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The nanofluids' viscosity prediction through particle-media interaction layer

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Abstract. This work aims to draw a more fundamental understanding of the rheology of nanofluids and the interpretation of the discrepancy in the literature. The rheology of dispersions based on SiO_2 and Al_2O_3 nanoparticles obtained by four different methods is experimentally investigated in the Newtonian range. It is shown that the viscosity dependence on concentration for nanofluids with particles of different synthesis methods has different values. The parameter of the associated liquid layer model describing the intensity of particles and dispersed medium interaction, as well as the ζ-potential of these liquids, were determined. The correlation between the ζ-potential and the thickness of the associated liquid layer is shown, and the possibility of their use for predicting the behavior of nanofluids is discussed.

Keywords: nanofluids, viscosity, colloid particles, particles distribution, particle-liquid interaction

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1. Introduction

Some of the nanotechnology areas are associated with the use of nanofluids (liquids containing nanoparticles) in various industrial processes. Even a small value of nanoscale additives can both provide special properties to materials and significantly change the conditions of their technological use. Therefore, research in this area is extremely important. In particular, the design of heat engines and heat transfer processes are directly related to the rheological properties of nanofluids due to the variety of their effect on heat transfer [1-3].

A significant number of experiments are presented in the literature regarding the variation of carrier fluid, concentration, size and shape of particles, and their chemical

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composition. However, despite such significant efforts, the formation of an acceptable unified model of the nanoparticles' influence and the possibility of predicting viscosity has not yet been obtained [4-8]. In general, it has been shown that an increase in concentration or a decrease in particle size leads to an increase in the viscosity value. At the same time, many experiments of different scientific groups came into contradiction with each other and gave results according to the following modified model.

Beginning with Einstein [9] and Brinkman, who developed the first theory [10], significant mathematical efforts have been made to reconcile conflicting data.

The main stages of reconciling can be marked as the semi-empirical relationship between Krieger and Doherty (KD model) [11], the Nielsen power-law model [12], the effect of Brownian particle motion performed by Batchelor [13], Lundgren's dependence in the power series form [14], and the Frankel-Akrivos model performed by Graeme [15], taking into account the radius of the particles and the distance between them. All these works make it possible to predict the liquid viscosity under different conditions, depending on the volume concentration of small particles.

However, the experimental values for the influence of nanoparticles are often much higher than the ones calculated by these models, and an example of this is the Batchelor formula [13], which is widely used for a suspension with microparticles. Moreover, as demonstrated in the experiments of different research groups, different types of nanoparticles – even ones of the same substances – can make different values of the effect. This indicates that the hydrodynamic description implemented in existing models, namely taking into account only one (concentration) or two (concentration and diameter) parameters, is insufficient, and it is necessary to consider the additional factors directly related to the nature of nanoparticles, taking into account the interaction of their surface with the environment.

Furthermore, an additional adhesion force of the liquid in the surroundings of the nanoparticles is proposed as a mechanism for such interactions. Concretely, in certain surroundings of the nanoparticles, the free mobility of the molecules of the dispersion medium decreases. From the model point of view, the three-phase Eshelby method can be used here [16], where an intermediate liquid layer is located between the undisturbed dispersion medium and the solid dispersed phase, which is directly connected with the particle. This layer has its own size and elastic properties. In different models, it is called the intermediate phase, hydrodynamic size, associated layer, etc.

In this model, the parameters of the interfacial layer (its size, elastic properties, and adhesion force to the solid phase) are determined by the effect of active centers present on the surface of nanoparticles, which can differ greatly depending on the method of their production. On this note, we can highlight some synthesis factors that affect the concentration and type of active centers, and these factors include the following: the changes in the stoichiometry of surface atoms, the particle size distribution, and the aggregation of nanoparticles.

It can be assumed that the process of developing many new methods is a tireless attempt to take these three synthesis factors into account; however, so far, these methods have not succeeded in understanding this phenomenon. The difficulty of the situation lies in the fact that the intensity of the interaction of the particles and the liquid is not known in advance. Even for nanoparticles of the same chemical composition but obtained by different methods, the adhesion force can differ due to variations in surface properties such as the presence of surface groups, porosity, etc. Therefore, direct consideration of these factors is hardly possible and will require additional use of spectroscopy and other methods not directly related to rheology. To simplify the analysis, researchers will be forced to use semi-empirical models that can implicitly consider these factors. Strictly speaking, the models mentioned above are semi-empirical and try to optimize the obtained experimental data with available mathematical dependences of a complex form.

One of the possible options for taking into account the properties of the surface and the aggregability of particles was undertaken in [17-19]. They variated the aggregation parameter in the KD model [11] and obtained the optimal value was 3.34, giving the good fitting of the experimental results of Ethylene glycol+TiO_2 and other nanofluids.

In our previous work, a simple mechanistic approach was presented to describe this mechanism [20]. We proposed using the Batchelor formula to calculate the viscosity, but we considered the particle diameter to be effectively increased by the region of interaction between the surface and the medium (an associated layer was introduced).

Moreover, as an additional complicating factor, it should be noted that at a specific concentration, which primarily depends on the interaction intensity between particles and liquid, the associated layers of neighboring particles can begin to intersect. Therefore, this may be one of the possible mechanisms for the appearance of nonlinear effects, when a nanofluid can become non-Newtonian, as noted by many authors [5,6].

On this note, this work is a study continuation of the nature of such interactions of nanoparticles and the medium based on experimental data on the viscosity of the epoxy resin and distilled water filled with nanoparticles of silicon dioxide and aluminum oxide synthesized by five different methods, which allows variation in the properties of the surface of nanoparticles. In return, this variability, among other aspects, illustrates the reasons for the significant discrepancy between the experimental data of other authors studying nanofluids based on nanoparticles of the same composition but different surface properties.

2. Materials and methods

In the experiments, aluminum oxide and silicon dioxide nanoparticles obtained by different methods were used. The first character in the name of the samples encodes the method of obtaining the second means the material: "s" – silica, "a" – alumina. One of the most developed gas-phase methods for the synthesis of nanoparticles is the combustion of a material tetrachloride in a hydrogen flame. Variations of this process allow the production of silicon, titanium, and aluminum oxides on an industrial scale. In this work, commercial nanoparticles (Evonik Industries), Aerosil (As series), and Aerooxide (Aa series) were used. The authors of this work participated actively in the development of a gas-phase method based on the evaporation by the continuous high-energy electron beam and the condensation of the substance in the form of nanoparticles in a carrier gas at atmospheric pressure [21]. Furthermore, silica powders obtained by this method are marketed under the Tarkosil brand and have been fully tested (Ts series) while aluminum oxide powders are produced in smaller volumes and have been partially tested (Ta series). Additionally, the work presents results for commercial nanoparticles (Nanjing XFNANO Materials Tech Co., China) obtained by the liquid-phase method (Ls and La series), and two more types of nanopowders obtained by high-temperature methods were used in this work: silicon dioxide powder obtained in high-temperature plasma method (Ps series) (Plasmotherm, Russia) and aluminum oxides obtained by wire electric explosion method (Ea series) [22].

The average particle size and particle size distribution for the powders were measured by the Nanosizer SALD-7500 (Shimadzu). Figure 1 demonstrates the particle size distribution functions (in form of probability density function) for the series of powders used to create suspensions. In this regard, it is seen that the powders used have similar distribution functions. Average sizes vary in the range of ~ 22–34 nm, and the only exception is the Ea series of the aluminum oxide nanoparticle, for which the average size is approximately twice as large and is ~ 64 nm.

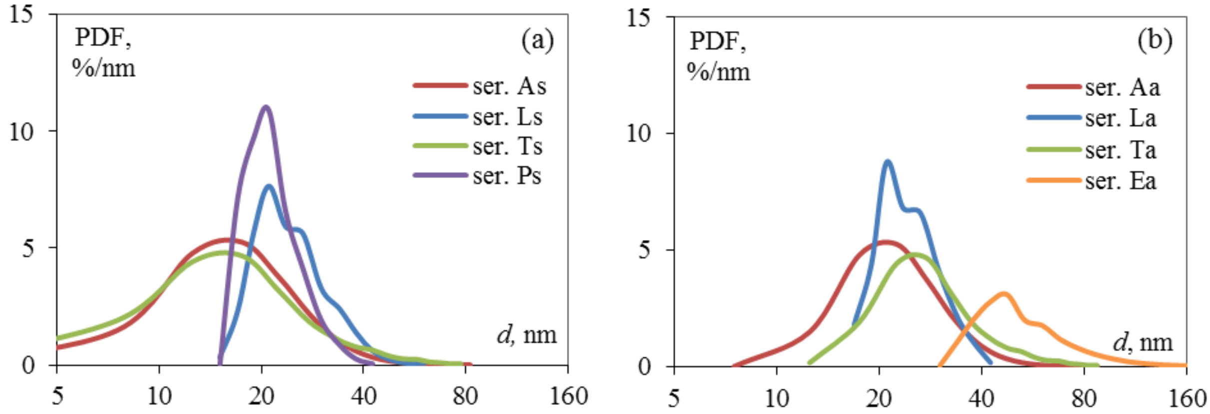

Fig. 1. Probability density function (PDF) for size nanoparticles distribution in series: (a) silica SiO_2; (b) alumina Al_2O_3

For nanofluid preparation, epoxy resins ED-20 and distilled water were used as a dispersion medium. For the deagglomeration of nanoparticles, the suspensions were sonicated for 30 minutes on a Sonopuls HD 3200 (Bandelin Electronic, Germany). This processing time was selected based on the data of the transparency measuring of an aqueous suspension of nanoparticles of the Ts and As series, partially shown in Fig. 2. It can be observed that the nanofluid transparency increases with the time of ultrasonic treatment, and this is due to the homogenization of the suspension and the transition from strong light blocking by large agglomerates to weak light scattering by nanoparticles [23]. Such deagglomeration process reaches saturation after 20 minutes, but since a more viscous liquid was also used in the experiments, the processing time was increased to 30 minutes. The measurements were carried out immediately after the deagglomeration procedure, during 30-60 minutes, when coagulation was insignificant, which was verified.

The viscosity was measured on an MCR 52 rheometer (Anton Paar, Austria) using the cone/plate (CP) measuring scheme. This system has greater accuracy in contrast to the immersed rotor schemes used in the rheometer. This method works well to combine the values and dependencies in a wide range of values, and there is no need to change the rotor for measurements. Viscosity measurements were carried out over a temperature range of 23 to 60°C for the epoxy resin, and measurements for water were carried out at a temperature of 23°C. The relative error of single measurements ranged from 1% at a temperature of 25°C to 5% at a temperature of 50°C and was determined by the technical characteristics of the rheometer. To reduce the coefficient of error, the viscosity was measured sequentially in four consecutive measurements, and the values were averaged. The temperature measurement accuracy was 0.1°C.

Additional preliminary measurements of relative viscosity μ/μ_0 were carried out to determine the effect of temperature and shear rate. Figure 3 shows the temperature dependences of the relative viscosity of the epoxy resin containing various mass concentrations of the Ts series nanoparticles. From Fig. 3, it can be noted that the increase in viscosity caused by the powder occurs proportionally for all presented values of concentration and temperature. On the other hand, Figure 4 shows the dependence of the relative viscosity on the shear rate in the range from 50 to 1000 s^{-1}. Accordingly, it can be observed that the viscosity coefficient is practically constant at a shear rate of more than 400 s^{-1} in the studied range of nanoparticle concentrations. Moreover, similar dependences were also obtained for water and other investigated nanoparticles.

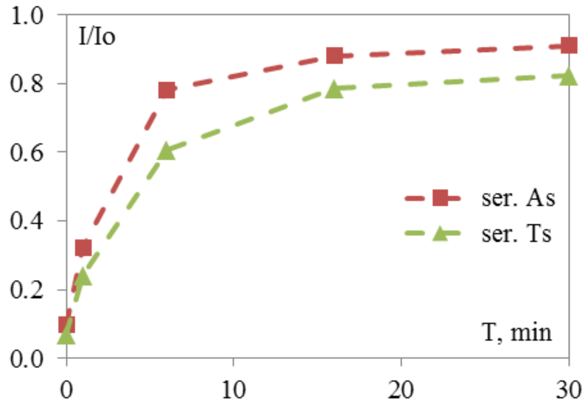

Fig. 2. Measurement of the transparency of the suspension dependence on the ultrasonic treatment time for samples of the Ts and As series

Thus, these measurements have demonstrated that for all suspensions, changes in the relative viscosity do not depend on temperature and shear rate within the current experimental variation limits of these parameters. This means that various nonlinear, thixotropic effects, or complex particle dynamics (for example, their agglomeration or sedimentation) did not manifest themselves under the conditions of the performed measurements. So, in the measuring range, the nanofluids showed the Newtonian behavior. Therefore, this issue is not covered further in this work.

On another note, to measure the zeta-potential, the method of electrophoretic light scattering was used. This method is based on dynamic light scattering in a Laser Doppler Anemometer (ELS) configuration. The measurements were carried out in distilled water on a NanoBrook Omni spectrometer (Brookhaven, USA), and the power of a solid-state laser with a 640 nm wavelength measured 35 mW. A special cell AQR2-0007 (Brookhaven Inst. Corp, USA) was used. The electrokinetic potential was calculated using the Smoluchowski formula: $\zeta = \eta\mu_e / \varepsilon\varepsilon_0$, where ζ – the zeta potential, ε – the dielectric constant of the liquid, ε_0 – the electrical constant, η – the viscosity of the initial liquid, and μ_e – the electrophoretic mobility.

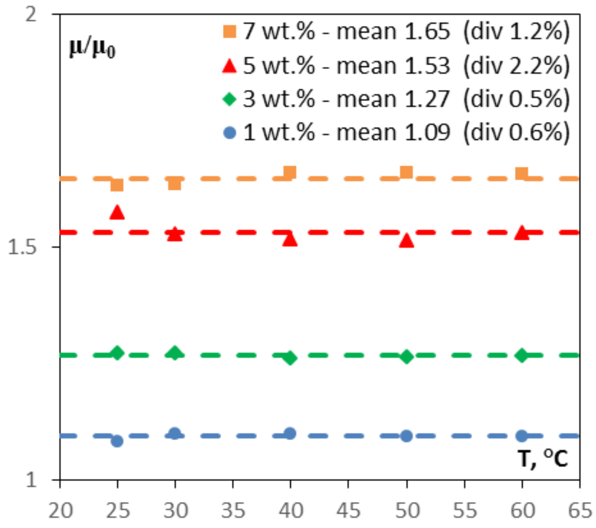

Fig. 3. Dependence of the relative viscosity. of the epoxy resin with different mass concentrations of Ts particles on temperature. Shear rate 500 s^{-1}

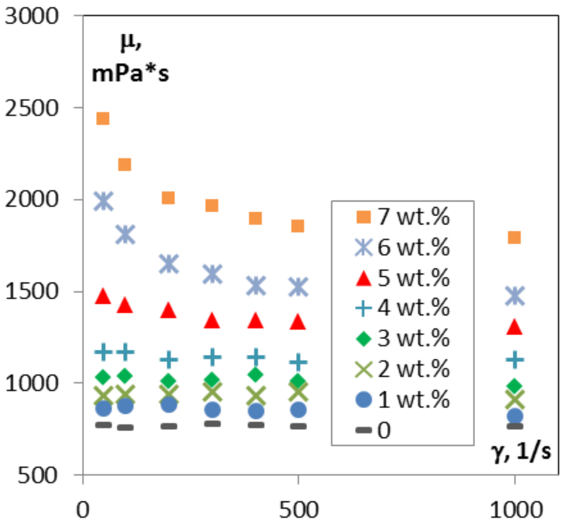

Fig. 4. Dependence of the viscosity of the epoxy resin with different mass concentrations of Ts particles on the shear rate. Temperature 50°C

Furthermore, the theoretical background for our calculations is presented in our previous work [20]. Here, we use some different notations, so we rewrite some formulas. We base our calculation model on the Batchelor formula [13], but we use increased volume concentration of the dispersed component due to the associated liquid layer with thickness δ (effective volume concentration θ_{eff} instead of "clear" volume concentration θ_0). With assumptions about independence of density and associated liquid layer thickness δ from particle size, we get the following equations (1-3):

$$\frac{\mu}{\mu_0} = 1 + 2.5\theta_{eff} + 6.25{\theta_{eff}}^2 = 1 + 2.5K\theta_0 + 6.25K^2{\theta_0}^2, \tag{1}$$

$$\theta_{eff} = \frac{V_{eff}}{V_\Sigma} = \frac{N_{part}}{V_\Sigma}\frac{\pi}{6}\int (d+2\delta)^3 f(d)\mathrm{d}d = \underbrace{\frac{N_{part}}{V_\Sigma}\frac{\pi}{6}E_3}_{\theta_0}\underbrace{\left(1 + 6\delta\frac{E_2}{E_3} + 12\delta^2\frac{E_1}{E_3} + 8\delta^3\frac{1}{E_3}\right)}_{K}, \tag{2}$$

$$\frac{8}{E_3}\delta^3 + 12\frac{E_1}{E_3}\delta^2 + 6\frac{E_2}{E_3}\delta + (1-K) = 0, \tag{3}$$

where K – the coefficient of "increase" in the volumetric concentration of the dispersed phase, V_{eff} – the effective volume of the dispersed phase in the total considered volume V_Σ, N_{part} – the total number of the dispersed phase particles in the volume V_Σ, $f(d)$ – the size distribution of the dispersed phase particles, and E_i – the i-th moment of particle size distribution.

The same notation is used in our previous work [20]. We calculate K from the experimental data for μ/μ_0 through the fitting by equation (1). As opposed to previous works [20], here we have the data for a particle size distribution function of a dispersed phase (Fig. 3) and directly calculate moments E_i for equations (1-3). The last calculation step is solving equation (3) for δ search (it has only one real root despite the cubic form due to positive coefficients for powers of δ).

3. Results and Discussion

Figure 5 shows the dependence of the relative viscosity on the volumetric concentration of nanoparticles of silicon dioxide and aluminum oxide produced by various synthesis methods. Additionally, given are the fitting curves (1) with the value of K. As observed in Fig. 5, our approximation, which is based on the model of the associated layer, is good for the experimental data while Batchelor's formula does not predict enough values of relative viscosity. The gained K value allows us to calculate the associated liquid layer thickness δ in each measurement's series.

As mentioned previously, nanoparticles dispersed in the medium are complex objects. The core of the system is a solid particle (or aggregate) with its own chemical characteristics and uncompensated charges. Consequently, it is covered by a diffuse layer of ions of the dispersion medium. The parameters of the diffuse layer directly determine the ζ-potential of the system. A preliminary study of the functional and chemical composition of the surface of nanoparticles [24,25] showed their difference, which is confirmed by the variation in the value of the ζ-potential (Table 1). As mentioned earlier, the distribution functions of the used nanopowders are close (Fig. 3), but some variation in the average particle size is observed. However, it is not possible to reveal the correlation between the particle size and ζ-potential. This means that the specific feature of the surface structure and the associated value of the ζ-potential is to a greater extent determined by the method of obtaining nanopowders than by the size of the particles.

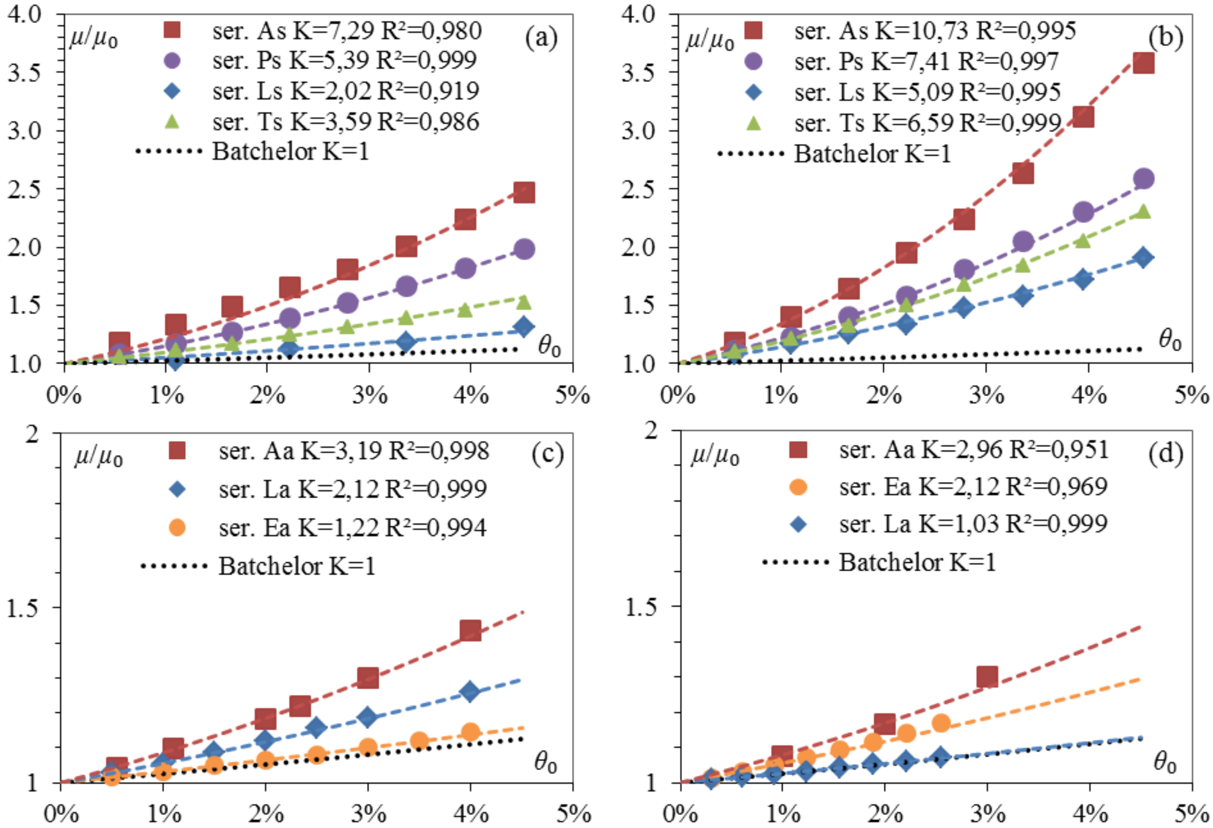

Fig. 5. Dependence of the relative viscosity of the medium with nanoparticles in comparison with Batchelor's formula and fitting curves by (1a) form for series: (a), (c) distilled water; (b), (d) epoxy resin; (a), (b) nanoparticles of silica; (c), (d) nanoparticles of alumina

Table 1. Comparative characteristics of the powders' series

Matter	Type of nanoparticles	Average particle size, <*d*>	*ζ*- potential	Associated liquid layer thickness, *δ*	
		nm	mV	nm	
				Resin	Water
SiO_2	ser. As	22.7	-36.2±0.5	3.7	3.2
	ser. Ls	28.2	-21.2±1.5	2.8	1.8
	ser. Ts	24.3	-30.2±0.6	3.3	2.5
	ser. Ps	23.8	-24.2±0.7	3.0	2.7
Al_2O_3	ser. Aa	27.7	-12.8±0.6	2.2	2.3
	ser. La	27.2	13.3±0.5	0.5	1.8
	ser. Ta	34.3	7.8±0.3	-	-
	ser. Ea	64.3	-17.5±0.4	2.5	1.5

In terms of a mechanistic approach, the nanoparticle moves together with a part of the diffuse layer [26,27]. The size of this part is determined by the structural features of the solid particle and the medium [28], and examples of these features are functional groups on the surface of particles, free bonds of the crystal lattice on the surface, the degree of ionization of the dispersion medium, surface porosity, etc. In the model described earlier (1-3), the parameter δ is responsible for the size of this layer. Therefore, the correlation between the value of the ζ-potential and the thickness of the associated liquid layer δ obtained from the experiments is considered in Fig. 6. For the case of aqueous dispersions, the correlation is not very high, and we can only speak of a general tendency towards a decrease in the associated layer thickness δ as the ζ-potential increases. This may be because the viscosity of water is low and its increase in absolute values is insignificant, which can lead to an error in the calculation of the associated layer thickness. For suspensions based on the epoxy resin, we can talk about the presence of an explicit linear relationship. That is, an increase in the ζ-potential leads to a decrease in the associated layer thickness. The strongest interaction of particles with the medium is formed by particles with the highest positive ζ-potential, and the particles with the highest negative ζ-potential have the least interaction with both resin and water.

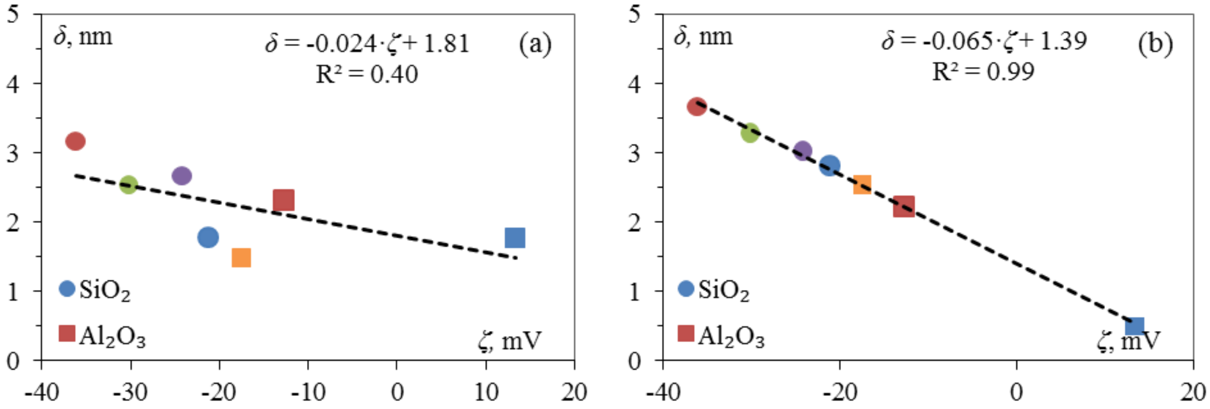

Fig. 6. Correlation of the ζ-potential and the thickness of the associated layer for suspensions based on: (a) distilled water; (b) epoxy resin

It should be noted that the L-series samples obtained by liquid-phase deposition demonstrate a different behavior compared to other samples obtained by methods of an intense heating of the starting materials. Both samples of the series (Ls and La) have the smallest values of the ζ-potential and the associated layer. In the case of alumina particles (La), this is one of the only two samples that show a positive ζ-potential. Thus, it can be concluded that the conditions for the synthesis of these samples lead to the creation of a charge structure on the surface of the particles, and this structure interacts with resin and water and is weaker than the one corresponding to the samples obtained by other methods.

Since the direct consideration of the variations in surface-active centers, the presence of porosity, and other surface features is an extremely complex problem, the determination of the ζ-potential and the thickness of the associated liquid layer can be an effective substitute for them in the rheology of nanofluids.

Having obtained such dependences of the attached layer thickness δ for each liquid and, consequently, the value of K on the value of the ζ-potential, we can predict its viscosity. The parameter in the Batchelor equation (1) will be the value of K, which directly depends (3) on the ζ-potential of dispersion and the moments of particle size distribution. This equation will be valid for a specific liquid with any nanoparticles, at least in the region where the suspension will retain Newtonian behavior.

To some extent, these factors can explain the occurrence of nonlinear effects [28,29]. If the physical concentration of particles becomes high enough that the associated layers of neighboring particles begin to intersect, then the unperturbed dispersed medium will disappear. Then, the value of the dispersed medium momentum will decrease in comparison with the sum of the momentum of the particles, and the effective viscosity will increase. That is the way for the appearance of the non-Newtonian effects. Calculated predictions here can be difficult since they require preliminary practical measurements to determine the intensity of the interaction between particles and between the associated layers of particles themselves.

It is necessary to note two special points in the obtained ratios of the associated layer thickness and the zeta potential value, which indicate the limitations of this model. The first is the zero value of the associated layer thickness, to which the dependence tends when the zeta potential is more than +20 mV. We assume that at large values of the zeta potential, the dependence ceases to be linear and becomes asymptotic to zero thickness of the associated layer. The second singular point corresponds to the zero value of the zeta potential, where the nanofluid is absolutely unstable and there is no electric double layer. At this point, our model does not apply. It is likely that the dependence will have a discontinuity at this point, with an asymptotic approach to the point (ζ=0, δ=0) on both sides. These features need to be experimentally verified using nanoparticles having the necessary zeta potential values.

4. Conclusion

This study demonstrates that dispersions based on SiO_2 and Al_2O_3 nanoparticles obtained by different methods have different viscosities, which are apparently determined by the value of the interaction between particles and medium. To determine the intensity of this interaction, a three-phase model of the associated layer is used, which describes the deviation of viscosity from Batchelor's law with high accuracy. The values of the thickness of the associated layer and the ζ-potential of the investigated liquids are determined and their correlation is shown. Furthermore, the possibility of using these parameters to predict the behavior of nanofluids is proposed.

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