

Analytical modeling of methane hydrate dissociation under thermal stimulation

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Abstract

In this study, a one-dimensional analytical model to describe heat and mass transfer during methane hydrate dissociation under thermal stimulation in porous media has been developed. The model is based on a similarity solution that considers a moving dissociation boundary which separates the dissociated zone containing produced gas and water from the un-dissociated zone containing only methane hydrate. The results of temperature distribution, pressure distribution, energy efficiency, and parametric study considering various initial and boundary conditions as well as various reservoir properties are presented and compared with previous studies. Sensitivity analysis of gas production on reservoir properties is also presented in this paper. The dissociation boundary moves faster by increasing the heat source temperature while decreasing the heat source pressure simultaneously, but the associated energy efficiency decreases. Increasing the well thickness has a negative effect on the energy efficiency of the process. Among the proposed thermal properties of the system, only the thermal diffusivities and conductivities of the reservoir as well as the porosity of the sediment affect the dissociation. The main contribution of this work is investigating analytically the hydrate dissociation using thermal stimulation by taking into account the effect of wellbore thickness and structure.

1 Introduction

Gas hydrates are cage-like substances (clathrates) in which a large amount of gas molecules is trapped by crystalline water molecules. They contain about 170 to 180 standard cubic meters of natural gas per cubic meter of hydrate (Kamath and Godbole, 1987). The high pressure and low

temperature underground with sufficient methane gas induce methane hydrate (MH) formation (Davie and Buffett, 2001). The estimation of the volume of the MH exceeds the whole volume of conventional gas resources worldwide (Englezos, 1993; Makogon, 1981). However, considering MH as a reliable and alternative source of energy for future depends on the availability of hydrates and the cost of gas production process from them.

The MH are mainly formed beneath the permafrost in arctic regions and in marine shelf sediments according to appropriate pressure and temperature conditions (Collett, 2001; Collett, 2008). As mentioned above, MH formation and stability require specific pressure and temperature conditions, so any change in temperature or pressure from equilibrium conditions would induce MH instability. The three main methods of MH dissociation that have been reported in previous works are as follows (Li et al., 2016; Pooladi-Darvish, 2004): i) thermal stimulation, in which the temperature of the reservoir is increased above the equilibrium temperature; ii) depressurization by decreasing the pressure inside the MH reservoir lower than the equilibrium pressure of the hydrate, so that the MH will start dissociating; and iii) depressurization in conjunction with thermal stimulation, that can improve the efficiency of the dissociation process (Wang et al., 2014). It should be noted that gas production aspects and potential using different methods is not yet fully investigated.

The mathematical backgrounds in the field of MH dissociation can be divided into two main categories: analytical and numerical. Analytical solutions are fast and simple, also they can provide better mechanistic view. On the other hand, the numerical studies are more comprehensive with smaller number of assumptions. In 1982, one of the first numerical studies was developed for MH dissociation under depressurization using a three dimensional (3D) model consisting of an MH layer and a free gas zone, in which conduction heat transfer and gas fluid flow were considered (Holder and Angert, 1982). This work was extended in 1986 by considering water movement, which is produced from MH dissociation (Burshears et al., 1986). In neither of the mentioned studies, the water/gas convective heat transfer was considered. In 1991, a numerical model was designed to predict MH dissociation in porous media by depressurization considering three phases of water, gas, and MH with gas-water flows and neglecting the effect of heat transfer (Yousif et al., 1991). Then, it was extended by considering water-gas flows and convective-conductive heat transfer (Masuda, 1997; MASUDA, 1999). In 2000, MH dissociation upon thermal stimulation

was simulated by assuming a moving dissociation boundary, which separates the dissociated zone from the undissociated zone, and considering different media permeability (Tsypkin, 2000). This moving boundary layer was assumed to be impermeable, so if the pressure and temperature conditions fall below the balance point the dissociation will be halted. Another numerical work employing finite difference method was performed to study gas production from MH dissociation using depressurization (Ahmadi et al., 2004). They considered the effect of heat transfer and reported that the process is a function of well pressure. At the same year, gas production from MHs using both depressurization and thermal stimulation was shown to be possible by using TOUGH2 simulator, which is capable of simulation by considering four components and up to nine phases in either kinetic or equilibrium models and upon different dissociation processes (Moridis, 2002). The feasibility of gas production from MH reservoirs in the Mackenzie Delta, Northwest Territories, Canada was also studied using TOUGH2 simulator (Moridis et al., 2004). The results revealed the gas production using depressurization and thermal stimulation, and higher efficiency achieved by using both the methods together. They additionally showed that increasing the initial formation temperature, the well temperature, and the formation thermal conductivity increased the amount of produced gas; while, it is not affected by the permeability of the formation and the specific heat of the rock and hydrate. The numerical simulations in another work showed that the kinetic reaction models should be taken into account in order to avoid under-prediction of recoverable MH, however, it requires more computational effort compared to equilibrium reaction model (Kowalsky and Moridis, 2007). Investigation on dispersed oceanic MH reservoirs with low hydrate saturation using TOUGH-Fx/HYDRATE simulator upon depressurization showed low gas production potential, which was not economically feasible, with high amount of water production (Moridis and Sloan, 2007). Nowadays, researchers are performing more mathematical investigations on this field accompanying with comparison with real field or experimental results or using parameters from real reservoirs (Chen and Hartman, 2018; Konno et al., 2017; Mardani et al., 2018; Wang et al., 2019; Wang et al., 2016a; Zhao et al., 2016).

In 1990, Selim et al. (Selim and Sloan, 1990) designed an analytical one-dimensional (1D) study on MH dissociation upon thermal stimulation method by considering a moving dissociation boundary. They took into account the gas convection heat transfer and its flow, while the water by-product remained motionless in the sediments. In 1982, a study on hydrate dissociation process upon hot water injection and depressurization employing two models has been designed: the

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frontal-sweep model, and the fracture-flow model. They reported that the gas production from hydrates using the depressurization method is feasible compared to thermal stimulation (McGuire, 1982). Makogon (Makogon, 1997) reported analytical expressions for the temperature and pressure profiles for the MH dissociation upon depressurization using a similar process of moving dissociation boundary including the effect of the throttling process in the energy equation. Tsypkin (Tsypkin, 2001) extended it by considering similarity solutions for temperature and pressure distributions and the water and gas movement. Another model was also presented in 2001 by including heat conduction to Makogon's model (Ji et al., 2001). An analytical work on dissociation induced by depressurization reported that the effect of the gas-water two-phase flow on MH dissociation is smaller than the effect of heat transfer and the intrinsic kinetics of MH decomposition (Hong et al., 2003). Recently, an analytical-experimental work has been reported to study MH decomposition by depressurization, thermal stimulation, and depressurization in conjunction with thermal stimulation (Wang et al., 2015). The analytical model in that work was based on performed experimental conditions, for example, heat transfer from outside the experimental reservoir into the reservoir is also considered.

In addition to mathematical approaches, experimental tools have also been employed to investigate MH dissociation upon different methods. One of the major challenges in experimental works in this field, is the size of the setup that significantly affects the outcome of tests (Wang et al., 2016b). For example, scale of experiment setup determines the main mechanism involving in the hydrate dissociation in porous media (Wang et al., 2016b), which could be one of the followings: i) the intrinsic kinetics of hydrate decomposition; ii) heat transfer in the decomposing zone; or iii) the multiphase flow (i.e., gas-water flow) during gas production (Hong et al., 2003). Tang et al. (Tang et al., 2007) reported that the intrinsic kinetics of hydrate decomposition is the determining factor in the core-scale experiments. They also showed that in larger scale works, such as field works, heat transfer in the decomposing zone becomes the controlling mechanism. An experimental investigation of the MH dissociation in a 3D cubic hydrate simulator (CHS) upon huff and puff thermal stimulation showed that MH dissociation process upon thermal stimulation proceeds as a moving boundary ablation process (Li et al., 2011b). Li et al. (Li et al., 2012b) experimentally investigated the MH dissociation upon depressurization using two hydrate simulators with different scales. They revealed that gas production period is longer for larger scale simulators. In another experimental work, conduction heat transfer was reported as the main mechanism for heat

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transfer to the dissociating zone (Zhao et al., 2012). Wang et al. (Wang et al., 2016b) studied the hydrate dissociation behavior below the quadruple point in the sandy sediment employing a 3D Pilot-Scale Hydrate Simulator (PHS). They revealed that MH dissociation below the quadruple point induces ice formation in pores, which in turn increases the dissociation rate. Another work, designed and performed by Wang et al. (Wang et al., 2018), reported that depressurization in conjunction with thermal stimulation is the optimum method to dissociate water-saturated hydrate samples using a pilot-scale hydrate simulator via the three abovementioned main methods. Studies over the past decades have provided important information on MH Dissociation and its gas production potential.

Apart from the interactions happening in the reservoir upon hydrate dissociation, the wellbore structure may affect the dissociation especially the heat transfer mechanism in thermal stimulation method. Furthermore, data from several studies suggest that the wellbores in the production region mainly consisted of three layers: casing, cement, and gravel (Florez Anaya and Osorio, 2014; Pucknell and Mason, 1992; Xu et al., 2014). On the other hand, among previous mathematical works that have been done in the field of hydrate dissociation up to now (Li et al., 2010; Li et al., 2012a; Wan et al., 2018; Zhao et al., 2016), no analytical study has investigated the impact of heat source structure (i.e., wellbore radius and the associated outer layers) on dissociation upon thermal stimulation using wellbore heating, which might induce unreliable outcomes while comparing to those from experiments or field works. Existing research addresses this gap and recognizes the critical role played by the wellbore structure in hydrate dissociation via wellbore heating.

In the present work, an analytical solution is generated to model a semi-infinite 1D hydrate reservoir in Cartesian coordinates using a flat heat source considering three main layers: casing, cement, and gravel. The model and results are validated and compared with the previous mathematical and experimental studies. The results of this work will be useful in assessing the gas production from MH reservoirs using thermal stimulation. The importance and originality of this study are that it analytically explores, for the first time, the effect of wellbore structure on the MH dissociation and the associated heat transfer process. Therefore, the findings should make an important contribution to the field of gas production from MH reservoirs by providing the outcomes obtained using conditions that are closer to the real-condition tests, which make them more valuable and reliable.

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2 Modeling

Figure 1 shows a schematic of hydrate dissociation using thermal stimulation method in the proposed 1D flat case. It should be mentioned that the reservoir is semi-infinite, and L denotes the overall well thickness including, cement, gravel, and casing layers. The fundamental steps of MH dissociation are as follows: in the beginning of the process, the reservoir with the porosity of ϕ is filled with MH in equilibrium conditions with initial temperature T_0 . At time $t = 0$, the temperature of the heat source at $x = 0$, increases to a new temperature of T_i , which is above the equilibrium temperature of the hydrate, and is kept constant during the process. Subsequently, MH starts dissociating and a sharp moving boundary surface is created as shown by dashed lines in Figure 1, which indicates the rate of hydrate dissociation and separates the water and gas produced in the dissociated zone (Zone I) from the un-dissociated zone (Zone II). The temperature of the dissociated zone is higher than the temperature in the hydrate zone inducing heat conduction from the dissociated zone to the hydrate zone and hydrate dissociation. The rate of hydrate dissociation is determined by the speed of moving interface, which decreases as the process continues and the dissociated zone thickens. Actually, it consumes a larger part of input heat to increase the temperature of the matrix material in the dissociated zone as well as the produced water and gas at the dissociation front as the dissociation interface moves forward. The remaining input energy is consumed for hydrate dissociation and temperature increase of the matrix materials in Zone II. As shown in Figure 2, the temperature and pressure distributions in the Zone I (T_I and P) are respectively $T_s < T_I < T_i$ and $P_i < P < P_s$, and in the Zone II, the temperature distribution is $T_0 < T_{II} < T_s$ and the pressure is in the equilibrium condition of the hydrate. The dissociation interface is shown by the dashed line and the associated pressure and temperature change due to time. It should be noted that T_I at the outer surface of the well changes with time, but it is always lower than T_i .

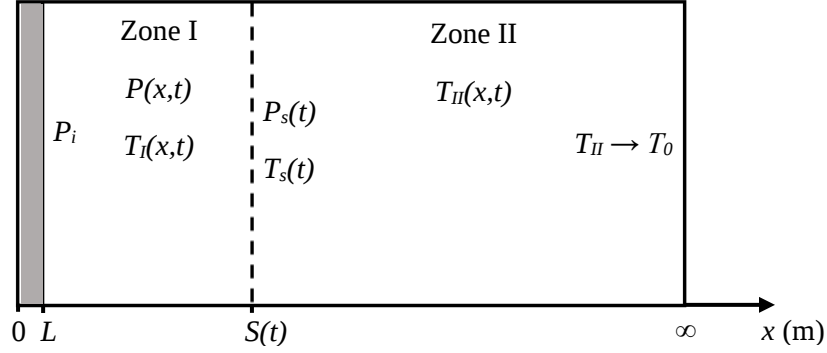


Figure 1. Schematic of hydrate dissociation in the semi-infinite flat reservoir. The dissociation interface is identified by the dashed line, and the grey region shows well thickness.

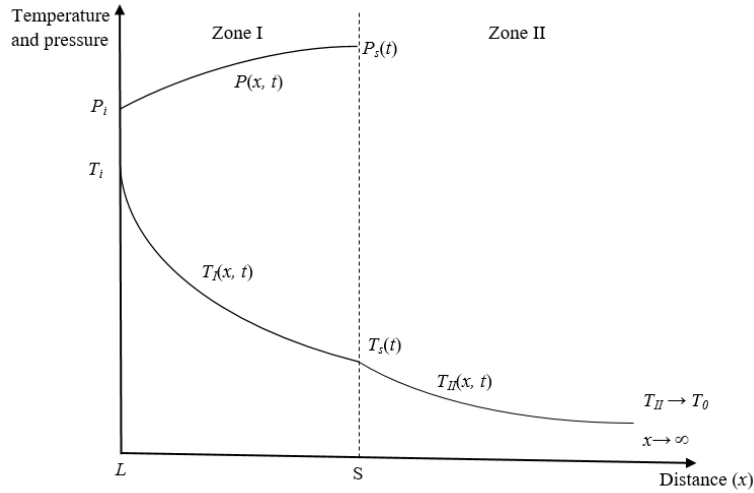


Figure 2. Schematic of pressure and temperature distribution in the reservoir upon hydrate dissociation.

According to the mass conservation law and the sudden change in density at the dissociation front due to gas production, the produced gas will be streaming towards the heat source. The assumptions considered in this study, which were in accordance with the previous analytical works (Selim and Sloan, 1990; Tsimpanogiannis and Lichtner, 2007), are as follows: i) hydrates filled the entire pores of the media; ii) temperature and pressure at the dissociation interface are at the thermodynamic equilibrium; iii) the produced water from the dissociation remains motionless in the pores; iv) thermophysical properties of the phases are constant; v) ideal behavior of gas is applied in the equation of state of gas; vi) the produced gas is in thermal equilibrium with the local

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sediments (with the same temperature); vii) viscous dissipation and inertia effects are neglected. It should be noted that the thermal properties of the system are assumed to remain constant during dissociation

The fundamental set of equations and the solution procedure for the flat case by considering a wellbore heat source are presented by the following expression:

The continuity equation of gas in Zone I is:

$$\phi \left(\frac{\partial \rho_g}{\partial t} \right) + \left(\frac{\partial \rho_g v_g}{\partial x} \right) = 0, L < x < S, t > 0 \quad (1)$$

where, ϕ is porosity, ρ_g is gas density (kg/m³), and v_g is gas velocity (m/s). The gas velocity in Zone I is calculated by the multiphase Darcy's law, which is as follows:

$$v_g = - \left(\frac{k}{\mu} \right) \left(\frac{\partial P}{\partial x} \right), L < x < S, t > 0 \quad (2)$$

where k is relative permeability of gas (md), μ is gas viscosity (mPa.s), and P is gas pressure (Pa). Equations 3 and 4 show the energy balance in Zones I and II, respectively:

$$\rho_I C_{pI} \frac{\partial T_I}{\partial t} + \frac{\partial \rho_g C_{pg} v_g T_I}{\partial x} = k_1 \frac{\partial^2 T_I}{\partial x^2}, L < x < S, t > 0 \quad (3)$$

$$\frac{\partial T_{II}}{\partial t} = \alpha_{II} \frac{\partial^2 T_{II}}{\partial x^2}, S < x < \infty, t > 0 \quad (4)$$

where ρ_I is density (kg/m³) of zone I, ρ_g is gas density (kg/m³), C_{pI} is specific heat capacity (J/(kg.K)) of Zone I, C_{pg} is specific heat capacity of gas (J/(kg.K)), T_I is the temperature (K) in Zone I, T_{II} is the temperature (K) in Zone II, α_{II} is thermal diffusivity (m²/s) of Zone II, and k_I is the thermal conductivity (W/(m.K)) of Zone I. In equation 3, the gas temperature in each location in Zone I is assumed to be equal to the local sediment temperature. This assumption has been proved by Weinbaum and Wheeler (Weinbaum and Wheeler Jr, 1949) and also employed in another work by Selim and Sloan (Selim and Sloan, 1990).

The gas behavior is assumed to obey the ideal gas law. So, the gas density in Zone I can be evaluated by the following equation:

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$$\rho_g = \frac{mP}{RT_i}, \quad L < x < S, t > 0 \quad (5)$$

where m is gas molecular mass (kg/mol), and R is the universal gas constant (J/(mol.K)). The above set of equations represents the fundamental concept of the process. In the following, the associated initial and boundary conditions are stated. Temperature and pressure at the inner wall of the well ($x = 0$) are kept constant and respectively equal to T_i and P_i . It is assumed that there is no pressure drop in the well wall so the pressure at the outer surface of the well ($x=L$) is P_i as well.

The assumed wellbore schematic in the production region is shown in Figure 3, in which the thickness of each of the casings is 0.7 cm, and the thickness for the cement and gravel respectively is 2.5 cm and 1.5 cm. Hence, the heat transfer equation in the wellbore could be stated by equation 6:

$$-k_I A_w \frac{\partial T_I}{\partial x} = \frac{(T_i - T_I)}{R_w}, \quad x = L, t > 0 \quad (6)$$

$$R_w = \frac{1}{A_w} \left(\frac{L_s}{k_s} + \frac{L_c}{k_c} + \frac{L_g}{k_g} \right) \quad (7)$$

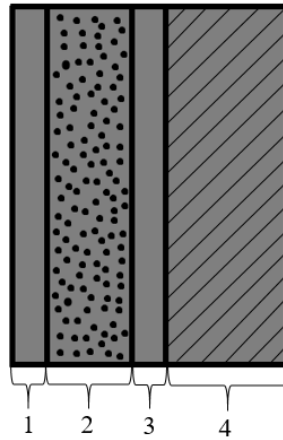


Figure 3. Schematic of the proposed wellbore structure. Different parts are as follows: 1) casing, 2) gravel pack, 3) casing, and 4) cement

where A_w is the wellbore area (m^2), R_w is thermal resistivity of the wellbore (W/K), k_x and L_x are respectively the thermal conductivity (J/(s.m.K)) and thickness (m) with s, c, and g subscripts respectively stands for steel (casing), cement, and gravel. It should be noted that this proposed

structure for wellbore is a general geometry and model, and there are many of the wellbore structures and the associated geometries in the literature that are different from this model.

The pressure at the dissociation interface is calculated from the Antoine equation, which is a thermodynamic relationship with the interface temperature:

$$P_s = e^{(A_a - B_a/T_s)}, \quad x = S, t > 0 \quad (8)$$

where P_s is the pressure (Pa) and temperature (K) at the moving interface, and A_a and B_a are constants. The mass and energy balances at the dissociation interface are respectively represented in equations 9 and 10. Also, the heat of MH dissociation is calculated by equation 11 (Selim and Sloan, 1990), and equations 12-14 represent the boundary conditions:

$$F_{gH} \rho_H \left(\frac{dS}{dt} \right) + \rho_g v_g = 0, \quad x = S, t > 0 \quad (9)$$

$$k_{II} \frac{\partial T_{II}}{\partial x} - k_I \frac{\partial T_I}{\partial x} = \phi \rho_H Q_{Hd} \frac{dS}{dt}, \quad x = S, t > 0 \quad (10)$$

$$Q_{hyd} = c + dT_s \quad (11)$$

$$T_I = T_{II} = T_s(t), \quad x = S, t > 0 \quad (12)$$

$$T_{II} = T_0, \quad x \rightarrow \infty, t > 0 \quad (13)$$

$$T_{II} = T_0, \quad 0 < x < \infty, t = 0, S = L \quad (14)$$

where ρ_H is the hydrate density (kg/m³), k_{II} is the thermal conductivity (W/(m.K)) of Zone II, Q_{Hd} is heat of MH dissociation (J/kg), and c and d are constants. F_{gH} in equation 8 is a constant, representing the ratio of mass of the methane gas trapped inside the MH to the mass of hydrate. In this work, the value of 0.1265 kg CH₄/kg hydrate is chosen for F_{gH} , which has been used in a previous study by Selim et al. (Selim and Sloan, 1990).

The following equations 15-17 are obtained respectively from equations 1, 3, and 8 by employing equations 2 and 5 in order to eliminate the gas velocity and density.

$$\phi \frac{\partial}{\partial t} \left(\frac{P}{T_I} \right) - \frac{k}{\mu} \frac{\partial}{\partial x} \left(\frac{P}{T_I} \frac{\partial P}{\partial x} \right) = 0 \quad (15)$$

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$$\rho_I C_{pl} \frac{\partial T_I}{\partial t} + \frac{kmC_{pg}}{\mu R} \frac{\partial}{\partial x} \left(P \frac{\partial P}{\partial x} \right) = k_1 \frac{\partial^2 T_I}{\partial x^2}, \quad L < x < S, t > 0 \quad (16)$$

$$F_{gH} \phi \sigma_H \left(\frac{dS}{dt} \right) - \frac{kmP}{\mu R T_I} \frac{\partial P}{\partial x} = 0, \quad x = S, t > 0 \quad (17)$$

To solve the above set of equations, the similarity solution introduced by Neumann (Ä–zisik et al., 1993; Carslaw and Jaeger, 1959) is employed. In this solution, the movement of the dissociation interface is assumed to be proportional with the square root of time ($t^{1/2}$). This assumption satisfies the initial and boundary conditions, and the following dimensionless transformation, shown in equation 17, is employed to simplify and solve the abovementioned equations.

$$\lambda = \frac{x}{\sqrt{4\alpha_{II}t}} \quad (18)$$

So, on the moving dissociation front, equation 18 becomes:

$$\beta = \frac{S}{\sqrt{4\alpha_{II}t}}, \quad x = S, t > 0 \quad (19)$$

Also, on the outer surface of the well:

$$\lambda_{os} = \frac{L}{\sqrt{4\alpha_{II}t}}, \quad x = L, t > 0 \quad (20)$$

The transformation of the above equations using equation 17 is developed and presented in the supplementary information.

By considering the procedure suggested by Özışık et al. (Ä–zisik et al., 1993), Carslaw et al. (Carslaw and Jaeger, 1959), and Selim et al. (Selim and Sloan, 1990), error function is employed to choose the following solutions for the temperature distributions:

$$\begin{cases} T_I = A \operatorname{erf}(a\lambda + b) - A \operatorname{erf}(b) + B \\ T_{II} = T_0 + C \operatorname{erfc}(\lambda) \end{cases} \quad (21)$$

A, B, a, and b constants are introduced in the supplementary information.

The pressure distribution (Pa) in Zone I can be calculated from equation S7 as follows:

$$P(\lambda) = \left(P_0^2 + \frac{4F_{gH} \phi \rho_H \alpha_{II} \mu R \beta}{km} \int_{\lambda_{os}}^{\lambda} T_I d\lambda \right)^{1/2} \quad (22)$$

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That by replacing and integrating T_1 from equation 21, becomes:

$$P = \left(P_0^2 + J(\beta)K(\beta)(\lambda - \lambda_{os}) + AJ(\beta)(L_1(\lambda) - L_1(\lambda_{os})) \right)^{1/2} \quad (23)$$

where $J(\beta)$, $K(\beta)$, and $L_1(\lambda)$ are defined in the supplementary information.

Heat flux at the boundary ($J/(s.m^2)$) as a function of time is calculated from the following formula:

$$u_f = -k_I \frac{\partial T_I}{\partial x}, \quad x = L \quad (24)$$

The transformed version of equation 24 considering equation 18 is provided in the supplementary information.

Cumulative heat input into the reservoir from the heat source (J/m^2) is calculated by integrating equation 23 with regard to time as follows:

$$Q_{ft} = -k_I \int_0^t \frac{\partial T_I}{\partial x} dt, \quad x = L \quad (25)$$

In order to calculate the total volume at standard temperature and pressure (STP) of dry gas, equation 26 is employed, where V_{fp} is the total volume of produced gas per surface area of the moving interface up to time t (m^3/m^2), n_{ft} is the total moles of gas produced per surface area of the moving interface up to time t (mole/ m^2), and T_{STP} and P_{STP} are temperature and pressure of gas at STP conditions, respectively.

$$V_{fp} = \frac{n_{ft} R T_{STP}}{P_{STP}} \quad (26)$$

More details of total volume calculation are provided in the supplementary information.

For the efficiency evaluation of gas production during the dissociation process using thermal stimulation method, the energy efficiency ratio is introduced as follows (Song et al., 2015):

$$\eta_f = \frac{V_{fp} Q_g}{Q_{ft}} \quad (27)$$

where η_f is the energy efficiency ratio in this case, and Q_g is the heating value of the gas at STP conditions (J/m^3). Actually, η_f represents the amount of energy that would be produced from

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combustion of the produced gas to the amount of input energy to the system from the heat source. It becomes apparent that an energy efficiency higher than one is preferred. It should be mentioned that the surface area of the well that is in contact with the reservoir is equal to 1m^2 .

The obtained solution for temperature and pressure distributions can satisfy the abovementioned basic equations and boundary conditions (equations 1-17) by direct substitution. MATLAB programming software is used for all calculations in this work.

3 Results and discussion

Equation 18 indicates that the location and movement of the dissociation interface are dependent on β . It should be noted that β represents both the dimensionless interface position and dimensionless interface velocity ($v_s / (4\alpha_{II}t)^{1/2}$). Thus, the higher the value of β , the faster the movement of the dissociation interface. As shown in the equation S23, the value of β is dependent on P_s and T_s values, which are calculated using the pressure and temperature at the heat source wall (outer boundary) according to the equations in the previous section. The temperature at the outer boundary of the heat source is time dependent, inducing P_s and T_s at the dissociation front to change over time. Figure 4 shows the value of β considering specific physical and boundary conditions presented in Table 1, which are obtained and set from the previous studies (Cheng et al., 2011; Dalla Santa et al., 2017; Remund, 1999; Selim and Sloan, 1990). β increases from a slightly lower value at the beginning but converges to a value as the temperature at the wall surface converges to temperature T_i of the inside of the well, which is kept constant.

Table 1. Parameters used in the modeling.

Total wellbore thickness, L , m	0.054
Cement thickness, L_c , m	0.025
Gravel thickness, L_g , m	0.015
Casing thickness, L_s , m	0.014
Contact area of wellbore with reservoir in production region, m^2	1
Thermal conductivity of cement, k_c , W/(m.K)	0.933
Thermal conductivity of gravel, k_g , W/(m.K)	0.4
Thermal conductivity of casing (steel), k_s , W/(m.K)	43.3
Porosity, ϕ	0.3
Permeability, k , μm^2	1

Thermal diffusivity of Zone I, α_I , $\mu\text{m}^2/\text{s}$	2.89×10^6
Thermal conductivity of Zone I, k_I , $\text{W}/(\text{m.K})$	5.57
Thermal diffusivity of Zone II, α_{II} , $\mu\text{m}^2/\text{s}$	6.97×10^5
Thermal conductivity of Zone II, k_{II} , $\text{W}/(\text{m.K})$	2.73
Hydrate density, ρ_H , kg/m^3	913
Heat of dissociation of hydrate, Q_{Hd} , J/kg	$446.12 \times 10^3 - 132.638T_s$
Gas heat capacity, C_{pg} , $\text{J}/(\text{kg.K})$	8766
Gas viscosity, μ , Pa.s	10^{-4}
Heating value of the gas at STP conditions, Q_g , MJ/m^3	37.6
Molecular mass of methane, m , g/mol	16.04
Mass ratio of the methane gas trapped inside the hydrate to the mass of hydrate, F_{gH}	0.1265
Universal gas constant, R , $\text{J}/(\text{mol.K})$	8.314

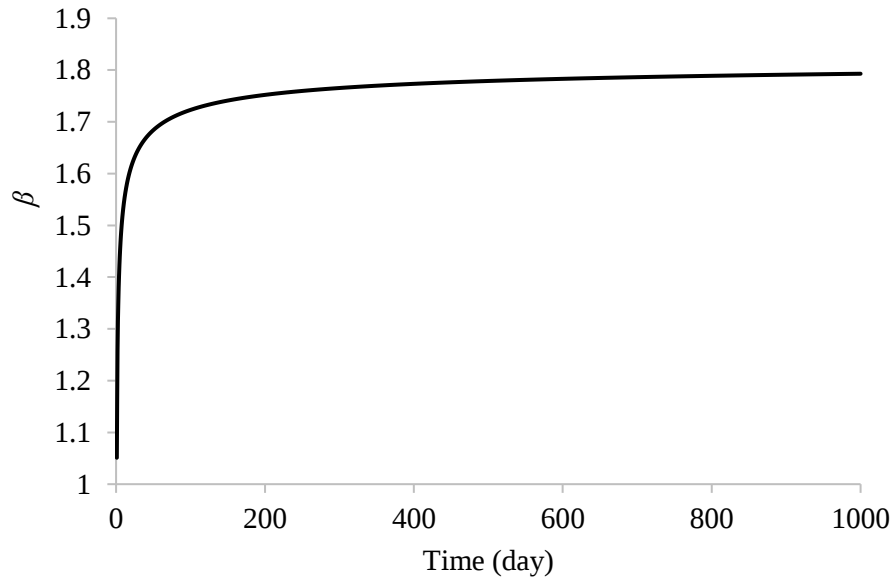


Figure 4. Dimensionless position of interface during 1000 days dissociation process at $T_i=563.5$ K, $P_i=7.6$ MPa, and $T_0=275$ K.

The effect of boundary conditions on the interface movement is presented in Figure 5, which shows the value of β with regards to different well pressure and temperature after 100 days of dissociation. The results shown in Figure 5 have the same trend as the results reported in the [Type here]

previous work by Selim et al. (Selim and Sloan, 1990), but with a slightly lower values of β (about 0.1 difference). This difference is due to the conduction heat transfer in the wellbore thickness, which causes the amount of transferred heat to the dissociation surface becomes lower compared to the previous work (Selim and Sloan, 1990), in which a flat heat source with constant temperature without wellbore thickness is considered. In all cases, decreasing the heat source pressure while increasing its temperature induces higher values of β . Figure 6 represents the relation between the interface velocity and MH temperature, T_0 . The higher the hydrate temperature, the higher the interface movement. These results are consistent with data obtained in a Numerical work by Liang et al. (Liang et al., 2010), who also found that decreasing the pressure and increasing the surrounding temperature would increase the rate of hydrate dissociation upon depressurization. They also validated their results against experiments performed by Masuda (MASUDA, 1999).

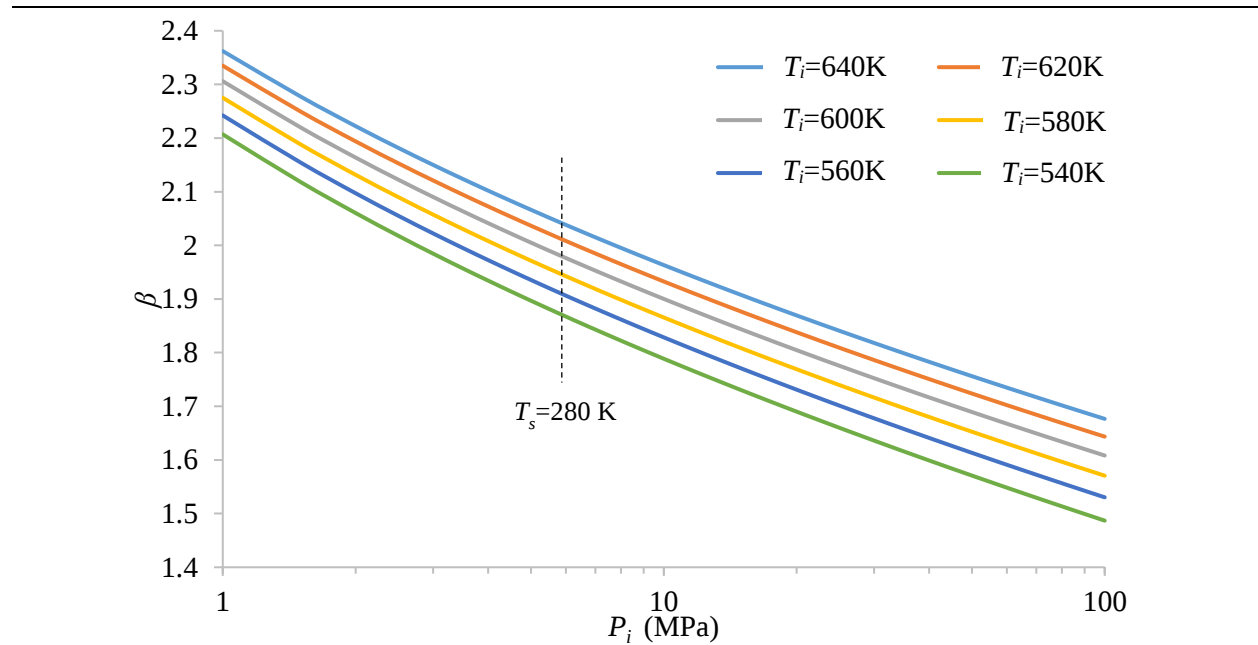


Figure 5. Dimensionless position of interface at $T_0=280$ K and various T_i and P_i values.

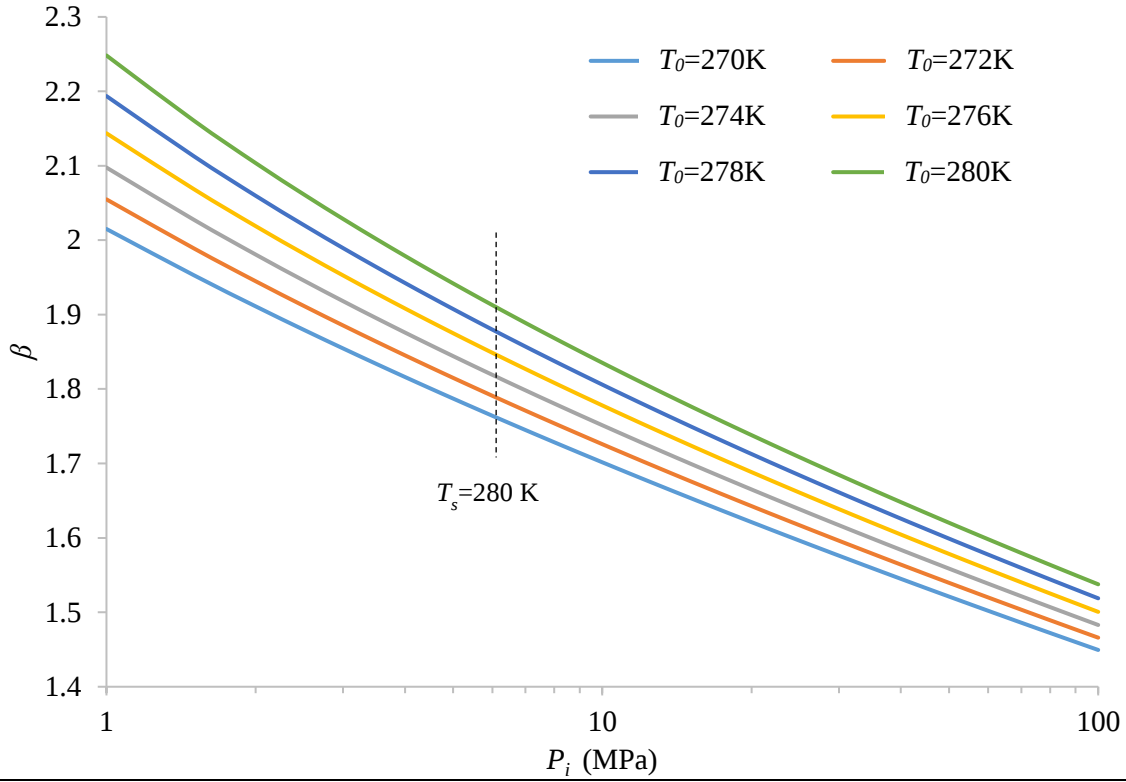


Figure 6. Dimensionless position of interface at $T_i=563.5$ K and various T_0 and P_i values.

Furthermore, in Figures 5 and 6, the associated temperature and pressure of the locus for which the dissociation temperature is equal to the temperature of MH (280 K) is shown. The dissociation temperature is mainly dependent on heat source pressure, while the effect of the temperature of the heat source and MH temperatures on T_s is almost negligible; higher P_i increases T_s , while lower P_i decreases T_s . This is also in agreement with the previous work by Selim et al. (Selim and Sloan, 1990), as they showed the direct dependency of T_s on P_i and mentioned that for lower pressures of 6 MPa, T_s may reduce to the freezing temperature of water which causes the interruption of the dissociation process due to ice generation. It should be mentioned that when T_s is equal to T_0 , all the transmitted heat to the interface is consumed for the dissociation and no heat is transferred to or from the hydrate zone, and the temperature of the hydrate zone remains constant. On the other hand, as T_s falls below T_0 , some part of the required heat for dissociation is absorbed from the hydrate zone, resulting in the temperature reduction of the hydrate zone.

Temperature and pressure distributions are calculated and presented in Figures 7 and 8 for the following two boundary conditions (BCs): BC 1) $T_i = 450$ K, $P_i = 10$ MPa, and $T_0 = 280$ K, and

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BC 2) $T_i = 563.5$ K, $P_i = 7.6$ MPa, and $T_0 = 275$ K considering three different time frames. The temperature distribution in Zones I and II are separated by a horizontal dashed line that shows the interface temperature. However, due to the small temperature increment at the well surface in longer time frames (as shown in the Figure 7), the interface temperature changes slightly. The temperature change at the well surface also causes the interface pressure to change in longer time frames. It should be noted that as the dissociation progresses and the temperature at the well surface converges to that inside the well, the temperature and pressure at the interface respectively tend to decrease and increase gradually and converge to the associated values reported Selim et al.'s work (Selim and Sloan, 1990), in which the associated values for the interface temperature and pressure for the two proposed boundary conditions were reported to be: BC 1) $T_s = 285$ K, $P_s = 290$ Pa; BC 2) $T_s = 282.5$ K, $P_s = 560$ Pa. Tsimpanogiannis et al. (Tsimpanogiannis and Lichtner, 2007) in their semi-analytical model showed that increasing the temperature of well would increase the pressure at the interface, which is also shown in our results of boundary conditions of BC 1 and BC 2.

The results of temperature distribution also illustrate that the distance between the dissociation surface and the heat source is higher in the previous study without considering the wellbore thickness (Selim and Sloan, 1990) compared to the present model due to the negative effect of heat conduction in the well, but this difference decreases in longer time frames as the dissociated zone thickens and the effect of heat source thickness disappears gradually. The results presented in Figure 7 for temperature distribution are also in good agreement with those reported by Li et al. (Li et al., 2011b), who experimentally investigated MH dissociation upon thermal stimulation. They also reported that the decomposition progressed by a moving boundary, which separates dissociate zone from the undissociated zone.

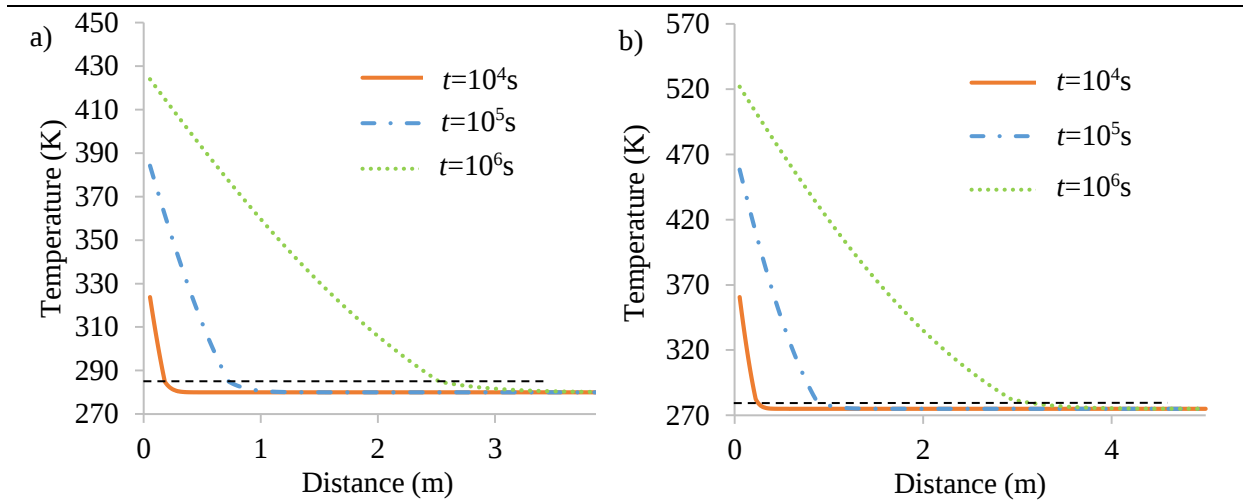


Figure 7. Temperature distribution for case 1 at different time frames for two initial and boundary conditions of a) BC 1 b) BC 2.

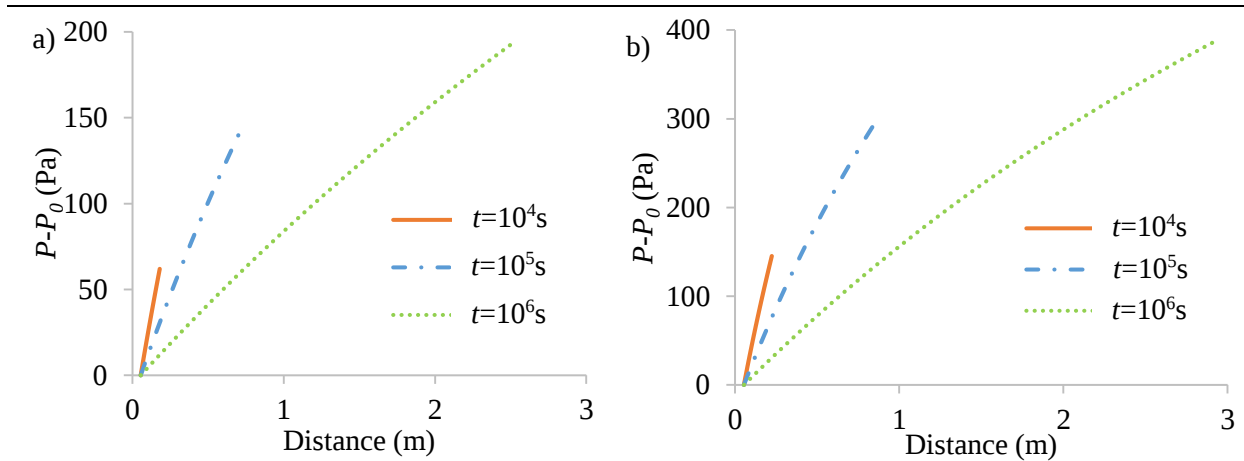


Figure 8. Pressure distribution for case 1 in the dissociated zone at different time frames for two initial and boundary conditions of a) BC 1 b) BC 2.

The volume of produced gas in STP conditions, amount of input heat, and energy efficiency upon hydrate dissociation considering the two initial and boundary conditions during 100 days of gas dissociation are shown in Figure 9. The volume of produced gas and the amount of input heat are higher considering BC 2 compared to BC 1. However, the energy efficiency is higher considering BC 2 compared to BC 1 due to the higher difference between the amounts of input heat and relatively smaller difference between the associated produced gas as shown in Figure 9. The results of energy efficiency indicate that increasing the heat source temperature and decreasing its

pressure would increase the rate of dissociation, but will not increase the total efficiency of the process. It is somewhat surprising that the energy efficiency increases to a peak point in the beginning then it decreases as dissociation progresses. This could be due to the constant surface area of the dissociation interface, which is equal to the surface area of the heat source, during the dissociation that restricts the amount of gas production during dissociation. On the other hand, the amount of input heat increases such that it cannot reach the dissociation front due to the sediment matrix of Zone I and dissociation products. Therefore, the speed of input heat increment exceeds the speed of the produced gas increment until the temperature at the heat source surface gradually converges to that inside the heat source, and the energy efficiency also converges to a constant value. The same trend for energy efficiency (i.e., significant increase at the beginning then decreasing smoothly) was also reported by Li et al. (Li et al., 2014; Li et al., 2011a) and Wang et al. (Wang et al., 2013), who investigated the MH dissociation behavior upon thermal stimulation using cubic hydrate simulators. They claimed that the sudden increase of the energy efficiency at the beginning is due to the rapid hydrate dissociation close to the wellbore at the beginning of the process. In another experimental work performed by Wang et al. (Wang et al., 2014) on the gas production from MH using thermal stimulation by hot water, the same trend for the energy efficiency and gas production was reported. It was claimed that the pre-depressurization before heat stimulation caused the sudden increase of the energy efficiency at the beginning, then it decreased and converged to lower value. They reported an energy efficiency of around 4 after 100 minutes of dissociation, which shows a good agreement with our results. In the work of Selim et al. (Selim and Sloan, 1990) an energy efficiency between 6.4-11.2 was reported, which is close to the results presented in the current paper and of high value because of the similar conditions considered in the two works. The energy efficiency reported in this work is consistent with that of Tang et al. (Tang et al., 2005), who experimentally investigated the gas production from MH reservoirs upon thermal stimulation. They reported an energy ratio, which was calculated in the same way as the energy efficiency in the present work, of about 5 upon conditions close to the BC 2. They also revealed that increasing the initial temperature and decreasing the initial pressure improved the energy ratio. Bayles et al. (Bayles et al., 1986) studied analytically MH dissociation upon cyclic steam injection. They reported the same trend for the energy efficiency for one year dissociation, which was converged between 4 to 9.6. Again, our results are in good agreement with

theirs, and the slight difference could be due to the direct steam injection into the reservoir and the cyclic pattern of the process.

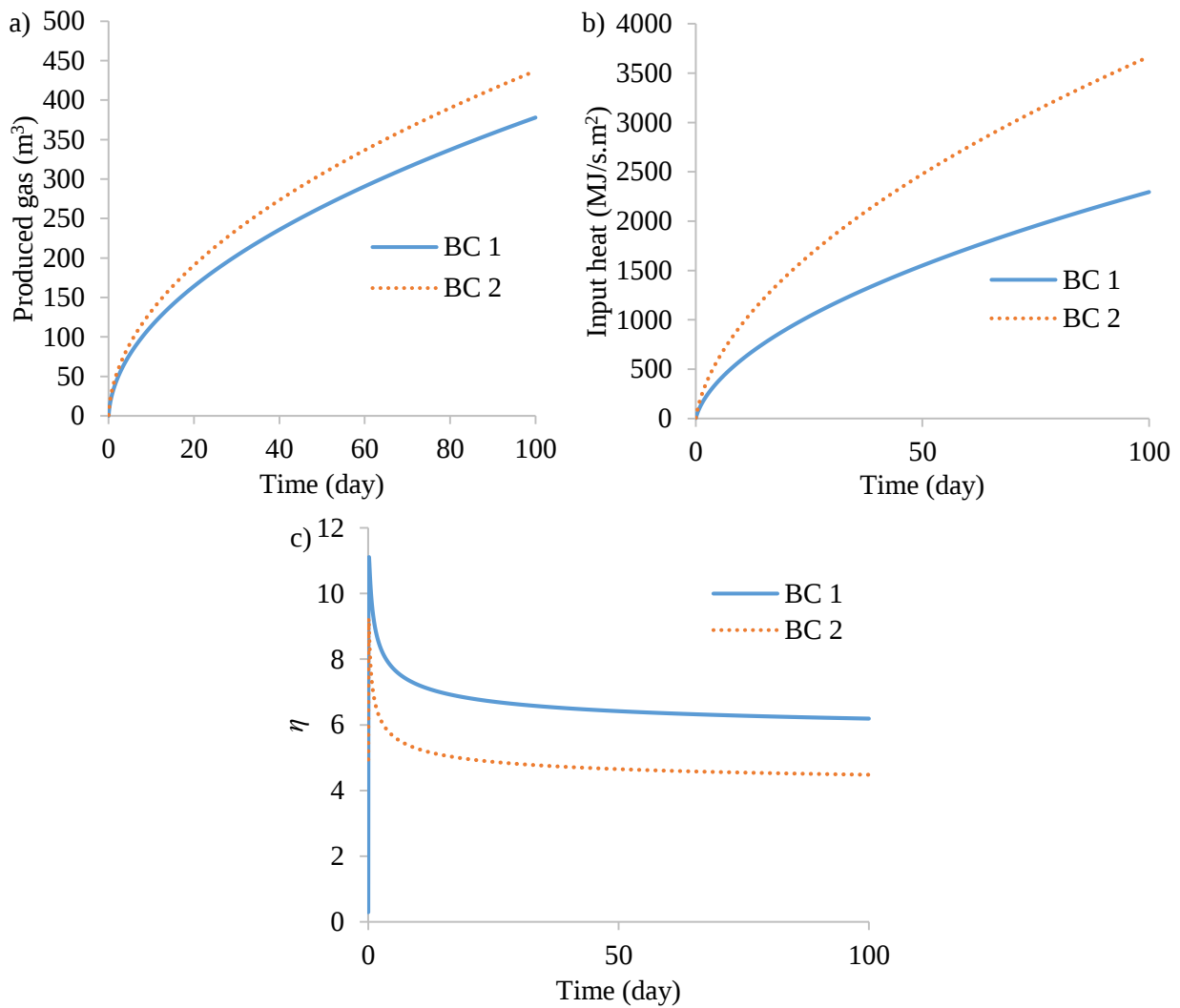


Figure 9. a) volume of produced gas, b) amount of input heat, and c) energy efficiency during hydrate dissociation for two BCs.

A parametric study is performed in order to investigate the effect of various parameters, stated in Table 2, on the dissociation process. The obtained results from the parametric study, shown in Figure 10, indicate the following points: a) the higher the thermal diffusivity and conductivity of Zone I, the higher the dissociation movement; b) the lower the thermal diffusivity and conductivity of Zone II, the higher the dissociation movement; and c) the permeability and gas viscosity almost have no effect on the dissociation process as the interface velocity does not change by various values of each. The same results were reported by Selim et al. (Selim and Sloan, 1990). Zhao et

al. (Zhao et al., 2015) investigated the numerical model of gas production from MH using thermal stimulation and showed that increasing the thermal conductivity promoted the dissociation process, on the other hand, different relative water-gas permeabilities have almost no effect on the gas production. They also numerically showed that increasing sediments' thermal conductivity would increase the rate of gas generation at the beginning of hydrate dissociation upon depressurization (Zhao et al., 2014). It should be mentioned that both of their works were validated by the experimental work of Masuda (MASUDA, 1999). Tsimpanogiannis et al. (Tsimpanogiannis and Lichtner, 2007) investigated a parametric study on the effect of different parameters on the MH dissociation upon thermal stimulation. They revealed that increasing the thermal conductivity of the porous media induced more MH dissociation.

Table 2. Range of parameters employed in the parametric study.

Porosity, ϕ	0.1 to 0.5
Permeability, k , μm^2	0.1 to 5
Thermal diffusivity of Zone I, α_I , $\mu\text{m}^2/\text{s}$	1×10^6 to 5×10^6
Thermal conductivity of Zone I, k_I , $\text{W}/(\text{m.K})$	3 to 7
Thermal diffusivity of Zone II, α_{II} , $\mu\text{m}^2/\text{s}$	4×10^5 to 8×10^5
Thermal conductivity of Zone II, k_{II} , $\text{W}/(\text{m.K})$	1 to 5
Gas viscosity, μ , Pa.s	10^{-4} to 10^{-6}

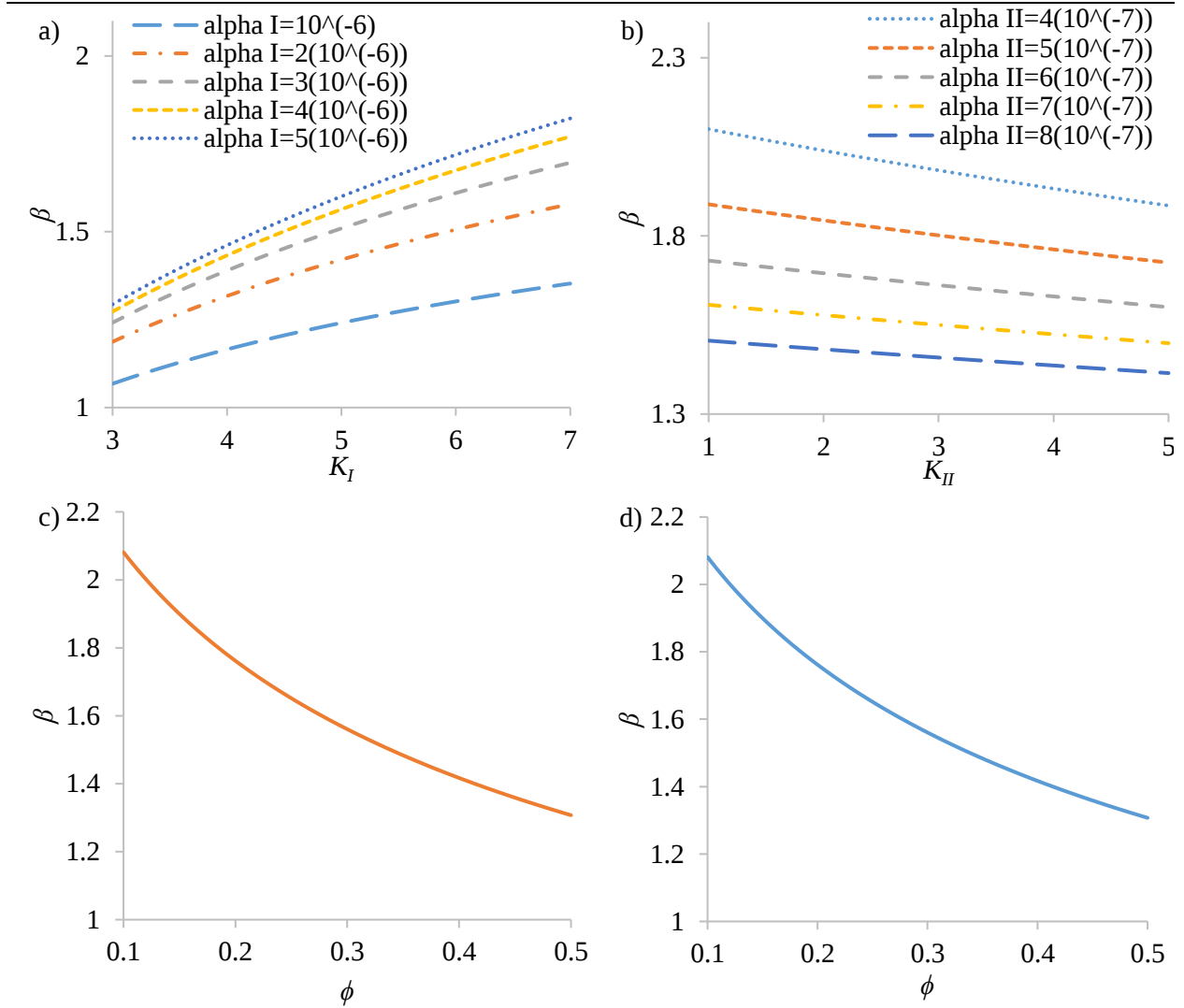


Figure 10. The effect of various values of the parameters stated in Table 2 on the interface movement after 100 days dissociation. a) thermal diffusivity and thermal conductivity of Zone I, b) thermal diffusivity and thermal conductivity of Zone II, c) various values of porosity with different permeabilities, and d) various values of porosity with different gas viscosities.

4 Conclusions

In this study an analytical model to investigate MH dissociation using thermal stimulation is presented. It takes into account the effect of wellbore thickness which may include multiple layers with different thermal properties. Evaluations of the effects of various reservoir parameters and boundary conditions on the dissociation process have been performed. The two main factors that have been considered for assessing the process are: a) moving dissociation boundary

position/velocity (λ), and b) energy efficiency of the process (η). These findings have significant implications for the understanding of how the wellbore structure affect the hydrate dissociation by providing an analytical solution for thermal stimulation method by taking into account heat conduction through the wellbore, which makes the models closer to the real conditions. This study appears to be the first study that examines associations between the wellbore structure and hydrate dissociation. In the limit when wellbore thickness approaches zero the model is in good agreement with previous similar analytical study. Additionally, the results of the present study match with those of previous mathematical and experimental investigations. Based on the results of this investigation, the following main conclusions can be made:

- The rate of dissociation and efficiency of reservoir heating are dependent on both well thickness and composition (which are design parameters). Well thickness causes a reduction in the dissociation front speed and produced gas amount compared to the cases without considering the well thickness.
- Temperature of the well boundary is not constant, which is also dependent on well thickness and composition.
- As a result, pressure and temperature at the dissociation front are not constant during dissociation.
- Increasing the well temperature, but decreasing its pressure, induce higher dissociation interface velocity and volume of produced gas, but reduces energy efficiency. Thus, in order to increase the energy efficiency of the process, a detailed study on the initial and boundary conditions including well composition (number of layers, their thickness and thermal properties) of the system should be performed.
- The good agreement between the results of this study and previous experimental and numerical studies provides validation of the assumptions made in the model development.

Acknowledgement

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Nomenclature			
A	Dimensionless constant in equation S9	n_{ft}	Total moles of produced gas per surface area of the moving interface up to time t
A_a	Dimensionless constant in equation 8	P	Pressure in Zone I
A_w	Wellbore area	P_s	Pressure at the interface
a	Dimensionless constant in equation S12	P_i	Pressure of the heat source
B	Dimensionless constant in equation S10	P_{STP}	Pressure of gas at STP conditions
B_a	Dimensionless constant in equation 8	Q_{Hd}	Heat of MH dissociation
BC	Boundary conditions	Q_g	Heating value of the gas at STP conditions
b	Dimensionless constant in equation S13	Q_{ft}	Total input heat to the reservoir from the heat source
C	Dimensionless constant in equation S11	R	Universal gas constant
C_{pI}	Specific heat capacity of Zone I	R_w	Thermal resistivity of the wellbore
C_{pg}	Specific heat capacity of gas		
c	Dimensionless constant in equation 11	S	Interface position
D	Dimensionless constant in equation S14	T_i	Temperature of the heat source
d	Dimensionless constant in equation 11	T_I	Temperature in Zone I
E	Dimensionless constant in equation S15	T_{II}	Temperature in Zone II
F	Dimensionless constant in equation S16	T_0	Initial temperature of hydrate
F_{gH}	Ratio of mass of the methane gas trapped inside the MH to the mass of hydrate	T_s	Temperature at the interface
$G(\beta)$	Dimensionless constant in equation S20	T_{STP}	Temperature of gas at STP conditions
$H(\beta)$	Dimensionless constant in equation S20	t	Time
$I(\beta)$	Dimensionless constant in equation S20	u_f	Heat flux from the well
$J(\beta)$	Function in equation S17	V_f	Volume of produced gas per surface area of the moving interface in the time fraction of “ $t, t-I$ ”
k	Permeability	V_{fp}	Total volume of produced gas per surface area of the moving interface up to time t
k_I	Thermal conductivity of Zone I	v_s	Interface velocity
k_{II}	Thermal conductivity of Zone II	X	Axial position
k_c	Thermal conductivity of cement	ρ_g	Gas density
k_g	Thermal conductivity of gravel	ρ_H	Hydrate density
k_s	Thermal conductivity of casing	μ	Gas viscosity
$K(\beta)$	Function in equation S18	λ	Dimensionless variable in equation 18

L	Thickness of the well	β	Dimensionless constant in equation 19
$L_1(\lambda)$	Function in equation S19	λ_{os}	Dimensionless variable in equation 20
L_c	Cement thickness	ϕ	Porosity
L_g	Gravel thickness	α_I	Thermal diffusivity of Zone I
L_s	Casing thickness	α_{II}	Thermal diffusivity of Zone II
MH	Methane hydrate	v_g	Gas velocity
m	Gas molecular mass	η_f	energy efficiency ratio
n_f	Total moles of produced gas per surface area of the moving interface in the time fraction of “ $t, t-l$ ”		

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Supplementary Information

• Materials and methods

In the following, the transformation of the fundamental equations, initial and boundary conditions in terms of λ is provided.

$$\text{Equation 4: } \frac{d^2 T_{II}}{d\lambda^2} + 2\lambda \frac{dT_{II}}{d\lambda} = 0, \quad \beta < \lambda < \infty \quad (\text{S1})$$

$$\text{Equation 12: } \frac{d^2 T_I}{d\lambda^2} + \frac{2\alpha_{II}\lambda}{\alpha_I} \frac{dT_I}{d\lambda} + \frac{C_{pg} km}{2k_I \mu R} \frac{d^2 P^2}{d\lambda^2} = 0, \quad \lambda_{os} < \lambda < \beta \quad (\text{S2})$$

$$\text{Equation 13: } \frac{P}{T_I} \frac{dP}{d\lambda} = \frac{2F_{gH} \phi \rho_H \alpha_{II} \mu R \beta}{km}, \quad \lambda = \beta \quad (\text{S3})$$

$$\text{Equation 14: } \frac{d}{d\lambda} \left(\frac{P}{T_I} \frac{dP}{d\lambda} \right) + \frac{2\phi \alpha_{II} \mu \lambda}{k} \frac{d}{d\lambda} \left(\frac{P}{T_I} \right) = 0, \quad \lambda_{os} < \lambda < \beta \quad (\text{S4})$$

$$\text{Equation 6: } \frac{-k_I A_w}{\sqrt{4\alpha_{II} t}} \frac{dT_I}{d\lambda} = \frac{(T_i - T_I)}{R_w}, \quad \lambda = \lambda_{os} \quad (\text{S5})$$

$$\text{Equation 7: } P_s = \exp(A - B/T_s), \quad \lambda = \beta \quad (\text{S6})$$

$$\text{Equation 9: } k_{II} \frac{dT_{II}}{d\lambda} - k_I \frac{dT_I}{d\lambda} = 2\phi \rho_H \alpha_{II} Q_{hd} \beta, \quad \lambda = \beta \quad (\text{S7})$$

$$\text{And, from equation 10: } T_{II} = T_0, \quad \lambda \rightarrow \infty \quad (\text{S8})$$

Furthermore, in a previous work reported by Selim et al. (Selim and Sloan, 1990), it is stated that

due to the very small ratio of $\frac{\rho_g}{\rho_H}$, the rate of hydrate dissociation (dS/dt) is too slow that the

transient term in the continuity equation could be neglected (second term in equation S4).

In the equation 21, A, B, and C constants are defined using equations S5 and 11 as follows:

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$$A = \frac{(T_s - T_i)F}{\text{Ferf}(a\beta + b) - \text{Ferf}(b) - D - E} \quad (\text{S9})$$

$$B = \frac{-A(D + E)}{F} + T_i \quad (\text{S10})$$

$$C = \frac{T_s - T_e}{\text{erfc}(\beta)} \quad (\text{S11})$$

, where a, b, D, E, and F are as follows:

$$a = \left(\frac{\alpha_{II}}{\alpha_I} \right)^{1/2} \quad (\text{S12})$$

$$b = \frac{C_{pg} F_{gH} \phi \rho_H \alpha_{II} \beta}{a k_I} \quad (\text{S13})$$

$$D = \frac{-k_I 2a \exp(-(a\lambda_{os} + b)^2)}{e\sqrt{\pi}} \quad (\text{S14})$$

$$E = -F(-\text{erf}(a\lambda_{os} + b) + \text{erf}(b)) \quad (\text{S15})$$

$$F = \frac{1}{R_w} \quad (\text{S16})$$

In the equation 22, pressure distribution in zone I, the $J(\beta)$, $K(\beta)$, and $L_1(\lambda)$ are defined as follows:

$$J(\beta) = \frac{4F_{gH} \phi \rho_H \alpha_{II} \mu R \beta}{km} \quad (\text{S17})$$

$$K(\beta) = B - A\text{erf}(b) \quad (\text{S18})$$

$$L_1(\lambda) = \lambda \text{erf}(a\lambda + b) + \frac{\text{berf}(a\lambda + b)}{a} + \frac{\exp(-(a\lambda + b)^2)}{a\sqrt{\pi}} \quad (\text{S19})$$

Now, equation S7 by insertion of the resulted T_I and T_{II} formulas (equation 21 and considering equation 10 becomes:

[Type here]

$$\frac{ak_I(T_i - T_s)}{k_{II}(T_s - T_0)} \left(\underbrace{\frac{F \exp(-(a\beta + b)^2)}{\text{F erf}(a\beta + b) - \text{F erf}(b) - D - E}}_{G(\beta)} \right) + \left(\underbrace{\frac{-\exp(-\beta^2)}{\text{erfc}(\beta)}}_{H(\beta)} \right) = \underbrace{\sqrt{\pi} \phi \rho_H \alpha_{II} \beta}_{I(\beta)} \frac{(c + dT_s)}{k_{II}(T_s - T_0)} \quad (\text{S20})$$

So, temperature at the interface is calculated as follows:

$$T_s = \frac{k_{II}T_0H(\beta) - ak_IT_iG(\beta) + cI(\beta)}{-ak_IG(\beta) - dI(\beta) + k_{II}H(\beta)} \quad (\text{S21})$$

From the equation 22, the pressure at the dissociation interface (P_s) would be:

$$P_s = \left(P_0^2 + J(\beta)K(\beta)(\beta - \lambda_{os}) + AJ(\beta)(L_1(\beta) - L_1(\lambda_{os})) \right)^{1/2} \quad (\text{S22})$$

Now, by implementing the equation 8 (Antoine equation), which is transformed to the following form (equation S23), the exact solution of temperature and pressure distributions will be achieved.

$$\frac{B_a}{A_a - \ln(P_s)} - T_s = 0 \quad (\text{S23})$$

Actually, β is the root of the equation S23 as all the parameters are known at each time except β . MATLAB programming software is employed to find the root of the equation S23 at each time and to perform all other calculations in this work. By finding β , the temperature and pressure distribution will also be calculated at each time.

The transformed form of the heat flux (equation 24) from the well due to the transformation term of equation 18 is as follows:

$$u_f = \frac{-k_I}{\sqrt{4\alpha_{II}t}} \frac{dT_I}{d\lambda}, \quad \lambda = \lambda_{os} \quad (\text{S24})$$

By inserting T_I from equation 21, equation S24 becomes:

$$u_f = \frac{-2ak_I A \exp(-(a\lambda_{os} + b)^2)}{\sqrt{4\alpha_{II} \pi t}} \quad (\text{S25})$$

In order to calculate the total volume of gas produced at STP conditions, the molar quantity of produced gas at each time should be calculated first as follows:

[Type here]

$$V_f = \phi(S_t - S_{t-1}) \quad (\text{S26})$$

$$n_f = F_{gH} \rho_H V_f / m \quad (\text{S27})$$

Equation S26 shows the volume of dissociated hydrate at each time step ($t-1, t$), and equation S27 represents the moles of produced gas at each time step ($t-1, t$). By summing the moles of produced gas at each time step, total moles of produced gas up to time t will be achieved as presented in equation S28.

$$n_{ft} = \sum_{t=0}^t n_f \quad (\text{S28})$$

Finally, the total volume of produced gas at STP conditions up to time t will be calculated by using equations S28 and 5 as presented by equation 25.