RHEOLOGICAL PROPERTIES OF FLOWS OF A DENSE GAS AND LIQUIDS IN NARROW SLOT-LIKE PORES

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The concentration dependences of the coefficients of shear viscosity of molecules in a wide range of pore fillings (from a rarefied gas to a liquid state) for different potentials of molecule–surface interactions have been theoretically investigated with the use of the lattice-gas model allowing for the volume occupied by atoms and the interaction between the atoms in the quasichemical approximation. This model allows one to find the self-consistent equilibrium characteristics of a vapor-liquid system and the coefficients of shear viscosity of molecules with the use of a single set of energy parameters. Molecular interpretation of the coefficient of sliding friction of the fluid near the pore walls is discussed.

It is very important to analyze the dynamics of flows of gas and liquid phases in narrow pores when processes of cleaning, wetting, impregnation, and drying of disperse systems are investigated [1–6]. In many practical situations, the density of the fluid in the pores can change over a wide range and cause capillary condensation.

The upper limit of the width of pores qualified as "narrow" pores is ~10 nm for the nonspecific short-range Lennard-Jones potential of molecular interaction [6–12], which corresponds to 25–30 single layers [7–9]. This value has been obtained from an analysis of the conditions under which the surface potential influences the conditions of capillary condensation (for long-range potentials it increases with increase in the interaction-potential radius).

In narrow pores, the potential of surface forces substantially influences the mobility of molecules and their distribution over the pore cross section. In them, all the transport characteristics of the adsorbate differ from those in the volumes of the vapor and liquid phases. Among the most important dynamic characteristics is the coefficient of shear viscosity. It should also be noted that the use of the Navier–Stokes equation in narrow pores is not correct; in this case, it is necessary to use the kinetic equations of transfer processes in condensed phases. A new molecular approach to description of flows in narrow pores in the case where the density and the temperature of fluids change over wide ranges has been proposed in [10, 11]. This method is based on the use of the simplest molecular model of condensed phases — the lattice-gas model that allows for the volume occupied by molecules and their interaction [12, 13] and can be used over wide ranges of fluid concentrations (from the gaseous state to the liquid state) and temperatures, including the critical region, which makes it possible to consider the dynamics of fluid flow under capillary-condensation conditions. The closed expressions for the transport equations and coefficients were constructed in the quasichemical approximation allowing for the intermolecular interactions and preserving the effects of short-range correlation. The results of the calculation of the equilibrium characteristics of porous systems on the basis of the lattice-gas model are in good agreement with the data obtained with the use of practically exact Monte Carlo methods and methods of molecular dynamics [14–16].

Microhydrodynamic equations [10, 11] are written for each cell whose dimension is of the order of the dimension of a molecule of the region considered. They have a discrete form that coincides with the "usual" difference form of representation of the Navier–Stokes differential equations. The obtained structure of the equations [10, 11] makes it possible to use the available methods of solving hydrodynamic problems; in this case, the transport coefficients are dependent on the local values of the density and the temperature.

In the present work, we have theoretically investigated the concentration dependences of the coefficients of shear viscosity of molecules over a wide range of pore fillings (from a rarefied gas to a liquid state) for different po-
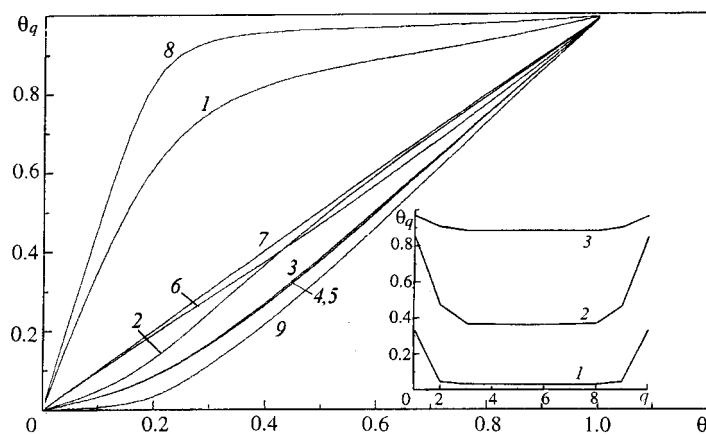

Fig. 1. Local layer fillings θ_q [1) $q = 1$, 2) 2, 3) 3, 4) 4, 5) 5, 6) 1, 7) 5, 8) 1, and 9) 5] of a slot-like pore of width 10 single layers with argon atoms at $T = 273$ K versus degree of filling of the pore with the adsorbate θ; 1–5 correspond to the system argon–carbon with $Q_1 = 9.24\varepsilon$, 6 and 7 correspond to the wall with the nonattractive potential $Q_1 = 0$, 8, and 9 corresponds to the wall with the strong attraction $Q = 16.5\varepsilon$. In the inset, the concentration profiles of argon atoms for $Q_1/\varepsilon = 9.24$ [1) $\theta = 0.095$, 2) 0.485, and 3) 0.905] are presented.

tentials of the molecule–wall of a slot-like pore interaction. Molecular interpretation of the expression for the coefficient of sliding friction near the pore wall is discussed.

The initial data in the approach of [10, 11] are the potentials of interaction between the adsorbate molecules and between the adsorbate and the adsorbent walls. The potential of interaction of the adsorbate with the homogeneous walls of a slot-like pore of width H single layers is represented as a set of discrete quantities Q_q. The binding energy for the argon atom in the layer q was calculated as $Q_q = U(q) + U(H - q + 1)$ and $U(q) = \varepsilon_a/q^3$ corresponds to the Mie-type potential [3–9]. The interaction between the argon atoms was modeled by the Lennard-Jones potential. It was assumed in the calculations that $Q_1 = Q_H = 9.24\varepsilon$, $Q_2 = Q_{H-1} = Q_1/8$, and the other $Q_q = 0$, which corresponds to the energetics of the argon atoms in a carbon pair at $\varepsilon = 1.0$ kJ/mole [17]. From the voluminous data [18, 19] on the concentration dependence of the compressibility coefficient, the quantity ε was determined as $\varepsilon = \varepsilon_0(1 - d_e\theta)(1 + u_1 T)$, where $\varepsilon_0 = 153.5$ cal·mole^{-1} and $d_e = 0.477$ [20, 21]. The second cofactor of the function allows for the contributions of the triplet interactions between the nearest neighboring molecules [12] which modify the pair interaction potential. The parameter u_1 (−0.002) allows for the temperature dependence of ε. Below we will use both variants for ε.

The procedure of calculation of the equilibrium concentration profile of molecules in the cross section of a pore (i.e., the distribution of the local fillings of the sites Q_q with an energy of the adsorbate–adsorbent interaction Q_q, $1 \le q \le t$, $t = H/2$ and $t = (H + 1)/2$ for even and odd H, respectively) and the adsorption isotherms in slot-like pores within the framework of the lattice-gas model has been considered in detail in [8, 14–16]; therefore, we will not discuss it here. Knowing the equilibrium distributions of molecules over the cross section of a pore, we can calculate the probabilities of jumps of molecules between different sites (which are dependent on their thermal velocities) and the coefficients of self-diffusion and viscosity [22, 23]. We restrict ourselves to consideration of the nearest neighbors z_q ($z_1 = 5$ and $z_{q>1} = 6$).

Figure 1 shows the concentration profiles of argon θ_q in a slot-like carbon pore of width $H = 10$ single layers (curves 1–5, q corresponds to the number of a single layer — counting is carried out from the pore wall) and θ_1 and θ_5 for systems with $Q_1/\varepsilon = 0$ and 16.5 (the second variant of ε). On the lower right, three concentration profiles of argon atoms at a total filling of the pore of 0.095, 0.485, and 0.905, respectively, are presented. Curves 1 and 8 show that the filling of the surface single layer differs significantly from that of other layers in the case of a strong attraction of the adsorbate to the pore walls. For the system argon–carbon the filling of the second single layer (curve 2) also differs markedly from the filling of the first layer, while the fillings of the third, fourth, and fifth layers are practically the same (curves 4 and 5 coincide). In the absence of the attraction of the wall, the surface layer (curve 6) is
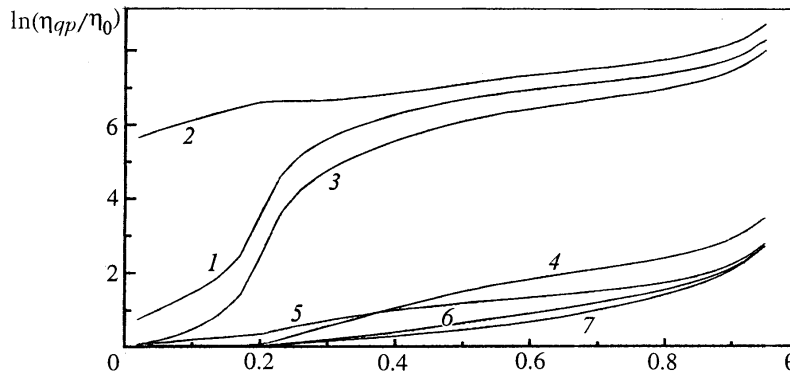
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Fig. 2. Normalized concentration dependences of the local coefficients of shear viscosity of argon atoms η_{qp} in a carbon pore of width 10 single layers (the first variant of ε and $\alpha_{11} = 0.1$): 1) $qp = 11$, 2) 12, 3) 21, 4) 22, 5) 23, 6) 33, and 7) 55.

filled to a lesser degree than the central layers (curve 7) although the differences between these curves are small. The anisotropy of the concentration profiles of the adsorbate significantly influences the concentration dependences of the viscosity coefficients.

The shear viscosity was calculated on the basis of the modified model [10] that makes it possible to extend the Eyring model [24] to the entire range of densities of the fluid. The initial Eyring model in which the structure of the liquid phase is assumed to be cellular (without vacancies) correctly describes the dependence of the shear viscosity of the liquid on the temperature, but it cannot be applied to rarefied fluids. The expression for the coefficient of viscosity of the gas η_{qp}, corresponding to the shift of the fluid in the cell p relative to the cell q, has the form (we restrict our consideration to the case of jumps to the nearest neighboring cells)

$$\eta_{qp} = \eta_0 \theta_q \exp{(\beta E_{qp})}/V_{qp}, \tag{1}$$

where $\eta_0 = (mkT/\pi)^{1/2}/(\pi\sigma^2)$ and $\beta = (kT)^{-1}$.

We have $E_{qp} \neq 0$ near the walls possessing adsorption potential and $E_{qp} = 0$ for cells positioned at a large distance from the pore wall; $V_{qp} = V_{qp}(\varepsilon, \varepsilon^*)$ is the concentration component of the velocity of the jump of a molecule from the cell q to the cell p (this quantity is described in detail in [10–12]) and $\theta_q/V_{qp} \to 1$ at $\theta_q \to 0$. It depends on the interactions between the neighboring molecules in the ground ε and transient ε^* states. The parameter $\varepsilon^* = \alpha\varepsilon$ was determined on the basis of the dependence of the thermal conductivity coefficient on the pressure in the bulk phase ($\alpha = -0.25 + 1.05\theta$). The indicated parameters reflect the properties of dense gaseous argon [18, 19] outside the rarefied-gas region. For the first variant of the parameter ε [17], the value $\alpha = 0.5$ was considered as the characteristic ratio corresponding to the experimental data in different surface processes [12].

At low densities, the viscosity is linearly dependent on the pore filling, as in the case of an ideal gas. As θ increases, the ratio between the parameters ε and ε^* begins to influence the behavior of $\eta(\theta)$. At low densities, the viscosity η is a function of the quantity $T^{1/2}$ and at high densities there takes place an exponential dependence of the viscosity on the temperature, as in the traditional Eyring model [24]. The volume viscosities of a number of gases (Ar, He, H_2, N_2, NH_3, CO_2), calculated in [20, 21] from (1), are in good agreement with experimental data [18, 19].

The concentration dependences of the local coefficients of viscosity for a slot-like pore of width 10 single layers are shown in Fig. 2. These coefficients characterize the deceleration of a flow (momentum dissipation) moving through the pore; their numerical values strongly depend on the direction of the local flow and on the distance from the pore wall. The system argon–carbon is characterized by a fairly large attraction of the walls. The position and behavior of curves 1–3 in the near-wall region is determined by the quantities Q_1 and E_{11} ($\alpha_{11} = E_{11}/Q_1$). In the central part of the pore (curves 4–7), the viscosity increases with filling of the pore. It changes from the values corresponding to the gas phase to the values corresponding to the liquid phase (the concentration dependences were normalized to the corresponding values of η_0 for the bulk phase at $\theta = 0$ and $Q_1 = 0$). Figure 3 shows the profiles of the coefficients of shear viscosity of argon atoms in a pore of width $H = 10$ single layers, corresponding to the profiles of the degrees
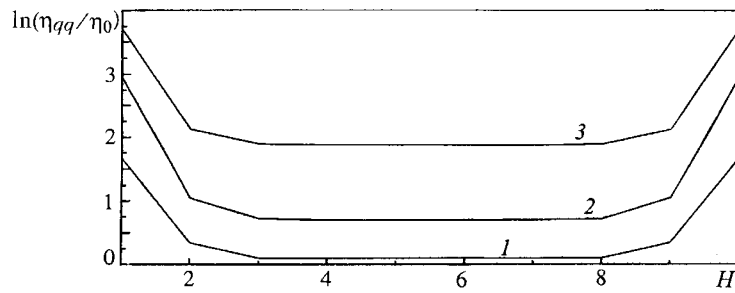
Fig. 3. Concentration profiles of the coefficient of shear viscosity of argon atoms η_{qq} ($1 \le q \le 10$) in a carbon pore of width 10 single layers (the second variant of ε and $\alpha_{11} = 0.33$): 1) $\theta = 0.095$, 2) 0.485, and 3) 0.905.
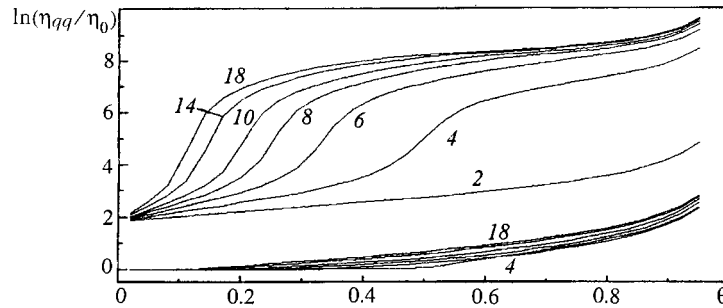

Fig. 4. Concentration dependences of the viscosity coefficient of argon in the surface layer η_{11} (the upper group of curves) and in the central layer η_{55} (the lower group of curves) for different pore widths. The numbers of the curves correspond to the pore width expressed as the number of single layers (the first variant of ε and $\alpha_{11} = 0.33$).

of filling, presented in the inset (see Fig. 1). The coefficients η_{qq} decrease as the distance between the layer and the pore wall increases and they increase monotonically with increase in the degree of filling of the pore. The influence of the pore width on the viscosity in the surface layer and in the layer found at its center is seen in Fig. 4. The shear viscosity of the adsorbate in the central layer depends only slightly on the pore width but its influence cannot be completely ignored. As the pore width H increases (at a fixed θ), the curves of the coefficient of viscosity in the surface layer η_{11} are positioned one above the other since the degree of filling of the surface layer θ_1 increases successively under these conditions. The results obtained within the framework of the lattice-gas model considered are in good agreement with the analogous results for the profiles of the concentration and the shear viscosity in slot-like pores of width 4 and 18 single layers [25], obtained by the method of nonequilibrium molecular dynamics and on the basis of the continual kinetic theory.

Figure 5 presents the concentration dependences of the coefficient of shear viscosity for different materials of pore walls. The type of the material influences first of all the value of the parameter Q_1. Its increase leads to an increase in the viscosity of the surface layer, while at $\theta = $ const the viscosity of the layer at the center of the pore decreases because of the decrease in the concentration of atoms (Fig. 1). The range of variation of the value of η_{11} is much larger than that of η_{55}. The nonmonotonicity of η_{11} at high values of θ is explained by the character of change in $\varepsilon^*(\theta)$. However, despite the large difference between the two variants of ε, the general regularities of the dependences $\eta_{qp}(\theta)$ are retained.

In [26, 27], two-dimensional flows of an argon fluid in narrow pores have been calculated on the basis of the equations from [10, 11]. The dynamics of establishment of the stationary state [26] and of interaction of regions with the gaseous and liquid phases [27] after the pulse disturbance was investigated. Figure 6 shows the distribution of the density of argon atoms in the cross section of a slot-like carbon pore of $H = 10$ at temperatures of 383.3 and 100.8 K for a dense gas at $\theta = 0.15$ and for a "rarefied" liquid phase at $\theta = 0.9$ (two upper fields) and the corresponding distributions of the velocity of the flow along the pore axis (two lower fields).
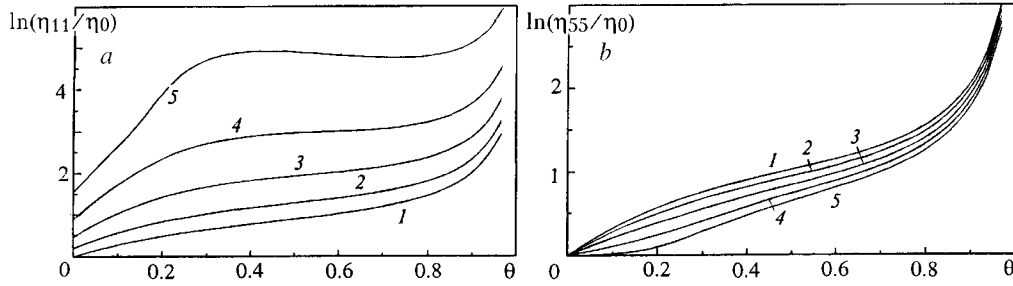
Fig. 5. Normalized concentration dependences of the local coefficients of shear viscosity of argon atoms in the surface layer η_{11} (a) and in the central layer η_{55} (b) in pores with different walls at $T = 273$ K, $H = 10$ single layers (the second variant of ε and $\alpha_{11} = 0.33$): 1) $Q_1/\varepsilon = 0$, 2) 2, 3) 5, 4) 9.24, and 5) 16.5.

At high temperatures, the gas atoms are practically uniformly distributed over the width of a pore in the central part, and their density is markedly increased on its walls. At low temperatures, a strongly anisotropic distribution of atoms over the normal to the surfaces of the walls is realized. The first layer is filled practically completely, and the gas in the central part of the pore is very rarefied (the distributions are symmetric relative to the center because of the identity of both walls of the pore). For the liquid phase, the local concentrations at the temperatures considered are comparable. The differences in conditions change the form of the concentration profiles: at a high temperature the concentration on the wall is lower and the concentration in the bulk part is higher than at a low temperature. The density distributions presented are typical. They are realized under equilibrium conditions (Fig. 1) and are retained at weak disturbances arising in flows with velocities of up to $u \sim 0.1$–1 m/sec.

In all the cases, the velocities near the pore wall are practically equal to zero, which is a result of the self-consistent calculation of the molecular characteristics of the fluid and not a postulated value, as is usually assumed in hydrodynamics when the boundary conditions are set [28] (see below). For the gas phase, the velocities change weakly (on the logarithmic scale) across the width of a pore in the temperature range (practically from 100 to 400 K). It is evident that the velocities are higher at its center than near the walls. A nearly parabolic curve is observed for the liquid phase at high temperatures, i.e., a viscous Poiseuille flow is realized. At low temperatures, the velocity distribution changes sharply. The region of a "frozen" fluid extends to three layers from each wall, and a high velocity of flow, as in the case of high temperatures, is retained at the center of the pore.

The molecular approach [10, 11] to calculation of the coefficient of shear viscosity allows us to give a molecular interpretation of the so-called "coefficient of sliding friction β_1" used in the calculation of the velocity profiles of fluids near the surfaces [29, 30]. The coefficient β_1 is determined as the ratio between the tangential force per unit surface and the *relative velocity* of the flow near the wall [29, 30]. This coefficient is present in the boundary condition on any solid wall:

$$\beta_1 u = -\eta \left.\frac{\partial u}{\partial r}\right|_{r=R}. \tag{2}$$

The expression $\eta/\beta_1 = \lambda$ has the dimensions of length. The cofactor λ follows from the dimensional relation since, unlike β_1, the coefficient of shear viscosity η represents the ratio between the tangential force per unit surface and the *velocity gradient* in the direction perpendicular to the direction of the flow [30].

As β_1 increases, the velocity

$$u = -\eta \left.\frac{\partial u}{\partial r}\right|_{r=R} \bigg/ \beta_1$$

on the wall of the channel vanishes, which corresponds to the traditional boundary condition of continuum mechanics [28]. However, this condition should be related to the molecular notions of the surface mobility of molecules and the
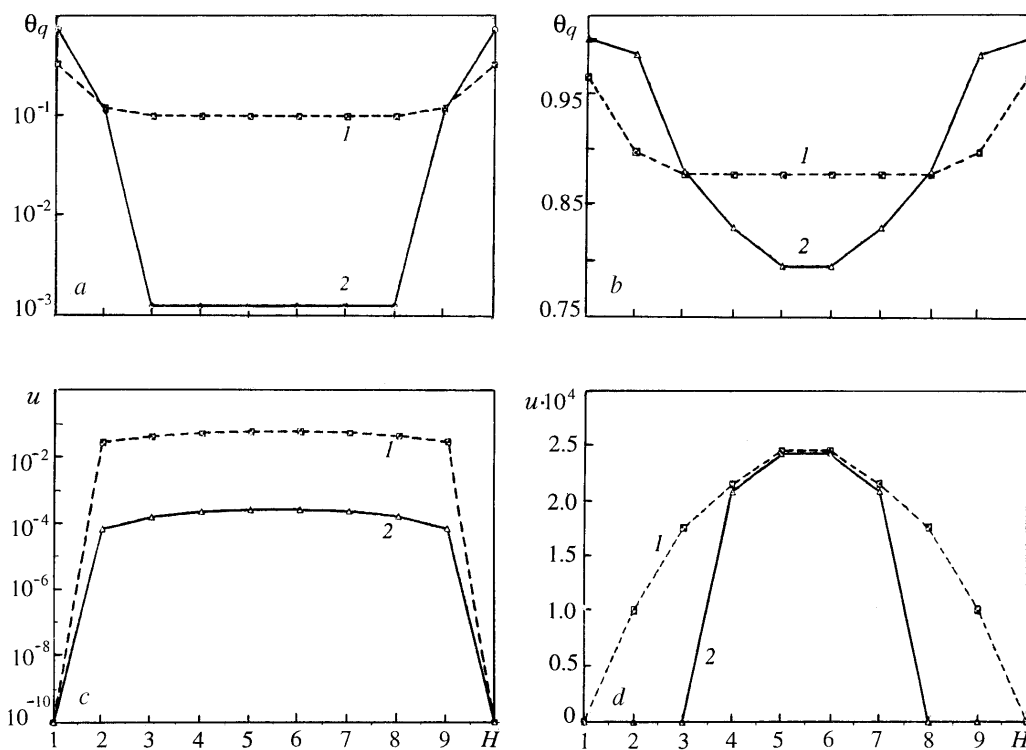
Fig. 6. Concentration profiles of argon atoms (a, b) and their velocities along the pore axis (c, d) in a carbon pair of width 10 single layers (the first variant of ε and $\alpha_{11} = 0.33$) at the mean density of argon $\theta = 0.15$ (a, c) and $\theta = 0.9$ (b, d) for the temperatures $T = 393.3$ K (1) and 100.8 K (2). u, m/sec.

surface flows, which were experimentally verified [2, 5]. The calculations of the contributions of the "surface" and "volume" transport of molecules to the total flow of labeled molecules along the pore axis (in the absence of a hydrodynamic flow), considered in [22, 23], point to the importance of the role of the surface mobility of molecules in their thermal motion characterized by the self-diffusion coefficient. The surface flow of molecules is predominant in the case of strong attraction of the walls at small densities of pore fillings and remains comparable to the volume transport of molecules nearly to the complete filling of the surface single layer. At a repulsive potential of the walls, the role of the surface transport of molecules increases with increase in the degree of filling of the pore. Of importance is the energy of activation of the surface migration of molecules E_{11}. The lower this energy, the larger the contributions of the surface flow of molecules. The effects of sliding were considered earlier only for rarefied gases. Unlike the determining role of the mirror reflection of molecules from the wall, for rarefied gases in dense fluids the effect of sliding is due to the surface mobility of molecules.

If the character of change in the viscosity coefficient of the surface layer is considered in accordance with (1), it follows from (2) that

$$\beta_1 = \eta_{11}/\lambda, \tag{3}$$

where the subscripts 11 correspond to the near-surface single layer. As a result, for the velocity of the flow in the near-surface layer we have

$$u = -\frac{\lambda\eta_{tt}}{\eta_{11}}\frac{\partial u}{\partial r}\bigg|_{r=R}$$

where the subscripts 11 correspond to the central part of a narrow pore or to the volume viscosity for broad channels. In the case of a strong adsorbate–adsorbent attraction or a decrease in the temperature, according to (1), the ratio

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η_{11}/η_{tt} increases sharply, which leads to an increase in the coefficient β_1 and a decrease in the velocity of the flow near the wall. Formula (3) clearly demonstrates the relation between the concentration dependences (see Figs. 2–5) and the results of calculation of the velocities (see Fig. 6).

CONCLUSIONS

1. The calculations carried out for slot-like pores show that the coefficients of shear viscosity of molecules in strong adsorption fields are anisotropic in character and substantially dependent on the distance from the fluid region considered to the wall pore and on the direction of the momentum transfer. They experience the largest changes near the pore walls. At the center of a pore, the values of η_{tt} depend on the contribution of the wall potential and on the total concentration of molecules.

2. It is shown that the traditional assumptions [2–4] of the constancy of the dynamic characteristics of molecules in narrow pores are incorrect. The experimental data must be analyzed with regard for the fairly strong concentration dependence of the dynamic characteristics of the adsorbate in narrow pores, which is due to the influence of the potential of the pore walls and the intermolecular interaction.

3. Molecular interpretation of the coefficient of shear viscosity is given. For dense fluids, the effect of sliding of a flow is due to the surface migration of molecules along the solid surfaces.

4. The molecular approach allows one to describe various flows of a fluid containing liquid and/or gaseous phases in complex porous systems, including pores with a variable cross section and the cases of coalescence of several pores into one pore or, on the contrary, separation of a pore into several smaller pores.

NOTATION

d_e, ratio between the contribution of triplet interactions and the contribution of pair interactions of the nearest neighbors; E_{qp}, activation energy of the jump of a molecule from the site q to the site p, kJ/mole; H, width of a slot-like pore, number of single layers; k, Boltzmann constant; m, mass of the solid sphere of a molecule; q, number of the pore layer in which the site considered is found; p, number of the cell neighboring the cell q; Q_q, energy of bond of a molecule found at the site of the layer q (or q-type layer) with the pore walls, kJ/mole; R, characteristic dimension (radius) of the channel, m; r, distance from the wall (channel) of the pore, m; t, number of different-type sites of the system; T, temperature, K; $U(q)$, potential of interaction of a molecule of the layer q with the pore wall, kJ/mole; u, velocity of the flow, m/sec; u_1, parameter of the temperature dependence ε; V_{qp}, concentration component of the velocity of jump of a molecule from the cell q to the cell p; z_q, number of nearest neighbors in the layer q; α and α_{11}, dimensionless parameters of $\varepsilon^*/\varepsilon$ and E_{11}/Q_1; β, reciprocal of the thermal energy $(kT)^{-1}$, mole/kJ; β_1, coefficient of sliding friction, kg/(m^2·sec); ε and ε^*, parameters of the lateral interaction of a molecule in the ground and transient states, kJ/mole; ε_a, parameter of the adsorbate–adsorbent potential; λ, dimension of a cell and width of a single layer ($\sim 1.12\delta$), m; η, viscosity coefficient at a large distance from the wall; η_0, viscosity coefficient of an ideal rarefied gas; η_{qp}, coefficient of shear viscosity, kg/(m·sec); δ, diameter of the solid sphere of a molecule; θ, degree of filling of a pore, dimensionless quantity, $0 \le \theta \le 1$; θ_q, degree of filling of the single layer q. Subscripts: a, adsorbate (adsorbent); 0, rarefied gas with a low density; e, external.

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