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THEORY OF RANDOM MOTION OF PARTICLES IN A SUSPENSION

A. N. Latkin

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We estimate the self-diffusion coefficient of particles in a moving suspension, taking into account pseudoturbulent and Brownian fluctuations.

Heat and mass transport in dispersed systems depend, to a large degree, on the random motion of the particles, which is caused by different physical factors. For quasilaminar motion fluctuations result from ordinary Brownian motion and from concentration fluctuations of the particles produced by the flow of the continuous phase (so-called pseudoturbulent motion). If the particles are sufficiently small, the only significant contribution to their random motion is isotropic Brownian motion, which depends on the concentration of the dispersed phase. Hence diffusion processes in suspensions are quite different from self-diffusion of a single particle in a pure liquid. Brownian motion is described in [1] for low concentrations. The results of [1] are generalized in [2] to higher concentrations. As the particle radius increases, pseudoturbulent motion begins to play an important role. Pseudoturbulent motion was considered in [3], neglecting Brownian fluctuations. From the results of [3] one can compute the mean square velocity fluctuation of the particles and the self-diffusion coefficients of the medium and the dispersed particles. However, because of nonlinear collective interactions in dispersions, Brownian motion (which smoothes out concentration fluctuations) can be important in diffusion, even when the amplitude of Brownian motion is relatively small. A simple superposition of Brownian and pseudoturbulent motion is not correct in concentrated suspensions because of the nonlinearity of the processes. In the present paper pseudoturbulent diffusion is considered for the same assumptions used in [3], but with the effect of the Brownian motion of the particles taken into account.

Because of the anisotropy of pseudoturbulence, one must consider the self-diffusion tensor. The principal components of this tensor can be represented in the form [2]

$$D_{11} = D_{11}^{(p)} + D^{(b)},\ D_{22} = D_{22}^{(p)} + D^{(b)}, \tag{1}$$

where the superscripts (p) and (b) refer to pseudoturbulent and Brownian motion, respectively. Here the first Cartesian coordinate is chosen to lie along the average relative velocity u of the phases of the suspension. Using the theory of [3], the following relations for the principal self-diffusion coefficients were obtained in [2]:

$$\begin{gathered} D_{11}^{(p)} = \frac{(au)^2}{D_{11}^{(p)} - D_{22}^{(p)}} S(\rho)\,[1-z)^2 I_0 + 2z(1-z)\,I_2 + z^2 I_4], \\ D_{22}^{(p)} = \frac{(au)^2}{2\,(D_{11}^{(p)} - D_{22}^{(p)})} z^2 S(\rho)\,[I_2 - I_4], \\ S(\rho) = 3\left[\frac{2}{9\pi}\right]^{2/3} \rho^{4/3}\left(1-\frac{\rho}{\rho_*}\right)\left[\frac{d\ln M(\rho)}{d\rho}\right]^2, \end{gathered} \tag{2}$$

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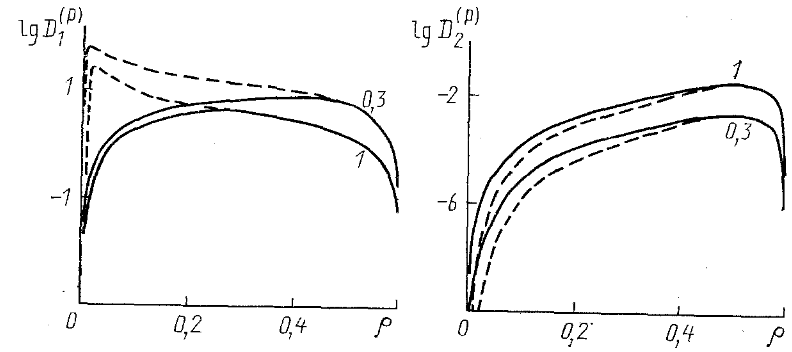

Fig. 1. Dependence of the dimensionless diffusion coefficients $D_1^{(p)}$ and $D_2^{(p)}$ on ρ; the numbers labelling the curves are the values of the parameter κ. Solid curves: with Brownian diffusion taken into account; dashed curves: with $D^{(b)} = 0$.

$$I_n = \int_0^1 \frac{t^n dt}{t^2+\gamma^2},\ \gamma^2 = \frac{D_{11}^{(p)}+D^{(b)}}{D_{11}^{(p)}-D_{22}^{(p)}},\ z = \frac{\rho d_1}{d},$$
$$d = (1-\rho)\, d_0 + \rho d_1,\ M(\rho) = (1-\rho)^{-5/2},$$

where ρ_* is the concentration of particles of the dispersed phase assuming close packing; d_0 and d_1 are the densities of the liquid and solid phases, respectively; a is the particle radius. The function $M(\rho)$ empirically describes the dependence of the effective viscosity of the suspension μ/μ_0 on the concentration of its dispersed phase (μ_0 is the viscosity of the pure liquid).

The self-diffusion coefficient due to Brownian motion is [2]

$$D^{(b)} = D_0 M^{-1}(\rho),\ D_0 = \frac{kT}{6\pi\mu_0 a}, \tag{3}$$

where kT is the temperature in energy units. This equation generalizes the result of [1] (valid only for dilute systems) to concentrated suspensions.

One of the important parameters of a suspension is the concentration of solid particles, therefore the behavior of the self-diffusion coefficients as functions of ρ is of particular interest. We introduce the following dimensionless coefficients:

$$D_1^{(p)} = \frac{D_{11}^{(p)}}{au},\ D_2^{(p)} = \frac{D_{22}^{(p)}}{au}.$$

Then from (2) and (3) we can find the dependence of the dimensionless quantities $D_1^{(p)}$ and $D_2^{(p)}$ on the concentration ρ for the entire interval $0 \le \rho \le \rho_*$ (Fig. 1). The effect of Brownian fluctuations on pseudoturbulent diffusion is evident from Fig. 1, as is the effect of the density ratio $\kappa = d_1/d_0$. The behavior of the self-diffusion coefficients shown in Fig. 1 is explained as follows. As the concentration ρ increases, steric interactions between the particles become more important and this stimulates a migration of particles into a region where the density is relatively small. The decrease of $D_1^{(p)}$ and $D_2^{(p)}$ as $\rho \to \rho_* = 0.6$ reflects the hindrance of self-diffusion in a suspension where the solid phase is nearly close-packed. The difference between the solid and dashed curves at small ρ shows the effect of Brownian diffusion on the smoothing of concentration fluctuations, which is the physical cause of pseudoturbulent motion of the particles (as noted above). It follows from (3) that as the concentration of the dispersed phase increases, the effect of Brownian fluctuations of the particles diminishes; this is also shown in Fig. 1.

A clearer illustration of the effect of Brownian self-diffusion on pseudoturbulent fluctuations is obtained by introducing the dimensionless quantity

$$\delta = \frac{\gamma^2}{1+\gamma^2} = \frac{D_{22}}{D_{11}} = \frac{D_{22}^{(p)}+D^{(b)}}{D_{11}^{(p)}+D^{(b)}},$$

which plays the role of an anisotropy factor for self-diffusion.

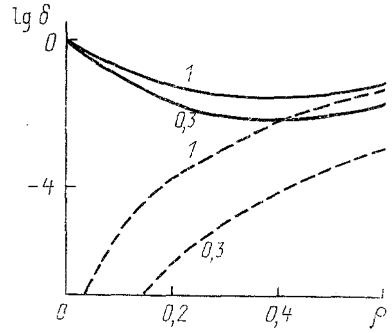

Fig. 2. Concentration dependence of the anisotropy factors δ for self-diffusion with both pseudoturbulent and Brownian fluctuations of the solid particles taken into account (solid curves) and δ_* for self-diffusion with Brownian fluctuations neglected (dashed curves); the numbers labelling the curves are values of the parameter κ.

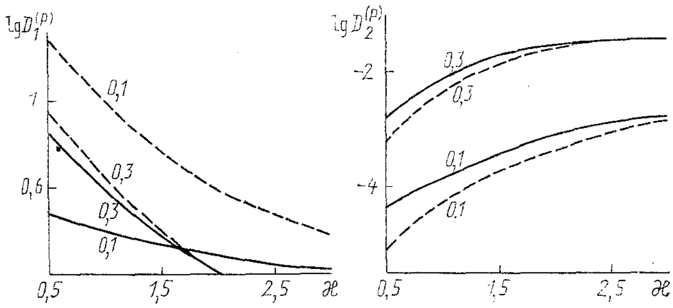

Fig. 3. Dependence of the coefficients $D_1^{(p)}$ and $D_2^{(p)}$ on κ; the numbers labelling the curves correspond to the concentrations of the dispersed phase ρ, the solid and dashed curves are as defined in Fig. 1.

The dependence of this quantity on the concentration of the dispersed phase is shown in Fig. 2 for different values of the parameter $\kappa = d_1/d_0$, together with the dependence of δ_* on ρ, where δ_* is δ when the effect of the Brownian motion of the particles on diffusion is neglected. The dependence of δ_* on ρ is the same as in [3]. The curves deviate at low ρ because the dimensionless anisotropy factor δ approaches unity at low particle cooncentrations, since $D_{11}^{(p)}$ and $D_{22}^{(p)}$ approach zero, while $D^{(b)}$ approaches D_0. Hence at low concentrations we have classical isotropic Brownian diffusion.

It is evident from (2) that there is a fundamental difference between pseudoturbulent and Brownian fluctuations because of the dependence of $D_1^{(p)}$ and $D_2^{(p)}$ on d_0 and d_1. The dependence of the principal dimensionless coefficients of the pseudoturbulent self-diffusion tensor on $\kappa = d_1/d_0$ is shown in Fig. 3.

The results of [2, 3] show that pseudoturbulence becomes more important as the particle radius increases. We see from (2) that the dependence on particle radius is not obvious, since the expression for the Brownian self-diffusion coefficient involves the classical Einstein coefficient, which in turn depends on the particle radius a.

The dependence of $D_{11}^{(p)}$ and $D_{22}^{(p)}$ on a is shown in Fig. 4 for ρ = 0.1 (we saw from Fig. 1 that the difference in the self-diffusion tensor with and without Brownian isotropic motion taken into account is striking for small ρ). The dashed ($D^{(b)} = 0$) and the solid curves approach one another as $a \to 10^{-6}$ because of the decreasing importance of the Brownian motion of the solid particles with increasing particle radius; this is consistent with simple estimates following from (2) and (3).

The expressions of [2, 3] for the mean-square velocities of longitudinal and transverse

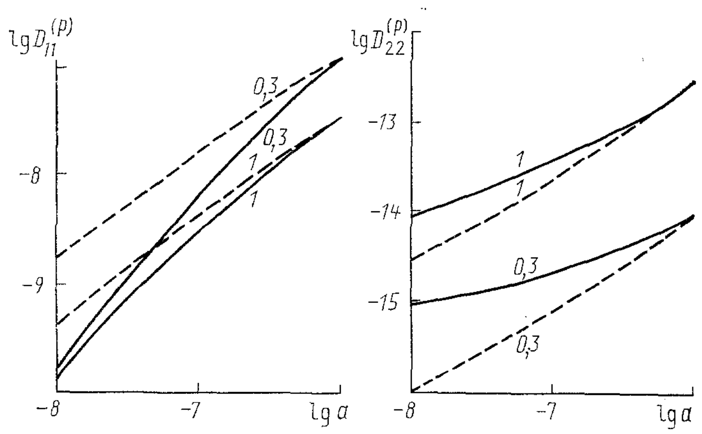

Fig. 4. Dependence of $D_{11}^{(p)}$ and $D_{22}^{(p)}$ on particle radius a; the numbers labelling the curves are values of the parameter κ. The dashed and solid curves are as defined in Fig. 1.

pseudoturbulent fluctuations are independent of the self-diffusion coefficients, in the framework of the assumptions made in [2, 3]. It was also noted in [2, 3] that because of the independence of the physical mechanisms responsible for the generation of Brownian and pseudoturbulent particle fluctuations, one can assume a simple superposition of these motions.

Hence the results of [2, 3] and the present paper describe the behavior of a suspension whose solid phase is subject to Brownian and pseudoturbulent motion.

NOTATIONS

a, particle radius of the solid phase of the suspension; d, average density of the suspension; D, self-diffusion tensor; k, Boltzmann constant; u, average relative velocity of the phases of the suspension; T, temperature; δ, anisotropy factor of the suspension; κ, ratio of the densities of the phases; ρ, volume concentration of the dispersed phase.

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