NUMERICAL STUDY ON PERMEABILITY HYSTERESIS DURING HYDRATE DISSOCIATION IN HOT WATER INJECTION

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ABSTRACT

Hot water injection is a production technique proposed to gas recovery from methane hydrate reservoirs. However, from a practical point of view, the injected water experiences a drop in temperature and re-formation of hydrates may occur in the reservoir. In this work, we proposed a model expressing permeability hysteresis in the processes between hydrate growth and dissociation, and studied hydrate dissociation behavior during hot water injection. The model of permeability hysteresis was incorporated into the simulator MH21-HYDRES (MH21 Hydrate Reservoir Simulator), where the decrease in permeability with hydrate saturation during hydrate growth process was assumed to be much larger than the decrease during hydrate dissociation process. Laboratory hydrate dissociation experiments were carried out for comparison. In each experiment, we injected hot water at a constant rate into a sand-packed core bearing hydrates, and the histories of injection pressure, core temperature, and gas/water production rates were measured. Numerical simulations for the core experiments showed the re-formation of hydrates led to the increase in injection pressure during hot water injection. The simulated tendencies of pressure increase varied markedly by considering permeability hysteresis. Since the experimental pressure increases could not be reproduced without the permeability hysteresis model, the influence of permeability hysteresis should be considered to apply hot water injection to hydrate reservoirs.

Keywords: methane hydrate, gas production, hot water injection, permeability hysteresis, numerical simulation

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NOMENCLATURE

 A_g Specific surface area of gas particles $[0.375\ \text{mm}^{-1}]$

 A_{HS} Specific surface area of hydrate particles $[0.375 \text{ mm}^{-1}]$

C Specific heat $[J \cdot kg^{-1} \cdot K^{-1}]$

D Depth from a reference level [m]

 f_e Gas fugacity at V-H-L_w equilibrium [MPa]

 f_g Gas fugacity in pore space [MPa]

g Acceleration of gravity [9.80665 $m^2 \cdot s^{-1}$]

h Enthalpy [J·kg⁻¹]

 K_B Hydrate dissociation rate constant [mol m⁻²·MPa⁻¹·s⁻¹]

 K_{gen} Hydrate formation rate constant [0.31 mol m⁻ 2 ·Pa⁻¹·s⁻¹]

 k_D Absolute permeability $[m^2]$

 k_{D0} Absolute permeability at $S_H=0$ [m²] krl Relative permeability to phase (1) [fraction]

krw° End-point water permeability [fraction]

krg° End-point gas permeability [fraction]

m Power of S_{wD}

N Permeability reduction index

 N_{hys} Permeability reduction index during hydrate re-formation / dissociation

n Power of $(1 - S_{wD})$

 \dot{n}_i Net generation rate of component (i) per 1m³ of sediment during hydrate dissociation /formation and water freezing/ice melting process (i = CH₄, H₂O) [mol·s⁻¹·m⁻³]

 $\dot{n}_{dissoc,i}$ Generation rate of component (i) due to hydrate dissociation per 1m³ of sediment (i = CH₄, H₂O) [mol·s⁻¹·m⁻³]

 $\dot{n}_{prod,l}$ Production rate of phase (1) per 1m³ of sediment [mol·s⁻¹·m⁻³]

p pressure [Pa]

 \dot{Q}_{H} Heat sink rate due to hydrate dissociation per 1m³ of sediment [W·m⁻³]

 \dot{Q}_{I} Heat sink rate with ice-water phase transition per 1m³ of sediment [W·m⁻³]

 \dot{Q}_{salts} Heat generation rate with salt dissolution into water per 1m³ of sediment [W·m⁻³]

 \dot{Q}_{ext} Heat sink rate to outside at the system boundary per 1m³ of sediment [W·m⁻³]

 $\dot{Q}_{prod,l}$ Heat sink rate with production of phase (l) per 1m³ of sediment [W·m⁻³]

R_{inj} Injection rate of hot water [ml/min]

 $S_{H_{hys}}$ The hydrate saturation at the reformation

 S_{H_norm} The normalized hydrate saturation in reformation / dissociation process

 $S_{iw} \ Irreducible \ water \ saturation \ [fraction]$

S₁ Saturation of phase (1) [fraction]

 S_{rg} Residual gas saturation [fraction]

T Temperature [K]

T_{inj.} Temperature of injected water [K]

t Time [s]

- U Internal energy [J kg⁻¹]
- x_i Mole fraction of component (i) in water phase (i
- = MeOH, salts) [fraction]

 λ_{eff} Effective thermal conductivity of sediment, $[W \cdot m^{-1} \cdot K^{-1}]$

 μ Viscosity [Pa·s]

 $\overline{\rho}$ Mass density [kg·m⁻³]

 ρ Molar density [mol·m⁻³]

Subscripts

g Gas phase H Hydrate phase I Ice phase R Sand grain (rock matrix) w Water phase

INTRODUCTION

Methane hydrate is a crystalline solid composed of water and methane. The total amount of methane gas in this solid form may surpass the total conventional gas reserve. And some individual methane hydrate accumulations may contain significant and concentrated resources [1]. That indicates the potential as a future energy resource. Hot water injection is one of gas production methods from hydrate reservoirs [2]. In this method, hydrate re-formation and permeability reduction may occur when the gas and water generated by dissociation of hydrates flow through the reservoir. These phenomena lead to the increase in injection pressure and may be the trigger of formation fracturing. So. the understanding of hydrate re-formation and permeability reduction is critical for application of the method. Sakamoto et al. reported permeability change by hydrate re-formation and concluded that the permeability changed rapidly when the hydrate saturation exceeded a threshold [3] [4]. In this study, we considered that the permeability change has hysteresis in the processes between hydrate formation and dissociation. We modeled the permeability hysteresis and incorporated the model into the original developed simulator: MH21HYDRES (MH21 Hydrate Reservoir Simulator). Thorough the comparison between numerical simulation and experimental data, the permeability hysteresis model was validated.

THEORY OF SIMULATOR Governing equations

The MH21-HYDRES is a compositional simulator solving the equations of mass balances for methane, water, methanol and salts, and one energy balance equation. The mass and energy balances equations are as follows:

Mass balance equations:

For methane components:

$$\nabla \cdot \left(\frac{k_D k_{rg} \rho_g}{\mu_g} \left(\nabla p_g - \overline{\rho}_g g \nabla D \right) \right) + \dot{n}_{CH_4} - \dot{n}_{prod,g}$$
$$= \frac{\partial}{\partial t} \left(\rho_g \phi S_g \right) \tag{1}$$

For water components:

$$\nabla \cdot \left(\frac{k_D k_{rw} \rho_w (1 - x_{MeOH} - x_{salts})}{\mu_w} (\nabla p_w - \overline{\rho}_w g \nabla D) \right) + \dot{n}_{H_2O} - (1 - x_{MeOH} - x_{salts}) \dot{n}_{prod,w} = \frac{\partial}{\partial t} \left[\rho_w \phi S_w (1 - x_{MeOH} - x_{salts}) \right]$$
(2)

For methanol components:

$$\nabla \cdot \left(\frac{k_D k_{rw} \rho_w x_{MeOH}}{\mu_w} (\nabla p_w - \overline{\rho}_w g \nabla D) \right)$$

$$- x_{MeOH} \dot{n}_{prod,w} = \frac{\partial}{\partial t} (\phi \rho_w x_{MeOH} S_w)$$
(3)

For salts components:

$$\nabla \cdot \left(\frac{k_D k_{rw} \rho_w x_{salts}}{\mu_w} \left(\nabla p_w - \overline{\rho}_w g \nabla D \right) \right) - x_{salts} \dot{n}_{prod,w}$$
$$= \frac{\partial}{\partial t} \left(\phi \rho_w x_{salts} S_w \right) \tag{4}$$

Energy balance equation:

$$\frac{\partial}{\partial t} \left\{ (1-\phi)\overline{\rho}_{R}C_{R}T + \phi \left(\sum_{l=I,H,salts} S_{l}\overline{\rho}_{l}C_{l}T + \sum_{l=w,g} S_{l}\overline{\rho}_{l}U_{l} \right) \right\}$$
$$= \nabla \cdot (\lambda_{eff}\nabla T) + \nabla \cdot \left(\sum_{l=w,g} h_{l} \frac{k_{D}k_{rl}\overline{\rho}_{l}}{\mu_{l}} (\nabla p_{l} - \overline{\rho}_{l}g\nabla D) \right)$$
$$- \dot{Q}_{H} - \dot{Q}_{I} + \dot{Q}_{salts} - \dot{Q}_{ext} + \sum_{l=w,g} \dot{Q}_{prod,l}$$
(5)

Permeability hysteresis during hydrate reformation / dissociation

The permeability of hydrate reservoirs can change due to presence of hydrates. Masuda et al. have proposed a following model to express this phenomenon [5].

$$k_{D} = k_{D0} \left(1 - S_{H} \right)^{N} \tag{6}$$

In dissociation process, as is common in depressurization method, this model is supported by experimental data [6]. On the other hand, in reformation / dissociation process, the permeability can change more rapidly than that of just dissociating process. We modeled this rapid change of permeability as:

$$k_{D} = k_{D0} \left(1 - S_{H_{hys}} \right)^{N} \left(1 - S_{H_{norm}} \right)^{N_{hys}}$$
(7)

$$S_{H_norm} = \frac{S_H - S_{H_hys}}{1 - S_{H_hys}}$$
(8)

where S_{H_hys} is the hydrate saturation at the reformation and S_{H_norm} is the normalized hydrate saturation in re-formation / dissociation process. In this model, the permeability reduction index of reformation/dissociation process N_{hys} is larger than N in Eq. (6). Figure 1 shows this permeability hysteresis in the processes between hydrate formation and dissociation.



Figure 1 Permeability reduction ratio vs. S_H.

Relative permeability

The gas and water relative permeabilities were modeled as follows:

Gas relative permeability:

$$k_{rg} = k_{rg}^{o} (1 - S_{wD})^{n}$$
⁽⁹⁾

Water relative permeability:

$$k_{rw} = k_{rw}^{o} S_{wD}^{m}$$
(10)

$$S_{wD} = \frac{\frac{S_w}{1 - S_H} - S_{iw}}{1 - S_{iw} - S_{rg}}$$
(11)

Figure 2 shows the gas and water relative permeability curves used for this study. Parameters in Eq. (9) and (10) were set as Table 1. Residual gas saturation was used as a matching parameter for gas production.



Figure 2 Relative permeability curves.

Table 1 Parameters for relative permeability.

Parameter	Value
krg°	1.0
krw°	0.2
n	2
m	3
S_{iw}	0.15
Srg	0.45

Hydrate formation / dissociation

Clarke-Bishnoi equation was used to model hydrate dissociation [7].

$$\dot{n}_{dissoc,CH_4} = K_B \phi S_H A_{HS} \left(f_e - f_g \right)$$
(12)

Hydrate formation was modeled based on the work of Malrgaonkar et al. [8].

$$\dot{n}_{form,CH_4} = K_{gen} \phi S_w S_g A_g (f_e - f_g)$$
(13)

EXPERIMENTAL

Apparatus and procedure

Artificial methane hydrate cores were prepared for experiments. The core length and diameter were 150 mm and 50 mm respectively. The core was packed into the rubber sleeve. The rubber sleeve thickness was 10 mm. The core holder as shown in Figure 3 was used for the dissociation experiments. The core was maintained at constant pressure and temperature. Hot water was injected from one side of the core and the pressure of the other end was kept constant. Figure 4 shows the position of the sensor. The volumes of gas and water produced, temperatures of inside and outside of the core and pressures at the both ends of the core were measured.





Figure 3 The schematic diagram of core holder.

Figure 4 The position of the sensor.

150 mm Rubber sleeve

SIMULATION DETAILS Grid system

A cylindrical coordinate system was used. The hydrate core and rubber sleeve were divided into 7 x 30 grids. Figure 5 shows the schematic grid

system. Boundary conditions such as heat transfer were set to reproduce the experimental conditions.



Figure 5 The schematic grid system.

Model settings

Parameter studies (Run 1-3) and comparison studies (Run e1, 2) were conducted. These simulations have differences in models: reformation and hysteresis models are considered or not. Table 2 shows the model differences between these simulations.

Table 2. Model settings.

Run No	Re-formation	Hysteresis
	model	model
Run 1	With	With
Run 2	With	Without
Run 3	Without	Without
Run e1	With	With
Run e2	With	Without

Input data

Table 3 shows the input data for simulations. The data were based on the experiment. The permeability reduction index N and N_{hys} were decided by comparison with the experiment.

	Run 1, 2, 3	Run e1, 2
φ	0.4	0.4
k _{D0} (mD)	3000	3000
S_{H}	0.70	0.76
$\mathbf{S}_{\mathbf{w}}$	0.30	0.20
Sg	0.0	0.04
P _i (MPa)	8.1	8.1
T _i (K)	281.15	281.15
R _{ini.} (ml/min)	10	9.43
T _{inj.} (K)	303.15	Follow the
·		schedule
		(about 300)
Ν	2	2
N _{hys}	8	8

Table 3. Input data.

RESULTS AND DISCUSSION

The effect of hydrate re-formation and permeability hysteresis

We studied the effect of hydrate re-formation and permeability hysteresis using parameter studies with three different conditions (Run 1-3). Figure 6 shows the comparison of injection pressures of these three Runs. Through the comparison between Run 2 and Run 3, it was found that the reduction of differential pressure was delayed due to re-formation of hydrates. But the value of differential pressure was almost the same whether hydrates re-form or not. On the other hand, in Run 1 with permeability hysteresis, the differential pressure increased and decreased rapidly since the processes between re-formation and dissociation of hydrates caused rapid change of permeability. The comparison of cumulative gas produced in Figure 7 shows that re-formation and permeability hysteresis had a limited effect on gas productivity as long as hot water could be injected. However, the large injection pressure may be critical problem for application of the method.



Figure 6 The comparison of injection pressures.



Figure 7 The comparison of cumulative gas produced.

Comparison between the simulation and the experiment

We conducted comparison study between the simulation and the experiment. Injection pressure, cumulative gas produced and temperatures of inside of the core were compared. Figure 8 shows the comparison of injection pressure. Measured injection pressure showed the rapid increase and decrease. The simulation with permeability hysteresis model (Run e1) reproduced the rapid change of the injection pressure. In contrast, the simulation without permeability hysteresis model (Run e2) could not reproduce this tendency. In the experiment, reductions of injection pressure were seen at 35 min and 55 min. It was considered that heterogeneity of the core caused the some reductions. Figure 9 shows the comparison of cumulative gas produced. The difference among models was not obvious, however, the simulation with the permeability hysteresis model reproduced the measured data. Figure 10 shows the comparison of temperatures of inside of the core. The simulation with permeability hysteresis model reproduced the tendency of measured data. Figure 11 shows the profiles of hydrate saturation, gas saturation, pressure and temperature at 20 min, 50 min, 100 min and 200 min. In Figure 11, hot water was injected from the left end of the core. Hydrates were dissociated from left to right side of the core and re-generated at low temperature region. At 50 min when the injection pressure reached maximum value, hydrate saturation rose up to 0.79, and pressure dropped rapidly in hydrate saturated region. At 100 min when the injection pressure dropped to initial value, hydrates of center of the core disappeared and gas broke through the core.



Figure 8 The comparison of injection pressures.



Figure 9 The comparison of cumulative gas produced.



Figure 10 The comparison of temperatures.



Figure 11 The profiles of hydrate saturation, gas saturation, pressure and temperature.

CONCLUSION

We modeled permeability hysteresis in the processes between formation and dissociation of hydrates. Developed model was incorporated into the simulator: MH21-HYDRES, and a comparison study between simulation and experiment was conducted to validate the model. The simulator with permeability hysteresis model reproduced the rapid change of injection pressure measured by the experiment. Injection pressure increased to value a few MPa higher than the initial pressure since reformation of hydrates caused permeability reduction. Simulations showed that re-formation and permeability hysteresis had limited effect on gas productivity as long as hot water could be injected. However, the large injection pressure may be a critical problem for application of the method.

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REFERENCES

[1] Milkov A.V. *Global estimates of hydratebound gas in marine sediments: how much is really out there?*. Earth-Science Reviews 2004; 66:183-197.

[2] Masuda Y, Kurihara M, Ohuchi H, Sato T. *A Field-Scale Simulation Study on Gas Productivity of Formations Containing Gas Hydrates*, In: Proceedings of the Fourth International Conference on Gas Hydrates, Yokohama, 2002.

[3] Sakamoto Y, Komai T, Kawamura T, Minagawa H, Tenma N, Yamaguchi T. Laboratory-scale experiment of methane hydrate dissociation by hot-water injection and numerical analysis for permeability estimation in reservoir: Part 1 - Numerical study for estimation of permeability in methane hydrate reservoir. INTERNATIONAL JOURNAL OF OFFSHORE AND POLAR ENGINEERING 2007; 17(1):47-56.

[4] Sakamoto Y, Komai T, Kawamura T, Minagawa H, Tenma N, Yamaguchi T. Modification of permeability model and history matching of laboratory-scale experiment for dissociation process of methane hydrate: Part 2 -Numerical study for estimation of permeability in methane hydrate reservoir. INTERNATIONAL JOURNAL OF OFFSHORE AND POLAR ENGINEERING 2007; 17(1):57-66.

[5] Masuda Y, Fujinaga Y, Naganawa S, Fujita K, Sato T, Hayashi Y. *Modeling and Experimental Studies on Dissociation of Methane Gas Hydrates in Berea Sandstone Cores,* In: Proceedings of the 3rd International Conference on Gas Hydrates, Salt Lake City, 1999.

[6] Masuda Y, Konno Y, Kurihara M, Ouchi H, Kamata Y, Ebinuma T, Narita H. Validation Study of Numerical Simulator Predicting Gas Production Performance from Sediments Containing Methane Hydrates, In: Proceedings of the Fifth International Conference on Gas Hydrates, Trondheim, 2005.

[7] Clarke M, Bishnoi P.R. Determination of the Activation Energy and Intrinsic Rate Constant of Methane Gas Hydrate Decomposition. The Canadian Journal of Chemical Engineering 2001; 79:143-147.

[8] Malegaonkar M.B., Dholabhai P.D., Bishnoi P.R. *Kinetics of Carbon Dioxide and Methane Hydrate Formation*. The Canadian Journal of Chemical Engineering 1997; 75:1090-1099.