

MODELLING THE PROCESS OF NON-EQUILIBRIUM HYDRATE FORMATION IN A POROUS RESERVOIR

M.K. Khasanov¹, S.R. Kildibaeva¹, M.V. Stolpovskiy²

¹Ufa University of Science and Technology, Sterlitamak, Russian Federation

²Ufa State Petroleum Technological University, Ufa, Russian Federation

E-mail: hasanovmk@mail.ru, freya.13@mail.ru, s_maxim.pmm@mail.ru

This paper presents a solution to the flat-dimensional problem of gas hydrate formation in a porous medium. Highly permeable reservoirs are considered, as a result of which it is assumed that the process accompanied by the transition of gas into the hydrate composition is nonequilibrium. Based on the numerical solution, the influence of injection pressure and formation permeability on the peculiarities of phase transitions process has been studied. It is shown that with an increase in the injection pressure, both the maximum possible temperature of the system and the length of the hydrate-containing region increase. It has been found that the maximum temperature realized in the system, depending on the permeability of the reservoir, has a non-monotonic character. The influence of the initial temperature of the porous reservoir on the dynamics of phase transitions has been studied.

Keywords: gas hydrate; phase transitions; nonequilibrium process; porous medium; kinetics.

Introduction

Natural deposits of hydrates are found at the bottom of seas and oceans, as well as in permafrost. The main aspect of the study of hydrates is the fact that the gas reserves contained in them are large enough and can provide the need for it for a very long period of time. According to estimates, the content of natural gas in gas hydrates is estimated at about 10^{15} m^3 [1, 2]. Another feature of the study is the fact that gas hydrates can accumulate a sufficiently large amount of industrial gases at sufficiently low economic costs [3, 4].

Experimental studies in the field of hydrate formation are presented, in particular, in [5, 6]. In these papers studies of gas hydrate formation were carried out in samples of small size and usually under thermal and barostatic conditions. Therefore, in these studies, due to the small size of the samples, the process of gas hydrate formation is limited only by the kinetics of the process. The works [7–9] consider mathematical models of the formation and decomposition of methane gas hydrates in porous reservoirs under thermal and depression effects. Studies on the replacement of methane in the hydrate composition of carbon dioxide are presented in [10, 11].

This paper presents a mathematical model of gas injection into a solid water-saturated porous medium, accompanied by a process of phase transitions in a nonequilibrium regime. This is based on the fact that the gas-solid gas hydrate transition process is considered for highly permeable formations, where in addition to heat and mass transfer, the kinetics of the process must be taken into account. Note that in contrast to [7–9], in the present work, the mathematical model takes into account the real properties of gas (to describe the dependence of the methane supercompressibility factor on pressure and temperature, the Latonov–Gurevich equation is used). It is shown that gas injection in the reservoir can

be divided into three characteristic regions. In addition, the effect of injection pressure and formation permeability on the maximum temperature realized in the system and the extent of the area saturated with gas hydrate has been studied. It is also shown that as the initial formation temperature increases, the extent of the region containing gas hydrate decreases.

1. Problem Statement

For small drops in pressure and temperature, values of the equilibrium pressure p_s as a function of the temperature T are approximated by [12]

$$p_s = p_{s0} \cdot \exp((T - T_{s0})/T_*), \quad (1)$$

where T_* is the empirical parameter. The system of basic equations for the real gas is the following [13]:

$$\begin{aligned} m \frac{\partial(\rho_g S_g)}{\partial t} + \frac{\partial}{\partial x}(m S_g v_g \rho_g) &= -J_g, \\ m \rho_l \frac{\partial S_l}{\partial t} = -J_l, m \rho_h \frac{\partial S_h}{\partial t} &= -J_h, \\ \rho_c \frac{\partial T}{\partial t} + \rho_g c_g m S_g v_g \left(\frac{\partial T}{\partial x} + \varepsilon_g \frac{\partial p}{\partial x} \right) &= \frac{\partial}{\partial x} \left(\lambda \frac{\partial T}{\partial x} \right) + \rho_g c_g m S_g \eta_g \frac{\partial p}{\partial t} + J_h L_h, \\ m S_g v_g &= -\frac{\kappa_g}{\mu_g} \frac{\partial p}{\partial x}, p = z_g \rho_g R_g T. \end{aligned} \quad (2)$$

Here t is the time; m is the porosity reservoir; S_i , ρ_i , J_i ($i = l, h, g$) are the pore volume saturation, density and intensity of the i -th phase consumption, respectively; v_g , κ_g , μ_g are the speed, permeability and dynamic viscosity of methane, respectively; L_h is the specific heat of hydrate formation; z_g is the methane supercompressibility coefficient; R_g is the reduced methane constant; ρ_c and λ are the specific volume heat capacity and thermal conductivity of the reservoir. Since the main contribution to the values of ρ_c and λ is made by the corresponding parameters of the porous medium skeleton, we consider them as constant values. Hereinafter, the lower indices $i = l, h, g$ refer to water, hydrate, and methane, respectively.

Let us represent the dependence of the gas supercompressibility coefficient z_g on pressure and temperature as follows [14]:

$$z_g = \left(0,4 \cdot \lg \left(\frac{T}{T_c} \right) + 0,73 \right)^{p/p_c} + 0,1p/p_c, \quad (3)$$

where p_c and T_c are the critical pressure and temperature of CH_4 , respectively. Thus, for the coefficients of differential throttling and adiabatic cooling of gas, we can write:

$$\varepsilon_g = -\frac{1}{\rho_g c_g} \frac{T}{z_g} \left(\frac{\partial z_g}{\partial T} \right)_p, \quad \eta_g = \frac{1}{\rho_g c_g} - \varepsilon_g,$$

where the derivative $\left(\frac{\partial z_g}{\partial T} \right)$ is taken at constant pressure.

Taking into account that a solid hydrate is a two-component system with a mass concentration of gas G , in order to relate the values of the rates of water flow J_l and gas J_g for hydrate formation with the value of the intensity J_h , we can write the following relations:

$$J_l = (1 - G) J_h, \quad J_g = G J_h. \quad (4)$$

From the second and third equations of system (2), taking into account (4), we can obtain:

$$m\rho_l (S_l - S_{l0}) = -m\rho_h (1 - G) S_h. \quad (5)$$

In deriving (5), it is assumed that in the initial state, the formation is saturated with CH_4 and H_2O , the volume fraction of which is equal to S_{l0} , and there is no hydrate. The volume fraction of gas hydrate in this case is equal to zero. From expression (5), we can get a limit on the maximum possible value of hydrate saturation $S_{h\max}$ realized at full transition of water ($S_l = 0$) in the hydrate state:

$$S_{h\max} = \rho_l S_{l0} / (\rho_h (1 - G)).$$

Dependence of the phase permeability coefficient on the current gas saturation S_g and the absolute permeability k_0 of the reservoir is as follows [15]:

$$k_g = k_0 S_g^3.$$

Intensity of gas consumption per unit volume for hydrate formation, according to [16], is as follows:

$$J_g = M_g K A_s (f - f_{eq}). \quad (6)$$

In (6), M_g is the mass of 1 mol of hydrate-forming gas CH_4 ; K is the reaction parameter of decomposition or hydrate formation; A_s is the surface area of the hydrate formation; f_{eq} and f is the CH_4 fugacity, determined for both equilibrium and for current system pressure. In the case of hydrate formation, according to [16], we assume that $K = 5,875 \cdot 10^{-12} mol / (m^2 \cdot Pa \cdot s)$, $A_s = \sqrt{\frac{m^3(S_g+S_l)^3}{2k}} (S_g S_l S_h)^{2/3}$. In the case of nonequilibrium decomposition of a solid hydrate $K = K_0 \exp(-\frac{\Delta E}{RT})$, where $K_0 = 3,6 \cdot 10^4 mol / (m^2 \cdot Pa \cdot s)$, $\Delta E/R = 9752,73K$; $A_s = \sqrt{\frac{m^3(S_g+S_l)^3}{2k}} S_h$.

We find the fugacity f of the real gas, corresponding to the pressure p , on the basis of the expression

$$\ln f = \ln p_{st} + \int_{p_{st}}^p \frac{z_g}{p} dp, \quad (7)$$

where $p_{st} = 10^5 Pa$.

Initial value conditions and conditions on the left boundary of the reservoir are represented in the form

$$\begin{aligned} t = 0 : p = p_0, T = T_0, S_h = 0, S_l = S_{l0}; \\ x = 0 : p = p_w, T = T_w. \end{aligned} \quad (8)$$

As conditions on the right boundary of the reservoir, we use conditions of the form

$$x = L : p = p_0, \frac{\partial T}{\partial x} = 0. \quad (9)$$

When solving the resulting system of equations, the following parameters are given: the skeleton porosity m , the water densities ρ_l and the gas hydrate ρ_h , the absolute reservoir permeability k_0 , the dynamic gas viscosity μ_g , the specific heat of hydrate formation L_h , the reduced gas constant R_g , the gas heat capacity c_g , the relative mass content gas in gas hydrate G , the specific volumetric heat capacity ρ_c and the thermal conductivity coefficient λ of the system. In this case, the following unknown functions are found: the pressure p , the temperature T , the hydrate saturation S_h and the water saturation S_l . These unknowns are functions of the coordinate x and the time t . An implicit finite-difference method was used to solve the system of differential equations [17]. For this, a uniform space-time grid was introduced.

2. Analysis of Results

Fig. 1 shows the distribution of the main parameters saturation of the porous medium for the case of gas injection under pressure $p_w = 8 \text{ MPa}$ and the temperature $T_w = 278 \text{ K}$ in a porous formation with the absolute permeability $k_0 = 10^{-13} \text{ m}^2$ for the time moment $t = 0,38 \text{ h}$. For the other parameters characterizing the system, the following values were taken: $p_0 = 5 \text{ MPa}$, $T_0 = 280 \text{ K}$, $S_{l0} = 0,15$ $m = 0,2$, $\lambda = 2 \text{ W}/(m \cdot K)$, $\rho_c = 2,5 \cdot 10^6 \text{ J}/(K \cdot m^3)$, $\rho_l = 1000 \text{ kg}/m^3$, $\rho_h = 900 \text{ kg}/m^3$, $G = 0,13$, $L_h = 5 \cdot 10^5 \text{ J}/kg$, $T_{s0} = 280 \text{ K}$, $T_* = 10 \text{ K}$, $p_{s0} = 5,5 \text{ MPa}$, $\mu_g = 10^{-5} \text{ Pa} \cdot s$, $M = 16 \cdot 10^{-3} \text{ kg}/mol$, $R_g = 520 \text{ J}/(kg \cdot K)$, $c_g = 1560 \text{ J}/(kg \cdot K)$, $l = 50 \text{ m}$. From Fig. 1 shows that the hydrate saturation of near area takes the maximum possible value corresponding to the full transition of water to the hydrate state. The pores of the far region contain gas and water. The intermediate region separating the above-mentioned areas is saturated with gas and water. In this area, hydrate saturation varies from maximum to zero. In the far region, only gas and water are present in the pores.

Fig. 2 shows the dependence of the maximum temperature realized in the porous medium (fragment *a*) and the coordinate of the area occupied by gas hydrate (fragment *b*) on the injection pressure p_w for the time moment $t = 10 \text{ min}$. The initial reservoir pressure was assumed to be $p_0 = 4 \text{ MPa}$. As follows from the fragment *a*, the maximum temperature of the porous medium increases with increasing injection pressure. This is explained due to increasing pressure of injection and, consequently, an increase in release of latent heat of phase transition.

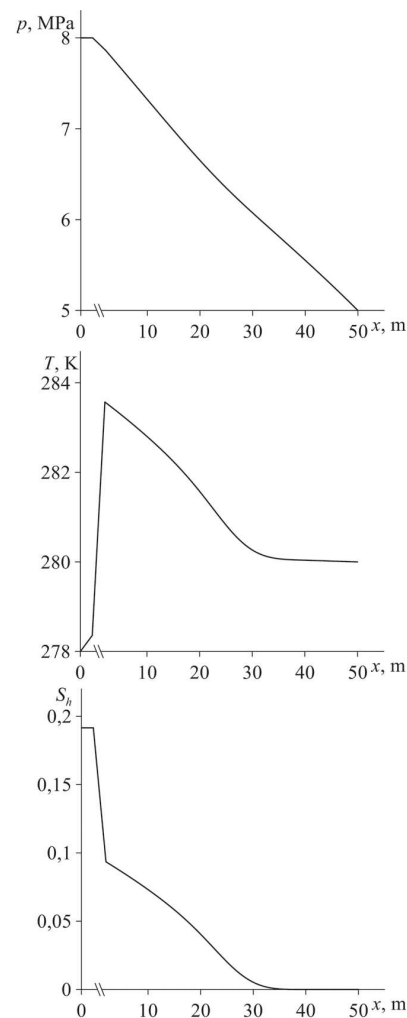


Fig. 1. Distribution of pressure, temperature and hydrate saturation of the porous medium

Fragment *b* of Fig. 2 also shows that with an increase in the gas pressure, the extent of the region, the extent of the area containing gas hydrate increases. This can be explained by the fact that with increasing values of p_w , the conditions corresponding to hydrate formation are satisfied in most of the reservoir.

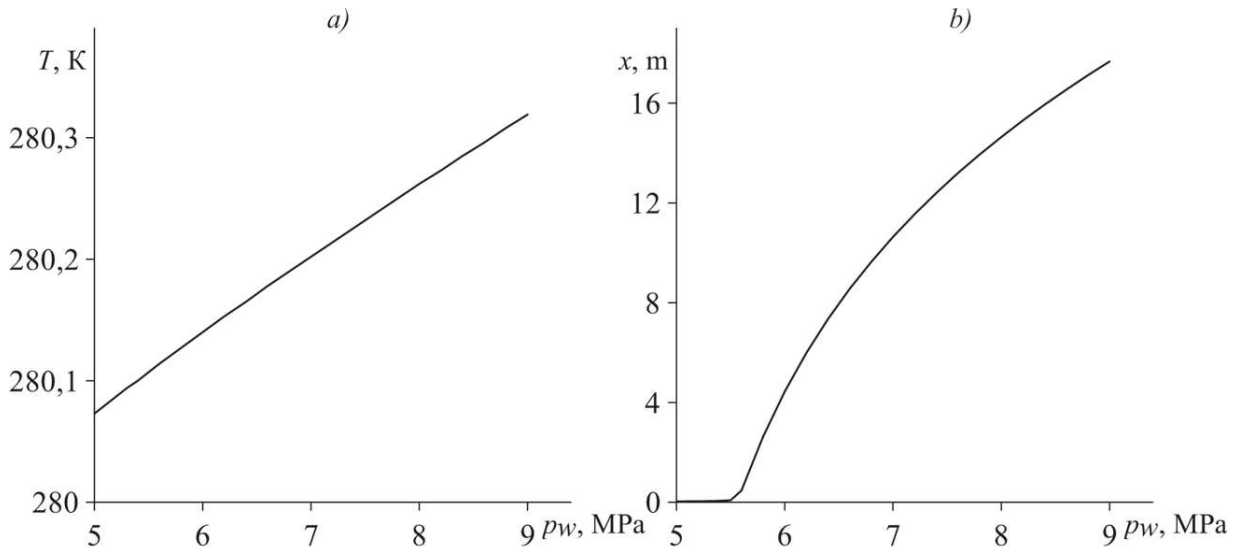


Fig. 2. Dependence of the maximum temperature (a) and the coordinate of the area occupied by the hydrate (b) on the injection pressure

Fig. 3 shows the dependence of the maximum possible temperature realized in the porous medium (fragment *a*) and the coordinate of the area occupied by hydrate (fragment *b*) on the formation permeability coefficient for the time moment $t = 10 \text{ min}$. The initial pressure and injection pressure were assumed to be $p_0 = 4 \text{ MPa}$ and $p_w = 6 \text{ MPa}$, respectively. As follows from fragment *a*, the maximum possible temperature of a porous medium, depending on of the system depending on formation permeability has a non-monotonic character. At the same time, in the region of low k_0 , with an increase in permeability, the maximum possible temperature of the porous medium increases. This is explained by the fact that with increasing permeability increases the intensity of hydrate formation and, consequently, the growth of system temperature. In this case, the adiabatic effect, associated with the real gas properties, appear rather weak. In the region of high values of permeability with growth of k_0 value the maximum temperature of the system is observed to decrease. In this case, the diabetic effect plays a more significant role, even in the case of an increase in the intensity of phase transitions. This leads to a decrease in the maximum possible system temperature with an increase in reservoir permeability.

From fragment *b* of Fig. 3. it follows that an increase in the permeability coefficient of a solid formation leads to a corresponding increase in the extent of the area occupied by the gas hydrate. This can be explained by the fact that with an increase in the value of k_0 , the area of propagation of the pressure disturbance also increases, which leads to a significant increase in the area occupied by the gas hydrate.

Fig. 4 shows the time dependence of the coordinate of the area occupied by gas hydrate. Lines 1 and 2 correspond to the initial temperature $T_0 = 280 \text{ K}$ and $T_0 = 280,5 \text{ K}$. The other parameters of the porous medium correspond to those in Fig. 1. From Fig. 4 it follows that in the initial moments of time after gas injection the length of the area occupied by gas hydrate increases to its maximum value, and then it practically does not change. With

increasing initial temperature of the system, the maximum extent of the area occupied by gas hydrate decreases. This is due to the fact that the conditions of gas hydrate existence are realized in smaller areas from the left boundary of the porous medium.

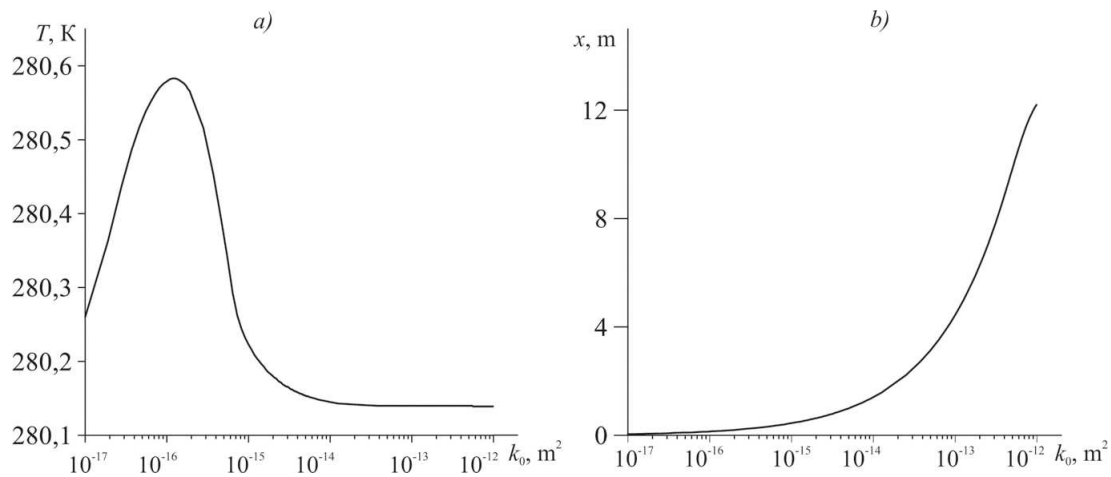


Fig. 3. Dependence of the maximum temperature (a) and the coordinate of the area occupied by hydrate (b) on formation permeability

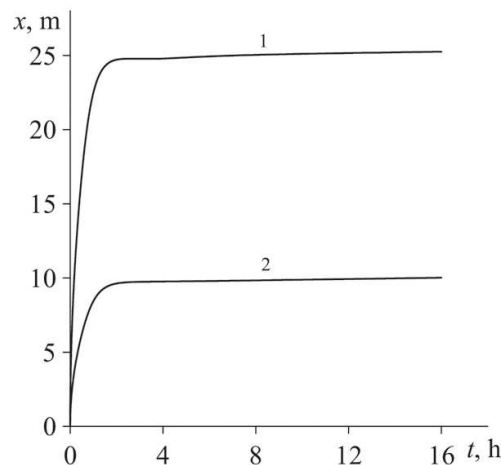


Fig. 4. Time dependence of the coordinate of the conditional boundary between the far field and intermediate regions

Conclusion

The paper presents a mathematical model of gas injection into a porous medium, accompanied by the process of phase transitions in a nonequilibrium regime. The distributions of the main parameters of the porous medium are constructed. It is shown that as a result of gas injection in the reservoir, three characteristic regions in which gas, hydrate and water are in different states can be distinguished. In addition, the effect of injection pressure and formation permeability on the system temperature and the intensity of phase transitions has been studied. It was shown that as the initial formation temperature increases, the length of the area saturated with gas hydrate decreases.

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МОДЕЛИРОВАНИЕ ПРОЦЕССА НЕРАВНОВЕСНОГО ГИДРАТООБРАЗОВАНИЯ В ПОРИСТОМ ПЛАСТЕ

М.К. Хасанов¹, С.Р. Кильдибаева¹, М.В. Столковский²

¹Стерлитамакский филиал Уфимского университета науки и технологий,
г. Стерлитамак, Российская Федерация

²Уфимский государственный нефтяной технический университет, г. Уфа,
Российская Федерация

В работе представлено решение плоскоодномерной задачи об образовании газогидрата в пористом пласте, изначально насыщенном метаном и водой. Рассматриваются высокопроницаемые пласты, вследствие чего полагается, что процесс гидратообразования является неравновесным. Система основных уравнений представляет собой законы сохранения масс, тепла и Дарси, которая дополняется заданием кинетики гидратообразования, а также начально-граничными условиями. Кроме того, представленная математическая модель учитывает параметры, которые приближены к реальным характеристикам системы: реальные свойства газа, эффекты Джоуля-Томсона и адиабатического сжатия и т.д. На основе численного решения исследовано влияние давления инъекции и проницаемости пласта на особенности протекания процесса гидратообразования. Показано, что с ростом давления инъекции возрастает как максимальная температура системы, так и протяженность области, насыщенная газогидратом. Установлено, что максимальная температура, реализуемая в системе, в зависимости от проницаемости пласта имеет немонотонный характер. Исследовано влияние исходной температуры пористого пласта на динамику образования газогидрата. Показано, что с увеличением исходной температуры пласта происходит уменьшение протяженности области, содержащей в своем составе газогидрат, уменьшается.

Ключевые слова: газовый гидрат; неравновесный процесс; кинетика.

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Марат Камилович Хасанов, кандидат физико-математических наук, доцент, кафедра прикладной информатики и программирования, Стерлитамакский филиал Уфимского университета науки и технологий (г. Стерлитамак, Российская Федерация), hasanovmk@mail.ru.

Светлана Рустамовна Кильдибаева, кандидат физико-математических наук, доцент, научный сотрудник, Стерлитамакский филиал Уфимского университета науки и технологий (г. Стерлитамак, Российская Федерация), freya.13@mail.ru.

Максим Владимирович Столповский, кандидат физико-математических наук, доцент, кафедра физики, Уфимский государственный нефтяной технический университет (г. Уфа, Российская Федерация), s_maxim.pmm@mail.ru.

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