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Estimation of the parameters of carbon dioxide injection into a saturated porous reservoir with heterogeneous permeability in the presence of hydrate formation

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One of the effective methods of combating the increase in the concentration of carbon dioxide in the atmosphere is its sequestration in porous media in the solid gas hydrate form. The unique properties of gas hydrates, such as their high gas capacity, low permeability and natural stability, make them an attractive option for long-term disposal of carbon dioxide. In the context of considering the problem of organizing geological gas hydrate storages of carbon dioxide, a mathematical model is written down that allows a theoretical study of the process of carbon dioxide hydrate formation during its injection into a reservoir whose pores are initially saturated with methane and water. The proposed mathematical model takes into account zonal heterogeneity of a porous reservoir, the flow in it in the presence of phase transformations (hydrate formation and solubility of carbon dioxide in water) of the gas (CH_4 and/or CO_2) and liquid (water and dissolved CO_2) phases, heat transfer from the considered region of a reservoir to the surrounding rocks; the hydrate formation process is considered as an equilibrium phase transition. Calculation equations for the studied process are presented and numerical solutions of the problem are constructed, describing the distribution of parameters (temperature, pressure, phase saturations) in a reservoir. It has been shown by calculations that when CO_2 is injected into a reservoir, several characteristic zones can form in it, differing in the composition of the fluids saturating them. It has been demonstrated that it is necessary to take into account such factors as heat released during phase transitions, the Joule-Thomson effect, and heat exchange between the porous reservoir and its surrounding rocks when describing the temperature field in the reservoir formed when carbon dioxide is injected into it. The results of computational experiments are presented and analyzed when placing an injection well in a high- or low-permeability zone of a porous reservoir. The conducted numerical study showed that for the organization of effective gas hydrate storage of carbon dioxide, porous media with sufficiently high permeability values are required.

Keywords: filtration, carbon dioxide, gas hydrate formation, zonal permeability, mathematical model

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Introduction

With the growth of global industrialization and the overexploitation of non-renewable energy sources, a large amount of greenhouse gases, primarily carbon dioxide, has been released (Xu et al., 2020). The works of a number of researchers have shown that currently one of the best options for large-scale reduction of these emissions is geological storage of CO_2 (Kim, Santamarina, 2014; Lu et al., 2021). According to the definition given in the article (Korzun et al., 2023), a natural geological object is a geological system that can keep a greenhouse gas in stable state for a sufficiently

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long period of time, without the risk of emergency situations associated with the release of utilized emissions from collectors to the surface. Measures for the utilization and storage of carbon dioxide in porous collectors include the capture of CO_2 emitted by industrial or energy manufacturers and its subsequent underground storage (Cao et al., 2022). Long-term storage of carbon dioxide in a gaseous state can lead to gas breakthrough to the surface and, accordingly, to unpredictable environmental problems (Mac Dowell et al., 2017). Therefore, the most acceptable option is geological storage of carbon dioxide in solid gas hydrate form (Kim, Santamarina, 2014; Hassanpouryouzband et al., 2019; Zhou et al., 2024). The efficiency of such CO_2 storage can be due to the fact that, under the same thermodynamic conditions, a unit volume of gas hydrate can contain several times more gas than in gaseous state (Istomin, Yakushev, 1992; Chuvilin, Guryeva, 2009; Makogon, 2010). Underground gas hydrate storage of carbon dioxide meets the main characteristics of a possible option for organizing such a storage facility, namely: large storage capacity, long-term CO_2 isolation (at least several hundred years), reasonable cost, minimal impact on the environment (Aminu et al., 2017). As indirect evidence of the above theses, one can cite the fact that, for example, in the northern Russian regions there are natural accumulations of methane hydrates, the estimated age of which reaches hundreds of thousands of years. It is important to note that the range of pressures and temperatures at which CO_2 hydrate can stably exist is much wider than that for CH_4 hydrate, which indicates the possibility of similar long-term storage of carbon dioxide in gas hydrate state.

Currently, large corporations in a number of countries are interested in geological storage projects for carbon dioxide. When organizing a CO_2 storage facility, an important issue is the selection of reservoirs suitable for underground gas hydrate storage of carbon dioxide (Lu et al., 2021). Such reservoirs can be depleted hydrocarbon deposits (Korzun et al., 2023; Zhou et al., 2024) located in relative proximity to manufacturers that are sources of CO2 emissions, such as electricity and heat producers.

Carbon dioxide storage in depleted oil and gas reservoirs is considered as one of the most effective storage options due to several reasons: 1) these reservoirs have been thoroughly studied before and during hydrocarbon production; 2) surface and underground infrastructure such as injection wells and pipelines are available and can be used for the storage process either directly or with minor modifications; 3) carbon dioxide injection into oil-saturated reservoir as an enhanced oil recovery method has already been applied in the oil and gas industry and hence such experience can be used for CO_2 storage process (Aminu et al., 2017; Ibragimov et al., 2024).

Rational formation of underground carbon dioxide gas hydrate storage dictates the need for an in-depth preliminary study of the aspects of non-isothermal gas-liquid flow in saturated porous reservoirs in the presence of phase transitions, as well as an analysis of the factors determining the efficiency of such CO_2 storage technology. For example, one of such factors is the heterogeneity in the reservoir permeability (Barenblatt et al., 2016; Hu et al., 2021). In this regard, mathematical modeling and numerical study of the processes occurring during non-isothermal flow of carbon dioxide in porous media with heterogeneous permeability, taking into account the formation of gas hydrates, seem important.

Methods

In this paper, mathematical modeling methods are used to study the features of non-isothermal filtration of a gas-liquid mixture, taking into account the gas hydrate formation when carbon dioxide is injected into a porous reservoir, the pores of which are initially filled with methane and water. First, let us consider the problem formulation.

Problem of the process of CO_2 injection into a porous layer of height H and radius R will be considered in a two-dimensional (radial axis r and vertical axis z) axisymmetric approximation (Figure 1).

Let gas be injected through a well of radius r_w, which penetrates the reservoir to its full height. The outer boundaries of the reservoir are impermeable to the gas and liquid, but heat exchange with the surrounding rocks can occur due to thermal conductivity. Let the porous medium initially consist of three phases, namely: a solid skeleton that does not participate in physical and chemical transformations, methane, and water with known saturations. We assume that the porous medium skeleton is immobile and incompressible. At the initial time moment the temperature in the reservoir and surrounding rocks is equal to T_0. Filtration is possible only in the porous reservoir, and the initial pressure in it

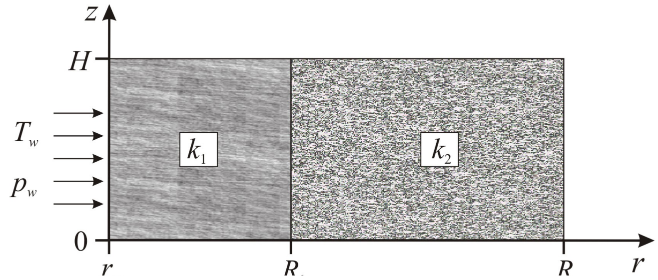

Figure 1. Schematic representation of the problem. z and r are the vertical and radial coordinates; H, r_w, R_b and R are the reservoir height, well radius, radius of the first reservoir zone and radius of the whole reservoir, respectively; k_1 and k_2 are the permeability of the first and second zones; T_w and p_w are the temperature and pressure of the injected carbon dioxide

is p_0. The reservoir consists of two zones with different permeability values k_1 and k_2. The injection of carbon dioxide occurs at a constant pressure p_w and temperature T_w, which is less than the equilibrium temperature of CO_2 hydrate formation for pressure p_w (Figure 1).

In mathematical modeling of two-phase filtration taking into account the formation of CO_2 hydrate, we will make the following main assumptions: gas hydrate is considered as a system consisting of water and gas, and the mass concentration of gas in the gas hydrate G is constant; methane does not dissolve in water; water evaporation is neglected; a single-temperature model is considered (the temperatures of all phases in a certain small volume are equal); the reservoir porosities for both zones are the same and constant; the density of water and the specific heats of phase transitions are constant values; capillary effects are not taken into account (Basniev et al., 1993; Bondarev et al., 2018). When studying the process of CO_2 injection into a reservoir, in contrast to the works (Tsypkin, 2014; Khasanov et al., 2019; Khasanov et al., 2020), the solubility of CO_2 in water, the filtration of water and gas, and the heat exchange of the porous reservoir with surrounding rocks are taken into account.

The equations for studied processes can be written on the basis of the mass conservation equations, Darcy's law, and the energy conservation equation (Nigmatulin, 1987; Basniev et al., 1993; Borodin, Belskikh, 2020; Musakaev, Borodin, 2023). In this case, the piezoconductivity equation and the energy equation, on the basis of which the change in pressure and temperature is calculated, respectively, unlike the work (Musakaev, Borodin, 2023), contain terms that take into account the intensity of carbon dioxide solubility in water J_{gl}. The ratio for finding this value can be written as follows:

$$J_{gl} = \varphi \rho_{wat} \frac{\partial}{\partial t}\left(S_l \frac{\omega_{l(d)}}{\omega_{l(wat)}} \right), \quad \sum_{j=d,wat} \omega_{l(j)} = 1.$$

Here and after, the indices $j = sk, g, l, h, d, m$ and wat refer to the parameters of the porous medium skeleton, gas phase, liquid phase, gas hydrate, carbon dioxide, methane and water, respectively; the subscripts in brackets denote the corresponding phase components. J_{gl} is the intensity of CO_2 solubility in water, kg/(m^3·s); S_j ($j = g, l, h$) is the saturation of pores with the j-th phase; φ is the reservoir porosity; ρ_{wat} is the density of water, kg/m^3; $\omega_{l(d)}$ and $\omega_{l(wat)}$ are the mass concentrations of carbon dioxide and water in the liquid phase, respectively.

The following equations of state are used for the liquid and gas phases (Shagapov et al., 2012; Borodin, Belskikh, 2020):

$$\rho_l = \rho_{wat} / \omega_{l(wat)}, \quad p = X_g \rho_g R_g T,$$

$$R_g = R_{un} / M_g, \quad M_g = \left(\frac{\omega_{g(d)}}{M_d} + \frac{\omega_{g(m)}}{M_m} \right)^{-1}, \quad \sum_{j=d,m} \omega_{g(j)} = 1,$$

$$X_g = \left(0.4 \cdot \lg\left(\frac{T}{T_{cr}} \right) + 0.73 \right)^{p/p_{cr}} + \frac{p}{10 p_{cr}},$$

$$p_{cr} = \sum_{j=d,m} \chi_{g(j)} p_{cr(j)}, \quad T_{cr} = \sum_{j=d,m} \chi_{g(j)} T_{cr(j)},$$

$$\chi_{g(j)} = \frac{\omega_{g(j)}}{M_j} M_g, \quad (j = d, m),$$

where p is the pressure, Pa; T is the temperature, K; ρ_j ($j = sk, g, l, h$) is the density of the j-th phase, kg/m^3; R_g is the specific gas "constant" (this parameter may vary depending on the gas phase composition), J/(kg·K); R_{un} is the universal gas constant, J/(mol·K); M_d and M_m are the molar masses of carbon dioxide and methane, kg/mol; $\omega_{g(d)}$ and $\omega_{g(m)}$ are the mass concentrations of CO_2 and CH_4 in the gas phase; T_{cr} and p_{cr} are the critical values of temperature (K) and pressure (Pa) for the gas phase; $T_{cr(d)}$ and $T_{cr(m)}$ are the critical values of temperature for carbon dioxide and methane, K; $p_{cr(d)}$ and $p_{cr(m)}$ are the critical values of pressure for CO_2 and CH_4, Pa; $\chi_{g(d)}$ and $\chi_{g(m)}$ are the mole concentrations of carbon dioxide and methane in the gas phase.

The reservoir permeability can be found from the relation (Zhang et al., 2022):

$$\mathbf{k} = \mathbf{k}_0 (1 - S_h)^n,$$

where $\mathbf{k_0}$ is the porous medium permeability tensor in the absence of gas hydrate, m^2; n is the exponent depending on the type of the pore space filling with gas hydrate. In the calculations, $n = 3$ is assumed.

The work assumes that the formation of CO_2 hydrate occurs in equilibrium mode (Borodin et al., 2022). To find the mass concentration of carbon dioxide in water, depending on its partial pressure in the gas phase, and common temperature, empirical relations from the work (Voronov et al., 2011) are used.

Results

The next step in studying the process of carbon dioxide storage in geological reservoirs in solid gas hydrate form is to conduct computational experiments to study the features of CO_2 hydrate formation when injecting carbon dioxide into a reservoir initially saturated with a water-methane mixture. These calculations were performed using the author's computer code that numerically implements the mathematical model. The methodology of calculations was presented by the authors at the First Russian Gas Hydrate Conference – 2024 (Borodin, Musakaev, 2024) and it is similar to the methodology described in (Musakaev et al., 2020). The parameters values used in the calculations (unless otherwise specified) are given in Table 1 (Istomin, Yakushev, 1992; Borodin, Belskikh, 2020; Misyura et al., 2023).

Figure 2 shows the distributions of pressure, temperature, CO_2 hydrate saturation, mass concentration of CO_2 in the gas phase and mass concentration of dissolved CO_2 in the liquid phase for three cases:

Parameter, unit of measurement	Symbol	Value
Calculation time, days	t	7
Radius of the whole reservoir, m	R	100
Radius of the first zone (in which the injection well is located), m	R_b	50
Reservoir height, m	H	10
Well radius, m	r_w	0.1
Initial reservoir pressure, MPa	p_0	1
Initial reservoir temperature, °C	T_0	5
Carbon dioxide injection pressure, MPa	p_w	3
Temperature of injected carbon dioxide, °C	T_w	5
Reservoir porosity	φ	0.2
Initial gas saturation (only methane)	S_{g0}	0.8
Initial saturation with liquid (only water, without dissolved CO_2)	S_{l0}	0.2
Latent heat of formation/decomposition of CO_2 hydrate, kJ/kg	L_{hd}	390
Latent heat of dissolution of CO_2 in water / evaporation of CO_2 from water, kJ/kg	L_{gl}	480
Mass concentration of CO_2 in gas hydrate	G	0.28

Table 1. Main calculations parameters

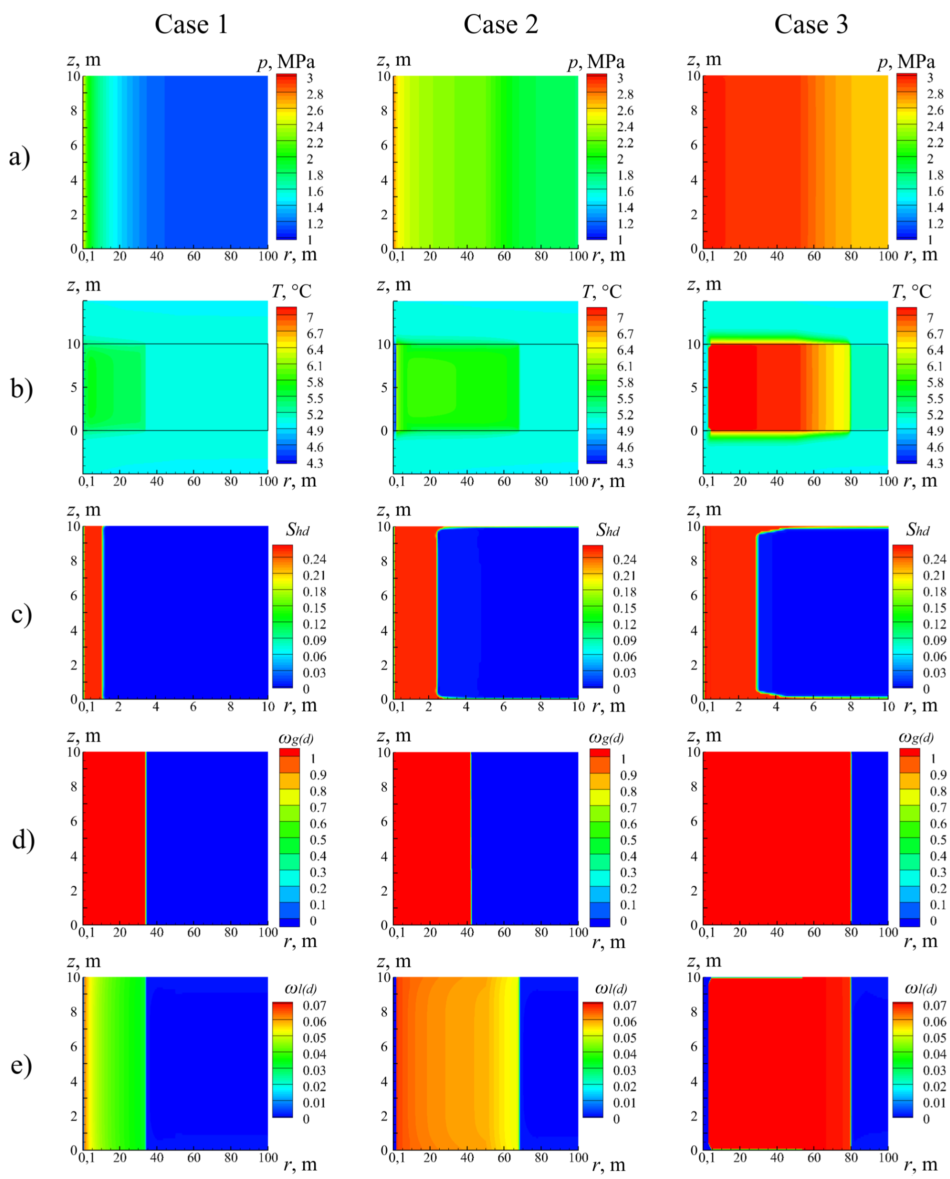

Figure 2. Distributions in the reservoir of pressure p (a), temperature T (b), hydrate saturation S_{hd} (c), mass concentration of CO_2 in the gas phase $\omega_{g(d)}$ (d) and mass concentration of dissolved CO_2 in the liquid phase $\omega_{l(d)}$ (e). The parameters distributions are plotted on the 7th day after the start of carbon dioxide injection into the reservoir

Case 1. The injection well through which carbon dioxide is injected into a reservoir is located in a low-permeability zone, with the permeability value $k_1 = 10^{-14}$ m^2, the permeability value for the second high-permeability zone $k_2 = 10^{-13}$ m^2 (Figure 1);

Case 2. The injection well is located in a high-permeability zone, and $k_1 = 10^{-13}$ m^2, $k_2 = 10^{-14}$ m^2.

Case 3. The injection well is located in a high-permeability zone, and $k_1 = 10^{-12}$ m^2, $k_2 = 10^{-14}$ m^2.

Figure 3 shows the distributions of parameters along the radial coordinate r for the average reservoir height (at $z = H/2$).

Figure 4 shows the change over time in the mass of CO_2 in the whole reservoir.

Discussion of the results

From the data presented in Figures 2 and 3, it is evident that carbon dioxide, injected into a porous medium, displaces methane deep into the reservoir. In the reservoir, the pores of which are saturated by methane and water before CO_2 injection, carbon dioxide is present in three states: as a part of gas hydrate, dissolved in water, and in gaseous state. Hydrate formation occurs mainly in the zone adjacent to the injection well; in this zone, carbon dioxide and its hydrate are present in the pores. Further (along the r coordinate), the pore space is filled with CO_2, CO_2 hydrate, and water; then with carbon dioxide and water; then with gas phase ($CO_2 + CH_4$) and water; and finally, with a water-methane mixture. In the

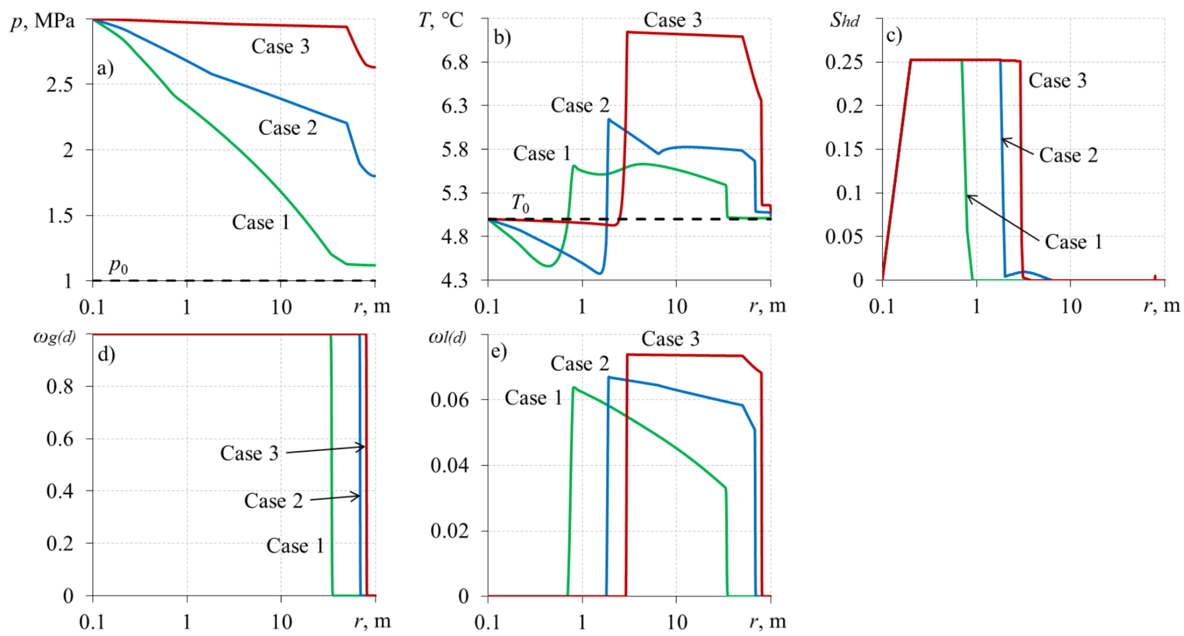

Figure 3. Distributions by the radial coordinate r at the average reservoir height z = H/2 of pressure p (a), temperature T (b), hydrate saturation S_{hd} (c), mass concentration of CO_2 in the gas phase $\omega_{g(d)}$ (d) and mass concentration of dissolved CO_2 in the liquid phase $\omega_{l(d)}$ (e) on the 7th day after the start of carbon dioxide injection into the reservoir.

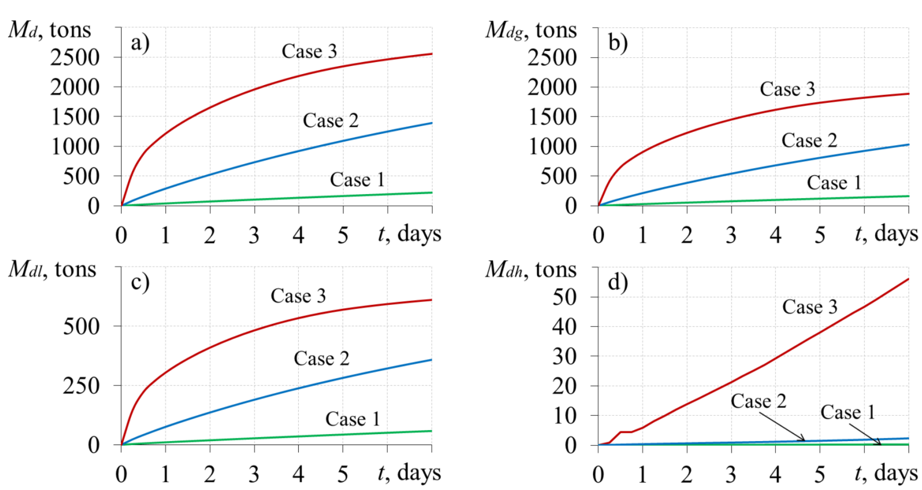

Figure 4. Change over time in the mass of carbon dioxide in the whole reservoir: a) total CO_2 mass; b) CO_2 in gaseous state; c) CO_2 dissolved in water; d) CO_2 as a part of gas hydrate.

first two noted zones, the reservoir properties deteriorate due to gas hydrate deposits, which lead to a decrease in the filtration rate from the well into the reservoir and a large pressure gradient in the zone with the presence of gas hydrate. For the case of the injection well located in a highly permeable zone (Cases 2 and 3), the above-described factor has a lesser effect, and the pressure in the entire reservoir increases significantly (even in the low-permeability zone remote from the well), which indicates better penetration of carbon dioxide into the reservoir. It is also evident from Figures 2 and 3 that the temperature in the reservoir is non-monotonous. This is due to the influence of the following factors: the Joule-Thomson effect (in the area of high pressure gradients, the temperature during filtration of real gas decreases significantly); heat release during the formation of CO_2 hydrate; heat release during the dissolution of carbon dioxide in water. It is evident that near the well the temperature decreases below the initial one, since the greatest pressure difference is observed in this area, which leads to the most intensive cooling due to the Joule-Thomson effect. Then the temperature increases above the initial one due to phase transitions. In the entire reservoir, where carbon dioxide and water are simultaneously present, the temperature increases above the initial one due to the release of heat during the dissolution of this gas in water. Moreover, near the upper and lower boundaries of the reservoir the temperatures are lower, and the amount of CO_2 dissolved in water is higher, which is explained by heat transfer to the impermeable surrounding rocks due to thermal conductivity. In the zone adjacent to the injection well, all the water passes into the gas hydrate, which is associated, first of all, with a significant decrease in temperature in this part of the reservoir due to the Joule-Thomson effect. Thus, for an adequate description of the temperature field in a reservoir, it is necessary to take into account such factors as the heat released during phase transitions, the Joule-Thomson effect, and the heat exchange of the porous reservoir with the surrounding impermeable rocks. Figures 2 and 3 also show that the displacement of methane by carbon dioxide occurs practically in a piston mode. The region length in which CO_2 and CH_4 are simultaneously in the gas phase are small compared to the reservoir length; the piston nature of the displacement is due to the fact that the viscosity of CO_2 is one and a half times higher than the viscosity of CH_4 (Tsypkin, 2014).

From the data presented in Figure 4, it is evident that the higher the permeability of the well bottomhole zone, the higher the mass of buried carbon dioxide, therefore, when organizing this method of CO_2 utilization, it is necessary to inject this gas into highly permeable zones, i.e., for the burial of carbon dioxide with greater intensity, it is desirable to locate the injection well in the reservoir area with the highest permeability. Analysis of Figures 2 and 3 allows us to conclude that for the parameters adopted in the work, for the organization of effective gas hydrate storage of carbon dioxide, porous media with sufficiently high permeability values are necessary. Otherwise, firstly, due to low permeability, the time for injecting the required volume of CO_2 will increase significantly and, secondly, the resulting gas hydrate will further reduce the porous medium permeability, which can lead to an almost complete stop of further CO_2 filtration.

For the considered set of parameters, when placing the injection well in a low-permeability zone ($k_1 = 10^{-14}$ m^2), the carbon dioxide in gas hydrate at the final point in time (7 days) is about 300 kg. If the injection well is located in a highly permeable zone ($k_1 = 10^{-13}$ m^2), this indicator is about 2,300 kg. With a further increase in permeability (Case 3), the accumulated mass of carbon dioxide in gas hydrate is already about 56,000 kg. However, as can be seen from Figure 4, for all the considered cases, the mass of CO_2 in the gas hydrate is less than its mass in free and dissolved state. This is due to the fact that the process of gas hydrate formation in a reservoir is significantly affected (in addition to the reservoir properties) by the initial thermodynamic parameters of the underground CO_2 storage facility. That is, additional studies are needed to determine reservoirs with the most favorable initial thermodynamic conditions for hydrate formation, as well as to identify the most effective parameters for carbon dioxide injection. And a comprehensive consideration of all these factors will ensure the most effective underground gas hydrate disposal of CO_2.

Conclusion

The article is devoted to studying the features of the process of carbon dioxide storage in geological reservoirs in the solid gas hydrate form. A mathematical model of non-isothermal filtration of a gas-liquid mixture is proposed in a two-dimensional axisymmetric approximation, in which, unlike previous works, additional consideration is given to such factors as the presence of zones with different permeability, two-phase filtration, CO_2 solubility in water, heat transfer (due to conductive heat exchange) from the considered area of the porous medium to the surrounding rocks, non-isothermal effects at filtration. It is shown by calculations that when carbon dioxide is injected into a porous reservoir, the temperature distribution along the reservoir length has a non-monotonic character, caused by the Joule-Thomson effect, heat release during the formation of CO_2 hydrate and the dissolution of this gas in water. Near the upper and lower reservoir boundaries, the temperatures are lower, and the amount of dissolved CO_2 in water is higher, which is explained by heat transfer to the impermeable surrounding rocks due to thermal conductivity. It is shown that for more efficient organization of gas hydrate storage of carbon dioxide in a zonally heterogeneous porous reservoir, it is necessary to locate the injection well in a zone with high permeability values.

Acknowledgements

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Оценка параметров закачки диоксида углерода в насыщенный пористый пласт с неоднородной проницаемостью при наличии гидратообразования

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Одним из эффективных методов борьбы с ростом концентрации диоксида углерода в атмосфере является его секвестрация в пористых средах в твердой газогидратной форме. Уникальные свойства газовых гидратов, такие как их высокая газоемкость, низкая проницаемость и естественная стабильность, делают их привлекательным вариантом для долговременного захоронения CO_2. В статье в рамках рассмотрения проблематики организации геологического газогидратного хранилища диоксида углерода записана математическая модель, позволяющая осуществить теоретическое изучение процесса образования гидрата CO_2 при его закачке в пласт, поры которого в исходном состоянии заняты метаном и водой. В предложенной математической модели осуществлен учет зональной неоднородности пористого коллектора, течения в нем при наличии фазовых превращений (гидратообразование и растворимость диоксида углерода в воде) газовой (CH_4 и/или CO_2) и жидкой (вода и растворенный CO_2) фаз, переноса тепла из рассматриваемой области пласта в окружающие горные породы; процесс гидратообразования рассматривается как равновесный фазовый переход. Представлены расчетные уравнения для изучаемого в работе процесса и построены численные решения задачи, описывающие распределения параметров (температуры, давления, насыщенностей фаз) в пласте. Расчетным путем показано, что при закачке CO_2 в пласт в нем возможно формирование нескольких характерных зон, отличающихся по составу насыщающих их флюидов. Продемонстрирована необходимость учета при описании температурного поля в пласте таких факторов, как теплота, выделяемая при фазовых переходах, эффект Джоуля-Томсона, теплообмен пористого коллектора с окружающими горными породами. Приведены и проанализированы результаты вычислительных экспериментов при размещении нагнетательной скважины в высоко- или низкопроницаемом участке пористого коллектора. Проведенное численное исследование показало, что для организации эффективного газогидратного хранения диоксида углерода необходимы пористые среды с достаточно высокими значениями проницаемости.

Ключевые слова: фильтрация, диоксид углерода, образование газового гидрата, зональная проницаемость, математическая модель

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