SIMULATION OF CH$_4$ RECOVERY FROM HYDRATE DEPOSITS BY INJECTION OF CO$_2$

by

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The paper presents a mathematical model of CH$_4$ recovery from gas hydrate deposits via injection of liquid CO$_2$. The process of CH$_4$ recovery is supposed to occur on the moving frontal boundary separating the deposit into two domains. The near domain is saturated with liquid CO$_2$ and its hydrate while the far one is saturated with CH$_4$ and its hydrate. Self-similar solutions describing the temperature and pressure fields were obtained. The effect of main parameters on the dynamics of the process under study was explored. It was revealed that at low temperatures of the CO$_2$ being injected the temperature in the far domain did not exceed that of equilibrium for decomposition of CH$_4$ gas hydrate. It was also found that with increasing pressure at which CO$_2$ was injected into the gas hydrate domain, the extension of the region saturated with CO$_2$ hydrate was increasing.

Keywords: *hydrate, CH$_4$ recovery, CO$_2$ injection*

Introduction

Recently, a rather serious alternative to traditional sources of natural gas has become gas hydrates. It is known that quite large volumes of CH$_4$ gas are concentrated in gas hydrate deposits located in permafrost zones and under the seafloor [1, 2]. Regarding to this, creating various technologies for extracting hydrocarbon gas from gas hydrate deposits is becoming urgent. Conventional methods of extracting CH$_4$ gas from hydrates are either heating or depressurization which is a rather complex technical problem under negative temperature conditions. Another problem not related to the extraction of natural gas from hydrates is constant emission of large amounts of CO$_2$ into the Earth's atmosphere [3, 4]. Consequently, the challenge is to seek for a fundamentally possible and promising method of CO$_2$ utilization. One of such methods is replacing CH$_4$ in its gas hydrate with liquid CO$_2$ [5-8]. Such a technology would allow both CH$_4$ to be recovered and CO$_2$ to be utilized. It is known that CO$_2$ hydrate is more stable than that of CH$_4$. Therefore, the process of replacing CO$_2$ molecules in its hydrate with CH$_4$ ones becomes possible [6].

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Belova, S. V., *et al.*: Simulation of CH_4 Recovery from Hydrate Deposits by …
THERMAL SCIENCE: Year 2019, Vol. 23, Suppl. 2, pp. S447-S454

S448

Experimental studies of the process of replacing CH_4 with CO_2 in its gas hydrate are discussed in [6-8]. In addition, in [8] the process of CO_2 hydrate formation is studied in a range of positive and negative temperatures. According to these, CH_4 replacement in its hydrate with liquid CO_2 will occur at temperatures and pressures that meet the conditions of stable hydrate [6, 7].

Mathematical models for formation and decomposition of gas hydrates when injecting the same gas which saturated the hydrate area in its initial state are considered in [9-13]. The papers [14, 15] present a mathematical model for injection of liquid CO_2 into a porous medium saturated with CH_4 and its gas hydrate.

The purpose of this work is to build a mathematical model of the process of CH_4 recovery from gas hydrate deposits via injection of liquid CO_2.

Problem statement and governing equations

Let a horizontal area at its initial time be saturated with a hydrate-forming mixture S_{hm0} (that of snow and CH_4) and CH_4 S_{gm0} at a negative temperature of, T_0, and a pressure of, p_0, that correspond to the stable state of CH_4 gas hydrate. We assume that liquid CO_2 is injected through the boundary $x = 0$ at the temperature, T_e, and pressure, p_e, that correspond to the stable state of CO_2 gas hydrate and are maintained constant on this boundary. The phase equilibrium diagram of the system is shown in fig. 1. To determine the dependence of the equilibrium temperature, $T_s(p)$, on the current pressure value, we apply the relation:

$$T_s\left(p\right) = T_{(s0)} + T_* \ln\left(\frac{p}{p_{(s0)}}\right) \tag{1}$$

where $T_{(s0)}$, $p_{(s0)}$ are the equilibrium temperature and pressure, respectively, and T_* – the empirical parameter.
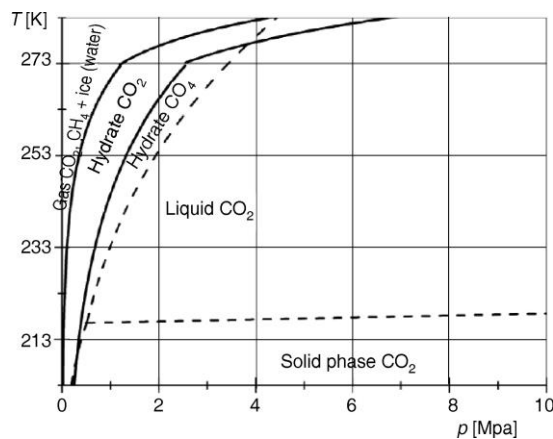

Figure 1. Phase equilibrium diagram of the H_2O-CH_4-CO_2 system

Given the assumptions, we write down a system of governing equations describing the processes of filtration and heat transfer in a gas-hydrate area for a 1-D flow. The process of replacement takes place on the front surface of $x(s)$ which divides the area into two domains. The near domain is saturated with liquid CO_2 and its hydrate. The far domain is saturated with CH_4 and its hydrate. Lower indices $i = d$ and $i = m$ correspond to the near domain, $0 < x < x(s)$, and the far one, $x(s) < x < \infty$, for CO_2 and CH_4, respectively.

We write the mass conservation equations for each domain:

$$\frac{\partial}{\partial t}(\rho_i S_i) + \frac{\partial}{\partial x}(\rho_i S_i v_i) = 0 \tag{2}$$

where ρ_i is the true density, S_i – the saturation, and v_i – the velocity of the ith phase.

The energy conservation equations are written:

Belova, S. V., *et al.*: Simulation of CH$_4$ Recovery from Hydrate Deposits by …
THERMAL SCIENCE: Year 2019, Vol. 23, Suppl. 2, pp. S447-S454

S449

$$\rho c \frac{\partial T_i}{\partial t} + \rho_i c_i S_i \upsilon_i \frac{\partial T_i}{\partial x} = \lambda \frac{\partial^2 T_i}{\partial x^2} \tag{3}$$

where c_i is the specific mass heat capacity of the ith phase, ρc – the specific volume heat capacity, and λ – the thermal conductivity of the system.

For the filtration process Darcy's law can be written:

$$S_i \upsilon_i = -\frac{k_i}{\mu_i} \frac{\partial p_i}{\partial x} \tag{4}$$

where μ_i is the gas dynamic viscosity and k_i is the gas permeability coefficient.

For CH$_4$ the Mendeleev-Clapeyron equation is written:

$$\rho_m = \frac{p_m}{R_g T_m}$$

For the liquid phase the equation of state is:

$$\rho_d = \rho_{0(d)} \exp\left[\beta(p_d - p_{0(d)})\right]$$

where β is the volume compression ratio of liquid CO$_2$.

Taking into account the conditions of mass and heat balance on the boundary between the two domains, the following relations are satisfied:

$$-\frac{k_i}{\mu_i} \frac{\partial p_i}{\partial x} = \left(S_m - \frac{\rho_{hm} G_m S_{hm}}{\rho_m}\right) \dot{x}_s \tag{5}$$

$$S_{hd} \rho_{hd} (1-G_d) \dot{x}_s = S_{hm} \rho_{hm} (1-G_m) \dot{x}_s \tag{6}$$

$$\lambda_d \frac{\partial T_d}{\partial x} - \lambda_m \frac{\partial T_m}{\partial x} = (\rho_{hd} S_{hd} l_{hd} - \rho_{hm} S_{hm} l_{hm}) \dot{x} \tag{7}$$

where l_{hi} is the specific heat of hydrate formation for the ith phase and $\dot{x}_s$ is the interface velocity.

The value of hydrate saturation in the CO$_2$ zone can be found from eq. (6) which is a condition of the hydrate-bound CO$_2$ mass balance:

$$S_{hd} = \frac{S_{hm} \rho_{hm} (1-G_m)}{\rho_{hd} (1-G_d)} \tag{8}$$

Let us introduce dimensionless quantities P_d and P_m in the following forms:

$$P_d = \frac{\rho_d}{\rho_{0(d)}} = \exp\left[\beta(p_d - p_{0(d)})\right], \quad P_m = \frac{p_d^2}{p_0^2} \tag{9}$$

Based on eqs. (2)-(4), the equations of piesoconductivity and thermal diffusivity are written:

$$\frac{\partial}{\partial t}(P_i) = \chi_i^p \frac{\partial}{\partial x}\left(\frac{\partial P_i}{\partial x}\right) \tag{10}$$

$$\frac{\partial T_i}{\partial t} = \chi^T Pe_d \frac{\partial p_i}{\partial x} \frac{\partial T_i}{\partial x} + \chi^T \frac{\partial^2 T_i}{\partial x^2} \tag{11}$$

where

$$\text{Pe}_\text{d} = \frac{\rho_{0\text{d}} c_\text{d}}{\lambda} \frac{k_\text{d}}{\mu_\text{d} \beta}, \quad \text{Pe}_\text{m} = \frac{\rho_{0\text{m}} c_\text{m}}{2\lambda} \frac{k_\text{m} p_0}{\mu_\text{m}}, \quad \chi_0^p = \frac{k_\text{ld}}{S_\text{ld} \mu_\text{ld} \beta},$$

$$\chi_0^p = \frac{k_\text{gm} p_\text{m0}}{S_\text{gm0} \mu_\text{gm0}}, \quad \varphi = \frac{\chi^T}{4\chi_0^p}, \quad \chi^T = \frac{\lambda}{\rho c}$$

This system is augmented with the following initial and boundary conditions:

$$t = 0 \ (x \geq 0): S_\text{g} = S_\text{gm0}, \quad S_\text{h} = S_\text{hm0}, \quad T = T_0, \quad p = p_0$$

$$x = 0 \ (t > 0): T = T_\text{e}, \quad p = p_\text{e}$$

$$x = \infty \ (t > 0): T = T_0, \quad p = p_0$$

Since the pressure at the interface between the domains is assumed to be continuous, the values of P_sd and P_sm are related by the ratio:

$$P_\text{sd} = \exp\left[\beta p_0 \left(\sqrt{P_\text{sm}} - 1 \right) \right] \tag{12}$$

Self-similar solution

Let us introduce the self-similar variable ξ:

$$\xi = \frac{x_\text{s}}{2\sqrt{\chi_0^p t}} \tag{13}$$

The equations of piesoconductivity eq. (10) and thermal diffusivity eq. (11) for this variable are written:

$$-\xi \frac{\text{d}P_\text{d}}{\text{d}\xi} = \frac{\chi_\text{d}^p}{2\chi_0^p} \frac{\text{d}^2 P_\text{d}}{\text{d}\xi^2}, \quad -\xi \frac{\text{d}P_\text{m}}{\text{d}\xi} = \frac{1}{2} \frac{\text{d}^2 P_\text{m}}{\text{d}\xi^2} \tag{14}$$

$$-\xi \frac{\text{d}T_\text{d}}{\text{d}\xi} = 2\text{Pe}_\text{d} \varphi \frac{\text{d}p_\text{d}^2}{\text{d}\xi} \frac{\text{d}T_\text{d}}{\text{d}\xi} + 2\varphi \frac{\text{d}^2 T_\text{d}}{\text{d}\xi^2} \tag{15}$$

On integrating eqs. (14) and (15), we can write the solution for the pressure and temperature distributions in each domain:

$$P_\text{d} = P_\text{sd} + (P_\text{e} - P_\text{sd}) \frac{\int_{\xi_\text{s}}^{\xi} \exp\left(-\frac{\xi^2}{\eta}\right) \text{d}\xi}{\int_0^{\xi_\text{s}} \exp\left(-\frac{\xi^2}{\eta}\right) \text{d}\xi}, \quad T_\text{d} = T_\text{s} + (T_\text{e} - T_\text{s}) \frac{\int_{\xi_\text{s}}^{\xi} \exp\left(-\frac{\xi^2}{4\varphi} - \text{Pe}_\text{d} P_\text{d}\right) \text{d}\xi}{\int_0^{\xi_\text{s}} \exp\left(-\frac{\xi^2}{4\varphi} - \text{Pe}_\text{d} P_\text{d}\right) \text{d}\xi} \tag{16}$$

$$P_\text{m} = l_0 + (P_\text{sm} - 1) \frac{\int_{\xi}^{\infty} \exp(-\xi^2) \text{d}\xi}{\int_{\xi_\text{s}}^{\infty} \exp(-\xi^2) \text{d}\xi}, \quad T_\text{m} = T_0 + (T_\text{s} - T_0) \frac{\int_{\xi}^{\infty} \exp\left(-\frac{\xi^2}{4\varphi} - \text{Pe}_\text{m} P_\text{m}\right) \text{d}\xi}{\int_{\xi_\text{s}}^{\infty} \exp\left(-\frac{\xi^2}{4\varphi} - \text{Pe}_\text{m} P_\text{m}\right) \text{d}\xi} \tag{17}$$

where

$$\eta = \frac{\chi_\text{d}^p}{\chi_0^p}$$

Belova, S. V., *et al.*: Simulation of CH$_4$ Recovery from Hydrate Deposits by …
THERMAL SCIENCE: Year 2019, Vol. 23, Suppl. 2, pp. S447-S454

S451

Based on the conditions eqs. (5)-(7) and taking into account the solutions eqs. (16) and (17), we obtain equations for determining the displacement front co-ordinate, ξ_s, and the values of p_s and T_s on it:

$$\frac{(P_e - P_{sd})\exp\left(-\dfrac{\xi^2}{\eta}\right)}{\displaystyle\int_0^{\xi_s}\exp\left(-\dfrac{\xi^2}{\eta}\right)\mathrm{d}\xi} = A_d \xi_s P_d \tag{18}$$

$$\frac{(P_{sm} - 1)\exp\left(-\xi^2\right)}{\displaystyle\int_{\xi_s}^{\infty}\exp\left(-\xi^2\right)\mathrm{d}\xi} = A_m \sqrt{P_m}\,\xi_s \tag{19}$$

$$\frac{(T_s - T_e)\exp\left(-\dfrac{\xi^2}{4\varphi} - \mathrm{Pe}_d P_d\right)}{\displaystyle\int_0^{\xi_s}\exp\left(-\dfrac{\xi^2}{4\varphi} - \mathrm{Pe}_d P_d\right)\mathrm{d}\xi} - \frac{(T_0 - T_s)\exp\left(-\dfrac{\xi^2}{4\varphi} - \mathrm{Pe}_d P_d\right)}{\displaystyle\int_{\xi_s}^{\infty}\exp\left(-\dfrac{\xi^2}{4\varphi} - \mathrm{Pe}_d P_d\right)\mathrm{d}\xi} = B\xi_s \tag{20}$$

where

$$A_d = \frac{\chi_0^p \mu_d \beta}{k_i}\left(1 - S_{hd} + \frac{\rho_{hd} G_d S_{hd}}{\rho_d}\right), \quad A_m = \frac{2\chi^p \mu_d}{p_0 k_m}\left(1 - S_{hm} + \frac{\rho_{hm} G_m S_{hm}}{\rho_m}\right)$$

$$B = \frac{2\chi_0^p}{\lambda}(\rho_{hd} S_{hd} l_{hd} - \rho_{hm} S_{hm} l_{hm})$$

Expressing the quantity of P_{sm} from eq. (18) and substituting it into the ratio eq. (12), we find P_{sd} as a function of ξ_s. Substituting $P_{sd}(\xi_s)$ into eq. (18), we result in a transcendental equation with one unknown ξ_s, which is solved by the bisection method. From eqs. (12), (18), and (20) we obtain the values of pressure, P_s, and temperature, T_s, on the boundary $x_{(s)}$.

Results of numerical analysis

For the thermophysical parameters, gas, CO$_2$, and hydrate the following values were taken: $G_m = 0.12$, $G_d = 0.28$, $R_g = 520$ [Jkg^{-1}K^{-1}], $\rho_{0d} = 890$ [kgm^{-3}], $\rho_{hm} = 900$ [kgm^{-3}], $\rho_{hd} = 1100$ [kgm^{-3}], $k_0 = 10^{-16}$ [m^2], $c_{gm} = 1560$ [Jkg^{-1}K^{-1}], $c_{ld} = 2600$ [Jkg^{-1}K^{-1}], $c_{hm} = 2500$ [Jkg^{-1}K^{-1}], $c_{hd} = 2500$ [Jkg^{-1}K^{-1}], $\mu_d = 10^{-4}$ [Pa·s], $\mu_m = 10^{-5}$ [Pa·s], $\lambda_{hm} = 0.5$ [Wm^{-1}K^{-1}], $\beta = 10^{-8}$ [Pa^{-1}], $l_{hd} = 1.5 \cdot 10^5$ [Jkg^{-1}] (at the $T < 0$ ℃), $l_{hm} = 1.45 \cdot 10^5$ [Jkg^{-1}] (at the $T < 0$ ℃).

For the equilibrium temperature and the corresponding pressure, as well as the empirical parameter included in the eq. (1), the following values were taken: the phase equilibrium condition for CH$_4 - T_{(s0)} = 263$ [K], $p_{(s0)} = 1.86$ [MPa], $T_* = 30$ [K], the phase equilibrium condition for CO$_2 - T_{(s0)} = 263$ [K], $p_{(s0)} = 0.67$ [MPa], $T_* = 16$ [K]. In the graphs, the equilibrium hydrate formation curve eq. (1) is indicated by a dash-dotted line.

For the CH$_4$ hydrate temperature, pressure, and saturation characterizing the initial state of the gas-CH$_4$ hydrate system, the following values were taken, respectively, $T_0 = 233$ [K], $p_0 = 1.5$ [MPa], and $S_{hm} = 0.3$.

Figure 2(a) illustrates the temperature and fig. 2(c) pressure field distributions in the computation domain at different pressure values of the liquid CO$_2$ being injected p_e, with scaled-up fragments of the temperature, 2(b), and pressure, 2(d), distributions being located on the right. In the figure, the dash-dotted line shows the values of the moving boundary ξ_s for the corresponding pressure values of the CO$_2$ being injected. At the pressure values of

$p_e = 2$ [MPa], $p_e = 3$ [MPa], $p_e = 5$ [MPa], the moving boundary takes the values of $\xi_s = 0.034$, $\xi_s = 0.084$, $\xi_s = 0.166$, respectively. It can be seen from the graphs that with increasing the pressure at which CO_2 is injected into the gas hydrate domain, the extension of the area saturated with CO_2 hydrate is increasing.
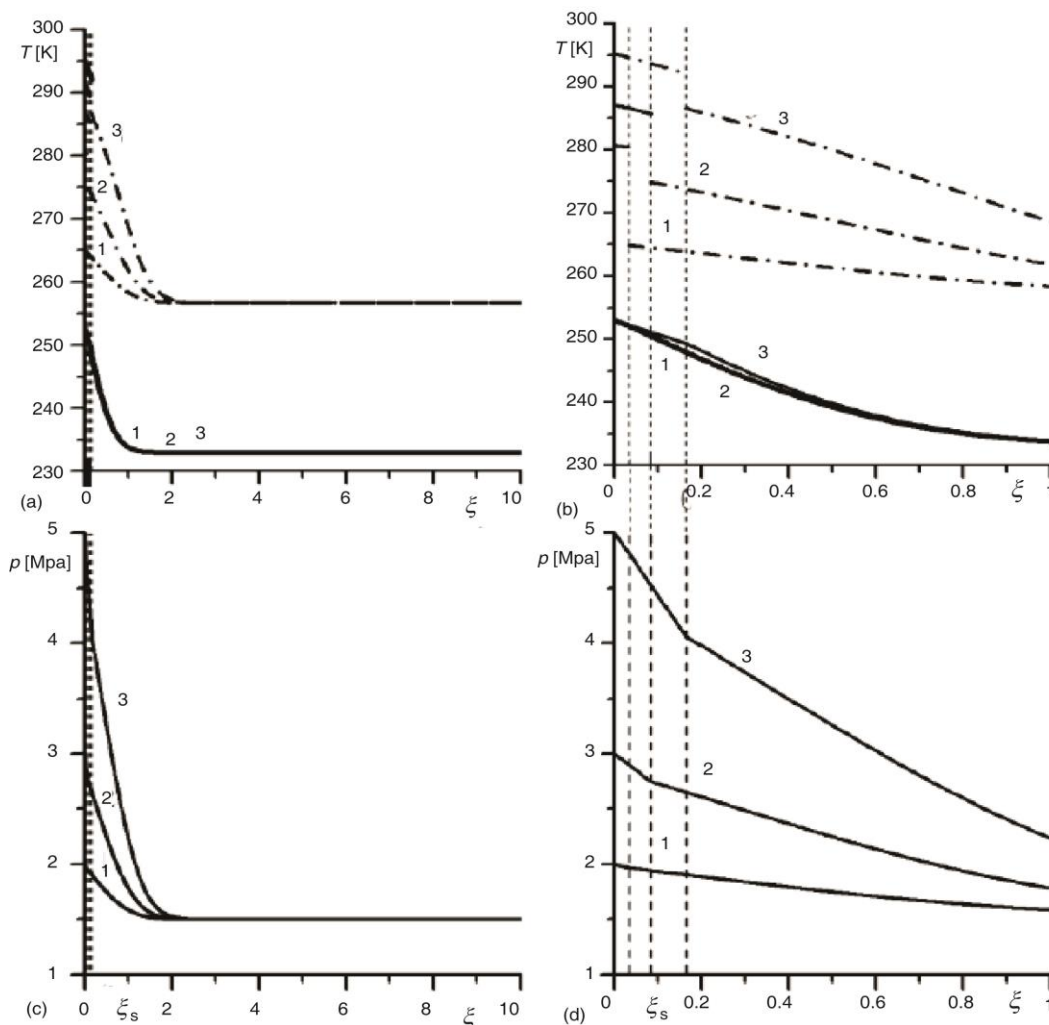

Figure 2. Temperature (a), (b) and pressure (c), (d) distributions in the domain at different pressures, p_e, of the liquid CO_2 being injected depending on the dimensionless self-similar variable, ξ; 1 − at $p_e = 2$ [MPa], 2 − at $p_e = 3$ [MPa], and 3 − at $p_e = 5$ [MPa]

Figure 3(a) shows the temperature and fig. 3(c) pressure field distribution in the computation domain at different temperatures of the liquid CO_2 being injected T_e, with scaled-up fragments of the temperature, 3(b), and pressure, 3(d), distributions being located on the right. At different temperatures of the CO_2 being injected the boundary coordinate ξ_s takes the value of $\xi_s = 0.034$. It can be seen from the graphs that if CO_2 is injected at a temperature that is lower than the initial one in the gas hydrate domain, the temperature rises to its initial value in the domain.
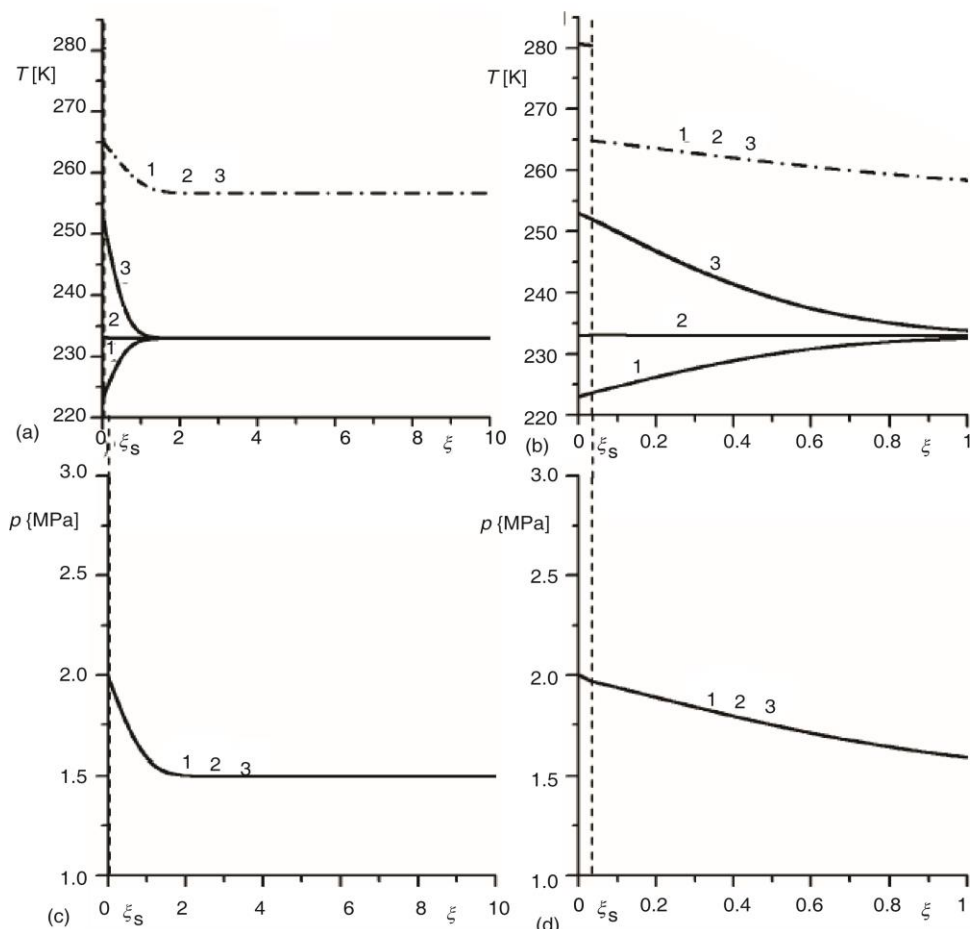
Belova, S. V., *et al.*: Simulation of CH$_4$ Recovery from Hydrate Deposits by …
THERMAL SCIENCE: Year 2019, Vol. 23, Suppl. 2, pp. S447-S454

S453

Figure 3. Temperature (a), (b) and pressure (c), (d) distributions in the domain at different temperatures, T_e, of the liquid CO$_2$ being injected depending on the dimensionless self-similar variable ξ; 1 − at T_e = 223 [K], 2 − at T_e = 233 [K], and 3 − at T_e = 253 [K]

Conclusion

A numerical analysis of the process of replacing CH$_4$ with liquid CO$_2$ in the gas hydrate domain at negative temperatures was conducted. Self-similar solutions describing the temperature and pressure fields were constructed. The impact of the main and initial parameters on the dynamics of the process under study was investigated. It was shown that at low temperatures of the CO$_2$ being injected, the temperature in the far domain did not exceed the equilibrium decomposition temperature for CH$_4$ gas hydrate. It was found that with increasing pressure at which CO$_2$ was injected into the gas hydrate domain, the extension of the region saturated with CO$_2$ hydrate was increasing.

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Belova, S. V., *et al.*: Simulation of CH$_4$ Recovery from Hydrate Deposits by …

S454   THERMAL SCIENCE: Year 2019, Vol. 23, Suppl. 2, pp. S447-S454

Nomenclature

c – specific mass heat, [Jkg^{-1}K^{-1}]
G – mass content of the phase in the hydrate
k – permeability coefficient, [m^2]
l – specific heat of hydrate formation, [Jkg^{-1}]
p – pressure, [Pa]
R – reduced gas constant, [Jkg^{-1}K^{-1}]
S – volume saturation of the phase
T – temperature, [K]
t – time
x – co-ordinate, [m]

Greek symbols

β – coefficient of volume compression of liquid CO$_2$, [Pa^{-1}]
λ – coefficient of thermal conductivity of the system, [Wm^{-1}k^{-1}]

μ – dynamic viscosity, [Pa·s]
ξ – dimensionless self-similar variable
ρ – true density, [kgm^{-3}]

Subscripts

0 – initial value of the parameter
d – liquid CO$_2$
e – value at the discharge boundary
g – gas phase
h – hydrate phase
i – phases corresponding to the areas of liquid CO$_2$ and CH$_4$
l – liquid phase
m – methane
s – mobile frontal boundary between the areas of CO$_2$ and CH$_4$

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