



## Theoretical research of the gas hydrate deposits development using the injection of carbon dioxide



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### ABSTRACT

In this paper the mathematical model of the carbon dioxide injection into a natural reservoir initially saturated with methane and its hydrate was constructed and also the research, using this model, was carried out. Self-similar solutions of this problem in the one-dimensional formulation were built. These solutions explain the distribution of the main parameters in a reservoir. It is shown that there are two possible regimes: the first – the recovery of methane from hydrate can occur without decomposition of CH<sub>4</sub> hydrate to gas and water, the second – with decomposition. In the first regime, the replacement of methane in hydrate with carbon dioxide is occurs. In the second regime the decomposition of CH<sub>4</sub> hydrate to gas and water with the subsequent formation of hydrate from water and carbon dioxide is occurs. The critical diagrams of the each regime existence were built and analyzed. The influence of the injection pressure of carbon dioxide and the reservoir permeability to the speed of methane recovery from hydrate for the each regime was researched.

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## 1. Introduction

Currently the natural deposits of gas hydrates due to significant resources and concentrated gas state are the serious alternative to conventional resources of natural gas [1–4]. Currently the main methods of the extraction of hydrocarbon gas (mostly methane) from a hydrate reservoir are the pressure drop and the heating of hydrate saturated rocks [1–4]. However, in some cases, these methods are inefficient because of large expenditure of energy. For example, when the pressure drop method of the gas hydrate deposits development, it is necessary that the reservoir pressure was below than the equilibrium decomposition pressure of hydrate [1–4]. This is achieved also due to the pumping-out of water from wells and this requires a large expenditure of energy. In addition, when the wells exploitation, the temperature decreasing due to the absorption of latent heat of hydrate decomposition, as well as adiabatic cooling and throttle effect. So the water which becomes free after the hydrate decomposition can freeze and plug the equipment [4].

When the thermal method of the gas hydrate deposits development the reservoir temperature increasing which necessary for the gas hydrate decomposition is achieved mainly by hot water circulation in a closed loop in a well or by injection into the reservoir of hot fluid (water or steam) [4]. However, this method also requires a large expenditure of energy for the hydrates decomposition and for the heating of the reservoir to the temperature that causes decomposition.

In connection with the foregoing, the actual problem is to research new methods of influence on gas hydrate reservoirs, which minimize expenditure of energy for the gas extraction. Promising in this respect is the technology of replacement, which consists in the displacement of methane from hydrates by filling their by other gas [5–11]. By several researches the most promising gas for the replacement process is carbon dioxide. Also its pumping into the hydrate saturated reservoir would at the same time to solve the problem of disposal of CO<sub>2</sub>, in connection with its negative role in the development of the greenhouse effect. This method is based on the fact that the hydrate of carbon dioxide is more stable than methane hydrate. Therefore the molecules of carbon dioxide can to replace the molecules of methane in the methane hydrate. Experimental researches proved that the replacement process CH<sub>4</sub>–CO<sub>2</sub> in the hydrate of methane does not require a supply of external energy, because the latent heat per unit volume of

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the CO<sub>2</sub> hydrate formation (from water and carbon dioxide) more than the latent heat of the CH<sub>4</sub> hydrate decomposition (to water and methane). In addition, a significant feature of the replacement process CH<sub>4</sub>-CO<sub>2</sub> in the methane hydrate is that this reaction occurs at pressures and temperatures corresponding to the conditions of stable existence of the methane hydrate [5,6] and not accompanied by the release of free water [5–8]. This eliminates the necessity to heat a reservoir to the temperatures causing the decomposition of methane hydrate to gas and water. These facts make this technology promising owing to the low energy costs. Technological ideas must be confirmed by the calculations based on valid theoretical models. Therefore the construction of the mathematical model of the dioxide carbon injection into the gas-hydrate reservoir is the actual problem.

Experimental researches of the replacement process of methane in hydrate with carbon dioxide are described, for example, in [5–11]. In these works, the researches were conducted in samples of small size and, as a rule, in the thermostatic and barostatic conditions. The replacement process in these researches is limited by the process kinetics due to the small sizes of the samples and the maintenance of constant temperature and pressure conditions. In the cases of long natural reservoirs at constant injection of carbon dioxide into these reservoirs the process of replacing methane with carbon dioxide in the hydrate will be limited by the mass transfer in a porous medium. Therefore, these researches do not give a complete picture of the processes that take place into natural reservoirs. This significantly complicates the comparison of the experimental data with the results of mathematical modeling of the processes for long reservoirs.

The mathematical models of the gas hydrate formation in extended porous mediums during the gas injection are formulated, in particular, in works [12–14], in which the process of the gas injection into the reservoir initially saturated with the same gas is researched. The mathematical model for the carbon dioxide injection into a reservoir, which contains methane and water in a free state, is presented in [15].

The mathematical model of the carbon dioxide injection into the natural reservoir saturated with methane and its hydrate is presented in this work. Note that some authors proposed to pump CO<sub>2</sub> into porous medium in the liquid state [6,8,9,11]. However, there are significant differences at the mathematical descriptions of the injection process of carbon dioxide in the gaseous or liquid state. It is known that the equilibrium temperature of the hydrate decomposition depends on pressure. Therefore, the possibility of the realization of the different regimes is determined by the overlay of the temperature and pressure fields. There are different equations of state when the different aggregate state of carbon dioxide. Also, difference in the coefficients of viscosity more than an order of magnitude. All this leads to substantial difference of pressure fields for the cases of the carbon dioxide injection in the liquid or gaseous state. This has a significant impact to the possibility of the realization of the different regimes. Let us explain this in more detail.

In the case of the liquid CO<sub>2</sub> injection the main hydraulic resistance has only the first reservoir zone, which is saturated with the liquid. In the case of the gaseous carbon dioxide injection the hydraulic resistance is uniformly by the all filtering area, because this area is saturated with gas. This is due, firstly, with the different equations of state for the liquid and gas, and, secondly, with a very significant difference in viscosity for the liquid and gas. Therefore, under the same conditions in the case of the CO<sub>2</sub> injection in the gaseous state the pressure drop in the near area is significantly lower than in the case of liquid CO<sub>2</sub>. In this connection, in the case of the gaseous CO<sub>2</sub> injection the dependence of pressure before the front (i.e., in the far area) on the injection pressure is much more significant than in the case of the liquid CO<sub>2</sub> injection. The realiza-

tion of the different regimes is defined by the equilibrium temperature in the far area, i.e. in fact by the pressure in the far area. When pumping liquid CO<sub>2</sub> a greater impact has the temperature of the injected CO<sub>2</sub> but not the injection pressure. In the case of the gaseous CO<sub>2</sub> injection a greater impact to the features of the process (in terms of the control of the process regimes) has the injection pressure. Therefore, in this work the effect of the pressures (the initial reservoir pressure and the injected fluid pressure) to the process features is researched. Also the concept of the critical pressure of the injected CO<sub>2</sub> was introduced and the value of this pressure was examined.

In addition, an essential feature of the heat and mass transfer processes with phase transitions is the release or absorption of the latent heat of phase transitions. When the injection of gaseous CO<sub>2</sub> the CO<sub>2</sub>-CH<sub>4</sub> replacement process in methane hydrate is exothermic. This process is endothermic when the injection of liquid CO<sub>2</sub>. This circumstance is taken into account in this work. Therefore, when the gaseous CO<sub>2</sub> injection the regime with the decomposition of CH<sub>4</sub> hydrate to gas and water can be realized even when the injected gas temperature is equal to the initial reservoir temperature. At the liquid CO<sub>2</sub> injection this regime can be realized only when the liquid CO<sub>2</sub> temperature is greater than the initial reservoir temperature. Therefore, the replacement regime while injecting liquid CO<sub>2</sub> more strongly depends on the temperature of the injected carbon dioxide than when the gaseous CO<sub>2</sub> injection.

## 2. The regime with the replacement of methane with carbon dioxide in hydrate

### 2.1. Problem statement and basic equations

Assume that the horizontal porous reservoir, initially saturated with methane hydrate (initial saturation  $v$ ) and methane, fills half-space  $x > 0$ . The top and bottom of the reservoir are impermeable. Initial pressure  $p_0$  and temperature  $T_0$  correspond to the thermodynamic conditions of the existence of mixture of methane and its hydrate:

$$t = 0, \quad x > 0: \quad T = T_0, \quad p = p_0, \quad S_h = v.$$

Assume that through the left boundary ( $x = 0$ ) gaseous carbon dioxide is being injected. The pressure of injection  $p_w$  and temperature of this gas  $T_w$  correspond to the conditions of the existence of heterogeneous mixture of carbon dioxide and its hydrate and are supported at this boundary at constant level:

$$t > 0, \quad x = 0: \quad T = T_w, \quad p = p_w.$$

Conditions of the existence of hydrates of carbon dioxide and methane are presented on the phase diagram (Fig. 1) [16]. In this chart the curves  $glh$  determine three-phase equilibrium “gas-water-hydrate”, and the curve  $lg$  – two-phase equilibrium “liquid-vapor”. The subscript 1 corresponds to CO<sub>2</sub> in gaseous or liquid state or its hydrate, and the subscript 2 – methane and its hydrate. In this problem the initial values of pressure and temperature of the reservoir are above the curve  $g_2lh_2$  (in the field of the existence of heterogeneous mixture of methane and its hydrate). The pressure and temperature of the injected carbon dioxide are above the curve  $g_1lh_1$  (in the field of the existence of carbon dioxide hydrate), but below the curve  $l_1g_1$  (in the field of the existence of gaseous carbon dioxide).

The values of temperature and pressure on the curve of three-phase equilibrium “gas-water-hydrate” are quite well described by the following equation [17]:

$$p = p_{s0} \exp\left(\frac{T - T_0}{T_*}\right), \quad (2.1)$$

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