

CONCENTRATION FLUCTUATIONS AND PARTICLE SELF-DIFFUSION IN MONODISPERSED SYSTEMS

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The dispersion of a random field of the volume concentration is calculated on the basis of the thermodynamic theory of fluctuations in limiting situations of finely and coarsely dispersed systems. A representation for the spectral density of this field is written, and concentration dependences of Brownian and isotropic pseudoturbulent self-diffusion of particles in the indicated situations are found.

Rheological properties and effective transfer coefficients in dispersed mixtures are largely determined by the properties of random pulsations of the particles and the liquid. Energy is brought to the pulsations due to the effect of the average liquid flow with respect to the particles on random fluctuations of the mixture concentration according to the model presented, in particular, in [1]. Therefore, the properties of such fluctuations, dealt with in this work, are of prime interest.

The statistical characteristics of a random concentration field were previously investigated on the basis of combinatorial analysis, generalizing the known Smoluchovski-Einstein theory for diluted mixtures to concentrated dispersions (see, for example, [2-4]). This analysis has a number of drawbacks which are generally inherent in lattice models of statistical physics, and therefore, its conclusions need refinement. The latter seems timely as concerns the formulation of systems of fundamental equations determining the motion of suspensions, fluidized beds, etc. (an example of such a system for a coarsely dispersed gas suspension can be found in [5]). Below, the general theory of thermodynamic fluctuations is used for this purpose [6].

Two limiting types of dispersion should be recognized. In the first of them an exchange of momentum and energy between suspended particles occurs by means of random velocity and pressure fields in the ambient liquid, and the role of direct collisions in this exchange is vanishingly small. In this case, which is characteristic of colloids and suspensions of fairly small particles in dropping liquids, a system of solute molecules which are in the field of an average force due to the solvent may serve as a statistical analog of the system of particles. Actually, it was such a case that was dealt with in [1]. In dispersions of the second type the interparticle exchange is determined, on the contrary, by direct collisions, while the influence of the interphase interaction on this process is negligible. Such a situation is characteristic of suspensions of large particles in gases, when the system of suspended particles can be considered as a certain pseudogas [5].

Elucidating the true role of collisions entails the analysis of the collective behavior of a system of a large number of particles complicated by hydrodynamic interactions between them, and hence presents a very difficult problem. This problem was recently discussed in [7], from which it follows that both of the situations mentioned are realized in practice.

Chemical Potentials of Particles. We will consider systems with identical spherical particles. To calculate the dispersion of the number of particles in some fixed volume, expressions for their chemical potential are primarily needed. These expressions turn out to be different for the two limiting types of mixtures indicated above. In both cases, however, one should proceed from a formal equation of state for the system of particles in two possible forms [8, 9]:

$$PV = NkTG(\rho) = V \left(\frac{\partial \ln(V^N Q_N)}{\partial V} \right)_{T,N} \quad (1)$$

The function $G(\rho)$ describes the steric interaction of particles due to the excluded volume effects (the impossibility of solid sphere overlap). It is desirable to obtain a simple analytical expression for this function, which can be done by using either variant of the Perkus-Yevik liquid state theory, for example, the Carnahan-Starling version [10] of this theory

$$G(\rho) = \frac{1 + \rho + \rho^2 - \rho^3}{(1 - \rho)^3}, \quad (2)$$

or the Enskog theory of dense gases [8]

$$G(\rho) = \frac{1}{1 - (\rho/\rho_*)^{1/3}}. \quad (3)$$

The first approach, previously applied in [11, 12], is in agreement with the data of numerical experiments performed by the method of molecular dynamics and the Monte-Carlo method; the second one was applied, for example, in [13].

A certain drawback of the second approach consists in using notions of a smoothed free volume of individual spheres and a priori introduction of the limiting value of the concentration ρ_* attained in the state of close packing, in which fluctuations are considered to be absent. In the second approach a possible phase transition to a crystallike phase at high concentrations is ignored.

For simplicity we disregarded the surface interactions of particles at the expense of molecular and electrostatic forces in (1)-(3). The latter may be important in colloid dispersions. The necessary generalization can be obtained by using the equation of state in the Braut form [14].

An equation for a configurational integral follows from (1), whose solution has the form

$$\ln Q_N = -N \int_0^\rho \frac{G(\rho) - 1}{\rho} d\rho. \quad (4)$$

The idea of subsequent calculations consists in using standard representations of the Gibbs thermodynamic potential for the system of dissolved particles and for the pseudogas in terms of Q_N . In the first case in the thermodynamic limit ($N \rightarrow \infty$) we have

$$\Phi = N_0\mu_0^\circ + N\mu^\circ + NkT \ln(\rho/e) - kT \ln Q_N,$$

and in the second case we have

$$\Phi = N\mu^\circ + F + PV, \quad F = NkT (\ln \rho - \psi(T)) - kT \ln Q_N,$$

where the degree symbol denotes the standard chemical potentials whose form is unimportant in what follows.

Chemical potentials of particles are hence obtained by differentiating with respect to the total number of particles N using the representation (4). In the first case differentiating is to be performed at constant temperature T and total number N_0 of liquid molecules and in the second - at constant T and P . The average volume concentration ρ is to be expressed in these cases as an $Nv + N_0v_0$ ratio and as a constant, following from (1), respectively.

As a result of calculations we arrive at the following formulas for the *system of dissolved particles*:

$$\mu = \text{const} + kT \left[(\ln \rho - \rho) + \rho \frac{8 - 5\rho}{(1 - \rho)^3} \right] \quad (5)$$

when using (2) and

$$\mu = \text{const} + kT \left[(\ln \rho - \rho) - 3 \ln \left\{ 1 - \left(\frac{\rho}{\rho_*} \right)^{1/3} \right\} + \frac{(1 - \rho)(\rho/\rho_*)^{1/3}}{1 - (\rho/\rho_*)^{1/3}} \right] \quad (6)$$

when using (3). Formula (5) was obtained previously in [11, 12].

For the *pseudogas of particles* we obtain

$$\mu = \text{const} + kT \left[\ln \rho + \rho \frac{4-3\rho}{(1-\rho)^2} + \frac{1+\rho+\rho^2-\rho^3}{(1-\rho)^3} \right] \quad (7)$$

when using (2) and

$$\mu = \text{const} + kT \left[\ln \rho - 3 \ln \left\{ 1 - \left(\frac{\rho}{\rho_*} \right)^{1/3} \right\} + \frac{1}{1 - (\rho/\rho_*)^{1/3}} \right] \quad (8)$$

when using (3). Formula (7) follows from [11].

As $\rho \rightarrow 0$ the known expressions which are characteristic of diluted solutions and ideal rarefied gases are obtained from (5)-(8).

Dispersion of Concentration Fluctuations. The dispersion of the number of particles in some volume A can be expressed in terms of a derivative of the chemical potential in the following manner [6]:

$$\langle N_A'^2 \rangle = kT (\partial \mu / \partial \langle N_A \rangle)_{T,A}^{-1},$$

whence an analogous formula for the dispersion of the volume concentration $\rho_A = vN_A/A$ determined by averaging over the volume A follows

$$\langle \rho_A'^2 \rangle = kT (v/A) (\partial \mu / \partial \langle \rho \rangle)_{T,A}^{-1}. \quad (9)$$

(The notation $\langle N_A \rangle$ and $\langle \rho_A \rangle$ for the average values is introduced in these formulas; for simplicity the angular brackets are omitted in (1)-(5).)

As is evident from [2, 4], the most detailed description of the random field of the concentration of the dispersed mixture, consistent with continuum theory, can be attained in the special case when the specific volume v/ρ in which there can be on the average one particle is chosen as the volume. In this case on the basis of calculations we arrive at the following results.

System of Dissolved Particles. Within the framework of the Perkus-Yevik theory

$$\langle \rho'^2 \rangle = \frac{\rho^2}{1-\rho} \left[1 + 2\rho \frac{4-\rho}{(1-\rho)^2} \right]^{-1} \quad (10)$$

and within the framework of the Enskog theory

$$\langle \rho'^2 \rangle = \frac{\rho^2}{1-\rho} \left[1 - \left(\frac{\rho}{\rho_*} \right)^{1/3} \right] \left[1 + \frac{1}{3} \frac{(\rho/\rho_*)^{1/3}}{1 - (\rho/\rho_*)^{1/3}} \right]^{-1}. \quad (11)$$

Pseudogas of Suspended Particles. In the Perkus-Yevik theory

$$\langle \rho'^2 \rangle = \rho^2 \left[1 + 2\rho \frac{4-\rho}{(1-\rho)^2} \right]^{-1} \quad (12)$$

and in the Enskog theory

$$\langle \rho'^2 \rangle = \rho^2 \left[1 - \left(\frac{\rho}{\rho_*} \right)^{1/3} \right] \left[1 + \frac{1}{3} \frac{(\rho/\rho_*)^{1/3}}{1 - (\rho/\rho_*)^{1/3}} \right]^{-1}. \quad (13)$$

Figure 1 gives the curves described by these formulas. It should be emphasized that the dispersions (10)-(13) turn out to be considerably smaller (several times smaller in the range of intermediate average concentrations) than

$$\langle \rho'^2 \rangle = \rho^2 (1 - \rho/\rho_*),$$

calculated in [2, 3] from the lattice model. The concentration fluctuation intensity of the pseudogas (a coarsely dispersed system) turns out to be somewhat lower than the analogous quantity for the dissolved particles (a colloid or a finely dispersed suspension). A substantial difference between the Enskog and Perkus-Yevik theories is observed, as was to be expected at large concentrations.

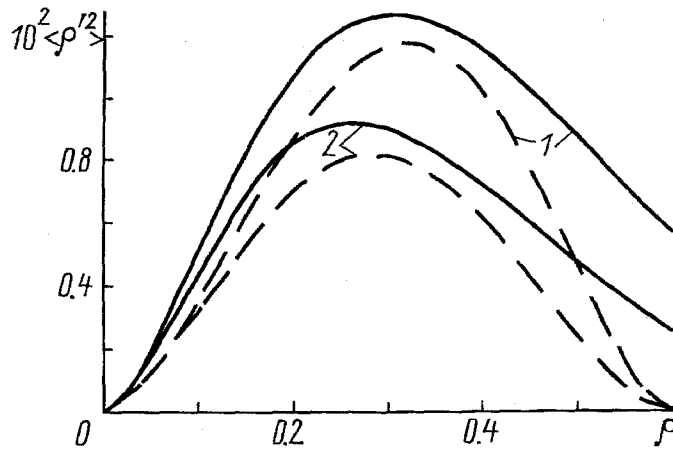


Fig. 1. Dispersion of the volume concentration for mixtures with small (1) and large (2) particles according to the Perkus-Yevik theory (formulas (10) and (12), solid curves) and the Enskog theory (formulas (11) and (13), dashed curves); $\rho_* = 0.6$.

Spectral Concentration Field Density. For applications it is important to know how to calculate not only the concentration dispersion but also the double-time two-point correlation functions. Within the framework of the correlation theory of stationary random processes this requires an explicit representation of the spectral density of concentration fluctuations. Without going into detail, we will give such a representation, following from [2].

Should the volume concentration fluctuation about its average value be represented in the form of the Fourier-Stieltjes integral

$$\rho'(t, \mathbf{x}) = \int e^{i(\omega t + \mathbf{kx})} dZ_\rho,$$

the correlation function may be represented as

$$\langle \rho'(t, \mathbf{x}) \rho'(t + \tau, \mathbf{x} + \mathbf{r}) \rangle = \int_{-\infty}^{\infty} d\omega \int d\mathbf{k} e^{i(\omega\tau + \mathbf{k}\mathbf{r})} \Psi_{\rho, \rho}(\omega, \mathbf{k}),$$

where the total spectral density is determined by:

$$\Psi_{\rho, \rho}(\omega, \mathbf{k}) = \lim_{d\omega \rightarrow 0, d\mathbf{k} \rightarrow 0} \langle dZ_\rho dZ_\rho^* \rangle / d\omega d\mathbf{k}.$$

By integrating this quantity over the entire frequency axis or over the entire wave space, we can hence obtain expressions for partial spectral densities, dependent solely on \mathbf{k} or solely on ω .

According to the theory [2]:

$$\Psi_{\rho, \rho}(\omega, \mathbf{k}) = \frac{kDk}{\pi} \frac{\Phi_{\rho, \rho}(k)}{\omega^2 + (kDk)^2}, \quad (14)$$

where $\Phi_{\rho, \rho}(k)$ is one of the partial densities. Expression (14) is written under the assumption that the evolution of the concentration fluctuations can be described with sufficient accuracy by an ordinary diffusion equation; if this is not the case, the finiteness of the velocity of diffusing particles is to be taken into account as was done in [2].

Furthermore, the relations

$$\Phi_{\rho, \rho}(k) = \Phi(k), \quad \int_0^{\infty} \Phi(k) k^2 dk = \frac{\langle \rho'^2 \rangle}{4\pi}, \quad (15)$$

are valid, where $\langle \rho'^2 \rangle$ is determined by one of formulas (10)-(13) and $\Phi(k)$ is the wave number function, substantially differing from zero only in the long-wave region $k \lesssim k_0 \sim \rho^{1/3}/a$. Depending on the method of roughly approximating the detailed numerical concentration of particles, initially represented by points, we obtain different expressions for $\Phi(k)$. In particular, if the particles are assumed to be uniformly distributed throughout their specific volume, we have [2]

$$\Phi(k) = \frac{\langle \rho'^2 \rangle}{2\pi^2} \frac{\sin kb - kb \cos kb}{k^3}, \quad b = \frac{a}{\rho^{1/3}}. \quad (16)$$

If it is assumed that the fluctuation spectrum is uniform in the region $k < k_0$ of the wave space and vanishes outside it, we obtain

$$\Phi(k) = \frac{3 \langle \rho'^2 \rangle}{4\pi k_0^3} H(k_0 - k), \quad k_0 = \left(\frac{9\pi\rho}{2a^3} \right)^{1/3}. \quad (17)$$

The functions (16) and (17) correspond to somewhat limiting situations, and the arbitrariness in their determination is in agreement with the possible accuracy of determining the position of any point within the framework of the continuum method of description.

The tensor in second dimensions \mathbf{D} describes self-diffusion coefficients of particles which are bound to appear in the evolution equation for a random concentration. In the general case this tensor may differ considerably from a spherical one owing to the anisotropy of random pulsations of particles [1]. However, two important exceptions occur when these pulsations are approximately isotropic and the diffusion of particles can be described by just the scalar coefficient. It is these exceptions that are dealt with below.

Self-Diffusion in a Pseudogas. If the determining role in the interparticle exchange of momentum and energy is played by direct collisions between particles the particle pulsations are bound to be close to isotropic ones, and the role of the temperature kT in power units is played by the double average kinetic energy, accounting for one progressive degree of freedom [5]. In this case the self-diffusion coefficient may be expressed using the well-known Enskog factor. This factor takes account of acceleration of transfer in concentrated systems due to the fact that its rate in the solid particles themselves is of the order of the velocity of sound and is considerably larger than the mean-square velocity of the pulsations. Furthermore, it also takes into account the negative influence on transfer of the effects of screening of any particle by its neighbors, which cause a relative decrease in the collision frequency. As a result the self-diffusion coefficient is [5, 8]

$$D = \frac{D_0}{\chi(\rho)}, \quad \chi(\rho) = \frac{G(\rho) - 1}{4\rho}, \quad D_0 = \frac{3}{32a^2 n} \left(\frac{kT}{\pi m} \right)^{1/2}. \quad (18)$$

Formulas (2) and (3) for $G(\rho)$ are in agreement with two different concentration dependences of the dimensionless coefficient D/D_0 , which are illustrated in Fig. 2. The dependence corresponding to the Enskog theory shows a discrepancy within $\rho \rightarrow 0$, which is associated with the inapplicability of this theory to the low concentration range [8].

Brownian Self-Diffusion. The second situation when the particle pulsations are approximately isotropic occurs in colloids and suspensions of very small particles in which anisotropic pseudoturbulent pulsations degenerate [1, 12] and only thermal Brownian motion occurs. In this case T denotes an ordinary temperature.

An abundance of works in which highly nontrivial models of statistical physics and methods of analyzing collective hydrodynamic interactions are commonly used concern investigations of Brownian diffusion (see model examples in [15-19]). However, even very elementary reasoning enables one to obtain simple representations for the coefficient of Brownian self-diffusion both at small and large times of the process.

Each particle can be represented as being in a "cage" formed by its nearest neighbors [18]. The characteristic time when the boundaries of such a cage are reached by the particle is of the order of magnitude $t_c = (a/\rho^{1/3})^2/D_0$, which, for small particles, is much larger than the hydrodynamic relaxation time of the particle $t_r = m/6\pi\eta_0 a$ [16]. If the time of random walk of the particle $t \gg t_r$ but simultaneously $t \ll t_c$ the particle undergoes a great many uncorrelated random displacements over this time as a result of its interaction with the molecules of the ambient liquid, and this walk can be described as a random process of the diffusion type. However, the screening action of the cage particles is not manifested during this time of walk; this being so, the presence of other particles has an effect on the motion of any individual particle merely in the sense that the rheological properties of the medium, in which the latter walks, differ from those of a pure liquid without particles. Thus, by using the model of an effective medium for the Brownian self-diffusion coefficient we have the usual expression

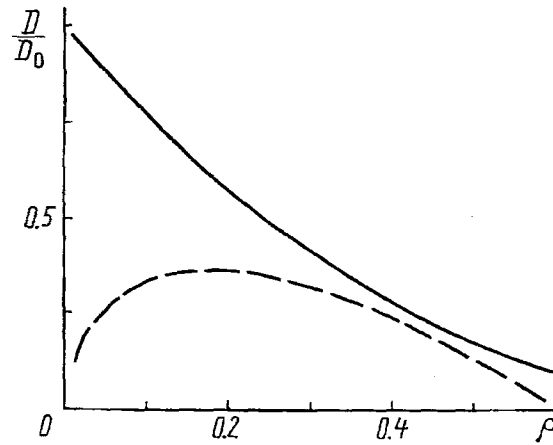


Fig. 2. Dimensionless coefficient of self-diffusion of pseudogas particles according to (18) from the Perkus-Yevik and Enskog theories (solid and dashed curves, respectively); $\rho^* = 0.6$.

$$D_- = \frac{D_0}{K(\rho)}, \quad D_0 = \frac{kT}{6\pi\eta_0 a}, \quad (19)$$

where $K(\rho)$ is the relative viscosity of the suspension. This formula refers, of course, only to small diffusion times, satisfying the inequality $t \ll t_c$.

For moderately concentrated suspensions $K = (1 - 5\rho/2)^{-1}$; this expression was proposed by many authors and was rigorously obtained in [20]. In order to extend it to the entire interval of the concentrations considered we will use the semiempirical formula $K = (1 - \rho)^{-5/2}$. Then the dimensionless coefficient D_-/D_0 of Brownian diffusion at small times is represented by the curve shown in Fig. 3.

Now we will deal with the other limiting case: $t \gg t_c$. When this inequality is satisfied, the effects described by the Enskog factor "join the game" and the particle leaves one cage to find itself in the next one. However, the number of these cages that the particle has a chance to enter over the time t is again sufficiently large for its walk to be a process of the diffusion type. Since the effects of acceleration of transfer in the particle material and the effects of screening are of purely geometric origin it seems unimportant whether the particle acts as a solute molecule or a gas molecule. This implies that a formula of the type of (18) continues to be applicable and the Brownian self-diffusion coefficient is consequently expressed by the following formula at large times:

$$D_+ = \frac{D_-}{\chi(\rho)}, \quad \chi(\rho) = \frac{G(\rho) - 1}{4\rho}. \quad (20)$$

Figure 3 also shows the concentration dependences of the quantity D_+/D_0 , which correspond to the models belonging to (2) and (3). It is also possible to obtain a unified curve for this quantity by using some composite expression for the Enskog factor, as is proposed in [5]. As follows from the curves in Fig. 3, the coefficient D_+ in concentrated dispersions can be tens of times smaller than D_- , which is in good agreement with observations. As an example, the same figure gives the experimental data obtained in investigations of light scattering by dispersions in [21, 22].

Since in the experiments the observation time is commonly fixed and the critical time scale t_c depends on the concentration of the dispersed system, increasing to infinity in the transition to extremely dilute dispersions, experimentally determined curves will asymptotically tend to $D_-(\rho)$ at low concentrations and to $D_+(\rho)$ at high ones. In fact, this tendency is also traced in the experiments given in Fig. 3. On the whole, taking into account the elementary character of the theory, its agreement with the experimental data can be regarded as very good and, in any case, sufficient for using its results in hydrodynamic modeling of suspensions.

Gradient Diffusion. To complete the picture we will briefly discuss the diffusion of particles in a nonuniform field of the average concentration. For a pseudogas, the directed motion of particles against the concentration gradient depends on the relationship between the effective pressure of the pseudogas, which is generated by pulsations and

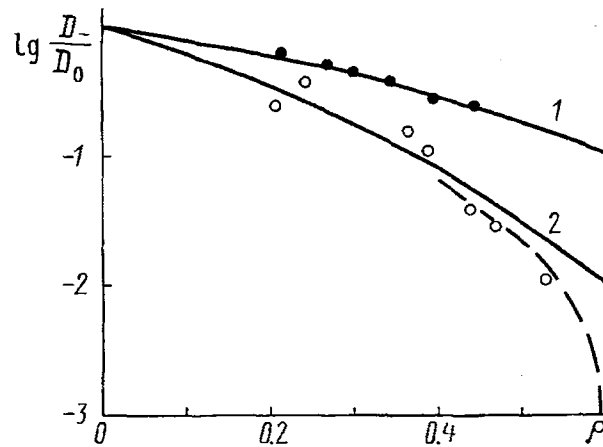


Fig. 3. Dimensionless coefficient of Brownian self-diffusion at small (1) and large (2) times (the solid and dashed curves are respectively from the Perkus-Yevik and Enskog theories) according to (19) and (20); points are the experiment for small and large diffusion times; $\rho^* = 0.6$.

is intricately dependent on the concentration, and the hydraulic and external mass forces affecting the particles, rather than on the pulsation intensity. The balance of forces determines, in particular, a stationary nonuniform distribution of particles along the height of fluidized beds [5]. It is difficult to introduce a universal coefficient of gradient diffusion for a coarsely dispersed medium since the above reasoning is insufficient for this.

For Brownian dispersions, however, the situation is completely different. In this case we can use a method based on the introduction of thermodynamic forces, which was first applied to dilute suspensions in [23] and was generalized to concentrated suspensions in [11, 14]. The general formula for the gradient diffusion coefficient has the form

$$D_m = \frac{1}{6\pi\eta_0 a K^*(\rho)} \frac{1}{1-\rho} \left(\frac{\partial \mu}{\partial \rho} \right), \quad (21)$$

where $K^{*-1}(\rho)$ has the meaning of the ratio of the particle sedimentation rate in the concentrated suspension to the analogous quantity for a single particle. If the particles did not pulsate, K^* would coincide with the relative viscosity of the suspension, in any case, within the framework of the model of an effective medium. The presence of pulsations gives rise to additional phase flows and to a change in the hydraulic resistance of the particles to the relative flow of the liquid [1]. Therefore, at this stage it makes sense to evaluate this function by using the results of experiments on constrained sedimentation of suspensions, which are well described by the empirical formula $K^* = (1-\rho)^{-j}$, where j varies from 5.5 to 6.5 [24]. Calculating the derivative in (21) using expression (5) for the chemical potential and taking $j = 6$, we have

$$D_m = (1-\rho)^6 \left[1 + 2\rho \frac{4-\rho}{(1-\rho)^4} \right] D_0, \quad D_0 = \frac{kT}{6\pi\eta_0 a}. \quad (22)$$

This dependence is plotted in Fig. 4, where the experimental points collected in Fig. 3.14 of the book [24] are also given. Agreement as a whole is good, especially taking into account that the coefficient of Brownian gradient diffusion strongly depends on the molecular and electric interactions between the particles [14, 16], which are very difficult to control in the experiments. Therefore, generally speaking, it cannot be completely guaranteed that these interactions were in fact absent in the experiments presented in [24] and in Fig. 4. We note that the difference of K^* from K was not taken into account in [11, 14], which resulted in a monotonically increasing theoretical dependence $D_m(\rho)$.

In conclusion we will make a number of tentative comments regarding the assumed conditions of validity of the formula obtained. By its definition and meaning the introduction of a smoothed free volume of particles in the Enskog theory of dense gases in the form of a sphere whose radius depends on the average concentration of a dispersed mixture eliminates the difference between regular and randomized packings of particles of the same

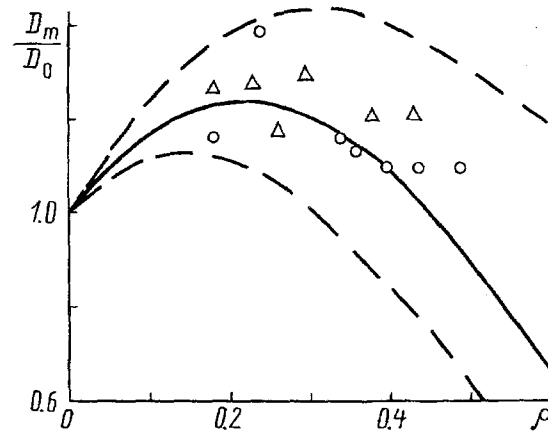


Fig. 4. Concentration dependence (22) of the dimensionless coefficient of Brownian gradient diffusion; points are experimental data [24], dashed curves are the dependence (22) with the exponent $j = 5.5$ and 6.5 in the first multiplier instead of $j = 6$.

concentration. This implies that the concentration fluctuations inherent in random packings are actually excluded from consideration. Therefore, in the high-concentration range formulas (11) and (13) based on the Enskog theory will yield very conservative results, which is confirmed by the curves in Fig. 1. Formulas (10) and (12) should be favored in this region.

On the other hand the Perkus-Yevik theory basically takes no account of the pulsation freeze-out on attaining some critical concentration, which can depend on the roughness of the particles and other accidental factors. Therefore, it should be expected that when calculating the diffusion coefficients in suspensions with a very high concentration the expression for $\chi(\rho)$ following from the Enskog theory would be more adequate (see Fig. 3). However, the latter is incorrect for dilute suspensions, where this theory loses its validity. In the range of small and apparently of moderate concentrations the quantity $\chi(\rho)$ following from the Perkus-Yevik theory should be favored.

NOTATION

A , sample volume; a , radius of a particle; b , radius of a specific volume; D , self-diffusion tensor; D_- , D_+ , D_m , coefficients of Brownian self-diffusion at small and large times and of Brownian gradient diffusion, respectively; H , Heaviside function; K , K^* , relative viscosity of the suspension and the ratio of the velocity of a single particle to the constrained sedimentation rate; k , wave vector; k , k_0 , wave number and its characteristic value (17); m , mass of a particle; N , N_0 , total number of particles and molecules of the liquid; n , numerical concentration of the particles; P , particle pressure in the equation of state; Q_N , configurational integral referred to V^N ; kT , temperature in power units; t_c , t_r , characteristic time scales; V , volume of the system; v , v_0 , particle and molecule volumes; dZ_ρ , random measure of concentration fluctuations; η_0 , viscosity of the liquid; μ , chemical potential; ρ , ρ_+ , particle concentration throughout the volume and its value in the close packing state; $\Psi_{\rho, \rho}$, $\Phi_{\rho, \rho}$, spectral measures of concentration fluctuations; Φ , chemical potential; $\chi(\rho)$, Enskog factor; ω , frequency; the degree symbol refers to the standard chemical potentials, angular brackets denote averaging, dashes denote fluctuations.

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