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EFFECT OF A SURFACE-ACTIVE SUBSTANCE ON NONEQUILIBRIUM PHENOMENA IN FILTRATION OF GAS-LIQUID SYSTEMS IN THE SUBCRITICAL REGION

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The influence of a surface-active substance (SAS) on the filtration of gas–liquid systems at a pressure higher than the saturation pressure is investigated. It is shown that the SAS makes it possible to control the rheological properties of gas–liquid systems in the subcritical region.

Numerous experiments with gas–liquid systems in the subcritical region [1,2] point to the presence of nonequilibrium phenomena, which is explained by intense nucleation. The mechanism of the phenomena is still not understood well; however, gas–liquid systems have gained wide use, especially in the petroleum industry [3, 4].

Taking into account the ability of surface-active substances (SAS) to change the surface energy of a system, which leads to important consequences for the formation of a new phase, and the fact that the available experimental data have been obtained mainly for gas–liquid systems with a hydrocarbon liquid used as a solvent, we investigate in the present paper the effect of an SAS on the filtration of water–gas mixtures in the subcritical region.

Experimental investigations have been carried out on an installation [5] that consisted of the following elements: column with a porous medium, hydraulic press, PVT bomb, ultrathermostat, and measuring equipment. The main measuring unit consisted of a Sapfir 22-DI strain gage, a B5-49 power supply, an R-333 resistance box, and a recorder.

Experiments were carried out according to the following scheme:

- A water–gas mixture (methan was used) was prepared with a molar concentration equal to 0.035 in the PVT bomb at a constant temperature ($T = 303$ K).
- The saturatuion pressure P_s (3.0 MPa) was determined by the volumetric method.
- The porous medium consisting of quartz sand with penetrability 0.15 μm^2 was saturated by the water–gas mixture ($P >> P_s$) subsequent to evacuation and under continuous thermostatic control ($T = 303$ K).
- At the inlet and the outlet of the column with the porous medium the required pressure was set, and the water–gas mixture was filtered under a constant pressure drop, until a constant flow rate was achieved, after which the main filtration parameters were fixed.

All the investigations were carried out with various weight-part concentrations of the SAS in the gas–liquid system ($C = 0$–0.04%). It should be noted that we used sulfanol as the SAS, and in the experiments its concentration did not exceed the critical concentration of micellation.

ABSATZ6 = In the first series of experiments the pressure-recovery curves (PRC) were measured at the outlet of the column at a constant pressure drop of 2.7 MPa.

In order to explain the results, the obtained PRCs were replotted in new coordinates [6] ln $[(P_0 - P_\infty)/(P_t - P_\infty)]$ vs. t; here $_0$, P_t, and P_∞ are the initial, current, and the final values of the pressure. The relaxation time can be determined from the tangent of the slope angle: $t = (\tan \alpha)^{-1}$.

Results of plotting in the new coordinates are shown in Fig. 1a, $(P_0 - P_\infty)/(P_t - P_\infty) - P^*$, from which is evident that while in the case when $P < 2P_s$ (curve 1) the dependence is a broken line, in the case when $P \geq 2P_s$ (curve 2) the dependence is a straight line, i.e., a change of models takes place from an exponential one to one

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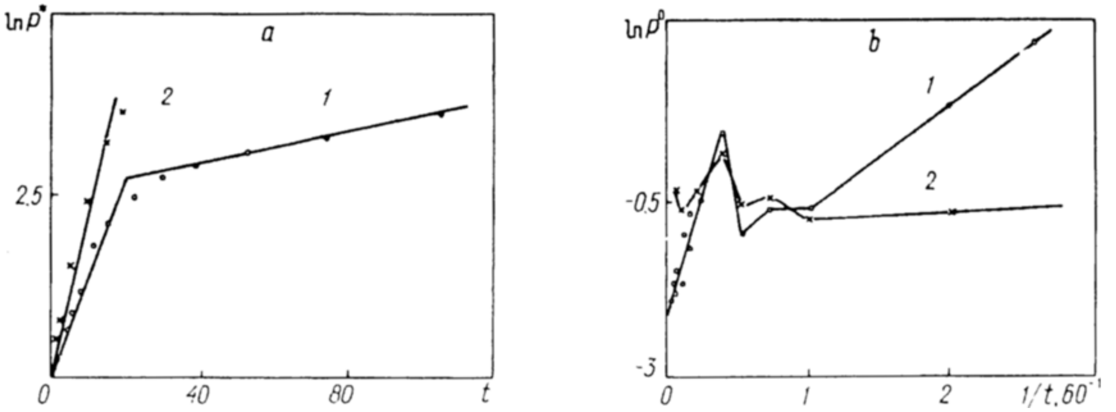


Fig. 1. PRC plotted in semilogarithmic (a) and "diffusion" (b) coordinates: 1) $P = 1.3P_s$; 2) $P > 2P_s$. t, sec.

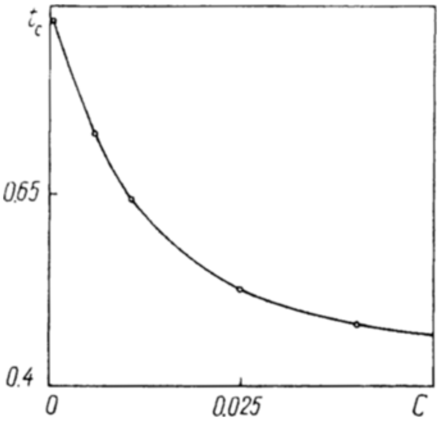


Fig. 2. Dependence of relative duration of diffusion process on SAS concentration. C, %.

described by a sum of exponentials. It should be noted that as the critical relaxation region is approached, the curves decline to the right, i.e., towards longer relaxation times.

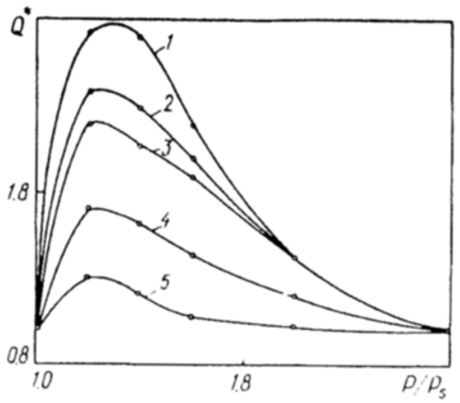
The first, shorter stage of pressure recovery is determined mainly by the compressibility of the system. For the second, longer stage, it is reasonable to assume that a concentration gradient apperars, and, as a result of duffusion, the system goes into a new equilibrium state characterized by a smaller size of gas nuclei.

In order to verify this assumption, according to [7] the PRCs were replotted in the new "diffusion" coordinates ln $(\sqrt{t}\,\partial P/\partial t)$ vs $1/t$. They are presented in Fig. 1b ($\sqrt{t}\,\partial P/\partial t = P^0$), from which it is evident that, while for an initial pressure close to the critical region ($P = 1.1P_s - 1.5P_s$), the final portion of the PRC is rectified in the specified coordinates (see Fig. 1b, curve 1), this does not take place for levels $P > 1.5P_s$ (Fig. 1b, curve 2), which substantiates our arguments.

Analysis of the PRC measured with addition of the SAS has shown that the relaxation time decreases with increasing SAS concentration for a constant initial pressure level at the outlet of the column. Thus, e.g., for $P = 1.3\ P_s$ at $C = 0\%$ $\tau = 5$ sec, while at $C = 0.04\%$ $\tau = 1.2$ sec.

Figure 2 shows the dependence of the relative duration of the diffusion process t_c (t_c is the ratio of the duration of the diffusion process to the total time of pressure recovery) on the SAS concentration in the solution at $P = 1.3P_s$, from which is evident that the duration of the diffusional component decreases by almost half with addition of the SAS, reaching 0.49 at $C = 0.04\%$.

The results obtained can be explained by the following mechanism. As is known, in the presence of a developed solid surface (in the particular case, the volume of the gas–liquid system in the porous medium is one-third the volume of the solid component, and the mean capillary diameter does not exceed 10^{-5} m), only



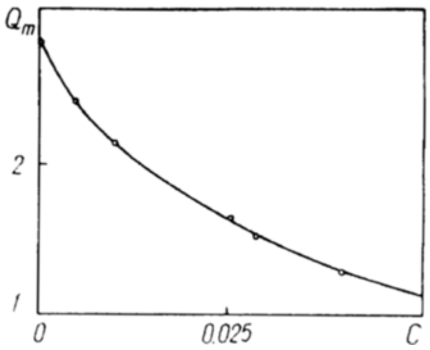


Fig. 3. Dependence of relative flow rate of gas–liquid system on relative pressure level: 1) $C = 0$; 2) 0.005; 3) 0.01; 4) 0.025; 5) 0.04.

Fig. 4. Dependence of maximum relative flow rate of gas–liquid system on SAS concentration.

heterogeneous nucleation takes place [8, 9]. With an increase in the wettability of the solid surface of the capillaries, which takes place with addition of the SAS due to its adsorption [10], the volume of the gas phase in the form of nuclei decreases substantially (as is shown in [8], for an edge wetting angle $\theta \to 0$, the nuclei will be virtually two-dimensional). This, with all other factors being the same, leads to a decrease in the concentration gradient that emerges with an increase in pressure and, correspondingly, to a decrease in the duration of the diffusional process as well as in the relaxation time.

In the second series of the experiments, for a constant pressure drop of 2.7 MPa, we determined the dependence of the flow rate of the gas–liquid system on the pressure level at the outlet of the column.

The experimentally measured dependence of the relative flow rate Q^* on the pressure level P/P_S ($Q^* = Q_t/Q_\infty$, where Q_t is the flow rate at a certain level of the pressure at the outlet, Q_∞ is the flow rate at $P > 2P_S$) is shown in Fig. 3 (curves 1 to 5 correspond to concentrations $C = 0$, 0.005, 0.01, 0.025, and 0.04%), from which is evident that for pressures at the outlet of the column close to the saturation pressure, a substantial (2.5–3 times) increase in the flow rate takes place. It should be noted that the flow rate of the gas–liquid system reduced to the pressure at the exit from the porous medium varies similarly to the flow rate of the liquid, which is substantiated by the practical constancy of the gas flow rate in the course of an experiment.

Figure 4 shows the dependence of the maximum relative flow rate Q_m on the SAS concentration. It is evident that the maximum value of the relative flow rate decreases by more than half with an increase in the SAS content, reaching a value of 1.3 at an SAS concentration of 0.04%.

In [1], the increase in the liquid flow rate in the vicinity of the saturation pressure is explained by a decrease in the absolute viscosity of the system observed upon approaching P_S. However, the decrease in the viscosity is negligible. According to data from [1], it does not exceed 10–15%, while the flow rate increases 2.5–3 times.

The results obtained can be explained within the framework of the above-discussed mechanism, according to which the volume of the gas phase in the form of nuclei decreases, which leads to a decrease in the thickness of the boundary layer (or, which is the same, the viscosity of the gas–liquid system with the SAS additive increases within the same boundary layer).

In order to substantiate this assumption, we consider the motion of the gas–liquid system in a single capillary and make use of the results of [11, 12], where the following function was obtained based on the Pontryagin maximum principle:

$$h(c) = \rho(c)/2\mu(c) + (A/(1 - r^2))\,c\,,$$

where $\rho(c)$ and $\mu(c)$ are the density and viscosity dependences on the gas concentration, respectively. We seek a concentration distribution over the capillary radius $c(r)$ such that $h(c)$ and, accordingly, the flow rate of the liquid are maximal. According to [11], A is a constant, and it should be noted that $A \in [0; \infty)$, $r \in [0; 1]$, and $c \in [0; 1]$.

The known expression can be adopted for the dependence of density on concentration: $\rho(c) = 1 - c$. For the concentrational dependence of the viscosity we adopt, according to [13], the following expression: $\mu(c) = 1 - \gamma c$ $(1/3 < \gamma < 1)$, and for relatively low mean concentrations F $(0 < F < 1/6\gamma)$ we obtain:

$$c(r) = \begin{cases} 0, & 0 \le r < \sqrt{\left(1 - \dfrac{2A}{1-\gamma}\right)}, \\ \dfrac{1}{\gamma} - \dfrac{1}{\gamma}\sqrt{\left(\dfrac{(1-\gamma)(1-r^2)}{2A}\right)}, & \sqrt{\left(1 - \dfrac{2A}{1-\gamma}\right)} < r < 1. \end{cases} \tag{1}$$

The mean concentration of the gas is determined, according to [11], from the relationship:

$$F = \int_0^1 c(r)\, r dr = \frac{A}{3\gamma(1-\gamma)},$$

from which it is obvious that the specified concentration distribution over the cross-section (1) will occur when $A \in (0; 1 - \gamma/2)$ and $F \in (0; 1/6\gamma)$.

We should note that at relatively high gas concentrations, when two-phase flow takes place, formation of a boundary gas layer is possible, as is shown in [11, 12].

In the case under investigation, motion of the gas as a separate phase is not realized, since this is possible only in the supercritical region.

Thus, maximization of the flow rate takes place upon layering of the gas–liquid system, when pure liquid flows in the center and liquid with gas nuclei (i.e., a system with a lower viscosity than in the center of the stream) flows in the boundary regions. It is evident that a relatively thin boundary layer saturated with heterogeneous gas nuclei will have much lower viscosity than a pure liquid; at the same time, the absolute viscosity of the entire system in the presence of gas nuclei decreases, according to [1], by only 10-15%.

It should be noted that upon addition of the SAS with the same concentration of dissolved gas, the viscosity of the gas–liquid system will be lower, due to the circumstances discussed, which manifests itself in a decrease in the coefficient γ. This, according to (1), leads to an increase in r and, correspondingly, to reduction of the boundary layer.

Let us consider how layering affects the flow rate of the system. We assume that the pure liquid moves in the center of the stream, which has viscosity μ_1, while the gas–liquid system, which for simplicity can be represented as an incompressible fluid with viscosity μ_2, $\mu_2 < \mu_1$, moves near the wall. Then the motion velocity in the cylindrical capillary is determined from the well-known formula

$$v_i = -\frac{\Delta P}{4 l \mu_i} r^2 + a_i \ln r + b_i, \tag{2}$$

where $\Delta P/l$ is the pressure gradient, and μ_i are the viscosities of the fluids (the parameters of the fluids flowing in the center and in the boundary region are denoted by indices 1 and 2). Let us use the following boundary conditions for determination of the constant coefficients a_i and b_i:

$$r = R, \quad v_2 = 0, \quad r = R_0, \quad \mu_2 \frac{dv_2}{dr} = \mu_1 \frac{dv_1}{dr}, \quad v_1 = v_2, \quad r = 0, \quad \frac{dv_1}{dr} = 0.$$

Substituting the conditions into (2) and rearranging the expression such as to yield the total flow rate, we finally obtain

$$Q = Q_1 + Q_2 = \frac{\pi \Delta P R^4}{8\mu_1 l} [\mu_0 + (1 - \delta)^4 (1 - \mu_0)], \tag{3}$$

where $\delta = 1 - R_0/R$, $\mu_0 = \mu_1/\mu_2$.

It is evident from Eq. (2) that the total flow rate increases with the growth of the less-viscous boundary layer, and it should be noted that the higher μ_0 for a constant thickness of the boundary layer, the higher the flow rate. Already at $\mu_0 = 3$ and $\delta = 0.2$ the flow rate is two times greater than that according to Poiseuille's law.

The estimates presented can serve as a qualitative explanation of the experimental results obtained.

The further drop in the flow rate takes place due to the fact that dimensions of the nuclei (and at $P \leq P_s$ already gas bubbles) become comparable with those of the pores, and this leads to a substantial increase in the filtration resistance [14, 15].

Analysis of the results obtained makes it possible to draw two important conclusions. Firstly, a greater increase in the flow rate of the fluid should be expected in hydrophobic porous media. Secondly, the penetrability of the porous medium will obviously affect the pressure level at which the maximum flow rate of the fluid is reached.

Thus, the investigations carried out have shown that SAS additives make it possible to control the rheological properties of gas–liquid systems in the subcritical region, which can be applied in technological processes, particularly, in the petroleum industry, where these systems are widely used.

NOTATION

P, pressure at the outlet; P_s, saturation pressure; Q, flow rate; τ, relaxation time; ρ, density; μ, viscosity; C, SAS weight concentration; c, gas concentration; F, mean gas concentration; l, length of capillary; R, radius of capillary; R_0, radius of fluid interface; δ, thickness of boundary layer.

REFERENCES

1. A. A. Bolotov, A. Kh. Mirzadzhanzade, and I. N. Nesterov, Izv. Akad. Nauk SSSR, Ser. Mekh. Zhidk. Gaza, No. 5, 172 (1988).
2. G. Kh. Melikhov, Investigation of the Effect of Nonequilibrium Conditions on Hydrodynamic Parameters of Gas–Liquid Systems at a Pressure Higher than the Pressure of Saturation, Candidate's Dissertation, Baku (1990).
3. A. Kh. Mirzadzhanzade, I. M. Ametov, and A. G.Kovalev, Physics of Oil and Gas Reservoirs [in Russian], Moscow (1992).
4. B. A. Suleimanov, E. M. Abbasov, and A. D. Ismailov, in: Abstracts of the V All-Union Scientific and Technical Conference Rock Fracture in Well-Drilling, Vol. 3, Ufa (1990), pp. 36-38.
5. B. A. Suleimanov and A. D. Ismailov, in: Collection of Scientific Papers of the M. Azizbekov Azerbidzhan Institute of Petroleum Chemistry, Baku (1991), pp. 32-35.
6. A. Kh. Mirzadzhanzade, A. K. Karaev, and S. A. Shirinzade, Hydraulics in Drilling and Cementing Oil and Gas Wells [in Russian], Moscow (1977).
7. N. B. Shtibina, Determination of Sorption Ability of Rocks from Results of Hydrodynamic Investigations, Candidate's Dissertation, Baku (1988).
8. A. I. Rusanov, Phase Equilibria and Surface Phenomena [in Russian], Leningrad (1967).
9. M. Volmer, Kinetics of New-Phase Formation [Russian translation], Moscow (1986).
10. N. V. Churaev, Physical Chemistry of Mass Transfer Processes in Porous Bodies [in Russian], Moscow (1990).
11. A. A. Abbasov, Investigation of Certain Inverse Problems of Heat and Mass Transfer in a Reservoir–Well System, Candidate's Dissertation, Baku (1990).
12. U. G. Abdullaev, in: Abstracts of the XIII School-Workshop on Problems of Pipeline Transport, Ufa (1990), p. 42.
13. R. M. Sattarov and P. Ya. Farzane, Inzh.-Fiz. Zh., 52, 765 (1987).
14. P. M. Ogibalov and A. Kh. Mirzadzhanzade, Mechanics of Physical Processes [in Russian], Moscow (1976).
15. N. E. Esipova, Z. M. Zorin, and N. V. Churaev, Kolloidn. Zh., No. 1, 9 (1981).