

Viscosity and Thermal Conductivity of Nanofluids

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Abstract The purpose of the present work was to study the momentum and energy transport processes in nanofluids by the molecular dynamics (MD) method. The MD-simulation results were compared with known formulae and experimental data. Unlike for suspensions with Brownian and other macroscopic particles, the viscosity and thermal conductivity of nanofluids were found to depend not only on the volume fraction of nanoparticles but also on the mass and radius of nanoparticles. The possible mechanisms of the nanoparticle effect on the transport coefficients of nanofluids are discussed.

Keywords: Nanofluids, Viscosity, Thermal Conductivity, Nanoparticle, Molecular Dynamics Method

1. Introduction

Recently, nanofluids have been extensively used in various MEMS technologies, including microflows. At present, however, there is no clear understanding of their transport properties. In particular, in spite of the experimental and theoretical studies of the last ten years (see, for example, Kwak and Kim, 2005; Prasher et al., 2006; Sharma and Singh, 2008), the problem of determining the viscosity and thermal conductivity of nanofluids has not been solved. It has been believed for a long time that the transport coefficients of nanofluids are depended, as in Einstein and Maxwell theories, only by the volume fractions of nanoparticles. However, the kinetic theory of rarefied gas nanosuspensions indicates that the transport coefficients have also to depend on the sizes and masses of nanoparticles (Rudyak and Krasnolutski, 2001; 2003). Recently, there has been experimental evidence for this (Prasher et al., 2006). The purpose of the present work was to study this dependence for viscosity and thermal conductivity coefficients.

Obtaining experimental data on the transport coefficients of nanofluids is complicated by a number of factors, such as difficulties in preparing monodisperse suspensions, problems in measuring particle size and concentration,

etc. The MD method is an alternative approach to study the transport processes in nanofluids. This method is used in present paper.

2. Viscosity Coefficient

The theory of effective viscosity for dilute coarse suspensions was constructed by Einstein (Einstein, 1906). He established that the effective viscosity coefficient η increased in proportion to the volume fraction ϕ of the dispersed particles

$$\eta(\phi) = \eta_0(1 + 2.5\phi), \quad (1)$$

where η_0 is the viscosity coefficient of the carrier liquid.

Subsequent experiments have shown that Einstein's formula is valid only for $\phi \leq 10^{-3}$. Many attempts have been made to extend Einstein's theory to the case of higher volume concentrations. Traditionally, research can be divided into three approaches. First, there are studies in which corrections to the velocity and pressure fields of the carrier liquid due to the interaction of dispersed particles have been determined (Batchelor, 1977). The second approach is concerned with the use of methods of nonequilibrium statistical mechanics (see, for example, Verberg et al., 1997). Finally, in

the third approach, the velocity and pressure fields are simulated using some stochastic laws (Felderhof, 1983). All these approaches yield relations of the form

$$\eta(\phi) = \eta_0(1 + 2.5\phi + k\phi^2), \quad (2)$$

in which the coefficient k differs in different studies, from 5.92 to 6.25.

Strictly speaking, the hydrodynamic approach cannot be used to describe the effective viscosity of nanofluids. Indeed, nanoparticles have characteristic sizes of the order of the hydrodynamic physically infinitesimal scale for the carrier liquid. Nevertheless, in any case, existing theories reduce the problem to a hydrodynamic one.

A small amount of systematic experimental data on the viscosity of nanofluids is available. There have been only a few experimental studies, in which contradictory results have been obtained. Thus, the viscosity of cyclohexane – SiO₂ monodisperse nanofluids with a particle diameter of 28 ÷ 76 nm was measured in Van der Werff et al. (1989). Verberg et al. (1997) argue that results of these measurements are well described by the equation (2). At the same time, the experimental data of Kwak and Kim (2005) obtained for an ethylene glycol – CuO nanofluid with an average particle diameter of 12 nm do not fit this theory. Finally, dependences of the effective viscosity coefficient on the volume fraction for ethylene glycol–Al₂O₃ and water–Al₂O₃ with an average particle size of about 28 nm were obtained in Wang et al. (1999). The results cited above, however, are so different that it is not possible to establish any correlation among them. The authors note that the measured values appear to depend greatly not only on the concentration but also on the method of preparing nanofluids.

In the absence of systematic experimental data for obtaining the effective viscosity coefficients of nanofluids, as an alternative one can use the molecular dynamics method. This was done in the present work. The viscosity coefficient was calculated using the relation (Alder and Wainwright, 1970)

$$\eta = \frac{1}{2VkTs} \left\langle \sum_{i=1}^N [m_i \dot{x}_i(t+s)y_i(t+s) - m_i \dot{x}_i(t)y_i(t)]^2 \right\rangle \quad (3)$$

where V is the volume of the simulation cell, T is the temperature of the system, k is Boltzmann's constant, m , x , and y are the mass and coordinates of a molecule or a nanoparticle, and s is the calculation time; the summation is performed over the entire number of molecules and particles N . Angle brackets denote the average values.

The equilibrium molecular dynamics algorithms and software package MDSib developed by the authors have been tested by solving various problems (see, for example, Rudyak et al., 2001; Rudyak and Belkin, 2003; 2004; Rudyak et al., 2008). The interaction of molecules with each other and with the particles was described by the hard spheres law. The density of the carrier fluid was defined by the parameter $\beta = nd^3$, where d is the diameter of molecules, n is the number density. The ratio of the particle diameter D to the molecule diameter was varied from 2 to 10. The nanoparticle volume fractions were varied from $2 \cdot 10^{-3}$ to $5 \cdot 10^{-2}$, and the ratio of the particle mass M to the mass of the carrier liquid molecule m was varied from 0.5 to 500.

The dependence of the effective viscosity coefficient on the particle volume fraction is well described by the quadratic function

$$\eta(\phi) = \eta_0(1 + k_1\phi + k_2\phi^2). \quad (4)$$

This correlation, however, is not universal; the coefficients k_1 and k_2 are not only functions of the density of the carrier medium but they also depend on the ratio of the masses and sizes of the nanoparticles and molecules. The effect of variation of the nanoparticle mass on the effective viscosity coefficient of the system is illustrated in Fig. 1, which shows dependences of the dimensionless viscosity coefficient η/η_0 (various marks) on the volume fraction of nanoparticles of various masses, here $\beta = 0.283$ and $D/d = 3$. The

solid curves correspond to the approximation (4) with the coefficients given in Table 1.

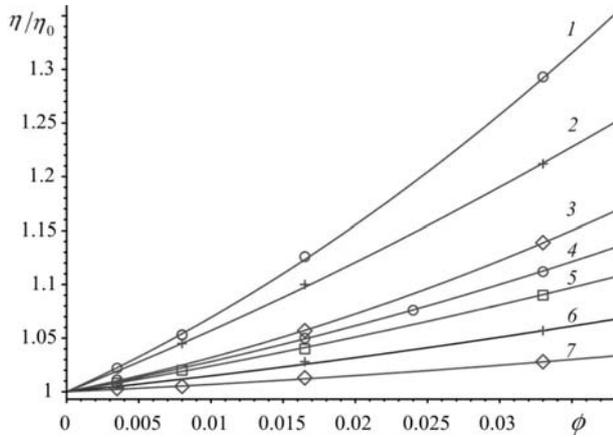


Fig. 1. Viscosity coefficient of nanofluids versus volume fraction of nanoparticles. Curves 1÷7 correspond to $M/m = 500, 300, 150, 100, 50, 10,$ and $0.5,$ respectively.

Table 1. Coefficients k_1 and k_2 of the approximation (4), $D = 3d$, $\beta = 0.283$

M/m	k_1	k_2
0.5	0.61	7.1
10	1.4	10.5
50	2.25	15.0
100	2.6	25.8
150	2.7	44.3
300	4.8	52.2
500	6.1	83.4

It is essential that Einstein's theory does not describe the behavior of the effective viscosity coefficient of nanofluids even for low nanoparticle concentrations. The effect of the mass ratio of nanoparticles to carrier fluid molecules has been previously studied by a nonequilibrium MD method (McPhie et al., 2006). They considered nanofluids with a size ratio $D/d = 1$. In those calculations, the coefficient k_1 was always smaller than the value predicted by Einstein's theory (see equation (1)). We obtained values of this coefficient smaller and larger than the Einstein coefficient for light and heavy nanoparticles, respectively (see Table 1). The possible cause of the discrepancy with the data of McPhie et al. (2006) may be the arbitrariness of

nanoparticle size determination noted by the authors of the paper cited (they used the Weeks–Chandler–Anderson (WCA) potential).

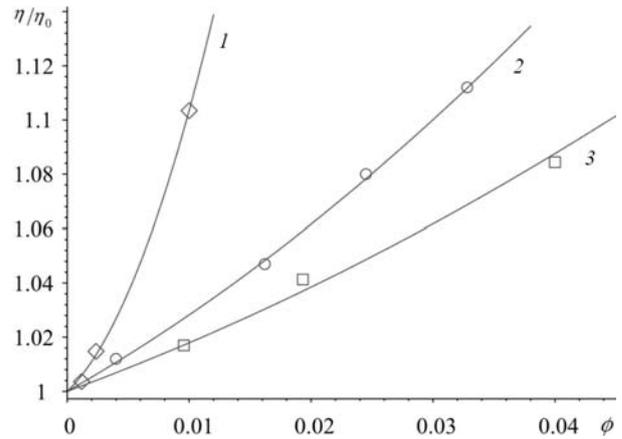


Fig. 2. Effective viscosity coefficient versus volume fraction. Curves 1÷3 correspond to $D/d = 2, 3,$ and $4,$ respectively. $M/m = 100.$

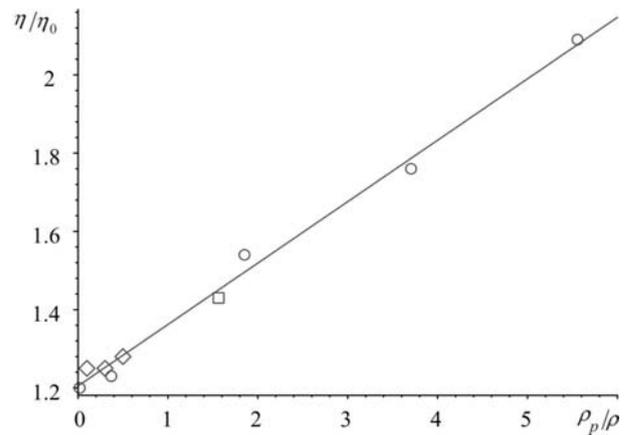


Fig. 3. Effective viscosity coefficient versus density ratio ρ_p / ρ . $\diamond - D/d = 10,$ $\square - D/d = 4,$ $\circ - D/d = 3.$ $\phi = 0.13 \%$.

The applicability of equation (4) is very limited because the coefficients k_i are functions of the diameter ratio D/d . Fig. 2 gives curves of the dimensionless viscosity η/η_0 versus particle volume fraction for a density $\beta = 0.471$ for particles of various sizes but identical masses $M/m = 100$. The nanofluid with larger nanoparticles has lower effective viscosity coefficient. Thus, the viscosity of nanofluids can be changed by varying the density of the nanoparticle material. Fig. 3 shows the effective viscosity

coefficient versus the ratio of the particle material density ρ_p to the molecule density ρ . It is obvious that exactly this ratio determines the viscosity of nanofluids.

The slope of the line presented in Fig. 3 is determined by the volume fraction of nanoparticles. However, the linear nature of the viscosity dependence on the nanoparticle material density is universal for any value of the parameter ϕ . Therefore, it is possible to propose the following simple correlation

$$\eta(\phi) = \eta_0 [a(\phi) + b(\phi)\tilde{\rho}], \quad (5)$$

$$a(\phi) = 1 + 1.25\phi + 2.1\phi^2, \quad b(\phi) = 0.32\phi + 7.2\phi^2,$$

where $\tilde{\rho} = \rho_p / \rho$.

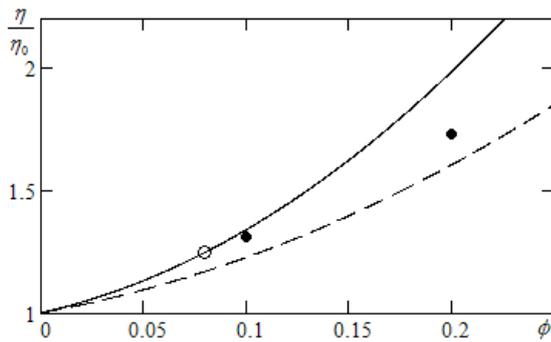


Fig. 4. Comparison of the experimental data of Van der Werff et al., 1989 (•) and Papir and Krieger, 1970 (○) with correlation (5). The solid and dashed curves correspond to $\tilde{\rho} = 1.85$ and $\tilde{\rho} = 0.77$, respectively.

In contrast to equation (4), correlation (5) depends on the volume fraction of nanoparticles and their material density. Therefore, it can be used to predict the viscosity of nanofluids with any size of nanoparticles. As an example, Fig. 4 gives a comparison of the experimental data obtained by Van der Werff et al. (1989) (filled circles, $\tilde{\rho} = 1.85$) and Papir and Krieger, 1970 (open circle, $\tilde{\rho} = 0.77$) and the results obtained using formula (5). Here the solid and dashed curves correspond to density ratios $\tilde{\rho} = 1.85$ and $\tilde{\rho} = 0.77$, respectively. The agreement between the experimental and MD data is not poor, but the accuracy of correlation (5) can be improved by increasing the number of

calculations. In addition, the accuracy of determination of the viscosity coefficient by correlation (5) depends on the accuracy of the density ratio $\tilde{\rho}$ (in particular, the accuracy of the molecule density).

3. Thermal Conductivity Coefficient

According to the Maxwell classical theory, the thermal conductivity coefficient λ of a suspension containing spherical particles at a low particle volume concentration ϕ is described by the equation $\lambda \cong \lambda_0(1 + 3\phi)$, where λ_0 is the thermal conductivity coefficient of the carrier liquid. However, the use of this relation to describe available experimental data from measurements of the thermal conductivity coefficient of nanofluids has not been successful. At present, the thermal conductivity of nanofluids has been the subject of a large number of both theoretical and experimental studies (see, for example, Wang et al. (1999); Patel et al. (2003); Das et al. (2003) and the references therein). Experiments have shown that the thermal conductivity coefficient of nanofluids is abnormally high even at a low volume concentration of nanoparticles. In addition, it depends greatly on the material of nanoparticles, their heat capacity, surface area, etc. However, despite the considerable number of experimental studies, there are still no systematic data to correctly estimate the effect of a particular factor and develop theory of thermal conductivity for nanofluids. In addition, experimental data are often contradictory. In this situation, it is reasonable to perform an "ideal" experiment in which it is possible to control various parameters of the system: the size and mass of nanoparticles, their volume concentration, and the parameters of the carrier liquid. This can be done using the MD simulation. Although this approach is natural, there are still no convincing data from the MD simulations of thermal conductivity for nanofluids. The present paper reports the first results obtained in our studies in this direction. In particular, the dependence of the thermal conductivity coefficient of nanofluids

on the volume concentration and mass of nanoparticles has been investigated. The volume concentration was assumed to be low ($\phi < 1\%$) because many experimental data indicate that nanofluids have abnormally high thermal conductivity at low particle volume concentrations.

The thermal conductivity coefficient was determined using the following relation similar to equation (3)

$$\lambda = \frac{1}{2VkT^2s} \left\langle \left[\sum_{i=1}^N \mathbf{r}_i(t+s)E_i(t+s) - \mathbf{r}_i(t)E_i(t) \right]^2 \right\rangle,$$

where E is the energy of a molecule or a particle. The simulation was carried out using the EMDSib software package, which has been tested by calculations of thermal conductivities of gases and liquids using the MD calculations obtained by Alder and Wainwright, 1970 and different experimental data.

The simulation results show that, similarly to the viscosity behavior, the thermal conductivity coefficient of a nanofluid with a fixed particle size depends on the mass ratio of the nanoparticles to carrier liquid molecules. This dependence is nonlinear and is illustrated in Fig. 5. The figure also gives the results of calculations for three volume concentrations of nanoparticles: $\phi = 0.12\%$ (circles), 0.24% (squares), 0.48% (crosses). Curves 1÷3 correspond to the same concentrations and are described by the following correlation

$$\lambda / \lambda_0 = 1 + b(M/m)^2, \quad (6)$$

where the coefficient b is a function of the volume concentration of nanoparticles and, generally, their radius.

The calculation results given in Fig. 5 show a very large increase in the thermal conductivity coefficient of nanofluids even at a low nanoparticle concentration. Each curve obtained using equation (6) in Fig. 5 is plotted for a fixed radius. This implies that, by simple renormalization, they can be reduced to a dependence of the thermal conductivity

coefficient on nanoparticle density. Thus, unlike viscosity, which increases linearly with density, the thermal conductivity coefficient of a nanofluid increases in proportion to the square of the density ratio: $(\lambda - \lambda_0) / \lambda_0 \sim (\rho_p / \rho)^2$.

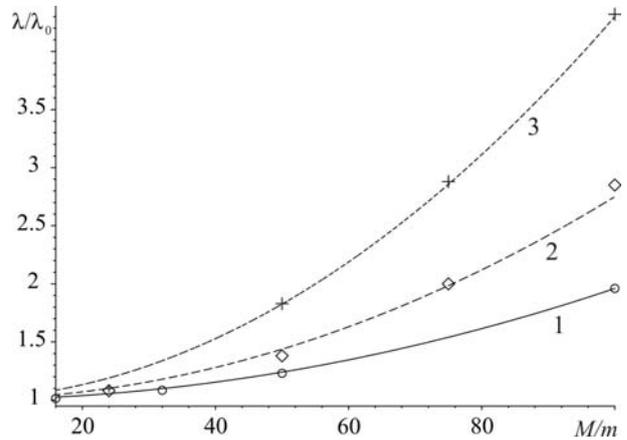


Fig. 5. Effective thermal conductivity coefficient versus mass ratio. Curves 1 ÷ 3 correspond to $\phi = 0.12\%$, 0.24% , and 0.48% , respectively. $\beta = 0.283$, $D/d = 3$.

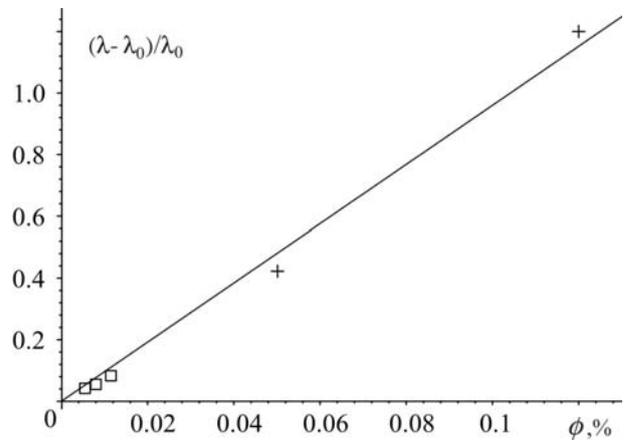


Fig. 6. Comparison of experimental data (Kumar et al., 2004.) (symbols \square) and simulation results, $D/d = 4$ (symbol $+$, $\phi = 0.12\%$), $D/d = 3$ (symbol $+$, $\phi = 0.05\%$)

The large increase in the thermal conductivity of suspensions even at $\phi < 1\%$ is consistent with experimental results. At rather small volume fraction of nanoparticles the thermal conductivity of nanofluid depends linearly on ϕ . This dependence for the toluene–Au nanofluid is presented in Fig. 6. Here the crosses correspond to our MD data and the line

is their approximation. The gold nanoparticle size was about 4 nm. The thermal conductivity of this nanofluid was measured by Kumar et al., 2004. The experimental data (symbols □ in Fig. 6) is good coincided with our calculations.

4. Effect of nanoparticles on transport processes

We see that the addition of a small amount of nanoparticles leads to a considerable increase in the viscosity and especially thermal conductivity nanofluids. The causes of this can be analyzed by considering thermal conductivity, as an example which has been studied in greater detail; at the same time, the indicated causes also refer to viscosity.

In (Kebllinski et al., 2002; Bhattacharya et al., 2004; Ren et al., 2005) several causes of the abnormally high thermal conductivity of nanofluids are noted. The first of them is the additional heat flux through the particle. However, the magnitude of this effect is determined by the particle volume fraction ϕ ; therefore, it cannot explain the observed strong dependence on particle size. The second cause is the thermal motion of nanoparticles. It has been investigated by different authors, and the results are contradictory (Kebllinski et al., 2002; Bhattacharya et al., 2004). The third cause is the effect of nanoparticles on the carrier medium. The simplest model of this effect is a layer with increased thermal conductivity surrounding a nanoparticle (Kebllinski et al., 2002; Bhattacharya et al., 2004; Ren, Xie and Choi, 2005).

The last important factor responsible for the increased thermal conductivity is the formation of clusters. However, for nanofluids with $\phi \leq 0.01$, the effect of this factor is insignificant (Kebllinski et al., 2002).

A very interesting model was suggested by Kumar et al., 2004. In this model it is assumed that there are two different path of heat flow, one though the liquid particles and the other through the nanoparticles. In additional, the second path of heat transfer (through the nanoparticles) is proportional to the nanoparticle surface. For this reason the

thermal conductivity of nanofluid is increased if the particle size is decreased.

In our opinion there are also other reasons which are discussed below. The perturbation of the carrier liquid by nanoparticles is likely to be the basic mechanism underlying the increase in the thermal conductivity for small ϕ . However, the model of a heat conducting layer has disadvantages. The nature of its formation remains unclear. The attempt to explain it by adhesion of the liquid to the surface has not been supported by experiments. The measured width of the liquid–surface contact area (Yu et al., 2000) is comparable to the molecular diameter, which is 2 ÷ 3 times smaller than the value required to explain the abnormal thermal conductivity.

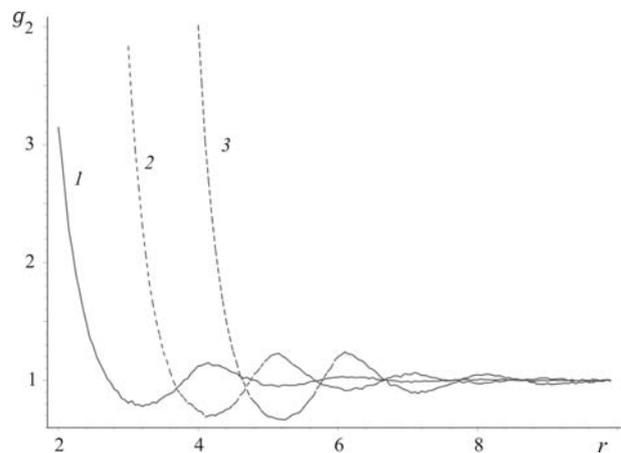


Fig. 7. Pair configuration functions of molecules (curve 1) and nanoparticle–molecule systems with $D/d = 2$ (curve 2) and $D/d = 3$ (curve 3). The distances between the centers are in molecular radii.

The effect of nanoparticles on the carrier liquid, in our opinion, is not reduced only to the formation of a heat-conducting layer. Nanoparticles significantly change the structure of the liquid and the nature of the short-range order, and these changes occur in a large region of space. Figure 7 gives the results of calculations of the pair configuration distribution functions $g_2(r)$ for a nanofluid with a particle volume fraction of 10^{-3} and a mass ratio of 100. The density of the carrier liquid is $\beta = 0.707$.

The heterogeneous medium is more ordered

than the homogeneous medium of the same density. Both the value of the first maximum of $g_2(r)$ and the difference between the subsequent maxima and minima increase. The size of the region of influence of a nanoparticle is comparable to its diameter, and the volume of this region is ten times larger than the volume of the particle. Thus, the effect of a nanoparticle on the structure of the liquid is not local. An increase in the short-range order leads to the fact that the carrier liquid becomes effectively denser and its properties become similar to those of solids. Naturally, its thermal conductivity and viscosity increase in this case.

On the other hand, a nanoparticle perturbs the velocity field of the carrier fluid molecules. Figure 8 shows the response function of molecular velocity $\mathbf{v}_f(t, r)$ to nanoparticles

$$\psi(t, r) = \mathbf{v}_p(0) \cdot \frac{\sum_{i=1}^{N-N_p} \mathbf{v}_f^i(t, r)}{(N - N_p) \langle v_p \rangle \langle v_f \rangle} \quad (7)$$

where r is the distance from the nanoparticle to the i -th molecule and $\mathbf{v}_p(0)$ is the nanoparticle velocity at the initial time.

The motion of the particle leads to the occurrence of a microfluctuation in which liquid molecules move in the direction of motion of the nanoparticle. It is important that the size of this region is not comparable to the diameter of the molecule, as in the case of contact interaction, but it is comparable to the nanoparticle diameter.

The interaction with microfluctuations largely determines the nanoparticle relaxation and diffusion coefficient (Rudyak and Belkin, 2003) and has a significant effect on the viscosity and thermal conductivity. The presence of the region with the preferred direction of the velocity of the molecules leads to slower attenuation of the correlation functions of the heat flux vector and stress tensor. Since the viscosity and thermal conductivity coefficients are time integrals of these functions, this should lead to an increase in the transport coefficients. The correlation

function of the heat flux vector is proportional to the velocity to the sixth power, and the correlation function of the stress tensor is proportional to the velocity to the fourth power. As a result, the interaction with microfluctuations has a stronger effect on the thermal conductivity coefficient.

The characteristic relaxation time of microfluctuations of the average velocity of fluid molecules depends greatly on the nanoparticle mass and size of nanoparticles (Rudyak and Belkin, 2003, 2004). This implies that the effective transport coefficients should depend not only on the volume fraction of nanoparticles but also on their mass and radius. The simulation results confirm this assumption.

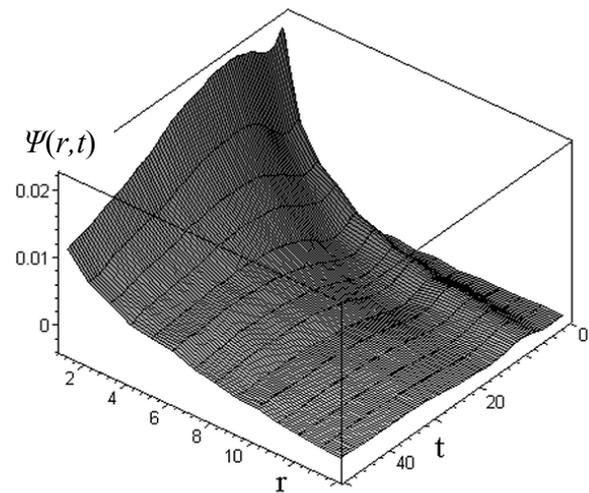


Fig. 8. Correlation function $\psi(r, t)$ in a system with $M/m=100$, $D/d=4$, $\beta=0.29$.

The difficulty in developing a model for the effective transport coefficients of nanofluids is due, in particular, to the absence of a consistent theory of transport processes for simple liquids. For dilute gases, theory is well developed and is easy to interpret qualitatively. In this case, momentum and heat transfer occurs mainly by molecular collisions, and the viscosity coefficient is proportional to the free path length of the molecules.

In studies of dilute gas nanosuspensions, it has been found that the viscosity coefficient is affected by the nanoparticle mass and size (Rudyak and Krasnolutskii, 2001, 2003). In this case, however, the viscosity can both increase and decrease compared to the

viscosity of the carrier gas. In gas nanosuspension, the momentum and energy dissipation processes are caused by collisions of molecules and particles and are determined by the corresponding scattering cross sections. In liquids, where there is short-range order, the momentum transfer mechanism is significantly different. In nanosuspensions, the kinetic collision mechanism of momentum transport makes a minor contribution to dissipative processes. The main mechanisms are those related to the formation of nonequilibrium microfluctuations and an increase in the short-range order. Both these mechanisms lead to an increase in the viscosity and thermal conductivity of the liquid with the addition of nanoparticles. Since these mechanisms are of substantially nonlocal nature, the effect of nanoparticles on the transport coefficients is extremely pronounced.

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