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# The role of associated liquid layer at nanoparticles and its influence on nanofluids viscosity

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ABSTRACT

This article describes the work which defined the relationship between the viscosity of nanofluids, containing silica nanoparticles, temperature and the concentration and the size of the particles. The nanopowders produced by two different methods have been used and the results compared. The difference in a way they affect the viscosity of the nanofluids has been studied and explained. It was identified that the viscosity depends on the value of a specific surface in accordance with the linear law. The mechanism to increase the value of the volume ratio of the solid phase by the means of associated layer of the colloidal particles has been proposed. The assessment has been made of the thickness of the associated layer of liquid on the surface of the silica nanoparticles which have been dispersed in water and epoxy resins. The application of the Bachelor formula has been demonstrated for the nanoparticles taking into account the volume ratio of the associated layer of liquid.



## 1. Introduction

One of the trends in nanotechnology is connected to the use of nanofluids in various industrial processes. Even small additives of the nanoparticles provide the special properties for materials – this makes the development of this field to be extremely important.

The problem of the nanofluids viscosity dependence on nanoparticles concentration and the possibility of viscosity prediction are still not clear, despite many experiments were undertaken to clarify this matter, e.g. [1–3]. In general it was shown that increasing the concentration and decreasing the particles size results in the viscosity growth. However, the experimental data for relative viscosity $\mu/\mu_0$ is significantly higher than the values calculated by the formula obtained by Batchelor [4], widely used for microparticles' suspension:

$$\frac{\mu}{\mu_0} = 1 + 2.5\psi_p + 6.25\psi_p^2, \quad (1)$$

where $\psi_p = \frac{V_p}{V_{tot}} = \frac{\varphi_p/\rho_p}{\varphi_p/\rho_p+(1-\varphi_p)/\rho_m}$ is volume concentration of particles, $\varphi_p$ is mass concentration of particles, $\rho_p$ is particle density, $\rho_m$ is density of liquid medium, $\mu$ is viscosity of nanofluid, $\mu_0$ is viscosity of primary liquid.

Alternatively, as it was shown by Mahbubul et al. [1], they attempted to describe the data by the other relationship of viscosity (power law, exponential law, etc.) from the particles volume concentration. However, the results were expressed as arbitrary selection of two-, three- parametric dependences in order to adapt the empirical data and did not provide an understanding of the mechanism of such a strong growth of viscosity due to the nanoparticles.

The authors believe that the studies of the mechanism of fluid viscosity with nanoscale powders should be carried out considering the interaction of dispersion medium and nanoparticles at the mesolevel. The adhesion of fluid at the hard spots surface is suggested as a mechanism of such an interaction, i.e. reducing mobility of dispersion medium molecules being near the particles. The attempts to consider this factor were done before by Graham [5], Kitano et al. [6], Masoumi et al. [7], but they did not lead to success in understanding of the phenomenon. In our opinion, to understand the influence of nanoparticles on the dispersion medium it is necessary to consider the structure of the nanoparticles surface.

This work is an attempt to correct the ratio (1) by experimental data on the viscosity of epoxy resin and distilled water filled with

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nanoparticles of silica varying their size, concentration and the characteristics of the nanoparticles surface. The latter is determined by the difference of the Tarkosil and Aerosil nanoscale powders.

## 2. Materials and methods

The viscosity measurements of three media with two kinds of silica were made. As the dispersion medium we used the epoxy resins ED-20 (Sverdlov's Factory, Russia) and DER 330 (The Dow Chemical), as well as the distilled water. The viscosity was measured at the rotary viscometer "Smart" (Fungilab) by standard procedure. The speed of rotation of spindles was ranged from 3 to 60 turn/min.

To reduce the factor of error the viscosity was measured sequentially in four equal tanks, with 80 mm diameter and 100 mm height, located into common thermostatic volume. The temperature was changed step by step in range from 25 up to 50 °C for resins. The measurements for the water were carried out at the temperature of 25 °C. After measurements the viscosity values from four tanks were averaged.

The factor of error in measurements was in the range from 1% at the temperature 25 °C and up to 5% at the temperature 50 °C. It was determined by the technical characteristics of the viscometer. The accuracy of temperature measurement was 0, 1 °C.

To check whether the fluid is Newtonian we measured the dependence of viscosity on the spindle speed of the viscometer. It emerged that when the spindle speed is higher than 1 turn/min., the viscosity does not depended on the rotation speed, i.e. the nanofluids studied can be classified as Newtonian.

For the experiments we used (a) the silica nanopowders of Tarkosil (T series) produced by the original technology of Bardakhanov et al. [8], Bardakhanov et al. [9]. These nanopowders have the specific surface 53, 74, 96, 123, 150 $m^2/g$. (b) commercial powders Aerosil (A Series) ("Evonik Industries") with specific surface 90, 120, 200, 300, 380 $m^2/g$.

Specific surface and average particle size (APS) for the powders were measured by liquid nitrogen adsorption method (Brunauer-Emmett-Teller analisys, BET) by standard procedure. The nitrogen was used as gas-adsorbent. The specific surface measurements were carried out at 6, 9, 15, 20 percent concentration of the nitrogen in the helium/nitrogen mixture. Transmission Electron Microscopy (TEM) by JEOL-2010 microscope (together with particle size analysis software for this instrument) was used for the measurements of particle size distribution (PSD).

A special attention was paid to the dispersion process of the powders into media. To avoid nanoparticle agglomeration all samples were subjected to ultrasound with power 250 W and frequency 22 kHz for 30 min by the disperser Sonopuls HD 3200 (Bandelin Electronic) after the powder was dispersed. All the measurements of the viscosity were carried out within three hours after this process.

Viscosity measurements $\mu(\varphi_p, T)$ were held at a fixed concentration of the powder and with the temperature of 5 °C steps. The values obtained were divided to the viscosity of the pure liquid $\mu(0, T)$ at the same temperature. Fig. 1 shows the viscosity ratio of the DER 330 containing the powder T150 on the temperature for various mass concentrations of the nanopowders. It is evident, that the relative viscosity caused by the powder does not practically change with the temperature.

This shows that nonlinear effects associated with the agglomeration of powders and thixotropic effect have been avoided. So we can separate the two main factors: nanopowders and temperature.

Similar dependences were obtained for other dispersion mediums such as ED-20 and water, as well as for Aerosil powders.

## 3. Results and discussion

### 3.1. Experimental data

Fig. 2 represents the dependences of relative viscosity on the mass concentration of powders in various combinations. As it was shown by Mahbubul et al. [1], the experimental values of viscosity for various powders have significant discrepancies with the formula (1) and between each other. In particular, Fig. 2 shows a significant discrepancy of viscosity values with Tarkosil and Aerosil fillers. We can assume the interaction between the dispersion medium and the nanoparticles to have a special feature in comparison with the microparticles ones because of surface forces.

The studies of the functional and chemical composition of the nanoparticles surface by Bardakhanov et al. [10] showed more absorption centers corresponding to the silicon atoms on the Aerosil nanoparticles surface and to the oxygen atoms in Tarkosil nanoparticles. So, Aerosil causes selective surface absorption of oxygen atoms, but Tarkosil's absorption is based on the weaker monovalent bonds.

This discrepancy is confirmed by the value of ζ-potential measured by Bulavchenko and Popovetsky [11]. Their measurements have shown −73 mV for Tarkosil aqueous dispersions, and −27 mV for Aerosil ones. In the case of Aerosil, the presence of surface quadrivalent ion increases its electrostatic attraction force, thus causing a greater compression of the diffusion layer and respectively reducing the ζ-potential. This fact results in establishing stronger bonds between the dispersion medium and the solid phase, so, the molecular diffusion decreases and the viscosity of dispersion increases as a whole.

For this reason one can distinguish Tarkosil and Aerosil as various kinds of silica nanopowders.

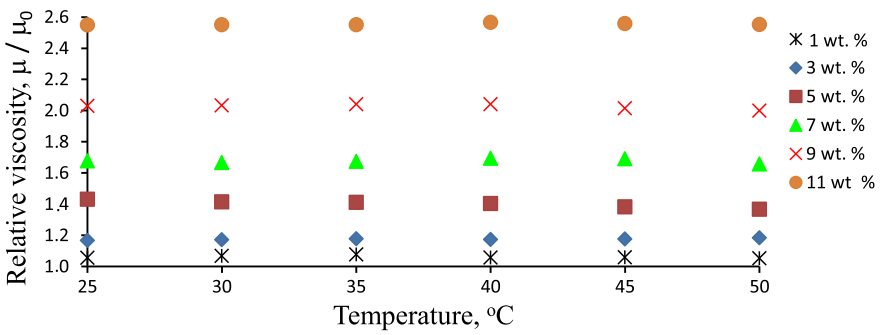


**Fig. 1.** Dependence of relative viscosity of the DER 330 resin with various mass concentration of the T150 powder on the temperature.