STRATIFICATION OF AN EQUIDENSE SUSPENSION OF BROWNIAN PARTICLES IN POISEUILLE FLOWS

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Nonuniform concentration profiles are formed in channels, and this nonuniformity has a significant effect on the observed hydraulic properties of a flow [1]. For suspensions with phases having identical densities, the initial reason for such structuring has to do with the effect of transverse inertial buoyancy on rotating particles in the flow. Depending on the direction of the relative velocity of the phases, this buoyancy helps particles migrate either toward the central region of the flow or toward the walls [2]. However, no one has yet definitively answered the question of what factors counteract this force, leading ultimately to the establishment of a certain stationary distribution of particles in the cross sections of the channel. There are only phenomenological models to explain the stratification of suepensions, these models having different foundations: the heuristic requirement of an energy dissipation minimum in actual flows [3, 4]; the introduction of diffusion and momentum-conservation equations by the methods of nonequilibrium thermodynamics or other methods [5, 6]; identification of the core of close-packed particles with an infinite cluster [7], etc.

Below, we assume that a stationary distribution of concentration is attained because the convective migration of particles in the transverse direction due to buoyancy is counterbalanced by an oppositely directed diffusion flow in a nonuniform concentration field. In accordance with Einstein's classical method, we will describe this diffusion by introducing a thermodynamic force that acts on the particles. This force is determined on the basis of the condition that the particle flow it creates is exactly equal to the diffusion flow. This force was introduced in [8] in a description of Brownian diffusion in dilute suspensions, while a generalization to concentrated suspensions was made in [9]. Various aspects of the hydromechanics of suspensions were discussed in [9-11] with allowance for this force.

The diffusion of particles can be caused by random fluctuations differing in their physical nature. Here, for the sake of definiteness we assume that the particles are small enough so that only isotropic Brownian motion makes a significant contribution to the fluctuations. The suspension is taken to be isothermal, and we adopt the hypothesis of local thermodynamic equilibrium. It follows in particular from this hypothesis that the mean energy of Brownian motion of a particle for each degree of freedom is the same as for the molecules of the surrounding medium.

1. We will examine a monodisperse suspension of small spherical particles of radius a, The density d of the particle is the same as the density of the fluid. Gravity and buoyancy do not cause phase slip in equidense suspensions, so they can be included in effective pressure in the usual manner. If the absolute value of slip velocity is much lower than the mean velocities of both phases — which is typical of the suspensions being examined here — then for incompressible phases we can write the mass and momentum conservation equations in the form

$$\operatorname{div} \mathbf{v} = 0, \quad -\nabla p + 2\nabla(\eta \nabla \mathbf{e}) = 0, \tag{1.1}$$

where $\mathbf{v}$ is the mean velocity of the suspension; p is pressure with allowance for external body forces; $\mathbf{e}$ is the strain-rate tensor constructed from the field of $\mathbf{v}$; $\eta = M(\rho)\eta_0$ is the effective viscosity of the suspension (η_0 is the viscosity of the pure fluid and $M(\rho)$ is an increasing function of the volume concentration of the disperse phase ρ, $M(0) = 1$).

For unidimensional Poiseuille flows, Eqs. (1.1) take the following form (the parameter m is equal to zero or unity for flows with planar or axial symmetry)

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$$-\frac{\partial p}{\partial z}+\eta_0\frac{1}{x^m}\frac{d}{dx}\left(x^m M\frac{dv}{dx}\right)=0,\quad -\frac{\partial p}{\partial x}=0 \tag{1.2}$$

(z, x are the longitudinal and transverse coordinates). Equations (1.2) must be supplemented by the corresponding equations for the phases of the suspension.

In the longitudinal direction, only the viscous Stokes force f_S and the Faxen force f_F are exerted on the particles by the fluid. Expressions for these forces in moderately concentrated suspensions were calculated in [12] with ρ independent of the coordinates. For all of the particles in a unit volume of the suspension, we have

$$f_S=\rho\frac{9}{2}\frac{\eta_0}{a^2}M(\rho)u,\ f_F=\rho\frac{3}{4}\eta_0 M(\rho)\Delta v$$

($u = v - w$ is the slip velocity if we take v and w to mean the average velocities of the fluid and the particles). It can be shown that the expression for f_F in a nonuniform suspension needs to be replaced by $\mathbf{f}_F = \rho(3/4)\eta_0 2\nabla(M\nabla\mathbf{e})$. Then the longitudinal component of the momentum conservation equation for the disperse phase is

$$u=\frac{a^2}{6M(\rho)}\frac{1}{x^m}\frac{d}{dx}\left(x^m M\frac{dv}{dx}\right). \tag{1.3}$$

The formulas for both forces conform to the well-known model of an effective medium: they coincide in form with the expressions for the analogous forces acting on non-hydrodynamically interacting particles in a homogeneous medium whose viscosity is the same as the viscosity of the suspension.

We proceed in the same manner to determine the transverse buoyancy force acting on all of the particles in a unit volume. Using the relation obtained for this quantity in [13] for a single particle, we write

$$f_L=\rho\frac{3\cdot 6{,}46}{4\pi a}d\left[\nu_0 M\left|\frac{dv}{dx}\right|\right]^{1/2}u\,\mathrm{sign}\left(\frac{dv}{dx}\right) \tag{1.4}$$

(ν_0 is the kinematic viscosity of the pure liquid). This force results in migration of the particles to the flow region where the shear rate is lower. It thus helps to form a nonuniform concentration profile. The nonuniformity in turn helps give rise to a thermodynamic force. Calculated for all of the particles in the unit volume for the case of unidimensional flows, this force is equal to

$$f_T=-\rho\frac{3}{4\pi a^3}\left(\frac{\partial\mu}{\partial\rho}\right)_{p,T}\frac{d\rho}{dx}, \tag{1.5}$$

where μ is the chemical potential of the particles. Differentiation is performed with constant pressure and temperature. This formula follows from basic principles of statistical mechanics [8].

Taking the forces (1.4) and (1.5) into account, we represent the transverse component of the momentum conservation equation for the disperse phase in the form

$$\left(\frac{\partial\mu}{\partial\rho}\right)_{p,T}\frac{d\rho}{dx}=6{,}46a^2 d\left[\nu_0 M\left|\frac{dv}{dx}\right|\right]^{1/2}u\,\mathrm{sign}\left(\frac{dv}{dx}\right), \tag{1.6}$$

where u is found from (1.3); (1.6) is the missing equation needed to determine the concentration profile.

Introducing thermodynamic forces into the momentum conservation equations of the phases of suspensions and other disperse systems should be regarded as the most natural method of accounting for diffusion phenomena in the hydrodynamic modeling of dispersions. Accounting for diffusion flows directly in the mass conservation equations would undermine the meaning of the mean velocities introduced in continuum mechanics [14].

2. We calculate the chemical potential of the particles of a concentrated suspension by using the Karnakhen–Starling variant of the approximate Percus–Yevick theory of a dense gas of hard spheres [15]. Within the framework of this variant, the equation of state of such a gas has the form

$$P^*V^* = NkTG(\rho),\ G(\rho) = \frac{1+\rho+\rho^2-\rho^3}{(1-\rho)^3},\ V = \frac{4\pi a^3}{3\rho}N. \tag{2.1}$$

Here, P* and V* are the pressure of the gas and the volume it occupies; N is the total number of particles; kT is the temperature in energy units; an ideal gas corresponds to G(0) = 1. This equation considers only the geometric interaction of the hard spheres due to excluded-volume effects.

Equation of state (2.1) makes it possible to use standard techniques [15] to construct the configuration integral for the gas and then use differentiation with respect to N to obtain the required representation for chemical potential μ. Here, it is important that, in accordance with the Gibbs method, such differentiation be done with constant p, T, and N_0 (but not V*). Here, N_0 is the total number of molecules of fluid [8]. It follows from this that a solution with a fixed amount of solvent and a variable gas volume can be studied as a molecular analog of the suspension. The results of corresponding calculations yield the following [9, 11]:

$$\mu = \text{const} + kTF(\rho),\ F(\rho) = \ln\rho - \rho + \rho\frac{8-5\rho}{(1-\rho)^2}. \tag{2.2}$$

Introducing $-\partial p/\partial z = P$ and the value of the coordinate x = R corresponding to the walls, we find it convenient to change over to dimensionless variables

$$\begin{Bmatrix}\xi\\ \eta\end{Bmatrix} = \frac{1}{R}\begin{Bmatrix}x\\ z\end{Bmatrix},\ \begin{Bmatrix}V\\ U\end{Bmatrix} = \frac{\eta_0}{PR^2}\begin{Bmatrix}v\\ u\end{Bmatrix}. \tag{2.3}$$

Then with allowance for (1.3), we use (1.2) and (1.6) to obtain a system of equations for the unknown functions V(ξ) and ρ(ξ):

$$\frac{1}{\xi^m}\frac{d}{d\xi}\left(\xi^m M\frac{dV}{d\xi}\right) = -1,\ P = \text{const},$$
$$M\frac{dF}{d\rho} = \Gamma\left|M\frac{dV}{d\xi}\right|^{1/2}\frac{1}{\xi^m}\frac{d}{d\xi}\left(\xi^m M\frac{dV}{d\xi}\right),$$
$$\Gamma = \frac{6{,}46}{6}\frac{a^4(PR)^{3/2}}{d^{1/2}v_0 kT}. \tag{2.4}$$

The boundary conditions for V and ρ follow from the conditions of flow symmetry and adhesion on the walls, i.e.,

$$V = 0,\ \xi = 1;\ dV/d\xi = d\rho/d\xi = 0,\ \xi = 0. \tag{2.5}$$

We find an additional condition by assuming that we already know either the particle concentration averaged over the cross section ⟨ρ⟩ of the flow-rate-mean concentration ρ_f (the fraction of disperse phase in the total volume of the suspension flowing in the channel). This leads to one of the following two conditions

$$(1+m)\int_0^1 \rho(\xi)\xi^m\,d\xi = \langle\rho\rangle,$$
$$\int_0^1 \rho(\xi)V(\xi)\xi^m\,d\xi\left(\int_0^1 V(\xi)\xi^m\,d\xi\right)^{-1} = \rho_f. \tag{2.6}$$

The function F(ρ) in (2.4) was determined in (2.2). To close system (2.4)-(2.6), it is necessary to also find the function M(ρ). Studying moderately concentrated suspensions, Buevich and Markov [12] obtained an expression which diverges at $\rho \to 0.4$: $M = (1 - 5\rho/2)^{-1}$. One of the numerous empirical representations for the ratio η/η_0 can be used as an approximation of M(ρ) that is suitable throughout the range of ρ. Here, we take

$$M(\rho) = (1-\rho)^{-5/2}, \tag{2.7}$$

This expression agrees quite well with the result in [12] in the region $\rho \lesssim 0.2$ for moderately concentrated suspensions and correctly describes the behavior of M(ρ) at large ρ.

A serious deficiency of the approximate theory of an ensemble of hard spheres [which leads to Eq. (2.2)] is the fact that it does not contain an order–disorder phase transformation and fails to reflect the formation of the close-packed (non-fluid) state. The functions $M(\rho)$ and $G(\rho)$ should approach infinity as this state is approached. Also, the structure of the close-packed state is not determined a priori – it may correspond to different types of regular lattices, sets of discrete ordered regions separated by randomized interlayers, etc. As a result, the volume concentration of particles ρ^* associated with the close-packed state turns out to be ambiguous, with the same given volume theoretically corresponding to topologically different structures. Statistical mechanics has yet to solve this problem. Thus, given the current level of knowledge, it is best if we follow [9] and regard ρ^* as an empirically assignable quantity. Here, we assume that the particles can be regarded as the disperse phase of a suspension at $\rho < \rho^*$, while they become completely immobile at $\rho = \rho^*$ and form a coherent close-packed system.

3. With allowance for [11], we find from the first equation in [10] that

$$MdV/d\xi = -\xi/(1+m), \tag{3.1}$$

which allows us to write the solution of the second equation of (2.4) in quadratures. If $\rho_0 < \rho^*$ (where ρ_0 is the concentration on the plane or axis of symmetry of the flow $\xi = 0$), then

$$I(\rho) - I(\rho_0) = \frac{2}{3}\frac{\Gamma}{\sqrt{1+m}}\xi^{3/2},\ 0 \leqslant \xi \leqslant 1. \tag{3.2}$$

Otherwise, when a close-packed core of particles occupying the region $\xi < \xi^*$ is formed in the flow, we have

$$I(\rho) = \frac{2}{3}\frac{\Gamma}{\sqrt{1+m}}(\xi^{3/2} - \xi_*^{3/2}),\ \xi_* \leqslant \xi \leqslant 1,$$
$$\rho = \rho_*,\quad 0 \leqslant \xi \leqslant \xi_*. \tag{3.3}$$

In (3.2) and (3.3) we introduced the function

$$I(\rho) = \int_{\rho}^{\rho_*} M(\rho)\frac{dF}{d\rho}d\rho, \tag{3.4}$$

which is expressed through known functions, with allowance for (2.4) and (2.7). However, it is simpler to construct this function numerically; the results of its calculation with $\rho^* = 0.6$ are shown in Fig. 1.

Using (3.2)-(3.3) and the curve in Fig. 1, we can construct profiles of concentration in planar and axisymmetric Poiseuille flows with different ρ_0 (or ξ^*) and Γ. Figure 2 also shows profiles for flow in a circular tube with different ρ_0 (corresponding to the points of intersection of the curves with the y-axis) and Γ (for all ρ_0, the curves starting from the top correspond to $\Gamma = 10$, 30, and 100, respectively); we took ρ^* in the calculations. It is evident that at sufficiently large Γ, the particles are concentrated in the central region of the flow. This effect diminishes with an increase in mean concentration. If a core of close-packed particles of the size ξ^* is formed, then – as shown by the calculations – concentration in the range $\xi^* \leq \xi < 1$ differs very little from the value for close packing.

With small ρ, we approximately have $F \approx \ln\rho$, $M \approx 1$, and it follows from (3.3) and (3.4) that

$$\rho \approx \rho_0 \exp\left(-\frac{2}{3}\frac{\Gamma}{\sqrt{1+m}}\xi^{3/2}\right),$$

This expression makes clear the effect of the parameter Γ on the particle distribution in sections of the Poiseuille flow.

Using the resulting function $\rho(\xi)$ in Eq. (3.1), we can construct profiles of dimensionless flow velocity

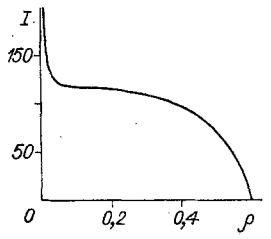

Fig. 1

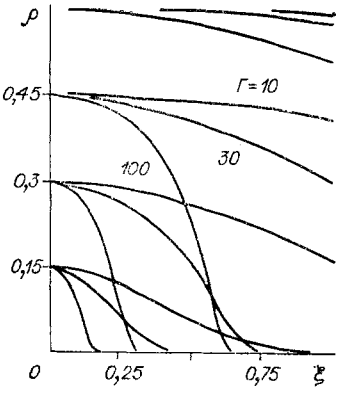

Fig. 2

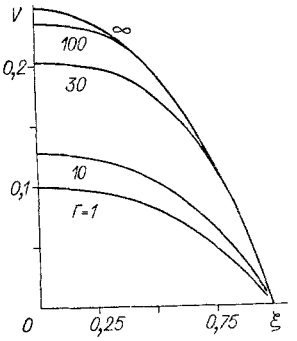

Fig. 3

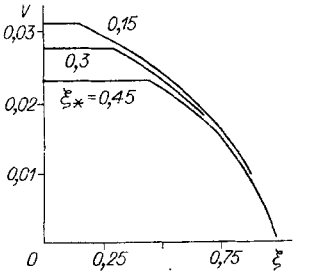

Fig. 4

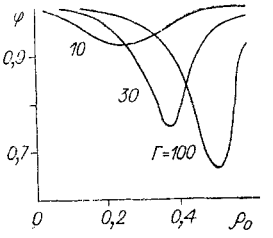

Fig. 5

$$V(\xi) = \frac{1}{1+m}\int_{\xi}^{1} \frac{\xi\, d\xi}{M(\rho)}, \quad \xi_* \leqslant \xi < 1,$$
$$V(\xi) = V(\xi_*) = V_*, \quad 0 \leqslant \xi < \xi_*. \qquad (3.5)$$

Figure 3 shows functions $V(\xi)$ corresponding to the concentration profiles in Fig. 2 for $\rho_0 = 0.30$. The top three profiles correspond to situations in which there are almost no particles in the peripheral region of the flow (see the concentration distribution in Fig. 2). Thus, the wall-region velocity gradients which correspond to these profiles coincide. However, the presence of particles in the central region leads to the manifestation of pseudo-plastic properties by the flow. Figure 4 shows profiles of dimensionless velocity for flows with a close-packed core.

Equations (2.6) make it possible to establish the relationship between mean values of concentration $\langle\rho\rangle$ or ρ_f and values of ρ_0 or ξ^* used to construct profiles of concentration and dimensionless velocity. Table 1 shows mean concentrations for conditions under which no close-packed core is formed. The critical values $\langle\rho\rangle$ and ρ_f, corresponding to the appearance of such a core, are shown in the column for $\rho_0 = \rho^* = 0.60$.

TABLE 1

Γ	$\langle\rho\rangle$				ρ_f			
	0,15	0,30	0,45	0,60	0,15	0,30	0,45	0,60
	ρ_0							
1	0,138	0,295	0,449	0,5997	0,142	0,296	0,449	0,5998
3	0,114	0,283	0,445	0,5990	0,120	0,287	0,446	0,5993
10	0,046	0,23	0,43	0,597	0,067	0,25	0,44	0,598
30	0,011	0,079	0,38	0,590	0,020	0,121	0,40	0,593
100	0,0021	0,016	0,12	0,56	0,0042	0,030	0,17	0,57

Equations (3.5) make it possible to calculate the dimensionless volumetric flow rate of the suspension for different Γ and ρ_0 (or ξ^*). Along with the flow rate Q, it is best to also examine the analogous flow rate Q^0 for the flow of a suspension with uniformly distributed particles ($\rho \equiv \langle\rho\rangle$), other conditions being equal. The relation

$$\varphi = \frac{Q^0}{Q} = \frac{1}{16M(\langle\rho\rangle)}\left(\int_0^1 V(\xi)\,\xi\,d\xi\right)^{-1}$$

is a coefficient which expresses the reduction in drag due to stratification of the suspension. It characterizes the pseudo-plastic properties of flows, which become stronger with an increase in Γ. Figure 5 shows the character of the dependence of φ on ρ_0 and Γ.

The results shown above also approximately describe stratification in vertical Poiseuille flows of suspensions with different phase densities if the effect of the weight of the particles (minus buoyancy) is small compared to the effect of the viscous force f_S exerted on the particles by the dispersion medium, i.e., if

$$\rho(d_1 - d)g = \rho(1 - \rho)(d_1 - d_0)g \ll \rho(9\eta_0/2a^2)M(\rho)u, \tag{3.6}$$

where d_0 and d_1 are the densities of the fluid and the particle material; $d = (1 - \rho)d_0 + \rho d_1$ is the density of the suspension. Strong inequality (3.6) is satisfied when the pressure gradient is sufficiently large.

When condition (3.6) is not met, gravity exerts a substantial effect on stratification of the suspension even in the case of vertical flow. Here, the magnitude and direction of slip velocity are determined not only by the Faxen force, but also by the gravitational force acting on the particles (minus buoyancy). In particular, in descending flows of suspensions with $d_1 > d_0$ and in ascending flows with $d_1 < d_0$, it is possible to have situations in which the transverse force changes sign, i.e., helps displace particles toward the boundaries of the flow rather than toward the center. In this case, particle concentration is reduced in the core of the flow and increased near the walls, while drag turns out to be greater than in a uniform flow with the same mean concentration. At high concentrations, the formation of a layer of close-packed particles adjacent to the walls becomes possible. The formulation and solution of the corresponding problems is obvious from the above discussion.

The same suspension stratification effects are also seen in other types of flows. For example, the familiar wall effect should be seen in a laminar boundary layer. This effect consists of the displacement of particles away from the surface of the body located in the flow. The necessary boundary conditions can be formulated by analogy with the problem examined in the present study. The profiles obtained above for concentration and velocity in a Poiseuille flow in a circular pipe agree (within the experimental error) with the empirical data (see [1], for example), although it remains unclear why certain authors have reported that the formation of pseudo-plastic velocity profiles has not been accompanied by particle redistribution across the flow.

In conclusion, we should point out certain limitations of the theory that has been developed. First of all, along with the thermodynamic force (which contains the concentration gradient), a component of the phase interaction force may be present – especially in the case of macroscopically nonuniform flows. The possibility of the creation of such a force in suspensions was emphasized in [12], which also presented a method of calculating

it. However, this calculation has yet to be performed. At the same time, there is data supporting the existence of such a force in systems with regular [16] or randomized [17] distributions of stationary particles. In light of the well-established fundamental difference between systems with stationary and moving particles and between the forces which act in ordered and randomized particle clouds, the results obtained in [16, 17] cannot be used within the present context. However, the above-mentioned force should in any event be proportional to the mean slip velocity. The thermodynamic force (1.5), meanwhile, is generally independent of slip velocity. Thus, the effect being discussed should be negligible for sufficiently small particles capable of intensive Brownian motion — the only type of particles we have examined here.

The thermodynamic force calculated above for particles which undergo Brownian motion can also be introduced into the analysis by a completely different approach. Using Eq. (2.2) for chemical potential and the equality $kT = m\langle w'_i{}^2\rangle$, where w'_i is one of the components of particle fluctuation velocity, it is not hard to write the force f_T from (1.5) as the derivative, with respect to the transverse coordinate, of the effective pressure in a concentrated system of pulsating particles $d_1\rho G\langle w'_i{}^2\rangle$. Meanwhile, the function $G(\rho)$ determined in (2.1) accounts for the existence of collective effects in this system — as in Enskog's well-known theory of dense gases. This corresponds to a situation in which f_T is represented in the general case in the form of the divergence of the momentum tensor due to pulsations of particles (regardless of their physical nature). The tensor is spherical in the case of brownian particles. This subject was discussed in detail in [11]. The only important restriction on the pulsation mechanism is the condition that the pulsations of individual particles not be correlated. An important example of random motions which approximately satisfy this condition is anisotropic pseudo-turbulent motion on suspended particles that are not excessively coarse.

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