir simulation for energy recovery from natural gas hydrates. Finally, further developments are suggested for improving the hydrate dissociation models relevant to practical applications of clathrate hydrates.

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1. Introduction

Gas hydrates are solid crystalline compounds that consist of water and gas molecules. Water molecules form cage-like crystal lattice through hydrogen bonds encaging guest gas molecules, which is stabilized by van der Waal's forces at certain temperature and pressure conditions (Davidson, 1973; Englezos, 1993; Sloan and Koh, 2008). In total, about 60 gas species form gas hydrates, including the most commonly seen methane, ethane, propane, and carbon dioxide etc. (Sloan, 2003b). The three main hydrate structures identified so far are cubic structure sI, cubic structure sII and hexagonal structure sH (Ripmeester et al., 1987; Sloan and Koh, 2008). Natural gas hydrates are usually found under deep sea and permafrost environment where the temperature is low and pressure is high. One key feature of clathrate hydrate is that it possesses the ability to contain large number of gas molecules effectively, resulting in a significantly high volumetric gas storage capacity: 1 volume of methane hydrate may contain ~170 volume of methane (STP) (Sloan and Koh, 2008) in theory. Thus, gas hydrates have given rise to numerous applications in major areas of water, energy and environment. Several novel applications are being explored in

recent decade by using gas hydrates as a technology enabler. These include CO₂ capture (Babu et al., 2013; Duc et al., 2007; Kang and Lee, 2000; Komatsu et al., 2013; Kumar and Kumar, 2015; Li et al., 2010; Linga et al., 2007; Park et al., 2013), seawater desalination (Babu et al., 2014; Javanmardi and Moshfeghian, 2003; Kang et al., 2014; Park et al., 2011), energy storage (Chatti et al., 2005; Englezos and Lee, 2005; Florusse et al., 2004; Gudmundsson et al., 1994; Sun et al., 2003; Veluswamy et al., 2016), refrigeration (Darbouret et al., 2005; Douzet et al., 2013; Fournaison et al., 2004; Obara et al., 2011; Xie et al., 2010), and other separation applications (Cha et al., 2010; Seo and Lee, 2001; Seo et al., 2004). Fundamental information on gas hydrates is available in a book (Sloan and Koh, 2008) and in some targeted specific review papers (Babu et al., 2015; Chong et al., 2016; Davidson, 1973; Englezos, 1993; Eslamimanesh et al., 2012; Koh, 2002; Koh et al., 2012; Kumar et al., 2015; Sum et al., 2009; Veluswamy et al., 2014; Xu and Li, 2014).

The study of gas hydrate is comprehensive, involving multi-scale analysis. The understanding of gas hydrates started from molecular level whereby researchers investigated the different types of gas hydrate structures; then their time-independent thermodynamic properties and phase equilibrium (Davidson, 1973; Englezos, 1993; Sloan and Koh, 2008). Subsequently, the investigation of gas hydrate formation and dissociation process is approached from both microscopic and macroscopic level to develop kinetic models to fully simulate the physics behind the

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process (Bishnoi and Natarajan, 1996; Englezos et al., 1987; Kim et al., 1987; Vysniauskas and Bishnoi, 1983). Production of natural gas from hydrate reservoir and application of gas hydrate as a medium, such as flow assurance, hydrate-based seawater desalination and energy storage, build upon the macroscopic-level understanding of hydrates formation and dissociation, which also involve other large-scale process design parameters.

Examining most hydrate related industrial and academic problems, most of the time it boils down to the most challenging and intriguing question – how gas hydrates form and dissociate against time. Compared with time-independent studies investigating hydrate thermodynamic property, molecular structures and phase equilibrium (Sloan, 1998, 2003a), the endeavour to use kinetic models to understand the time-dependent behaviour of hydrate formation and dissociation only started in mid 1980s (Englezos et al., 1987; Kim et al., 1987; Vysniauskas and Bishnoi, 1983, 1985). Even until now, the kinetic behaviour of hydrate formation and dissociation is still not completely understood compared with its thermodynamic behaviour. Besides, the mechanisms behind the kinetic behaviour of hydrate formation and dissociation still need to be elucidated at multiple scales.

A good knowledge of hydrate formation kinetics could benefit us in two folds: to promote the hydrate formation rate, which is favourable from energy storage and other technological applications perspective; to inhibit the hydrate formation rate, which is beneficial in hydrate plug formation and hydrate risk management from flow assurance perspective. Similarly, a better understanding of hydrate dissociation kinetics can assist in natural gas production from energy recovery perspective and hydrate plug remediation from flow assurance perspective. In comparison, the study of hydrate formation process is more complicated than hydrate dissociation process, partly because hydrate formation involves two inter-related processes: a stochastic hydrate nucleation process and a hydrate growth process controlled by kinetics, heat and mass transfer limitations (Sloan and Koh, 2008). A complete understanding of hydrate formation requires the incorporation of nucleation theory into traditional heat and mass transfer problem; whereas hydrate dissociation process is essentially a multiphase-multicomponent heat and mass transfer problem coupled with intrinsic kinetics and fluid flow behaviour (Sloan and Koh, 2008).

The focus of this review is on the development of gas hydrate dissociation kinetic models towards the evolution of hydrate reservoir simulators in the recent decades. General concept of hydrate dissociation and the benefit of understanding the process will be introduced first before proceeding to the detailed review on hydrate dissociation kinetic models and reservoir simulators. The formulation of various kinetic models, key assumptions, governing equations, solution method, and limitations are summarized with the relationship between various models identified according to the main driving mechanism. Key issues and knowledge gaps for currently available kinetic models are pointed out with possible directions for further work suggested. Upon appreciating the development of various kinetic models, the review proceeds towards the development of reservoir simulation tools, which includes an overview of their theoretical basis, computational algorithm, and application in various production tests.

2. Hydrate dissociation

2.1. General concept and modelling of hydrate dissociation

Hydrate dissociation is an endothermic process in which heat must be supplied to break the hydrogen bonds between water molecules and the van der Waals interaction forces between guest molecule and water molecule of the hydrate lattice. The product of

the hydrate dissociation process is water (host) and gas (guest) molecules. Several conventional methods have been proposed for the production of natural gas from hydrate reservoir, including depressurization, thermal stimulation, chemical inhibitor injection, or a combination of these methods (Collett et al., 2015; Englezos, 1993; Haligva et al., 2010; Makogon, 1997; Sloan and Koh, 2008; Sum et al., 2009). The first and most popular method is depressurization, which involves reducing the pressure below the hydrate stability zone. The second method is thermal stimulation, which requires increasing the temperature above the natural gas hydrate equilibrium temperature at the prevailing pressure. The third method is chemical inhibitor injection, which can be differentiated into two different types: thermodynamic inhibitor and kinetic inhibitor injection (Sloan and Koh, 2008). Chemical inhibitor injection calls for the addition of strong hydrogen-bonding chemicals (such as methanol or ethylene glycol), leading to a shift in gas hydrate equilibrium curve. In contrast, kinetic inhibition allows system to exist within the hydrate stability zone, while small crystals are stabilized without agglomerating into larger hydrate mass (Koh et al., 2002; Lederhos et al., 1996; Notz et al., 1996; Sloan et al., 1998). Current research is shifting from hydrate avoidance using thermodynamic inhibitors towards hydrate risk management, whereby kinetic hydrate inhibitors (KHI) and anti-agglomerates (AA) are preferred to delay hydrate formation and prevent the adherence of hydrate particles respectively (Sloan, 2005; Sum et al., 2009).

Hydrate reservoir exploration and field production tests from hydrate reservoirs are another major area related to hydrate dissociation process. Field tests have been carried out in both onshore and offshore environment. These hydrate field tests include the Mount Elbert well test (Hunter et al., 2003), Ignik Sikumi field trials in Alaska North Slope by CH₄–CO₂ exchange (White and Lee, 2014), Malik gas hydrate site test in Canada (Yamamoto and Dallimore, 2008a) and MH-21 Japan Nankai Trough hydrate well test by depressurization (Tsuji et al., 2004). These field trials involve high capital investment, long planning and execution period, arduous working environment and unpredictable operations, therefore require strong collaboration effort between government, corporations and research institutions. Therefore, there is a critical need to pursue cutting edge research developments in a laboratory setting at multi-scale levels to understand the hydrate dissociation characteristics to devise the methods and equipment for realizing the potential of energy recovery from natural gas hydrates. The most critical issue before the execution of any field test is the selection of appropriate hydrate bearing zones and the estimation of gas production behaviour under different production strategies. Therefore, fundamental understanding of hydrate dissociation kinetic models is essential to model hydrate reservoir dissociation process for energy recovery from natural gas hydrates.

Over the past four decades, several mathematical models and numerical codes have been developed to investigate methane hydrate decomposition and methane gas production behaviour. For a mathematical model to be representative, it should couple all the primary dissociation mechanisms, including heat and mass transfer, gas and water flow behaviour, intrinsic decomposition kinetics in pure hydrate system or effective decomposition rate for hydrate dissociation in porous medium. These three mechanisms involved during hydrate dissociation are of equal importance; coupled with each other and either of them could be the limiting factor under different conditions. Intrinsic decomposition rate of methane hydrate is controlled by temperature, pressure, interfacial area and the intrinsic rate constant (Kim et al., 1987). It affects the heat transfer rate from surrounding to hydrate core due to the endothermic characteristics of the reaction. In addition, flow of the decomposed gas and water through the porous media could also be

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