



# Mathematical model for carbon dioxide injection into porous medium saturated with methane and gas hydrate

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## ABSTRACT

A mathematical model of the injection of warm carbon dioxide into a layer of finite length the pores of which are saturated in the initial state with methane and its gas hydrate is constructed. A solution of the problem is presented for the case of the formation of two moving interphase boundaries. In this case, at two different frontal boundaries there occurs the decomposition of the CH<sub>4</sub> hydrate into methane, water and the formation of CO<sub>2</sub> hydrate from carbon dioxide and liberated water. The dependence of the rate of decomposition of methane gas hydrate on the temperature of the injected carbon dioxide and the initial temperature of the formation is numerically studied, as well as on the pressure values on the right and left boundaries of the formation. It is shown that forming two different boundaries of phase transitions is realized at high temperatures of the formation and injected carbon dioxide. A critical diagram is constructed that determines the “injection temperature–initial reservoir temperature” and “injection pressure–initial pressure of the formation” on the parameter planes in the region of existence of two different process flow regimes. The dependence of the coordinates of both boundaries of phase transitions on time is investigated. It has been established that over time the formation boundaries of CO<sub>2</sub> hydrate and the decomposition boundaries of the CH<sub>4</sub> hydrate can be merged. The influence of heat released during the formation of the CO<sub>2</sub> hydrate on the acceleration of the decomposition of the CH<sub>4</sub> hydrate was studied. It was found that the latent heat of formation of the CO<sub>2</sub> hydrate markedly increases the rate of decomposition of CH<sub>4</sub> hydrate, and the contribution of this heat to the rate of decomposition of the hydrate increases with decreasing temperature of the injected gas.

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## 1. Introduction

At the moment, natural gas hydrates are a promising object of uncommon natural gas resources [1–3]. As of right now, pressure reduction and heating of hydrate-bearing rocks are considered the main methods of obtaining gas from the gas hydrate formation. However, these methods are in many cases ineffective due to high energy costs. Thus, with the depressive method (tested, for example, at the Messoyakhskiye field [4]), the reduction in reservoir pressure necessary to decompose the gas hydrate is achieved by pumping water from the well. This requires so much energy that a significant portion of the gas produced at Messoyakha has to be used to compensate for the energy expended at the mining site. With the thermal method for the development of gas hydrate deposits (tested, for example, at the Canadian experimental drilling rig Mallik [5]), the increase in formation temperature necessary to

decompose the gas hydrate is achieved mainly by pumping hot water. In this case, the energy consumption is comparable to the energy capacity of the released gas. Due to high energy consumption, the economic costs of developing gas hydrate deposits exceed the costs of developing traditional gas fields. This is one of the main deterrents to gas production from gas hydrates on an industrial scale.

One of the new ways of recovering methane from gas hydrate is the method of injecting carbon dioxide, which can replace methane in the initial gas hydrate [6–13]. As experimental studies have shown, the methane replacement reaction with carbon dioxide in methane hydrate does not require the supply of energy from outside. This is due to the fact that the latent heat of formation of CO<sub>2</sub> hydrate from water and carbon dioxide per unit volume of gas hydrate is greater than the latent heat of decomposition of CO<sub>2</sub> hydrate into water and methane. It should be noted that studies [6–13] and other similar experimental studies were carried out in samples of small size. Therefore, in these studies, the process of gas hydrate replacement because of small sample sizes is limited

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by the process kinetics (gas diffusion through the hydrate crust). In the case of real long natural formations, the formation of gas hydrate is limited not so much by the kinetics of the process as by heat and mass transfer in the porous medium itself. These factors do not allow us to compare the results of mathematical modeling of gas hydrate replacement processes in natural formations with experimental data. This makes this technology promising in terms of energy costs, which was convincingly proved in 2012–2013 during the experiment in Alaska (testing well Ignik Sikumi [14]).

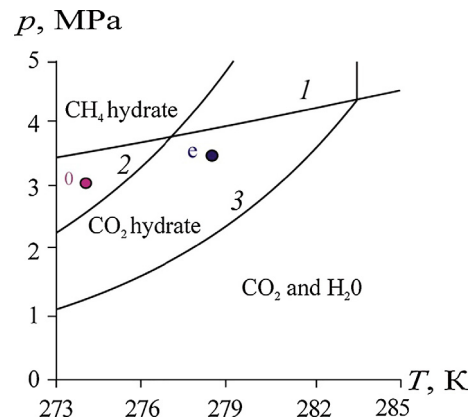
The results of the experimental study of the formation of carbon dioxide gas hydrate are given, in particular, in studies [15–17]. In these and other similar experimental studies, research was done in providing a rapid increase in pressure reactors. Therefore, the results of these studies describe the features of such hydrate formation processes, which are limited by their kinetics.

According to the data obtained in experimental studies, the process of transition of the hydrate of  $\text{CH}_4$  to  $\text{CO}_2$  hydrate during the injection of carbon dioxide can occur in two different modes [10–12]. In the first mode, there is no release of free water. This mode is realized at pressures and temperatures corresponding to the stable existence of methane hydrate. In the second mode, intermediate decomposition of methane hydrate to gas and water occurs, as does the sequential formation of  $\text{CO}_2$  hydrate from carbon dioxide and water. This mode is realized at thermodynamic parameters corresponding to the existence of a mixture of methane and water. According to experimental data in the first mode, the process of transition of the hydrate of  $\text{CH}_4$  to the  $\text{CO}_2$  hydrate is much slower than in the second mode [8]. One of the reasons for this is that in the first mode there is no destruction of the solid gas hydrate and therefore the replacement rate in the methane hydrate to carbon dioxide is determined by the diffusion of these gases in the hydrate. In the second mode, the rate of phase transformations is determined by the diffusion of these gases in water. However, the diffusion coefficient of gases in water is much higher than in the hydrate [13].

There are several articles devoted to the mathematical modeling of the processes of developing gas-hydrate deposits by pumping carbon dioxide. Approximate analytical solutions of the problem of injecting carbon dioxide into a reservoir containing methane and its gas hydrate are presented, for example, in [18–20]. In these papers, self-similar solutions of the problem for a semi-infinite formation are constructed. However, these solutions can adequately describe the process only at the initial time stage of the process (i.e., until the pressure perturbation reaches the right boundary of the formation). Later on, with the passage of time, the conditions on the right boundary will have an increasingly more significant effect on the process. In the present work, in contrast to [18–20], the finite extent of the formation is taken into account and the influence of pressure on its right boundary on the features of the process are investigated. It was shown in [18–20] that the process of injection of warm carbon dioxide can be accompanied not only by the substitution of methane for carbon dioxide in the gas hydrate, but also by decomposition of methane gas hydrate into gas and water. This case is interesting because the heat released during the formation of  $\text{CO}_2$  hydrate (approximately 400 kJ per 1 kg of hydrate) can accelerate the decomposition of methane hydrate. In this paper, the case is examined when, upon the injection of warm carbon dioxide, decomposition of  $\text{CH}_4$  hydrate into methane and water and the formation of  $\text{CO}_2$  hydrate from carbon dioxide and liberated water occurs.

## 2. Mathematical model

A horizontal porous formation of a finite length  $L$ , is initially saturated with methane hydrate with the saturation  $v$ . Pressure  $p_0$  and temperature  $T_0$  of the formation in their initial states on the



**Fig. 1.** Initial formation parameters (point 0) and parameters of injected carbon dioxide (point e) in the phase diagram of the « $\text{CO}_2\text{-H}_2\text{O}$ » and « $\text{CH}_4\text{-H}_2\text{O}$ » systems. Curve 1 is a two-phase equilibrium line “liquid-vapor” for carbon dioxide. Curves 2 and 3 are three-phase equilibrium lines “ $\text{CH}_4\text{-water-hydrate CH}_4$ ” and “ $\text{CO}_2\text{-water-hydrate CO}_2$ ”, accordingly.

phase diagram (point 0, Fig. 1) lie in the area of stable existence of methane gas hydrate [21]. A gaseous carbon dioxide is injected through the left boundary of the formation ( $x = 0$ ). Pressure  $p_e$  and temperature  $T_e$  of the injected carbon dioxide on the phase diagram (point e in Fig. 1) correspond to the conditions of the existence of a mixture of carbon dioxide gas and its hydrate, as well as a mixture of methane and water.

Then, as a result of the injection of carbon dioxide,  $\text{CH}_4$  hydrate decomposes into methane and water, and further formation of  $\text{CO}_2$  hydrate from carbon dioxide and liberated water occurs.

In this article, we will neglect the mixing of gases and assume the stability of the displacement front of methane by carbon dioxide. This is due to the laminar nature of the flows in natural formations and the higher value of the viscosity of carbon dioxide compared to methane. In addition, it was shown in [22] that in the natural porous formations the filtration (convective) transport significantly prevails over the diffusion transfer. Therefore, the diffusion mixing of gases in this work will be neglected.

The experimental values of the kinetics of the formation and decomposition of gas hydrates in porous media strongly depend on the characteristic pore sizes, but on average it is on the order of several hours. Thus, in [23], in which the formation and decomposition of  $\text{CH}_4$  hydrate in a porous medium with a grain size of 110  $\mu\text{m}$  was studied, this time was of the order of one hour. In this paper, we will assume that the reservoir has a sufficiently long extension (100 m). We will also consider the case where the formation has a low or medium permeability ( $k < 10^{-13} \text{ m}^2$ ). Then the characteristic time for reaching the front of the displacement of methane by the carbon dioxide of the right boundary of the reservoir will significantly exceed the characteristic time of the kinetics of formation and decomposition of gas hydrates. Consequently, considering sufficiently large values of time after the onset of gas injection (of the order of several days), the kinetics of the formation and decomposition of gas hydrates can be neglected.

Thus, we can assume, that under said conditions three corresponding areas materialize. In the first (the nearer) region, the pores are saturated with carbon dioxide and its hydrate, in the second (intermediate) porous medium contains methane and water, and in the third (the further) region the pores are saturated with methane and its hydrate (Fig. 2). Thus, the decomposition of  $\text{CH}_4$  hydrate occurs on a movable frontal surface that separates the second and third regions (the far boundary of the phase transitions). The formation of  $\text{CO}_2$  hydrate occurs on a movable frontal surface

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