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Underground Storage of Natural Gas in Hydrate State: Numerical Experiment

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Abstract. The paper is devoted to simulation of the initial stage of natural gas hydrate underground storage: gas injection into aquifer just below permafrost rocks. It is based on the mathematical model of multiphase non-isothermal real gas and water flow in porous media. The model takes into account the transformation of gas and water into hydrate at certain temperature which depends on gas flow pressure. The dynamics of hydrate and water saturation as well as the pressure and temperature fields in a reservoir with given porosity, permeability and initial values of pressure, temperature and water saturation have been studied. An implicit finite-difference scheme is used to approximate the original boundary-value problem. The finite-difference equations have been solved using simple iteration and sweeping algorithms. Several examples of calculations corresponding to real cases are given. Calculations have revealed that the final result strongly depends on the combination of porosity and permeability of a reservoir.

1. Introduction

Nowadays underground gas storages are built in the depleted gas reservoirs or aquifers situated near gas pipelines or large centers of gas consumption. They are used to meet load variations, that is, gas is injected into storage during periods of low demand and is withdrawn during periods of peak one, which is especially important for the Northern regions where they can be used as distinctive accumulators of natural gas.

One of the alternatives to common gas storages would be those of hydrates compounds formed when natural gas is injected into porous reservoirs under certain thermodynamic conditions (at specific temperature – pressure relations controlled by the gas composition). Subpermafrost aquifers in the areas of continuous permafrost act readily as such reservoirs. For example, in Central Yakutia, they can occur directly beneath the permafrost base at depths of 500-600 m [1], with permeability ranging between 10^{-12} and 10^{-14} m^2.

In [2] it is shown that carbon dioxide sequestration in the subpermafrost horizons is possible through CO_2 injections into reservoirs located beneath the carbon dioxide hydrate stability zone.

Major advantages of this method consist in greater compactness and stability of the repository, given that gas in a solid state occupies a considerably smaller volume versus its being in free state at equal temperature and pressure, and that during gas transition into the hydrate state all free reservoir water becomes bound. The authors proved conceptual possibility of gas underground storage in hydrate state via mathematical modeling of gas injection into water saturated reservoir at shallow depths corresponding to the permafrost base in the central part of Eastern Siberia [3]. Gas injection time was limited to 10 days. Here time of gas injection is extended up to 100 days, which corresponds to period of lower gas consumption during the summer. Approximate models of the problem are described in publications [4 - 12].

2. Problem formulation

To assess the concept of storing natural gas in hydrate state consider a standard axisymmetric problem of gas injection into a horizontal aquifer, with impermeable and thermally insulated top and bottom, through a single well. Let us assume that gas flows in the reservoir initially saturated with water. Porous matrix is considered rigid, gas is only in the gaseous/hydrate state, whereas water – only in the liquid/hydrate states, that is neither ice nor vapor are formed.

In [13] it was shown that, the role of thermal conductivity is negligible versus forced convection in overall heat transfer balance and, therefore, the heat conductivity term in the energy equation is can be set to zero. Then, in the frame of multiphase flow mechanics [14] and subject to the generalized Darcy's law, the energy equation in cylindrical coordinates takes the following form:

$$(\rho c)_e \frac{\partial T}{\partial t} - mq\rho_h \frac{\partial \nu}{\partial t} - m(1-\nu-\sigma)\left(1+\frac{T}{z}\frac{\partial z}{\partial T}\right)\frac{\partial p}{\partial t} - k(1-\nu)\left(\rho_w c_w \frac{f_w}{\mu_w} + \rho_g c_g \frac{f_g}{\mu_g}\right)\frac{\partial p}{\partial r}\frac{\partial T}{\partial r} +$$

$$+ k(1-\nu)\rho_g c_g \frac{f_g}{\mu_g}\frac{RT^2}{c_p p}\frac{\partial z}{\partial T}\left(\frac{\partial p}{\partial r}\right)^2 = 0, \tag{1}$$

where $(\rho c)_e = (1-m)\rho_s c_s + m(1-\nu-\sigma)\rho_g c_g + m\nu\rho_h c_h + m\sigma\rho_w c_w$ – is effective value of specific volume heat capacity of porous medium saturated with gas, hydrate and water.

The equations of gas and water filtration are to be written down in these same coordinates:

$$m\frac{\partial}{\partial t}\left((1-\nu-\sigma)\frac{p}{zT}\right) = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{k(1-\nu)f_g}{\mu_g}\frac{p}{zT}\frac{\partial p}{\partial r}\right) - m\rho_h \varepsilon R\frac{\partial \nu}{\partial t}, \tag{2}$$

$$m\frac{\partial \sigma}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{k(1-\nu)f_w}{\mu_w}\frac{\partial p}{\partial r}\right) - m(1-\varepsilon)\frac{\rho_h}{\rho_w}\frac{\partial \nu}{\partial t}. \tag{3}$$

Here and elsewhere the following notations are used: c – specific heat capacity; f – relative permeability; k – absolute permeability; m – porosity; p – pressure; q – latent heat of "hydrate–gas+water" phase change; R – gas constant; r – space coordinate; r_b – well radius; r_k – external boundary radius; T – temperature; t – time; z – gas compressibility function; ε – gas content per hydrate unit volume; μ – dynamic viscosity; ρ – density; σ – water saturation; ν – hydrate saturation. The lower indices g, h, s, w, 0 stand for gas, hydrate, solid matrix, water and reference state, respectively.

To find a single-valued solution of the (1) - (3) set, initial and boundary conditions should be formulated. Constant values of pressure, temperature, hydrate saturation and water content have been chosen as initial conditions:

$$p(r,0) = p_0,\ T(r,0) = T_0,\ \nu(r,0) = \nu_0,\ \sigma(r,0) = \sigma_0. \tag{4}$$

At gas injection point (bottom hole), the following conditions are set:
constant temperature

$$T(r_b, t) = T_b; \tag{5}$$

and bottom hole gas pressure or its volume flow rate (modified to normal physical conditions)

$$2\pi\, r_b H \frac{\rho_g}{\rho_n} \frac{k(1-\nu) f_g}{\mu_g} \frac{\partial p(r_b, t)}{\partial r} = -Q\,, \tag{6}$$

where H – reservoir thickness; ρ_n – gas density at ρ_n = 101325 Pa and T_n = 273.15 K.

Instead of impermeability condition at reservoir boundary used in [3] here the possibility of water flow outside storage boundary is stated

$$-\frac{\partial p(r_k, t)}{\partial r} = \frac{f_w(p(r_k, t) - p_0)}{r_k \ln(r_{out}/r_k)}, \tag{7}$$

where r_{out} – radial distance of hydrodynamic influence.

Equations of the problem are closed by:

1) the relations for gas and water relative permeabilities [15]

$$f_g(\sigma) = \begin{cases} \left(1 - \dfrac{\sigma}{0.9}\right)^{3.5} (1 + 3\sigma),\ 0 \le \sigma < 0.9 \\ 0,\ 0.9 \le \sigma \ge 1 \end{cases};\quad f_w(\sigma) = \begin{cases} \left(\dfrac{\sigma - 0.2}{0.8}\right)^{3.5},\ 0.2 < \sigma \le 1 \\ 0,\ 0 \le \sigma \le 0.2 \end{cases}; \tag{8}$$

2) gas-hydrate-water thermodynamic equilibrium condition

$$T = \alpha_1 \ln p + \alpha_2\,, \tag{9}$$

where α_1, α_2 – empirical constants determined through experimental data or calculated for gas of a given composition, on the basis of methods described in [16, 17];

3) equation of state for real gas

$$\rho_g = p / zRT\,, \tag{10}$$

where $z = (0.17376 \ln(T/T_c) + 0.73)^{p/p_c} + 0.1\, p/p_c$ – gas compressibility function approximated by the empirical equation from [18]. Critical parameters of natural gas are determined according to its composition by the Kay's Rule (for non-ideal gas mixtures) [19].

The method of finite differences is applied to solve the problem (1) - (10), that is, the original equations, boundary and initial conditions are replaced by their mesh analogues [13], whereas the proposed by [20] algorithm for implementation of simple iterations method is applied for solving the corresponding system of algebraic equations at each time step.

3. Results of computational experiment

The effects of gas injection rate on the dynamics of the fields of temperature, pressure, water and hydrate saturation were studied in computational experiment. Other initial parameters were the following:

ρ_w = 1000 kg/m^3, ρ_s = 2650 kg/m^3, ρ_h = 920 kg/m^3, c_w = 4200 J/(kg·K), c_s = 700 J/(kg·K), c_h = 3210 J/(kg·K), c_g = 2093 J/(kg·K), q = 510 000 J/kg, ε = 0.147, $\mu_w = 1.8\cdot10^{-3}$ Pa·s, $\mu_g = 1.3\cdot10^{-5}$ Pa·s, $p_0 = 3\cdot10^6$ Pa, T_0 = 274.15 K, T_b = 279.15 K, H = 10 m, r_b = 0.1 m, r_k = 300.1 m, r_{out} =1000.1 m, m = 0.15, $k = 8\cdot10^{-14}$ m^2.

The composition of the injected natural gas and calculated gas constant, critical pressure and temperature, and empirical coefficients in equation (9) correspond to the Sredne-Botuobinskoye field in the Republic of Sakha (Yakutia): CH_4 – 85.90, C_2H_6 – 7.32, C_3H_8 – 2.24, iC_4H_{10} – 0.26, nC_4H_{10} – 0.68, iC_5H_{12} – 0.17, nC_5H_{12} – 0.24, C_6H_{14} – 0.08, CO_2 – 0.05, N_2 – 2.64, H_2 – 0.14, He – 0.28 (volume percents); R = 445.6 J/(kg·K), p_c = 4.555·106 Pa, T_c = 204.134 K, α_1 = 7.82 K, α_2 = 166.64 K.

The computational experiment was carried out to evaluate the role of gas injection flow rate (1 m^3/s and 5 m^3/s) in dynamics of hydrate and water saturation fields as well as temperature and pressure ones. Initially the aquifer does not contain hydrates and its water saturation equals 0.9. The most essential results of calculations can be seen at Fig. 1 - 7. Their analysis leads to the following conclusions.

At first, consider dynamics of gas temperature because of its determinative role in hydrate formation.

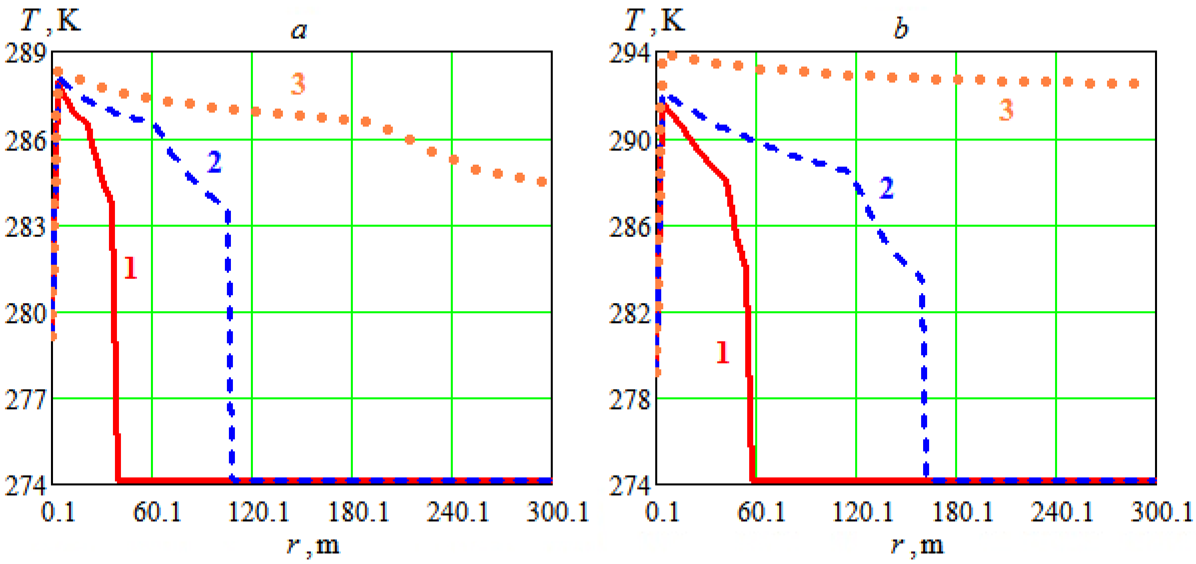

Figure 1. Temperature distribution in reservoir: *a, b* – volume flow rate equals to 1 and 5 m^3/s, correspondingly (1 – t = 1.25 days, 2 – t = 10 days, 3 – t = 100 days).

It is seen from Figures 1 and 2*a* that over a short time interval temperature grows up significantly: at high flow rate – about 20 degrees, at low one - about 15 degrees (cf. curves 1 and 2 with 3 and 4 in Fig. 2*a*). At the end of gas injection for high flow rate the temperature is almost leveled throughout the reservoir (curve 3 in Fig. 1*b*).

Near gas well pressure is growing with the same speed as temperature but at low flow rate it almost reaches its limit of 6 MPa (curve 3 in Fig. 2*b*), while for high flow rate it is growing progressively (curves 1 and 2 in Fig. 2*b*). Pressure growth at high flow rate is twice as much as at low one (cf. Fig. 3*a* and Fig. 3*b* as well as curves 1 and 2 with 3 and 4 in Fig. 2*b*). For some geological conditions, such high pressure may lead to rock fractures.

Now consider the influence of dynamics of pressure and temperature fields on water displacement and hydrate formation in the storage. Here we limit the analyses to the case of low permeability because it corresponds to higher temperature and pressure values, which may lead to *a priory* unpredictable dynamics of hydrate formation.

Comparison of curves 1 and 2 in Fig. 1 and in Fig. 4 shows that velocity of water saturation front is significantly lower than that of the temperature front. At Fig. 4 and Fig. 5 it is clearly seen that water saturation distribution is in qualitative agreement with the solution of Buckley-Leverett problem [15]. The effect of hydrate formation, i.e. transition of water into the immobile phase, is manifested in non-monotonic water distribution behind the front and in the fact that water saturation before the front is always lower than 1 (curves 1 and 2 in Fig. 4). Naturally, velocity of front propagation is strongly dependent on rate of gas injection. However, in accordance with the theory of two-phase flow in porous media [15] gas injection cannot displaces all reservoir water (see curve 3 in Fig. 4*b*).

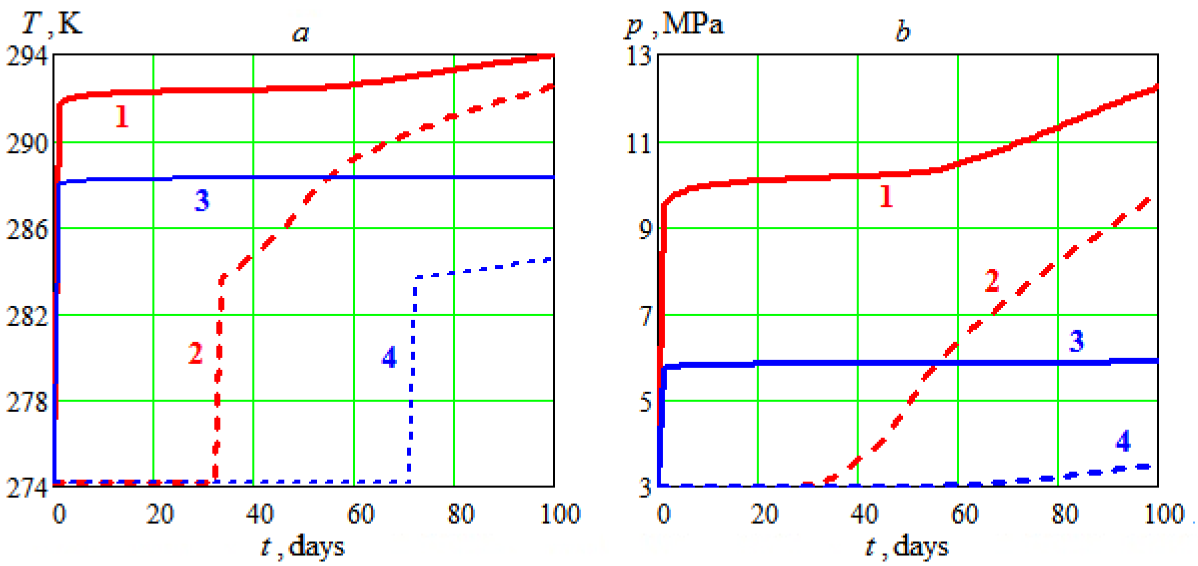

Figure 2. Temperature (*a*) and pressure (*b)* dynamics; 1 and 2 – volume flow rate equals 5 m^3/s; 3 and 4 – volume flow rate equals 1 m^3/s (solid lines – $r = 0.1$ m, dashed lines – $r = 300.1$ m).

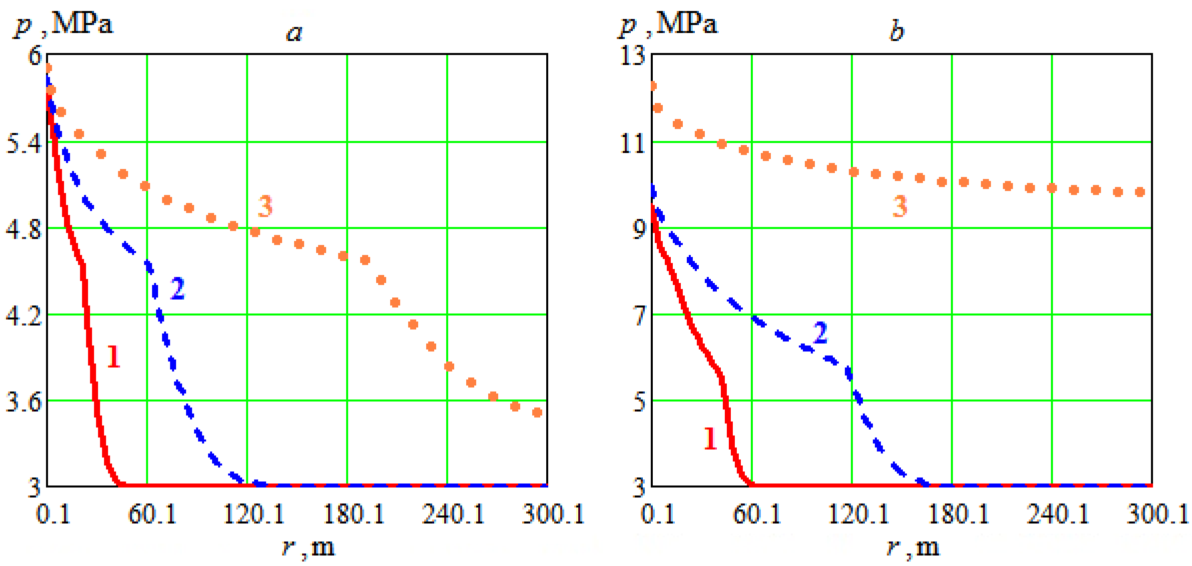

Figure 3. Pressure distribution in reservoir: *a, b* – volume flow rate equals to 1 and 5 m^3/s, correspondingly (1 – $t = 1.25$ days, 2 – $t = 10$ days, 3 – $t = 100$ days).

Calculations of hydrate formation show the complicated influence of such competitive factors as reservoir conditions and technology of gas injection. First of all, it is seen that higher rate of injection is more favourable for hydrate formation in a reservoir with lower permeability. It is clear from the fact that high pressure is favourable for hydrate formation. The statement is also supported by comparison of curves 3 in Fig. 6*a* and Fig. 6*b*. Promising is the growth of hydrate saturation at the reservoir boundary with time (curve 3 in Fig. 7*b*).

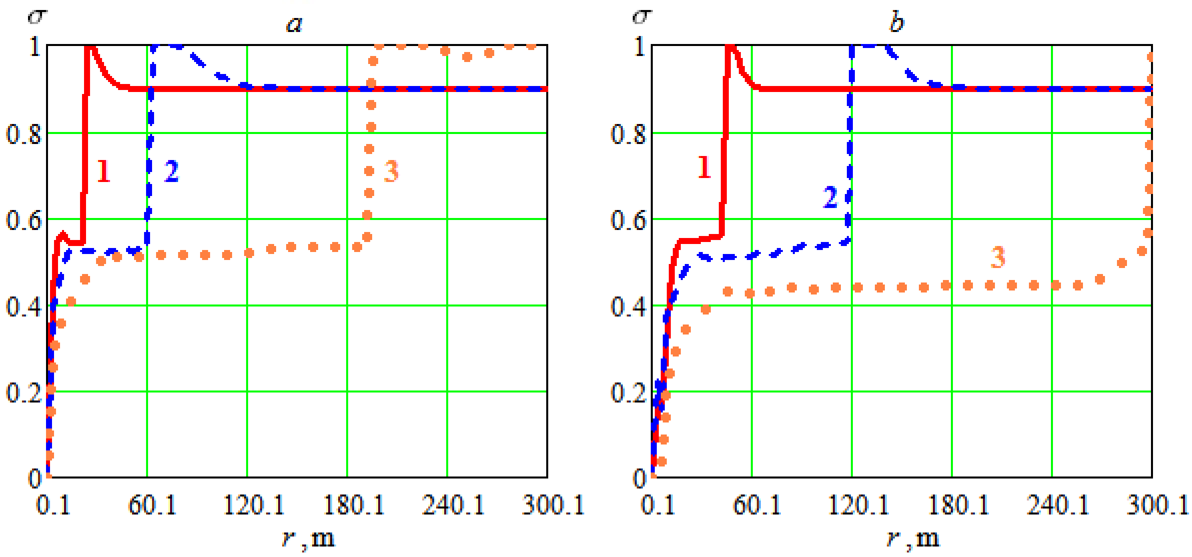

Figure 4. Water saturation distribution in reservoir: *a* – volume flow rate equals 1 m^3/s; *b* – volume flow rate equals 5 m^3/s (1 – t = 1.25 days, 2 – t = 10 days, 3 – t = 100 days).

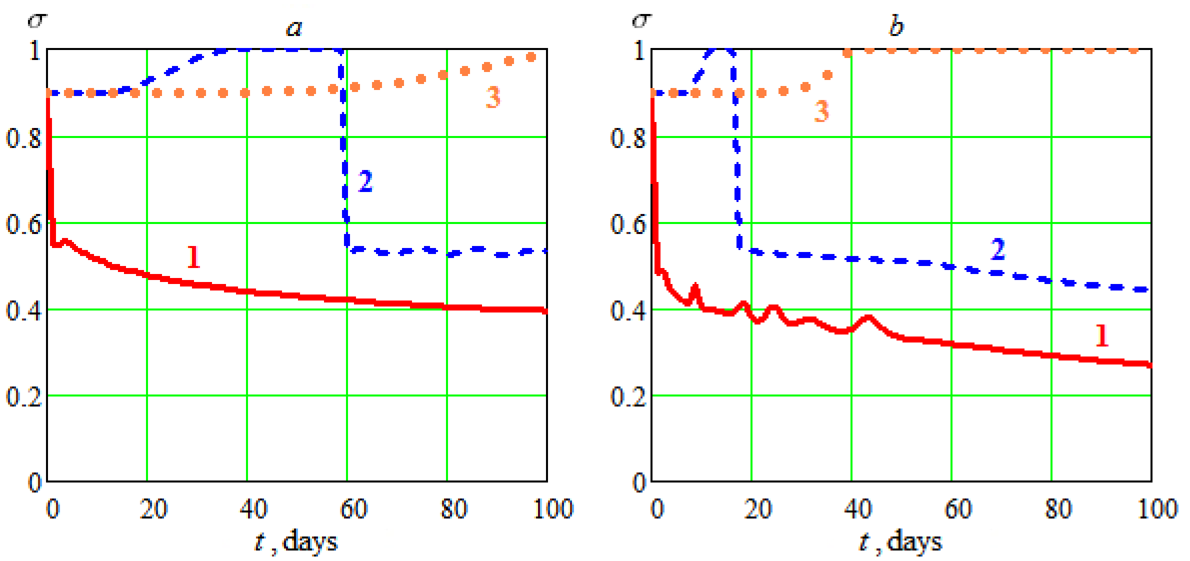

Figure 5. Water saturation dynamics: *a* – volume flow rate equals 1 m^3/s; *b* – volume flow rate equals 5 m^3/s (1 – r = 12.4 m, 2 – r = 150.1 m, 3 – r = 300.1 m).

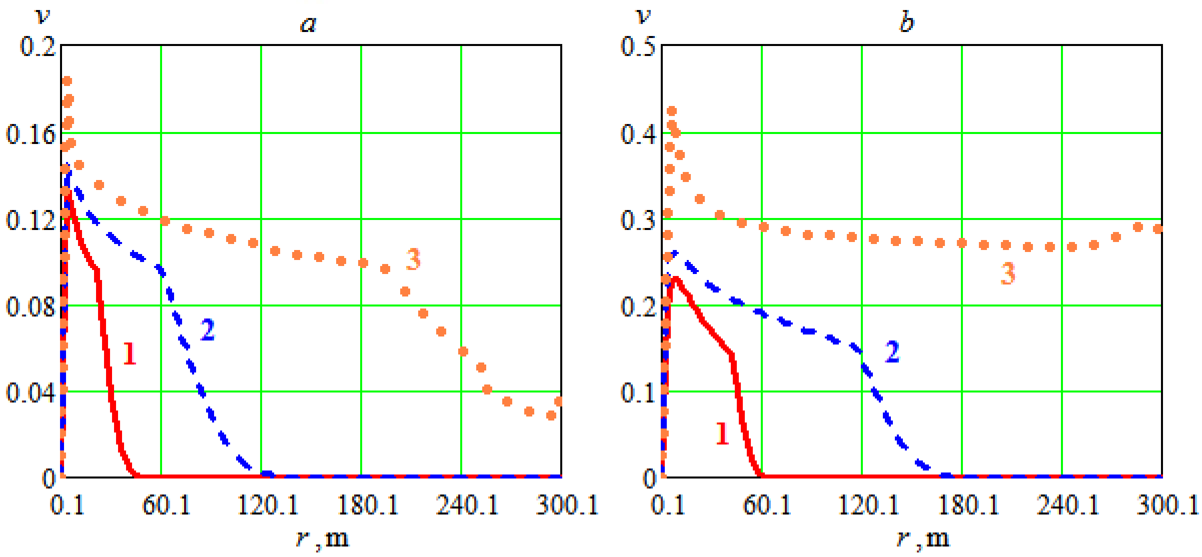

Figure 6. Hydrate saturation distribution in reservoir: *a* – volume flow rate equals 1 m^3/s; *b* – volume flow rate equals 5 m^3/s (1 – t = 1.25 days, 2 – t = 10 days, 3 – t = 100 days).

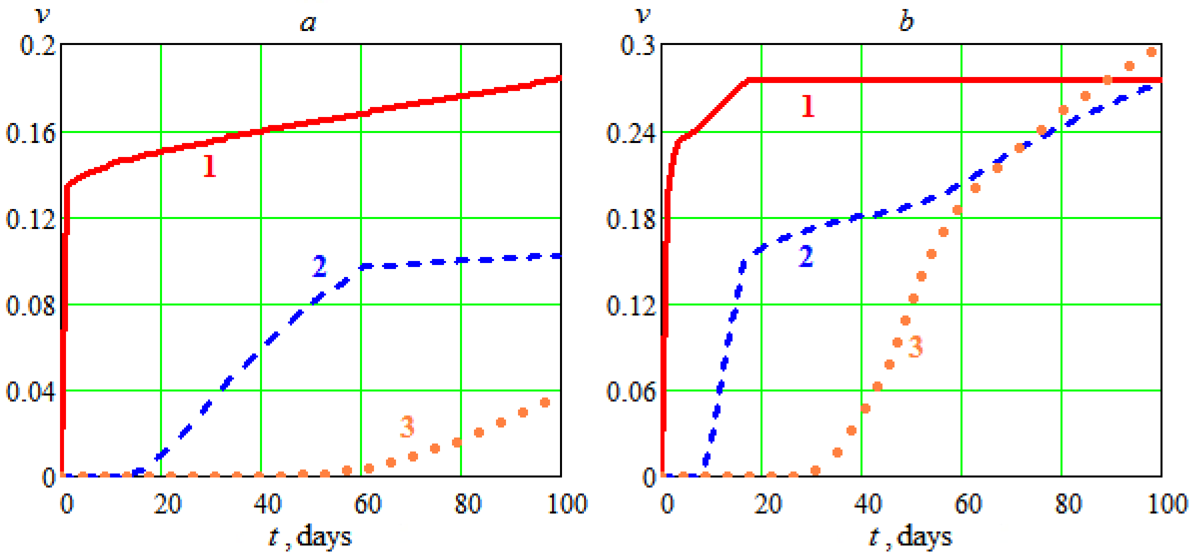

Figure 7. Hydrate saturation dynamics: *a* – volume flow rate equals 1 m^3/s; *b* – volume flow rate equals 5 m^3/s (1 – r = 12.4 m, 2 – r = 150.1 m, 3 – r = 300.1 m).

4. Conclusions

The results of computational experiment show that underground gas hydrate storage development in the subpermafrost aquifers requires a careful analysis of the reservoir geological characteristics and well test data. Specifically, reservoirs with porosity less than 0.2 should be preferred because it ensures a uniform filling of the storage by hydrate. Permeability higher than 10^{-14} m^2 is advantageous to prevent loss of sealing properties of the reservoir top and bottom at high rate of gas injection.

Additional research is needed to estimate thermal interaction of gas storages with the surrounding rocks and hydrate formation after an injection period.

The results of numerical experiment and proposed mathematical model can be used in the development of scientific bases substantiation of underground hydrate storage of natural gas, as well as carbon dioxide and other toxic gases.

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