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MATHEMATICAL MODELING OF DISPERSION OF ARGILLACEOUS MINERALS IN MUDDED OIL RESERVOIR BEDS

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The reasons for the technogenic decrease in the filtering capacity of oil reservoirs are considered. It is shown that the processes of transformation of the structure of argillaceous minerals are determining in this case. They lead to the dispersion of argillaceous aggregates and the segregation of smectite and mica components. The latter, being carried by a fluid flow and being mechanically fixed on the inhomogeneities of the pore channels, can form a reverse electroosmotic flow (retarding filtration) due to the high surface charge having no time to be compensated for. A mathematical model of the process has been constructed. The results of the calculations are presented.

Introduction. The experience gained over many years in exploiting the largest oil deposits in Tatarstan and other regions has made it possible to reveal a number of phenomena that are difficult to explain within the framework of the traditional conceptions of oil production. Every so often the injection of fresh water from the surface basins into an oil bed decreases the rate of filtration both in the face zones of injection wells and within the entire terrigenic reservoir, especially in the case of its increased mudding. This process not infrequently becomes disastrous in the productive Devonian strata in Tatarstan. It has been established that the decrease in the density of the stratal water to 1.09 g/cm^3 usually leads to the fact that the oil-saturated sandstone or siltstone with a content of the clay component of more than 5% practically ceases to participate in the production [1]. Phenomena analogous in their consequences, even though smaller in scale, are observed in some regions of Western Siberia.

Investigations concerning the evaluation of the character and scale of the phenomena involved in the decrease in the mineralization of the stratal water within the oil-producing areas and oil pools in the territory of Tatarstan in connection with the injection of fresh surface water into a bed were begun in 1982 and were continued during all the ensuing years. In this case, it was initially suggested that the main reason for the decrease in the filtering properties of an oil reservoir was the effect of swelling of the argillaceous cement of rocks, involved in the change of the hydrochemical regime of the bed. However, permeable rocks buried at depths of more than 1.7 km usually do not contain montmorillonite (the main swelling component of clays) as an independent phase capable of swelling. This contradiction stimulated the search for other causes and mechanisms of the negative effects involved in the technogenic change in the hydromechanical regime of productive oil beds.

In [2], it has been established that here the main effect is the transformation of some argillaceous minerals to others, involved in the change of the hydrodynamic regime of an oil bed in injection of fresh surface water. The main method of investigation was x-ray diffractometry of oriented preparations obtained by precipitation of mineral particles from the aqueous suspension on the surface of a glass plate. Visualization of the results of the technogenic transformation of the mineral composition of rocks has been performed with the use of a Hitachi HSM-2 scanning electron microscope. We also carried out ordinary investigations of rocks in transparent metallographic sections by the method of crystal optics [2]. Our main concern in the experiments was with the specimens of rocks from oil wells duplicating injection wells and wells compacting the operation net on areas with a high degree of water encroachment of oil pools. The main objective of the experiments was to detect the newly formed argillaceous minerals, which has been realized in investigations of specimens from duplicate wells, and to demonstrate the mobility of the fundamental particles of the newly formed mica by analyzing the sediment from the settling basins of the plants for primary prepara-

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tion of oil where these particles have been detected along with the traditional minerals of the sediment. A characteristic feature of the process of transformation of minerals and of subsequent transfer of the dispersed phase is its inhomogeneity over the bed section. The effect under study must lead to the removal of mica particles from the central zone of the bed and to the appearance of an inhomogeneity in the filtrating properties of the initially homogeneous bed. An analysis of the specimens of rocks has confirmed this conclusion. Our experiments have been described in more detail in [2].

Thus, it has been established that the injection of weakly mineralized surface water into an oil bed leads to the formation of secondary smectite (swelling component of clay) from illite. The field consequences of formation of smectite (swelling) phases in an oil reservoir cannot be reduced to a simple increase in the capability of clays to swell. Below we give calculations confirming this conclusion. The reverse transformation of illite, occurring in the volume of a reservoir with time, represents a process directly dependent on the regimes of oil production. Since the crystalline structure of micas is heterogeneous, the formation of smectite blocks cannot proceed simultaneously and homogeneously throughout their volume. As a result, there arises a system consisting of weakly connected mica plates–fundamental particles which are extended in a two-dimensional manner and are separated by regions with a smectite-like structure. The introduction of water molecules into the newly formed labile interlayer spaces of smectite produces intense mechanical stresses [3]. All this decreases the connection of the fundamental particles with the solid skeleton of a rock. As a result, the reaction products can be mobilized by a fluid flow, forming a moving suspension.

A crystallochemical feature of micas, in contrast to other minerals forming the skeleton of rocks, is the existence of a high negative charge distributed over the entire volume of the silicate layer. In a homogeneous structure, it is compensated for with cations K^+. This charge forms a negative potential across the surface of mica particles, equal to $4.25 \cdot 10^{-4}$ e/nm^2 (e is the electron charge) in the limiting case (where compensators are absent). Its value usually does not exceed $0.8 \cdot 10^{-4}$ e/nm^2 for montmorillonite. Whereas smectites compensate for their excessive charge with any hydrated cations of the solution, which actually takes place when labile interlayer spaces are formed, the layer charge of micas can locally be compensated for only with anti-Stokes cations (K^{1+}, Rb^{1+}, and Cs^{1+}) which do not have hydrated shells. On the geological scale of time, the surfaces of mica particles are practically completely neutralized by the processes of chemisorption in the case where they are in contact with the stratal water. But when technogenic phases having a mica-like structure appear in a reservoir, the surface of these particles must preserve to a large degree its high negative potential if the water injected into the bed is fresh and does not contain anti-Stokes cations.

Physical Model and Calculations. The experimental data obtained make it possible to construct a physical model and to evaluate the role of each of the possible technogenic processes decreasing oil production from the productive Devonian strata. For this purpose it is necessary first of all to determine the scale of the change in the pore space of the reservoirs caused by the swelling of their clay component. The thermodynamics of montmorillonite clay (or clay containing smectite-like blocks and being in contact with the electrolyte solution) is determined by the condition of equilibrium of its colloidal particles [4]:

$$\Pi (H) = P_{\mathrm{m}} + P_{\mathrm{i}} + P_{\mathrm{a}} = 0 \, . \tag{1}$$

The components P_{m} and P_{i} are determined in the standard manner; P_{a} is due to the excess of the dissolved component in the interlayer because of the cations adsorbed by particles, and it is determined from the formula [4]

$$P_{\mathrm{a}} = RT \int_0^{C_0} \left(\frac{\partial \Gamma}{\partial H} \right) \frac{dC}{C} \, , \tag{2}$$

where

$$\Gamma = \nu_0^{-1} \int_{\delta}^{H-\delta} [C - C_0] \, dx \tag{3}$$

is determined from the conditions of chemical equilibrium of the cations and anions of the porous and free solutions with concentration C_0:

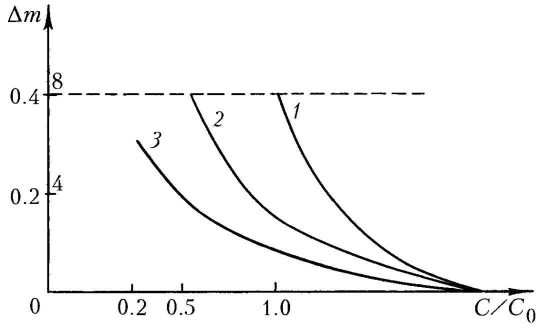

Fig. 1. Change in the relative porosity Δm of an oil reservoir due to the decrease in the mineralization of stratal water in the process of injection: 1) for an 8% content of smectite in the rock (right vertical scale), 2) for an 0.8% content, and 3) for 0.5% content of smectite in the rock (left vertical scale). The dashed line denotes the actual decrease in the porosity according to the data of [6]. Δm, %.

$$\bar{\mu}_i = \mu_i\,, \quad i = 1, 2\,. \tag{4}$$

With allowance for the condition of electroneutrality of the system particle–pore solution we have

$$\bar{C}_1 - \bar{C}_2 = \frac{2\sigma}{H}\,. \tag{5}$$

For $\bar{C}_1$ and $\bar{C}_2$ we obtain

$$\bar{C}_1 = \frac{\sigma}{H} + \sqrt{\frac{\sigma^2}{H^2} + C_0^2}\,, \quad \bar{C}_2 = -\frac{\sigma}{H} + \sqrt{\frac{\sigma^2}{H^2} + C_0^2}\,. \tag{6}$$

On substitution of the expression for P_a calculated from formulas (2)–(6) and P_m and P_i from [4] into (1), we obtain an equation consistent with the data of [5]:

$$\frac{H}{D} = \frac{1}{\sqrt{C_0}}\,. \tag{7}$$

To relate the total porosity m, the transport porosity m_p, and the porosity of the clay component m_c we use the obvious relation

$$m = m_c + m_p\,, \quad m = \text{const}\,. \tag{8}$$

The calculations carried out by the formulas obtained have shown that the porosity decreases only slightly (by less than 1%) due to the swelling of the additionally appearing smectite component of the bed D_0–D_1 (Romashkino deposit, Tatarstan). Such values cannot explain the significant changes in the character of filtration in the bed. The swelling of the clay cement can represent a serious problem for some deposits in Western Siberia, for example, the productive bed A_1^{1+2} of the Nizhnevartovsk vault (Western Siberia): the total clayiness is 25% and the concentration of the illite-smectite phases in it is 30% in the case where the content of the smectite components is 60–80%. The calculations give a value of Δm = 7–9%, which is fairly close to the characteristic values of 8–15% obtained from the analysis of the field data [6]. The results of the calculations are presented in Fig. 1.

Quite a different mechanism acts, in our view, in the above-discussed situation. The fundamental particles of 1M mica are carried by a flow until each of them is fixed at a certain point of the porous space because of its geometric inhomogeneity. A part of them, as we already know, reaches the operating wells. Nonetheless, in a certain volume of the reservoir, there is formed an additional surface of the solid phase with a high negative potential having no time to be compensated for with the processes of adsorption in desalinated stratal water. In the process of filtration in such a porous medium there arises a flow potential which causes an electroosmotic counterflow retarding the filtration. The filtration rate corrected for the electroosmosis is described by the equation [7]

$$\mathbf{V} = -\frac{k\nabla P}{\eta}\left(1 - \frac{\varepsilon_0^2\psi^2}{2\pi^2\eta K_0\nu_0 r_c^2 m}\right), \tag{9}$$

where

$$\sigma = \frac{\varepsilon_0\chi\psi}{4\pi}, \quad \frac{1}{\chi} = \sqrt{\frac{\varepsilon_0 RT}{8\pi z^2 F^2 C_0}}\ .$$

To a first approximation, the specific conductivity K_0 and the concentration of the solution C_0 may be thought of as functionally related to each other in terms of the equivalent electrical conductivity Λ_0 according to the Arrhenius formula

$$K_0 = \alpha\Lambda_0 C_0\ . \tag{10}$$

Then Eq. (9) takes the form

$$\mathbf{V} = \frac{k\nabla P}{\eta}\left(1 - \frac{a}{C_0^2}\right),$$

where

$$a = \frac{RT\sigma^2\varepsilon_0}{F^2 z^2\pi\eta\Lambda_0\nu_0 r_c^2 m}\ ,$$

and the relative decrease in the permeability can be calculated versus the degree of desalination of the stratal water $C_0/C_{\max}$, where $C_{\max}$ is the critical concentration for which (at the given parameters) the filtration is completely terminated. It is evident that the condition for this is the equality $a/C_0^2 = 1$.

More conservative estimates [7] point to the fact that the filtration decreases by at least 50% in this case. To evaluate the effect we use values of the parameters which are reasonable for the Devonian sandstones: volume clayiness 5%, content of the mica component of polytype 1M, two-thirds of which turned out to be mobilized by the flow in the form of plates of size 5 × 200 nm and be blocked in the porous medium of the reservoir, in the argillaceous fraction (including the mixed-layer phases) 30%, porosity of the rock 20%, and specific surface $10^5\ \mathrm{m}^{-1}$. The initial salinity of the stratal water was taken to be corresponding to a specific conductivity of 7 $\Omega^{-1}\cdot\mathrm{m}^{-1}$. The true values of the surface charge of mica are unknown to us and it is doubtful whether they can correctly be determined in experiments under the real conditions of a flooded oil bed. Therefore, in the calculations we used boundary values of 0.05 and 0.02 $\mathrm{C/m^2}$, the first of which approximately corresponds to the theoretical value of the charge of a mica layer and the second of which is somewhat lower than half of it. The second value is apparently more real, since the dispersion of the mica blocks must occur first of all in smectite-type interlayer spaces carrying a half-charge. We cannot completely exclude a partial compensation for the surface charge with the dipole moments of the H_2O molecules and with the hydrated cations. To evaluate the reality of the adopted value of σ we calculated the value of the surface potential necessary to completely terminate the filtration at the same prescribed parameters, using equalities (9) and (10). As has been noted, this takes place when the density of the diluted stratal water reaches a value of 1.09 $\mathrm{g/cm^3}$. The calculations give a value of $\sigma = 0.04\ \mathrm{C/m^2}$, which falls within the range under study. It is apparent that the same effect can also be obtained for a value of 0.02 $\mathrm{C/m^2}$ if the total clayiness is increased to 10% or the content of the dispersed mica is doubled. Both are entirely possible for producing levels that practically have ceased to participate in oil production.

We have developed an algorithm and a program for calculation of an unsteady two-dimensional, two-phase filtration with allowance for the secondary hydrodynamic inhomogeneity appearing in an oil bed. The algorithm is based on the following assumptions:

1. The dispersion of free mica particles is limited by the equilibrium concentration of the mica particles c_e for which the solution in the bed will be "saturated" (equilibrium at a given mineralization of the solution) and will not occur from this point on. This process becomes physically equivalent to dissolution. The value of c_e is calculated in terms of the quantity H for a known concentration of the solution from formula (7). The rate of dispersion is assumed to be infinitely high (except for special cases), so that the solution will be "saturated" instantaneously. This assumption has been introduced since the real values of the controlling kinetic constants for the process of dispersion of argillaceous mica particles are unknown. There are reliable data on only the equilibrium concentrations of the suspension formed and on the dependences of these concentrations on the mineralization of the aqueous-phase solution. This circumstance was taken into account in developing the algorithm.

2. The impurity in the bed becomes active and creates a so-called anisotropic factor of resistance to the flow due to the reverse electroosmotic flow. In this case, the resistance is high in the direction of bedding and is practically absent in the transverse direction.

3. The related and limiting water saturations are independent of the suspension concentration.

4. The concentration of the suspension is not so high as to take into account the change in the volume of water as a result of the dispersion and release of mica particles.

5. The process of dispersion does not lead to a marked change in the absolute permeability and porosity of the skeleton.

We give the differential equations describing the process of two-phase, three-component filtration:

$$L(\mathbf{V}) = \operatorname{div}(\mathbf{V}) = 0\,, \quad \mathbf{V} = -\,\omega \nabla P\,, \quad L(f\mathbf{V}) = -\,m\frac{\partial s}{\partial t}\,, \quad L(cf\mathbf{V}) = -\,m\frac{\partial sc}{\partial t} + b\,. \tag{11}$$

For b we have

$$\begin{gathered} b = qm\frac{\partial s}{\partial t}\,, \quad t^0 \le t \le \bar{t}\,, \quad \frac{\partial s}{\partial t} > 0\,; \\ b = qm\,(s - s\,(\bar{t})) + \int_{\bar{t}}^{t} L\,((c - \bar{c})\,f\mathbf{V})\,d\tau\,, \quad \bar{t} \le t \le \bar{t}^{\,0}\,, \quad \frac{\partial s}{\partial t} > 0\,; \\ b = 0\,, \quad \frac{\partial s}{\partial t} \le 0\,, \end{gathered} \tag{12}$$

where t^0 is the instant at which water reaches the point under consideration, $\bar{t}$ is the instant at which the limiting concentration c_e is attained at the point under consideration, i.e., the instant at which $c(\bar{t}) = c_e$ and $c(\bar{t} - \varepsilon_0) \le c_e$, and $\bar{t}^{\,0}$ is the instant at which the trailing edge of the impurity begins to pass through the given point; it is determined from the conditions

$$c\,(\bar{t}^{\,0}) \le c_e\,, \quad c\,(\bar{t} - \varepsilon_0) = c_e\,. \tag{13}$$

The quantity q takes into account the fraction of dispersed particles in the composition of the solid phase. The constants characterizing the process in this algorithm are μ, q, and c_e. The obtained nonlinear system of integrodifferential equations can be solved by numerical methods. We note the important points of numerical solution. First and foremost, let us note the characteristic features of the sought function c, which influence the numerical method of determination of c. The field of c is formed due to the right-hand side of the dispersion equation, i.e., the initial concentration of the impurity is equal to zero and the injection of water occurs without an impurity. This determines the special properties of the formation of the leading edge of the impurity concentration and, as a consequence, of the use of the Courant criterion for evaluation of the permissible time step of calculation. Here the leading edge of the concentration is determined by the water flow. Moreover, after a certain time or at a certain distance the leading edge can have a jump from zero

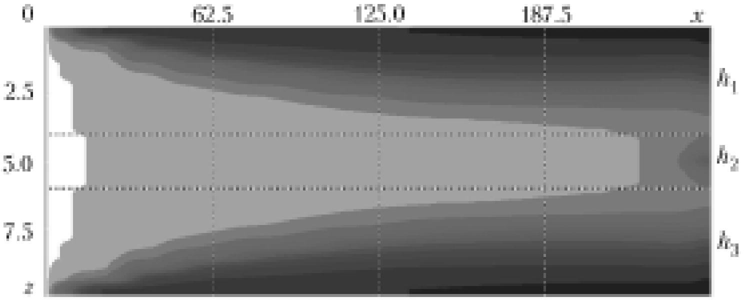

Fig. 2. Formation of the break of water through the central zone (layer h_2) of an oil bed due to the electroosmotic effect. The oil pillars are in the upper and lower parts (h_1 and h_2) of the bed. The white color denotes water, the black color denotes oil, and the tints of gray denote the two-phase zone. x and z, m.

to the limiting value of c_e. The trailing edge can be of two types: it can move over the region that still has an "insoluble" impurity or over the region that already does not have an insoluble impurity in the pores filled with water. At the trailing edge, c formally does not vanish since the concentration tends to zero asymptotically (as s tends to s^*).

Another important feature is due to the existence of the limiting concentration c_e. Not only does this complicate the form of the function b, but it also complicates the algorithm of calculation of the concentration. At the same time, the anisotropic factor of resistance has a significant influence on the process of filtration and the distribution of the water saturation in the bed, but it does not lead to a marked complication of the algorithm of solution of the problem.

An "undissolved" component (dispersion product) can be in two forms: on the one hand, its amount is determined by the quantity $(1 - s)q$, and on the hand, part of the component does not pass into solution because of the existence of the limiting concentration of the suspension of free mica particles in the pores where there is a solution with this concentration. Let us denote this part as a. Thus, the total amount of "undissolved" mica (i.e., mica transformed and retained after the dispersion in the pores with an equilibrium value of the suspension concentration) is equal to

$$Q = a + (1 - s)\, q \,. \tag{14}$$

This factor also has a significant influence on the algorithm of calculation of c.

System (11) is completely approximated by a conservative difference scheme. In this case, the equation of the total flow is implicit relative to the pressure and the values of the water saturation and the suspension concentration are taken to be equal to those for the previous layer. The water saturation in the balance equation of conservation of water is determined by an explicit scheme. In this equation, the function $f(s)$ at seminodes is determined with allowance for the direction of the flow by a scheme that is based on the fractional-linear approximation and allows for the anisotropy of the resistance factor. The suspension concentration in the equation of conservation of the impurity content is determined by an explicit scheme. In this case, the concentration in the flow terms at seminodes is determined with allowance for the direction of the flows by the "corner" scheme. If the impurity flow through the boundary of an elementary cell is denoted as $L(cf\mathbf{V})$, the algorithm for calculating the concentration in an arbitrary cell will take the form

$$r^{(1)} = s^t c^t + \frac{\tau}{m} L\,(cf\mathbf{V})\,, \quad r^{(2)} = r^{(1)} + a + (s^{t+\tau} - s^t)\, q\,, \quad r = \frac{r^{(2)}}{s^{t+\tau}}\,.$$

If $r \le c_e$, then $c^{t+\tau} = r$ and $a^{t+\tau} = 0$. If $r < c_e$, then $c^{t+\tau} = c_e$ and $a^{t+\tau} = r^{(2)} - c_e s^{t+\tau}$. Here s^t, $s^{t+\tau}$, c^t, and $c^{t+\tau}$ are the water saturation and the impurity concentration in the suspension at the instants of time t and $t + \tau$ respectively. It is evident that, in this case, the content of the sorbate component of the second kind a is determined simultaneously with the concentration. This is necessary for controlling the calculations by the balance relations. Figure 2 shows the pattern constructed by the model proposed. It is clearly seen as water breaks through the central (flashed) zone of the bed; this break is technogenic in character, i.e., the bed was initially homogeneous.

CONCLUSIONS

Our investigations have shown that the reasons for the decrease in the filtering capacity of mudded oil reservoir beds are mineralogical and not hydrodynamic in nature. It has been shown that under our conditions the swelling of argillaceous cement plays no significant role in the decrease in permeability in the case where the hydrochemical regime of a bed changes in the process of exploitation. It has been established that the processes of transformation of the structure of argillaceous minerals are determining in this case. They lead to the dispersion of aggregates and the segregation of the smectite and mica components carried by the fluid flow. In contrast to montmorillonite, which rapidly coagulates in the smallest pores of the reservoir in the vicinity of the injection wells, deteriorating their acceleration characteristics, particles of secondary micas can move in the bed to large distances. They are mechanically fixed on the inhomogeneities of the pore channels and can create, due to the high surface charge having no time to be compensated for, a reverse electroosmotic flow retarding filtration to its complete termination. The calculations made on the basis of the mathematical model developed give the pattern of the process observed under field conditions.

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NOTATION

$\Pi(H)$, disjoining pressure; H, thickness of the water film between clay particles; $P_{\rm m}$, $P_{\rm i}$, and $P_{\rm a}$, molecular, electrostatic, and adsorption components of the disjoining pressure; Γ, adsorption; C, concentration of the solution between clay particles; C_0, concentration of the solution in the pores; μ, chemical potential of the dissolved substance; σ, surface density of the charge of a clay particle; D, thickness of a clay particle; m, porosity; $m_{\rm c}$, porosity of clays; $m_{\rm p}$, transport porosity; $\mathbf{V}$, rate of filtration; k, absolute permeability; P, pressure; t, time; η, fluid viscosity; ε_0, permittivity; ψ, surface potential of a clay particle; K_0, electrical conductivity of the solution; Λ_0, equivalent electrical conductivity of the solution; R, universal gas constant; T, temperature; F, Faraday number; α, degree of dissociation; s, water saturation; s^*, residual water saturation; c, concentration of dispersed particles; $\omega = k\left(\frac{k_{\rm w}^*}{r\mu_{\rm w}} + \frac{k_{\rm o}^*}{\mu_{\rm o}}\right)$, hydraulic conductivity; $k_{\rm w}^*$ and $k_{\rm o}^*$, relative permeabilities of water and oil respectively; $\mu_{\rm w}$ and $\mu_{\rm o}$, viscosities of water and oil respectively; $r = 1 + \bar{\mu}[1 - \delta(x - x')]c$, resistance factor; $\bar{\mu}$, constant (large number); $\delta(x - x')$, function equal to unity for $x = x'$ and to zero in all the other cases; x, horizontal coordinate; $f = \frac{kk_{\rm w}^*}{r\mu_{\rm w}\omega}$, fraction of water in the flow; $c_{\rm e}$, equilibrium concentration of dispersed particles in the pores; τ, time step; ν_0, partial molar volume of water. Subscripts: a, adsorbed; c, clay; e, equilibrium; i, ionic; m, molecular; o, oil; w, water; 0, initial value; p, transport; 1, belonging to cations; 2, belonging to anions; overscribed bar, belonging to the threshold solution; (1) and (2), belonging to the computational grid for numerical solution of the problem.

REFERENCES

1. R. Kh. Muslimov, R. Z. Mukhametshin, V. N. Dolzhenkov, É. I. Suleimanov, G. A. Krinari, and E. A. Yudintsev, in: *Experience in Exploration and Exploitation of the Romashkino and Other Large Oil Deposits of the Volga-Kama Region* [in Russian], Kazan' (1998), pp. 207–211.
2. G. A. Krinari, M. G. Khramchenkov, and R. Kh. Muslimov, *Georesources*, No. 2, 30–37 (2000).
3. G. A. Krinari, M. G. Khramchenkov, and R. Z. Mukhametshin, *Geoékologiya*, No. 4, 357–364 (2001).
4. B. V. Deryagin, N. V. Churaev, and V. M. Muller, *Surface Forces* [in Russian], Moscow (1987).
5. K. Norrish, *Disc. Faraday Soc.*, **18**, 120–134 (1954).
6. A. Ya. Khavkin, *Role of the Dispersivity of the Oil–Water–Rock System in the Processes of Displacement of Oil from Porous Media* [in Russian], Moscow (1998).
7. N. V. Churaev, *Physics and Chemistry of the Processes of Mass Transfer in Porous Bodies* [in Russian], Moscow (1990).