



Microscale effects on methane hydrate dissociation at low temperature in the micro porous media channels by depressurization



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ABSTRACT

As for the dissociation of methane hydrate accompanied by multiphase multicomponent flow through micro porous media channels, it is necessary to investigate the microscale effects on the multiphase flow behavior as well as the mass and heat transfer between phases. Meanwhile, the interaction between hydrate dissociation or reformation and ice formation or melting is one of the most important factors. In this paper, with the consideration of ice formation/melting and heat transfer between phases, the momentum and energy equations for all phases and a comprehensive Users' Defined Subroutine (UDS) are solved in a proposed numerical model to investigate methane hydrate dissociation. The reliability of current model has been proved by comparing with the experiment data and previous simulation results in a good agreement, and then the model was used to study the methane hydrate dissociation at low temperature in the micro porous media channels. The microscale effects on methane hydrate dissociation was studied in two cases (with/without microscale effects), and our results showed that water temperature, volume fraction of phases, ice distribution, hydrate dissociation rate, methane velocity, mass transfer rate between water and ice and saturation of phases are affected seriously by the microscale effects. Especially the water temperature, which is one of the critical factors for hydrate dissociation, shows an unstable change when the microscale effects is ignored.

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1. Introduction

Methane hydrate, which is a solid compound containing water and methane gas, has been viewed as a potentially new type of energy resource for the future [1,2]. A huge number of methane hydrate is formed in permafrost and under ocean regions under suitable conditions of high pressure and low temperature [3–5].

In recent years, considerable efforts have been made to understand the hydrate dissociation and find suitable hydrate mining methods [6–12]. Methane hydrate dissociation involves heat and mass transfer among four phases (solid methane hydrate phase, gaseous methane phase, liquid water phase, and solid ice phase) and three components (methane hydrate, methane, water, ice). Moreover, it always takes place inside the porous media. Transport properties of the hydrate-bearing porous media and heat and mass transfer between phases are significantly affected by the fluid dynamic behaviors of water and methane. The dissociated gaseous methane significantly increases the pressure inside the pore space,

and the methane also increases the interactive force between fluid and hydrate-bearing porous media, these phenomena may result in the reformation of hydrate [13,14], and the deformation of hydrate-bearing sediments [15,16]. The endothermic methane hydrate dissociation can lead to ice formation, and the ice can destroy or block the micro porous media channels. In addition, since ice formation is an exothermic process, and it makes a contribution to methane hydrate dissociation to some extent. These interactions make the study for methane hydrate dissociation become quite complex. Furthermore, when the characteristic length of the porous media is small, the flow can fall into the slip region, so the microscale effect should be taken into account, this raises the difficulties in understanding the dissociation of methane hydrate. Three popular methods are accepted to extract methane gas from hydrate reservoirs: thermal stimulation [17] (increasing the temperature), depressurization [18] (reducing the pressure) and inhibitor injection [2,4]. Restricted by high energy consumption of the thermal stimulation and environmental problem of the inhibitor injection, depressurization method is currently considered to be the most promising approach for methane hydrate mining [8].

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Nomenclature

A_d	surface area of hydrate per unit volume (m^2)	Q_{sp}	heat transfer between solids surface and phase q
h_g	specific enthalpy of the methane (J/kg)	R	universal gas constant ($\text{J}/(\text{mol k})$)
h_w	specific enthalpy of water (J/kg)	$S_{\emptyset,q}$	source term of phase q
h_h	specific enthalpy of hydrate (J/kg)	S_g	saturation of methane
h_{pq}	interphase enthalpy between the p and q phases (J/kg)	S_w	saturation of water
h_{eff}	effective convective heat transfer coefficient	S_h	saturation of hydrate
K_{DO}	absolute permeability of porous medium (md)	S_{gr}	irreducible saturations of methane
k_d^0	intrinsic dissociation constant ($\text{mol}/(\text{m}^2 \text{Pa s})$)	S_{wr}	irreducible saturations of water
k_B	Boltzmann constant, 1.38066×10^{-23} (J/K)	S_{sh}	source term due to the endothermic reaction
L	characteristic geometric length of the flow domain	T_s	solids surface temperature (K)
\dot{m}_{hg}	mass transfer rate from hydrate to methane ($\text{kg}/(\text{m}^3 \text{ s})$)	T_f	water freezing temperature (K)
\dot{m}_{hw}	mass transfer rate from hydrate to water ($\text{kg}/(\text{m}^3 \text{ s})$)	UDS	Users' Defined Subroutine
\dot{m}_h	mass dissociation rate for methane hydrate ($\text{kg}/(\text{m}^3 \text{ s})$)	u	velocity components that are parallel to the wall
\dot{m}_w	mass generation rate of water ($\text{kg}/(\text{m}^3 \text{ s})$)	v	velocity components that are normal to the wall
\dot{m}_g	mass generation rate of methane ($\text{kg}/(\text{m}^3 \text{ s})$)	\vec{v}_g	velocity of methane (m/s)
M_g	molecular weight of methane (kg/mol)	\vec{v}_w	velocity of water (m/s)
M_w	molecular weight of water (kg/mol)	\vec{v}_{hg}	interphase velocity between hydrate and methane (m/s)
M_h	molecular weight of hydrate (kg/mol)	\vec{v}_{wg}	interphase velocity between water and methane (m/s)
N	permeability reduction index	ρ_q	phase density of phase q (kg/m^3)
P_g	methane pressure (Pa)	α_q	volume fraction of phase q
P_{eh}	equilibrium pressure (Pa)	ΔH_d	latent heat of hydrate dissociation (kJ/kg)
Q_{pq}	intensity of heat exchange between p and q phases	ΔE	activation energy
		λ	mean free path of the molecules

Methane hydrate dissociation has been a hot topic of both experimental and numerical studies for many years [19]. Due to the limitations of real-scale studies and the complex of real reservoir conditions, many studies are mainly focus on the lab-scale research. In terms of experimental research, investigations are performed both in field laboratories and conventional laboratories on natural samples [20–22] and laboratory-synthesized [23–25] samples to understand the thermodynamic properties (e.g. the equilibrium curve, effect of pore size, effect of inhibitor, hydrate structure), gas composition, mechanical, thermal and hydrologic properties of hydrate-bearing sediment. However, due to the limitations of experimental conditions and lack of appropriate experimental methods, it is difficult to investigate some critical factors mentioned above inside the micro porous media channels. As for the numerical investigations, Kim et al. [26] studied the kinetic methane hydrate dissociation by using a semi batch stirred-tank reactor, and the hydrate dissociation rate model was developed based on the experimental results. Then, Yousif et al. [22] developed a pioneering one-dimension numerical model for lab-scale by extending the Kim-Bishnoi's kinetics equation to simulate the process of isothermal hydrate dissociation by depressurization. They also predicted the evolutions of pressure and dissociation location. With the application of kinetic model of Kim et al. [26],

Masuda et al. [27] developed a finite difference simulator to study the methane hydrate dissociation. And they also conducted an experiment using Berea sandstone cores. The experimental data was employed as a classic database to validate the modeling results by other researchers. Besides, the methane hydrate dissociation in the porous media was widely studied by various models [16,27–33]. The models of Masuda et al. [27], Yousif et al. [28], Clarke and Bishnoi [29,30], Nazridoust and Ahmadi [31], Zhao et al. [32], and Sun et al. [16,33] have been used to simulate. A comparison among these models is summarized in Table 1. It can be seen that several simplifications are made to reduce the complexities of the research. One of the simplifications is the application of Darcy's Law for multiphase flow to decouple the calculation of fluid velocities in porous media. However, the accuracy of the velocity calculation is limited because slip flow of gas cannot be considered in the model based on Darcy's law [34]. As a result, these model based on Darcy's law is not suitable when the flow lies in slip region. In addition, the single energy equation shared among phases also affects the accuracy of the temperature calculation.

As stated previously, methane hydrate dissociation always takes place in the porous media, and actually, the hydrate always decomposes inside the micro porous media channels. Sometimes,

Table 1
Comparison between our model and representative models of available studies.

	Kim et al. [26]; Clarke and Bishnoi [29,30]	Ruan et al. [1]; Zhao et al. [18]; Masuda et al. [27]; Nazridoust and Ahmadi [31]; Sun et al. [16,33]	Current study
Dissociation kinetics	✓	✓	✓
Heat transfer	×	✓	✓
Calculation of fluid flow	×	Darcy's law	Momentum equation
Energy equation	×	A single equation	Equations for all phases
Heat transfer between phases	×	×	✓
Effect of ice phase	×	×	✓
Microscale effects	×	×	✓

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