

THEORETICAL MODELING OF THE CARBON DIOXIDE INJECTION INTO THE POROUS MEDIUM SATURATED WITH METHANE AND WATER TAKING INTO ACCOUNT THE CO₂ HYDRATE FORMATION

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Abstract. In this work the mathematical model is constructed and the features of the injection of warm carbon dioxide (with the temperature higher than the initial reservoir temperature) into the porous reservoir initially saturated with methane gas and water are investigated. Self-similar solutions of the one-dimensional problem describing the distributions of the main parameters in the reservoir are constructed. The effect of the parameters of the injected carbon dioxide and the reservoir on the intensity of the CO₂ hydrate formation is analyzed.

Keywords: gas hydrates, hydrate formation, filtration, porous medium, carbon dioxide.

1. INTRODUCTION

The most probable cause of global warming is an increase in the concentration of greenhouse gases, in particular CO₂. To combat this phenomenon, it is necessary to take measures to reduce the carbon dioxide concentration by regularly removing it from the atmosphere [1–7]. One of such ways of utilization of carbon dioxide is its disposal in porous reservoirs in the gas hydrate form [8,9]. Since the gas hydrate contains much more gas under identical conditions than in a free state, the gas hydrate state allows storing a large amount of carbon dioxide at relatively low pressures [10,11]. This reduces the risk of carbon dioxide release to the surface and actually increases the capacity of underground storage tanks for carbon dioxide. In addition, in natural porous reservoirs, the area of contact between water and gas takes enormous values. This property of porous media is

favorable for intensifying the process of hydrate formation and allows relatively quickly and economically to convert carbon dioxide into the gas hydrate state, for example, by pumping it into spent oil or gas reservoirs partially saturated with water.

It is worth noting that the organization of the carbon dioxide utilization in the gas hydrate form is a very complex technological problem, so a thorough and comprehensive theoretical study is needed [12, 13]. The theoretical study of the carbon dioxide hydrate formation in porous mediums is presented in [12–15]. In these papers mathematical models are proposed, analytical solutions of the problem are constructed in a plane-parallel formulation. In this paper the mathematical model of the warm carbon dioxide injection (with the temperature higher than the initial reservoir temperature) into a natural porous reservoir accompanied by the CO₂ hydrate formation is presented in a radially symmetric formulation.

2. MATHEMATICAL MODEL

Consider the process of injecting through the well of a radius r_e of carbon dioxide with known specific mass flow rate Q_e (per unit thickness of the reservoir) and temperature T_e . The well opens the reservoir over its entire thickness.

Let the horizontal porous reservoir initially saturated with methane gas and water, and the pressure p_0 and temperature T_0 in the reservoir correspond to the thermodynamic conditions of their existence in a free state: $t = 0, r > r_e : T = T_0, p = p_0, S_w = S_{w0}, S_g = 1 - S_{w0}$, where t is the time; r is the radial coordinate; S_i ($i = w, h, g$) is the pore saturation by the i -th phase (w is water, h is hydrate, g is gas); S_{w0} is the initial water saturation.

The coordinate r is measured from the central axis of the well. The initial pressure and temperature correspond to the thermodynamic conditions of the existence of the carbon dioxide hydrate.

As a result of the CO₂ injection into a porous reservoir, a region saturated with carbon dioxide and its hydrate is formed. We exclude the effect of the value of r_e on the features of the carbon dioxide hydrate formation in the porous medium. To do this, we will consider sufficiently large time values after the start of the CO₂ injecting, in which the radius of the region saturated with carbon dioxide and its hydrate is much greater than the well radius. Then the conditions on the left boundary of the reservoir can be written in the form: $t > 0, r = r_e \rightarrow 0 : Q = Q_e = \text{const}, T = T_e$.

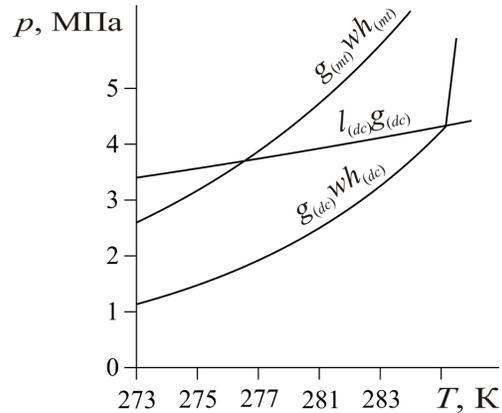


Fig. 1. Phase diagrams of the “CO₂-H₂O” and “CH₄-H₂O” systems

Fig. 1 shows the phase diagram of the systems “CO₂-H₂O” and “CH₄-H₂O” [16]. In this diagram, the curves gwh determine the three-phase equilibria “gas-water-hydrate”,

and the curve lg is the two-phase equilibrium “liquid-vapor”. The subscripts in parentheses (dc) and (mt) refer respectively to the parameters of carbon dioxide and methane. In the considered problem the initial reservoir pressure and temperature lie above the curve $g_{(dc)}wh_{(dc)}$ (in the region of existence of a mixture of carbon dioxide and its hydrate). The pressure and temperature of the injected carbon dioxide lie below the curve $l_{(dc)}g_{(dc)}$ (in the region of existence of CO₂ in gaseous state).

The values of temperature and pressure on the curve of the three-phase gas-water-hydrate equilibrium are quite well described by the equation [11]

$$T = T_0 + T_* \ln(p/p_{s0}), \quad (1)$$

where p_{s0} is the equilibrium pressure corresponding to the temperature T_0 ; T_* is an empirical parameter depending on the type of gas hydrate. In this problem, the following condition is satisfied: $p_{s0} < p_0$.

When studying the carbon dioxide hydrate formation at the CO₂ injection into a porous medium, we will assume that the intensity of the phase transition is limited by the filtration mass transfer rate in the porous medium. This is true for times significantly exceeding the characteristic time of the process kinetics. The experimental values of the hydrate formation kinetics in porous media strongly depend on the characteristic pore sizes, but on average they are on the order of several hours. For example, in the work [16], in which the formation and decomposition of carbon dioxide hydrate in a porous medium was studied, this time was of the order of one hour. These characteristic times are usually very small in comparison with the time of the carbon dioxide injection into a natural reservoir.

We will also neglect the diffusion mixing of carbon dioxide and methane, because at the continuous injection of carbon dioxide into a porous medium the mass transfer intensity due to filtration in the porous medium is significantly higher than the mass transfer intensity due to diffusion [12]. In addition, the front of the displacement of methane by carbon dioxide can be considered stable, because in natural reservoirs the flows are laminar and the viscosity of carbon dioxide is about one and a half times higher than the viscosity of methane (for the considered range of pressures and temperatures) [12, 13].

Then, considering that in the problem the initial state of the reservoir corresponds to the conditions of the carbon dioxide hydrate formation, it can be assumed that this formation occurs at the boundary of methane displacement by carbon dioxide. Consequently, in the considered case, two characteristic zones are formed in a reservoir (Fig. 2). In the near (first) zone of the reservoir (near to the well), the porous medium is filled with carbon dioxide and its hydrate. In the far (second) zone, the pores are saturated with methane and water. Correspondingly, the carbon dioxide hydrate is formed from carbon dioxide and water at the mobile interface ($r = r_s$) separating these zones.

To describe the processes of heat and mass transfer when CO₂ is injected into a porous reservoir, we assume the following assumptions. The process is one-temperature, i.e. the temperature of the porous medium and the saturating agents are the same. The CO₂ hydrate is a two-component system with mass concentration of gas $G_{(dc)}$. The porous medium skeleton and the gas hydrate are incompressible and immobile, the porosity m is constant, methane and carbon dioxide are calorically perfect gases.

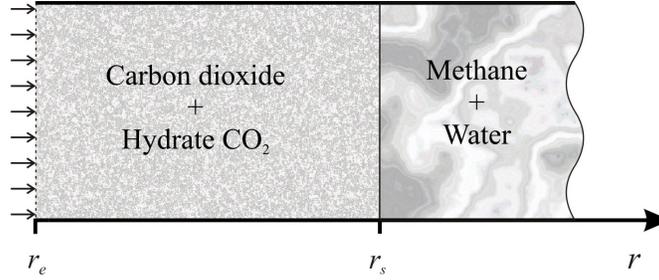


Fig. 2. The scheme of the problem of the carbon dioxide injection into the natural reservoir initially saturated with water and methane

The system of basic equations describing the processes of filtration and heat transfer in a porous medium include the balance of mass, momentum (the Darcy law) and energy equations, and the equations of state for the gases. This system in the radially symmetric case taking into account the assumptions has the form [12, 17, 18]

$$\begin{aligned}
 \frac{\partial}{\partial t} (m\rho_{(i)}S_{(i)}) + \frac{1}{r} \frac{\partial}{\partial r} (rm\rho_{(i)}S_{(i)}v_{(i)}) &= 0, (i = dc, mt) \\
 mS_{(i)}v_{(i)} &= -\frac{k_{(i)}}{\mu_{(i)}} \frac{\partial p}{\partial r}, \\
 \frac{\partial}{\partial t} (\rho c T) &= \frac{1}{r} \frac{\partial}{\partial r} \left(r\lambda \frac{\partial T}{\partial r} \right) - m\rho_{(i)}c_{(i)}S_{(i)}v_{(i)} \frac{\partial T}{\partial r}, \\
 p &= \rho_{(i)}R_{g(i)}T,
 \end{aligned} \tag{2}$$

$\rho_{(i)}$, $c_{(i)}$, $k_{(i)}$ and $\mu_{(i)}$ ($i = dc, mt$) are the density, specific heat, phase permeability and dynamic viscosity of a gas; $v_{(i)}$ is the real mean gas velocity; $R_{g(i)}$ ($i = dc, mt$) is the specific gas constant; $S_{(i)}$ is the saturation of the pores by a gas; ρc and λ are the specific volume heat capacity and the coefficient of thermal conductivity of the system. Since the main contribution to the values of ρc and λ is made by the parameters of the porous medium skeleton, we shall consider them to be constant quantities.

Taking into account the Darcy law and the equation of state for carbon dioxide, conditions on the left boundary of the reservoir can be rewritten as: $t > 0, r = r_e \rightarrow 0$:

$$r \frac{\partial p^2}{\partial r} = -\frac{Q_e \mu_{(dc)} R_{g(dc)} T_e}{\pi k_{(dc)}}, T = T_e.$$

In this work we consider the case when the initial water saturation does not exceed 0.2, therefore the water can be assumed immobile. It is also possible to neglect the compressibility of water, i.e. $\rho_w = \text{const}$.

To determine the gas permeability coefficient from the gas saturation, we use the Kozeny equation [12, 17]: $k_{(i)} = k_0 S_{(i)}^3$, ($i = dc, mt$) where k_0 is the absolute reservoir permeability.

At the boundary $r = r_s$ the following relations, obtained from the mass balance and heat balance conditions, are satisfied

$$\begin{aligned}
-\frac{k_{(dc)}}{\mu_{(dc)}} \frac{\partial p_1}{\partial r} &= m \left(\frac{\rho_h}{\rho_{(dc)}} S_h G_{(dc)} + S_{(dc)} \right) \dot{r}_s, \\
-\frac{k_{(mt)}}{\mu_{(mt)}} \frac{\partial p_2}{\partial r} &= m S_{(mt)} \dot{r}_s, \\
m \rho_h S_h (1 - G_{(dc)}) \dot{r}_s &= m \rho_w S_{w0} \dot{r}_s, \\
\lambda \left(\frac{\partial T_1}{\partial r} - \frac{\partial T_2}{\partial r} \right) &= m \rho_h S_h L_h \dot{r}_s,
\end{aligned} \tag{3}$$

subscripts 1 and 2 refer to parameters in the near and far zones; the subscript s refers to the parameters at the boundary between these zones; ρ_h and L_h are the density of the CO₂ hydrate and its specific heat of formation; \dot{r}_s is the velocity of the hydrate formation boundary. The temperature and pressure at this boundary will be assumed to be continuous.

From the third equation of the system (3) for the hydrate saturation value in the first zone can be written following: $S_h = \frac{\rho_w S_{w0}}{\rho_h (1 - G_{(dc)})}$.

On the basis of the system (2) it is possible to obtain the heat and pressure conductivity equations

$$\begin{aligned}
\frac{\partial p_i^2}{\partial t} &= \chi_i^{(p)} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial p_i^2}{\partial r} \right), \quad (i = 1, 2) \\
\frac{1}{\chi^{(T)}} \frac{\partial T_i}{\partial t} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_i}{\partial r} \right) + \eta_i \frac{\partial p_i^2}{\partial r} \frac{\partial T_i}{\partial r}, \\
\chi_1^{(p)} &= \frac{k_{(dc)} p_1}{m \mu_{(dc)} (1 - S_h)}, \quad \chi_2^{(p)} = \frac{k_{(mt)} p_2}{m \mu_{(mt)} (1 - S_{w0})}, \quad \chi^{(T)} = \frac{\lambda}{\rho c}, \\
\eta_1 &= \frac{\rho_{(dc)} c_{(dc)} k_{(dc)}}{2 \mu_{(dc)} \lambda p_1}, \quad \eta_2 = \frac{\rho_{(mt)} c_{(mt)} k_{(mt)}}{2 \mu_{(mt)} \lambda p_2}.
\end{aligned} \tag{4}$$

3. SELF-SIMILAR SOLUTION

To obtain the self-similar solutions of the considered problem we introduce following variable: $\xi = r / \sqrt{\chi^{(T)} t}$.

For this variable the equations of the system (4) take the form:

$$\begin{aligned}
-\frac{\xi}{2} \frac{d p_i^2}{d \xi} &= \frac{\chi_i^{(p)}}{\chi^{(T)}} \frac{1}{\xi} \frac{d}{d \xi} \left(\xi \frac{d p_i^2}{d \xi} \right), \quad (i = 1, 2) \\
-\frac{\xi}{2} \frac{d T_i}{d \xi} &= \frac{1}{\xi} \frac{d}{d \xi} \left(\xi \frac{d T_i}{d \xi} \right) + \eta_i \frac{d p_i^2}{d \xi} \frac{d T_i}{d \xi}.
\end{aligned} \tag{5}$$

Here, the piezoconductivity equation is a nonlinear equation since the piezoconductivity coefficient $\chi_i^{(p)}$ ($i = 1, 2$) include unknown function p_i . Considering the case of small pressure differences in the reservoir the Leibenzon's linearization can be applied [19]. And the variable pressure p_i in the parameter $\chi_i^{(p)}$ can be assumed constant and equal to the initial reservoir pressure p_0 .

Integrating Eq. (5), let us write the following relations for the distributions of pressure and temperature in each of the reservoir zones

$$p_1^2 = p_s^2 + \frac{Qe\mu_{(dc)}R_{g(dc)}T_s}{k_{(dc)}\pi} \int_{\xi}^{\xi_s} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4\theta_1}\right) d\xi, \quad (0 < \xi < \xi_s) \tag{6}$$

$$T_1 = T_s + (T_e - T_s) \int_{\xi}^{\xi_s} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_1 p_1^2\right) d\xi \bigg/ \int_0^{\xi_s} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_1 p_1^2\right) d\xi,$$

$$p_2^2 = p_0^2 + (p_s^2 - p_0^2) \frac{\int_{\xi}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4\theta_2}\right) d\xi}{\int_{\xi_s}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4\theta_2}\right) d\xi}, \quad (\xi_s < \xi < \infty) \tag{7}$$

$$T_2 = T_0 + (T_s - T_0) \int_{\xi}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_2 p_2^2\right) d\xi \bigg/ \int_{\xi_s}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_2 p_2^2\right) d\xi,$$

where $\theta_i = \chi_i^{(p)} / \chi^{(T)}$ ($i = 1, 2$).

Based on the relations (3), taking into account the solutions for pressure and temperature (6) and (7), we obtain equations for finding the coordinate of the hydrate formation boundary $\xi = \xi_s$, and the values of pressure p_s and temperature T_s

$$\frac{Qe\mu_{(dc)}R_{g(dc)}T_e}{k_{(dc)}\pi} \exp\left(-\frac{\xi_s^2}{4\theta_1}\right) = F \cdot p_s \xi_s^2, \tag{8}$$

$$(p_s^2 - p_0^2) \exp\left(-\frac{\xi_s^2}{4\theta_2}\right) = E \cdot p_s \xi_s^2 \int_{\xi_s}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4\theta_2}\right) d\xi, \tag{9}$$

$$\frac{(T_s - T_e) \exp\left(-\frac{\xi_s^2}{4} - \eta_1 p_s^2\right)}{\int_0^{\xi_s} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_1 p_s^2\right) d\xi} - \frac{(T_0 - T_s) \exp\left(-\frac{\xi_s^2}{4} - \eta_2 p_s^2\right)}{\int_{\xi_s}^{\infty} \frac{1}{\xi} \exp\left(-\frac{\xi^2}{4} - \eta_2 p_s^2\right) d\xi} = B \xi_s^2, \tag{10}$$

$$F = \frac{m\mu_{(dc)}\chi^{(T)}}{k_{(dc)}} \left(1 + \frac{\rho_h G_{(dc)} S_h}{\rho_0(d_c)} - S_h\right), \quad E = \frac{m\mu_{(mt)}\chi^{(T)}(1 - S_{w0})}{k_{(mt)}}, \quad B = \frac{m\rho_h L_h S_h}{2\rho c}.$$

This system of equations was solved as follows. We express p_s from Eq. (8) as a function of the self-similar coordinate of the hydrate formation boundary ξ_s . Then we put

p_s in Eq. (9) and obtain a transcendental equation with one unknown ζ_s . This equation was solved by the bisection method. Then from Eqs. (8) and (10) we determine the values of the parameters p_s and T_s . And then, taking into account the values found at the boundary of the phase transitions ($\zeta = \zeta_s$), the distributions of the parameters in the first ($0 < \zeta < \zeta_s$) and second ($\zeta_s < \zeta < \infty$) zones of the reservoir can be found from (6) and (7).

4. CALCULATION RESULTS

The solutions, obtained in the work, were investigated for compliance with the condition of thermodynamic consistency. It means that the local pressure in the first zone should be higher than the equilibrium pressure of the carbon dioxide hydrate formation. This pressure is determined from Eq. (1) using the temperature distribution in this zone.

Fig. 3 shows the distributions of temperature and pressure in the reservoir at two different values of the specific mass flow rate of the carbon dioxide injection. For other parameters the following values are used: $m = 0.2$; $k_0 = 10^{-14} m^2$; $S_{w0} = 0.1$; $G = 0.28$; $p_0 = 2.9$ MPa; $T_0 = 276$ K; $T_e = 278$ K; $\lambda = 2$ W/(m·K); $\rho c = 2.5 \cdot 10^6$ J/(K·m³); $\rho_h = 1100$ kg/m³; $c_{(dc)} = 890$ J/(K·kg); $c_{(mt)} = 1560$ J/(K·kg); $R_{g(dc)} = 189$ J/(K·kg); $R_{g(mt)} = 520$

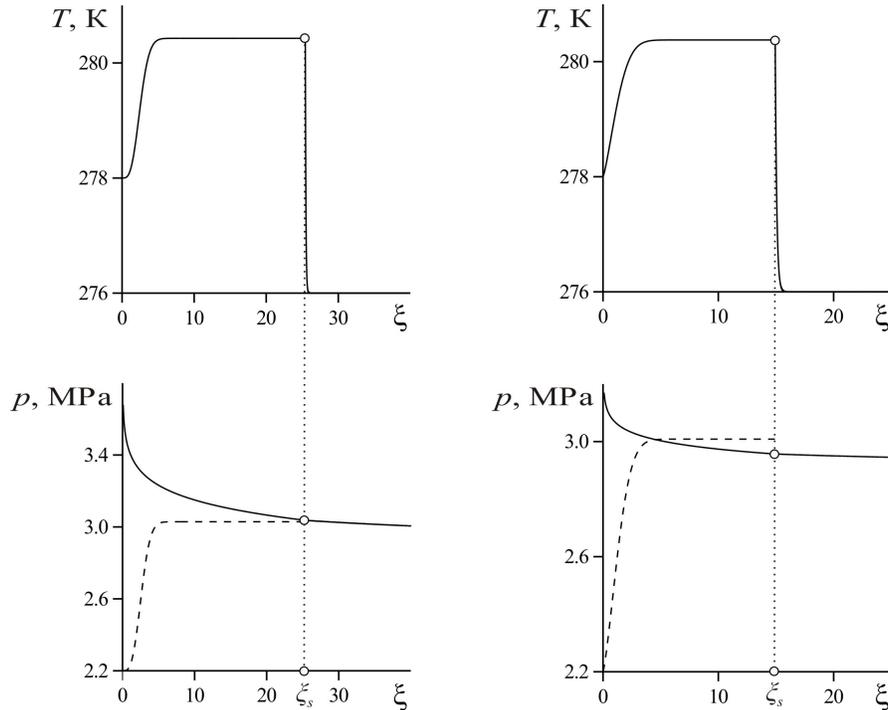


Fig. 3. Change in the self-similar coordinate ζ of the reservoir pressure and temperature at $Q_e = 0.03$ kg/(m·s) (left) and $Q_e = 0.01$ kg/(m·s) (right). The dashed line is the equilibrium pressure of the carbon dioxide hydrate formation

J/(K·kg); $\mu_{(dc)} = 1.3 \cdot 10^{-5}$ Pa·s; $\mu_{(mt)} = 10^{-5}$ Pa·s; $L_h = 4 \cdot 10^5$ J/kg; $T_* = 7.6$ K, $p_{s0} = 1.68$ MPa [8, 11, 12, 17].

Fig. 3 shows that at carbon dioxide injection rate $Q_e = 0.03$ kg/(m·s) the pressure in the first zone is higher than the equilibrium pressure of the CO₂ hydrate formation. So, in this case the solution with the frontal surface of the phase transitions adequately describes the process. With a lower carbon dioxide injection rate ($Q_e = 0.01$ kg/(m·s)) the pressure in the reservoir at a portion of the first zone falls below the equilibrium pressure and consequently it is necessary to consider the extended zone of phase transitions.

The condition of the realization of a regime with an extended zone of the carbon dioxide hydrate formation is determined by the inequality: $T_0 + T_* \ln(p_0/p_{s0}) < T_s$, the temperature at the boundary of the phase transitions T_s is determined from Eq. (10).

On the basis this inequality calculations were carried out to determine the value of the carbon dioxide injection flow rate Q_{cr} (we call it the limiting flow rate), below which the regime with an extended region of the CO₂ hydrate formation is realized.

Fig. 4 shows the dependence of Q_{cr} on the absolute reservoir permeability. It can be seen that as the permeability increases, the limiting value of the flow rate increases, and the faster, the higher the initial water saturation of the reservoir. This means, that the regime with an extended region of the carbon dioxide hydrate formation is realized in high permeability reservoirs, as well as in the cases of high initial water saturation. This is due to the fact that, on the one hand, an increase in permeability at a constant mass flow rate requires a reduction in the well pressure, which leads to a decrease in the reservoir pressure. On the other hand, an increase in the initial water saturation leads to an increase in heat release at the phase transition boundary and, accordingly, to an increase in the reservoir temperature and the equilibrium pressure of hydrate formation. Thus, the realization of a regime with the extended zone of hydrate formation in cases of high values of the reservoir permeability and its initial water saturation is due to the fact that the pressure at the phase transitions boundary is below the equilibrium pressure of the carbon dioxide hydrate formation.

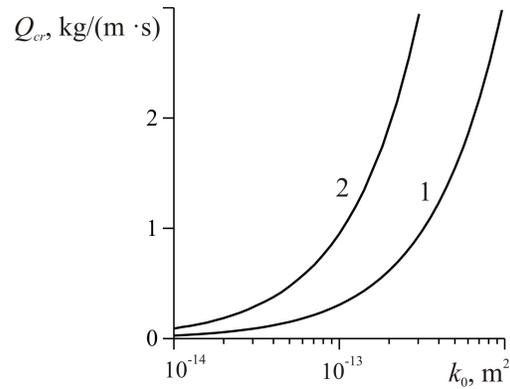


Fig. 4. Dependence of the limiting flow rate Q_{cr} on the absolute reservoir permeability k_0 for different values of the initial water saturation S_{w0} . Lines 1 and 2 correspond to $S_{w0} = 0.1$ and 0.11

5. CONCLUSIONS

On the basis of the continuous medium mechanics methods, the mathematical model of the process of the carbon dioxide injection into the porous reservoir, initially saturated with water and methane, is constructed. It has been established that the carbon dioxide hydrate formation can occur either on a frontal surface or in an extended zone. It is shown that the regime with the carbon dioxide hydrate formation in an extended zone

is realized at low values of mass flow rate of carbon dioxide injection, as well as high reservoir permeability and high initial water saturation.

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