



Theoretical Analysis of Gas Hydrate Dissociation in Sediment

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Abstract. Theoretical analysis was carried out to investigate the dissociation of gas hydrate in stiff sediment. First the mathematical model for gas hydrate dissociation was decoupled by asymptotic expansion method considering the order differences of time scales among seepage, dissociation of gas hydrate and heat conduction. The multi-scale perturbation method was used to solve the problem. It is shown that seepage is the fastest process. The heat conduction is the slowest process. With the pressure decreases at the boundary, pressure changes first while no hydrate dissociation and heat conduction occur. Gradually, dissociation causes the decrease of temperature. After a long time, heat can conduct to cause the increase of temperature. Otherwise, the decreased temperature will cause the stop of dissociation if no heat is supplied in time.

Keywords: Gas hydrate · Dissociation · Mathematical model

1 Introduction

Natural gas hydrate (NGH) is treated as a potential energy resource for the 21st century because a large amount of NGH is trapped in hydrate reservoirs though it can induce the large deformation [1] and even kinds of hazards in some cases such as marine landslide, crack extensions [2]. In the past 30 years, considerable effort has been made for commercial production of NGH from hydrate reservoirs. Until now, all methods are still limited to experimental scale, except for one gas-hydrate field in western Siberia, which was exploited successfully [3].

To recover NGH from hydrate reservoir, depressurization, temperature falling and replacing are three presented methods. Extensive reviews of NGH reported by Englezos [4] and Sloan [5]. Selim and Sloan [6], Tsyppkin [7] and Lu et al. [8] presented different methods for analysis of the dissociation process of NGH by depressurization. In those models, various assumptions were adopted in order to obtain the analytical solution, such as the water in the reservoir remained stationary and the well temperature was kept constant. Moridis et al. [9] added a module for NGH dissociation into the TOUGH2 (general-purpose reservoir simulator). Swinkels and Drenth [10], Zhang et al. [11] and

Ji et al. [12] used thermal stimulation for NGH dissociation and studied the enthalpy of dissociation for hydrates formed by different gases.

Either the formation or the recovery of NGH is related with the thermal conduction/convection, migration of gas and water, dissociation of gas hydrate, stress/deformation variation of stratum. These processes are very complex because of the multi-components and complex sediment's conditions such as fracture system [13, 14]. To analyze the recovery of gas hydrate, these four processes must be clarified.

In this paper, theoretical analysis on NGH dissociation was studied. The governing equations were first decoupled into three courses by asymptotic expansion method: seepage, dissociation of NGH and heat conduction, considering the order differences of the time scales. Then solutions were given under two-dimensional conditions.

2 Formation of Problem

We consider the hydrate-bearing sediment constituted of free methane (gas), methane hydrate (solid), water (liquid) and soil/rock skeleton (stiff solid). The four components are in thermodynamic equilibrium. The formation is assumed to be uniform and the flow of free methane and water can be regarded as flow through a porous media with porosity $\varepsilon_g + \varepsilon_w$, which are fractional volume of gas and water. Soil/rock skeleton is assumed to be stationary. The flow of gas and water in pores obeys the Darcy law. Though the permeability of gas and water changes with the properties of media such as porosity, components, they are assumed as constants for simplicity of analysis.

Dissociation will happen once the pressure and temperature changes. Seepage and temperature change inside the sediment will occur also once the pressure changes at the boundary.

The fractional volume of each component is ε_g , ε_h , ε_w and ε_m respectively and

$$\varepsilon_g + \varepsilon_w + \varepsilon_h + \varepsilon_m = 1 \quad (1)$$

in which ε_h , ε_g , ε_w , ε_m are fractions of hydrate, gas, water and skeleton, respectively. For a unit volume, ε_m is a constant, ε_g , ε_h , ε_w will change with the dissociation of gas hydrate.

Methane (gas) satisfies the ideal gas law, i.e.

$$pV = NRT \quad (2)$$

in which p is the pressure, V is the volume, N is the Mole number of gas, R is the ideal gas constants, T is the temperature.

Mass conservation equations are as follows by neglecting diffusion term:

$$\begin{aligned} \frac{\partial \varepsilon_g \rho_g}{\partial t} + \nabla \cdot \varepsilon_g \rho_g \bar{u}_g &= -\lambda \rho_h \frac{\partial \varepsilon_h}{\partial t} \\ \frac{\partial \varepsilon_w \rho_w}{\partial t} + \nabla \cdot \varepsilon_w \rho_w \bar{u}_w &= -(1 - \lambda) \rho_h \frac{\partial \varepsilon_h}{\partial t} \end{aligned} \quad (3)$$

in which ρ_g , u_g are density and velocity of gas, ρ_w and u_w are density and velocity of water, $\chi = M_g/M_h$, M_h and M_g are Mole numbers of gas hydrate and methane gas, respectively.

Momentum conservation equations are as follows by neglecting inertia,

$$\begin{aligned}\bar{u}_g &= -\frac{K_g}{\varepsilon_g \mu_g} \nabla p \\ \bar{u}_w &= -\frac{K_w}{\varepsilon_w \mu_w} \nabla p\end{aligned}\quad (4)$$

in which K_g and K_w are relative permeabilities of gas and water, respectively, μ_g and μ_w are cohesions of gas and water, respectively.

The energy conservation equation is

$$\begin{aligned}\varepsilon_g \rho_g \frac{\partial C_g T}{\partial t} + (\varepsilon_w \rho_w C_w + \varepsilon_h \rho_h C_h + \varepsilon_m \rho_m C_m) \frac{\partial T}{\partial t} + \varepsilon_g \rho_g u_g \cdot \nabla C_g T + \varepsilon_w \rho_w C_w \bar{u}_w \cdot \nabla T \\ = [\Delta H + \chi C_g T + (1 - \chi) C_w T] \rho_h \frac{\partial \varepsilon_h}{\partial t} + K \nabla^2 T\end{aligned}\quad (5)$$

in which C_g , C_w , C_h , C_m are thermal capacities of gas, water, hydrate and skeleton respectively, ΔH is the latent heat, K is the heat conduction coefficient which is assumed to be a constant.

The dissociation rate of NGH is

$$\frac{\partial \varepsilon_h}{\partial t} = -k_d M_g A_s (f_e - f) \quad (6)$$

in which A_s is the area, k_d are coefficients, f_e and f are the three-phase equilibrium fugacity and methane fugacity, respectively. In the following analysis, they are substituted by the phase equilibrium pressure of methane hydrate and pore pressure in the hydrate-bearing sediment. Generally, k_d changes with temperature or pore pressure. For simplicity, it is assumed as a constant here. The above equations can be normalized by a characteristic specific heat C , T_0 and ρ_h and p_0 . T_0 and p_0 can be chosen as the initial temperature and pressure of the media.

$$\begin{aligned}\frac{\partial \varepsilon_g \bar{\rho}_g}{\partial \tau} + \nabla (\bar{\rho}_g \cdot \nabla \bar{p}) &= -\chi \bar{\rho}_h \frac{\partial \varepsilon_h}{\partial \tau} \\ \frac{\partial \varepsilon_w}{\partial \tau} + \nabla^2 \bar{p} &= -(1 - \chi) \bar{\rho}_h \frac{\partial \varepsilon_h}{\partial \tau} \\ \frac{\partial \varepsilon_h}{\partial \tau} &= \bar{k}_d M_g \bar{A}_s (\bar{f}_e - \bar{f})\end{aligned}\quad (7)$$

$$\begin{aligned}
& \varepsilon_g \bar{\rho}_g \frac{\partial \bar{C}_p \bar{T}}{\partial t} + (\varepsilon_w \bar{\rho}_w \bar{C}_w + \varepsilon_h \bar{\rho}_h \bar{C}_h + \varepsilon_m \bar{\rho}_m \bar{C}_m) \frac{\partial \bar{T}}{\partial t} \\
& \quad - \frac{p_0 k_g}{\mu_g} \bar{\rho}_g \nabla \bar{p} \cdot \nabla \bar{C}_p \bar{T} - \frac{p_0 k_w}{\mu_w} \bar{\rho}_w \bar{C}_w \nabla \bar{p} \cdot \nabla \bar{T} \\
& = \left[\frac{\Delta H}{C T_0} + \chi \bar{C}_p \bar{T} + (1 - \chi) \bar{C}_w \bar{T} \right] \frac{\partial \varepsilon_h}{\partial t} + \frac{K}{C \rho_h} \nabla^2 \bar{T}
\end{aligned} \tag{8}$$

For simplicity, the “-” in the equations are omitted. In this problem, let $\tau = p_0 k_g t / (\mu_g r_0^2)$, $\zeta = x / r_0$. Since $\eta_1 = C \rho_h \mu_g / (K p_0 k_g)$ and $\eta_2 = K \alpha A_s \mu_g / (p_0 k_g)$ are two small parameters, we can give the asymptotic expansions by using of multi-scale method:

$$\begin{cases} f = f^{(0)}(x_i, \tau_0, \tau_1, \tau_2) + \sum_{n=1}^{\infty} \eta_2^n f^{(n)}(x_i, \tau_0, \tau_1, \tau_2) \\ f^{(0)} = f^{(0)}(0)(x_i, \tau_0, \tau_1, \tau_2) + \sum_{n=1}^{\infty} \eta_1^n f^{(0)(n)}(x_i, \tau_0, \tau_1, \tau_2) \end{cases} \tag{9}$$

in which $\tau_0 = \tau$, $\tau_1 = \eta_1 \tau$, $\tau_2 = \eta_2 \tau$. Instituting these expressions into Eqs. (7) and (8), we can decouple the problem. It is interesting to note that the equations of the first three orders indicate the three physical courses: 0th order, the development of seepage, the fast course; 1st order, the dissociation of gas hydrate, the second fast course; 2nd, heat conduction, the third fast course. The 0th order equations are as follows:

$$\begin{cases} \frac{\partial \varepsilon_g^{(0)(0)} \rho_g^{(0)(0)}}{\partial \tau_0} - \nabla (\rho_g^{(0)(0)} \nabla P^{(0)(0)}) = 0 \\ \frac{\partial \varepsilon_w^{(0)(0)}}{\partial \tau_0} - \nabla^2 P^{(0)(0)} = 0 \\ \frac{\partial \varepsilon_h^{(0)(0)}}{\partial \tau_0} = 0 \\ \varepsilon_g^{(0)(0)} \rho_g^{(0)(0)} \frac{\partial C_g T^{(0)(0)}}{\partial \tau_0} + \varepsilon \rho C \frac{\partial T^{(0)(0)}}{\partial \tau_0} + \left[\frac{k_w \mu_g}{k_g \mu_w} \rho_w C_w + \rho_g^{(0)(0)} C_g \right] \nabla P^{(0)(0)} \nabla T^{(0)(0)} = 0 \end{cases} \tag{10}$$

Summing the last three equations together and neglecting the smallness, the controlling equation of $P^{(0)(0)}$ can be obtained

$$\varepsilon_{g0} \frac{\partial P^{(0)(0)}}{\partial \tau_0} - 2P^{(0)(0)} \nabla^2 P^{(0)(0)} = 0 \tag{11}$$

We can obtain the following simplified equation when the pore pressure difference is small [15].

$$\varepsilon_{g0} \frac{\partial P^{(0)(0)}}{\partial \tau_0} - 2p_0 \nabla^2 P^{(0)(0)} = 0 \tag{12}$$

The 0th order solution can be obtained as follows:

$$\left\{ \begin{array}{l} T^{(0)(0)} = T_0 \\ p^{(0)(0)} = p_1 + \frac{p_0 - p_l}{l} x + \sum_{n=0}^{\infty} \frac{2(p_0 - p_l)}{n\pi} e^{-\frac{n^2 \pi^2 a^2}{l^2} \tau_0} \sin \frac{n\pi}{l} x \\ \varepsilon_h^{(0)(0)} = \varepsilon_{h0} \\ \varepsilon_w^{(0)(0)} = \sum_{n=1}^{\infty} \frac{2(p_0 - p_l)}{n\pi} \frac{1}{a^2} e^{-\frac{n^2 \pi^2 a^2}{l^2} \tau_0} \sin \frac{n\pi}{l} x + \varepsilon_{w0} - \frac{p_0 - p_l}{a^2 l} (l - x) \end{array} \right. \quad (13)$$

in which p_0 is the initial pore pressure. The pore pressure at the end l is kept as p_0 , p_1 is the pore pressure at the end $x = 0$, $a^2 = 2\rho_0/\varepsilon_{g0}$, initially $\varepsilon_w = \varepsilon_{w0}$ at $t = 0$.

1st order equations are as follows:

$$\left\{ \begin{array}{l} \left(\varepsilon_g^{(0)(0)} \rho_g^{(0)(0)} \frac{\partial C_g T^{(0)(1)}}{\partial \tau_0} + \varepsilon \rho C \frac{\partial T^{(0)(1)}}{\partial \tau_0} \right) + \left[\frac{k_w \mu_g}{k_g \mu_w} \rho_w C_w + \rho_g^{(0)(0)} C_g \right] \nabla p^{(0)(0)} \nabla T^{(0)(1)} \\ = \left[\frac{\Delta H}{CT_0} + \chi C_g + (1 - \chi) C_w T^{(0)(0)} \right] A (p^{(0)(0)} - p_e) \\ \frac{\partial (\varepsilon_g^{(0)(1)} \rho_g^{(0)(0)} + \varepsilon_g^{(0)(0)} \rho_g^{(0)(1)})}{\partial \tau_0} - \nabla (\rho_g^{(0)(1)} \nabla P^{(0)(0)}) + \rho_g^{(0)(0)} \nabla P^{(0)(1)} = -\chi A (p^{(0)(0)} - p_e) \\ \frac{\partial \varepsilon_w^{(0)(1)}}{\partial \tau_0} - \nabla^2 p^{(0)(1)} = -(1 - \chi) A_1 (p^{(0)(0)} - p_e) \\ \frac{\partial \varepsilon_h^{(0)(1)}}{\partial \tau_0} = A_2 (p^{(0)(0)} - p_e) \end{array} \right. \quad (14)$$

in which $A = k_d M_g A_s \rho_h$, $A_1 = k_d M_g A_s \rho_h / \rho_w$, $A_2 = k_d M_g A_s$. Neglecting the effects of convection on the variation of temperature, then the variation of pore pressure $p^{(0)(0)}$ is mainly due to dissociation of GH, the 1st order solution solution can be obtained as

$$\left\{ \begin{array}{l} T^{(0)(1)} = \left[\frac{\Delta H}{CT_0} + \chi C_g + (1 - \chi) C_w T_0 \right] \frac{A}{\varepsilon \rho C + \varepsilon_g^{(0)(0)} \rho_g^{(0)(0)}} \\ \left(T_1 \tau_0 + \frac{T_0 - T_l}{l} x \tau_0 - \sum_{n=0}^{\infty} \frac{2(T_0 - T_l)}{n\pi} \frac{l^2}{n^2 \pi^2 a^2} e^{-\frac{n^2 \pi^2 a^2}{l^2} \tau_0} \sin \frac{n\pi}{l} x - T_e \tau_0 \right) \\ p^{(0)(1)} = [A_2 - \chi A - (1 - \chi) A_1] \left(p_1 x^2 + \frac{p_0 - p_l}{3l} x^3 - \sum_{n=0}^{\infty} \frac{2(p_0 - p_l) l^2}{n^3 \pi^3} e^{-\frac{n^2 \pi^2 a^2}{l^2} \tau_0} \sin \frac{n\pi}{l} x - p_e x^2 \right) \\ \frac{\partial \varepsilon_w^{(0)(1)}}{\partial \tau_0} = -(1 - \chi) A_1 (p^{(0)(0)} - p_e) + \nabla^2 p^{(0)(1)} \\ \varepsilon_h^{(0)(1)} = A_2 \left(p_1 \tau_0 + \frac{p_0 - p_l}{l} x \tau_0 - \sum_{n=0}^{\infty} \frac{2(p_0 - p_l)}{n\pi} \frac{l^2}{n^2 \pi^2 a^2} e^{-\frac{n^2 \pi^2 a^2}{l^2} \tau_0} \sin \frac{n\pi}{l} x - p_e \tau_0 \right) \end{array} \right. \quad (15)$$

$T^{(1)}$ and $p^{(1)}$ are small parameters. Neglecting the product term of these two smallness and considering $\varepsilon_g^{(1)}\rho_g^{(0)} \gg \varepsilon_g^{(0)}\rho_g^{(1)}$, $\rho_g^{(1)}\nabla P^{(0)} \gg \rho_g^{(0)}\nabla P^{(1)}$, 2nd order solution can be simplified as

$$\begin{cases} \left(\varepsilon_g^{(0)}\rho_g^{(0)} + \varepsilon\rho C \right) \frac{\partial T^{(1)}}{\partial \tau_1} = \nabla^2 T^{(1)} \\ \frac{\partial \varepsilon_g^{(1)}\rho_g^{(0)}}{\partial \tau_0} - \nabla\rho_g^{(0)}\nabla P^{(1)} = -\chi A p^{(1)} \\ \frac{\partial \varepsilon_w^{(1)}}{\partial \tau_0} - \nabla^2 p^{(1)} = -(1-\chi)A_1 p^{(1)} \\ \frac{\partial \varepsilon_n^{(1)}}{\partial \tau_0} = -(1-\chi)A_2 p^{(1)} \end{cases} \quad (16)$$

Then the solution for $T^{(1)}$ can be obtained as follows

$$T^{(1)} = T_1 + \frac{T_0 - T_l}{l}x + \sum_{n=1}^{\infty} \frac{2(T_0 - T_l)}{n\pi} e^{-\frac{n^2\pi^2 b^2}{l^2}\tau_1} \sin \frac{n\pi}{l}x \quad (17)$$

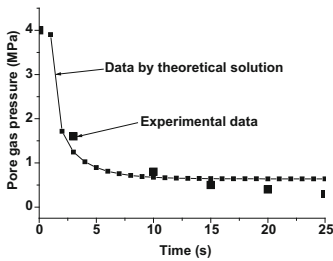
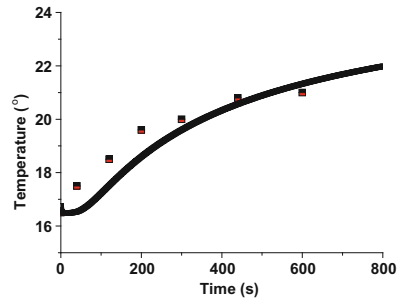
T_0 is the initial temperature, T_1 is the temperature at the end $x = 0$, the temperature at the end $x = 1$ is kept as T_0 , $b^2 = 1 / \left(\varepsilon_g^{(0)}\rho_g^{(0)}C_g + \varepsilon\rho C \right)$. In the same way, $p^{(1)}$, $\varepsilon_w^{(1)}$ and $\varepsilon_s^{(1)}$ can be solved.

It can be seen clearly from the above analysis that the dissociation process can be divided into three decoupled courses: seepage, phase change and thermal conduct in the sequence of the development speed of each course in a GH reservoir with low permeability. Thermal conduction is the slowest course while seepage is the fastest. The analytical solution can be obtained easily by the decoupled courses relative to obtain the solution directly from the original equation. The solution shows that no matter what kind of method to induce the dissociation of GH, thermal conduction is the key in dissociation of GH because it is the slowest course and phase change needs heat consumption. In other words, without enough heat supply phase change will undoubtedly stop no matter what the method we adopted so the fast heat supply is very important in exploitation of GH.

For certification, we compute the development of pore pressure and temperature with time by the above analytical solutions first and, compare them with experimental results in literature [16]. The parameters adopted here are shown in Table 1. Initial pore pressure is 4.0 MPa. At one end the pore pressure is 0.1 MPa and at the other end the pressure is kept as the initial value. Initial temperature is 16.5 °C. At one end temperature increases to 40 °C while at the other end it keeps the initial value. It can be seen from Figs. 1 and 2 that the analytical solution is close to the experiments'. The measurement of pore pressure is relatively accurate. The temperature is measured by thermocouple whose accuracy is 1°, thus the measured value of temperature is not as precise as pore pressure. That is why the differences between theoretical and experimental temperature is a little large. The fact that the theoretical value is only approximation leads to error.

Table 1. Parameters adopted in computation

Parameter	Value	Parameter	Value	Parameter	Value	Parameter	Value
C_d /kJ/kg/K	-1.05	N	6	$\rho_w/10^3$ kg/m ³	1.00	a_0 /m	0.007
P_{gc} /MPa	3.5	λ_g /W/m/K	0.06	$\rho_h/10^3$ kg/m ³	0.92	$k_0/10^4$ mol/m ² /Pa/s	3.6
P_{g0} /MPa	0.1	λ_w /W/m/K	0.50	$\rho_s/10^3$ kg/m ³	2.60	n_w	4
T_0 /K	289.5	λ_l /W/m/K	0.46	$\mu_w/10^{-3}$ Pa·s	1.00	n_g	2
T_e /K	413.0	λ_s /W/m/K	2.90	λ_a /W/m ² /K	0.26	n_c	0.65
S_{h0}	0.0	C_g /kJ/kg/K	2.18	D /m	0.038	ϕ	0.27
S_{g0}	0.32	C_w /kJ/kg/K	4.20	L /m	0.78		
S_{w0}	0.0	C_l /kJ/kg/K	2.22	$H_0/10^6$ J/kg	3.53		
K_0 /mD	86.1	C_s /kJ/kg/K	0.89	ΔE /kJ/mol	81		

**Fig. 1.** Comparison of theoretical and experimental results in literature (Liu, 2013)**Fig. 2.** Comparison theoretical and experimental results in literature (Zhang, et al., 2010)

3 Conclusions

A new method to obtain the analytical solution of equations describing the dissociation of NGH in stiff sediment was presented. By this method it can be seen clearly that the dissociation process can be divided into three decoupled courses which are thermal conduct, phase change and seepage by development speed in a NGH reservoir with low permeability. It is easily to obtain the analytical solution by the decoupled courses. The solution shows that no matter what kind of method to induce the dissociation of GH, thermal conduction is the key because it is the slowest course while phase change needs heat consumption. In other words, without enough heat supply phase change will undoubtedly stop no matter what the method we adopted. The analytical solution is close to that of experimental results by comparison.

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