Surface and optical properties of ethylene glycol-based nanofluids containing silicon dioxide nanoparticles: an experimental study

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Abstract

In this paper, the results of the experimental study on density, surface tension and optical properties of ethylene glycol-based nanofluids containing silicon dioxide have been presented. The nanofluid-air surface tension values were determined in the temperature range from 283.15 K to 318.15 K with Du Noüy ring method with the commercial equipment and self-made station. To determine the surface tension of a fluid, the information about its density is required; therefore, density of all examined nanofluids was measured in the same temperature range, and results were summarized in the paper. The refractive index of SiO_2-EG nanofluids was determined in the same temperature range in which the surface tension was examined. Finally, the extinction (UV–Vis spectra) of these materials was examined and presented.

Keywords Nanofluid · Silicon dioxide · Ethylene glycol · Surface tension · Optical properties

Introduction

Nanofluids, which are suspensions of nanoparticles (with at least one diameter less than 100 nm), exhibit unique physical properties as it was summarized in some recent review papers [1–6]. This kind of properties makes nanofluids an interesting material for the numerous possible applications [7–11]. One of the promising possible application of nanofluids, based on its complex optical properties, is using them in the renewable energy industry, especially in solar collectors and PV/T systems [12–18]. Beside the fact that nanofluids are known for over 25 years know, the scientific community still does not have a coherent theoretical model describing the physical properties of nanofluids. Numerous scientific teams tries to develop theoretical models; however, such works require detailed, reliable experimental data which in many cases are missing in the literature. This paper brings a contribution on this field and presents results of experimental study on density, surface tension and optical properties of ethylene glycol (EG) based nanofluids containing silicon dioxide (SiO_2) nanoparticles.

The physical properties of ethylene glycol-based nanofluids containing SiO_2 were already the subject of research. Żyła and Fal [19] presented results of the study on the dynamic viscosity, thermal and electrical conductivity of SiO_2-EG nanofluids containing up to 5.0 mass% of nanoparticles at 298.15 K. They showed that in examined mass fraction range nanofluids exhibit Newtonian nature and thermal conductivity could be described with classical Maxwell model. Li et al. [20] examined the dependence of thermal conductivity of SiO_2-EG nanofluids on the time of sonication during preparation process. The specific heat of various types of nanofluids with SiO_2 nanoparticles was examined by Akilu et al. [21]. It was summarized that there is a decrease of nanofluids specific heat with increasing mass fraction of nanoparticles in a SiO_2-EG nanofluids. Recently, Alazwari et al. [22] used ANN and RSM techniques to evaluate the thermal conductivity of SiO_2-EG nanofluids and possible applications of it. Fal et al. deepened the understanding of SiO_2-EG properties with information on the effect of temperature on electrical conductivity [23]. Additionally, they developed a ethylene glycol-based silicon oxide-lignin nanofluids, and presented a comprehensive study on its electrical and dielectric properties [24].

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According to the best of the authors' knowledge, there are no literature data on the surface tension of SiO_2-EG nanofluids and there are only a few papers on the surface tension (ST) of ethylene glycol-based nanofluids available in the literature. Moosavi et al. [25] show an increase in surface tension of ZnO-EG nanofluids with increasing volume concentration of nanoparticles. Harikrishnan et al. [26] used pendant drop method to measure surface tension of Bi_2O_3-EG and CuO-EG at 303.15 K. They observed that the surface tension of the tested nanofluids did not increase with the mass concentration of nanoparticles in suspension. Wanic et. al. [27] presented results of surface tension for the pendant drop and Du Noüy method for TiN-EG, AlN-EG and Si_3N_4-EG nanofluids and both methods show that ST increases slightly with the concentration of nanoadditions.

The optical properties of nanofluids are interesting from the point of view of possible applications of such materials and from the point of view of basic science. Again, according to the best authors' knowledge SiO_2-EG nanofluids were not subject of the evaluation of optical properties. However, one might find some papers on the optical properties of other ethylene glycol-based nanofluids. Żyła et al. [28] showed that for Si_3N_4-EG nanofluids, the transmittance of the nanofluid increases with increasing wavelength and decreases sharply with increasing nanoparticles volume fraction. Sani et. al. [29] studied nanodiamond particles in ethylene glycol and showed a much lower transmittance than pure base fluid, leading to high sunlight absorption by these nanofluids with promising potential in direct absorption solar collectors. Di Rosa et al. [30] examined optical properties of EG-based nanofluids containing nanodiamonds with two various purities. Torres et. al. [31] compared samples of TiN-EG nanofluid prepared with pulse laser ablation in liquids (PLAL) and with two-step method. It was presented that the nanofluid synthesized by PLAL showed a lower transmission spectrum for the visible wavelengths. The refractive index of ethylene glycol-based nanofluids was subject of studies performed by Żyła et al. [28] and Sobczak et al. [32] for Si_3N_4-EG and carbon black nanofluids, respectively.

In this experimental work, the surface and optical properties of SiO_2-EG nanofluids with mass fraction change from 1 mass% to 5 mass% at the temperature range of 283.15–318.15K were investigated to fill this gap in nanofluids research.

Materials and methods

In this section, the sample preparation and measuring procedures were described along with relative uncertainty values.

Sample preparation

The nanoparticles used in this study were manufactured by PlasmaChem GmbH (Berlin, Germany); according to manufacturer declaration, nanopowder exhibit 99.8% purity. The detailed information, about physical properties of this material, was presented elsewhere [19]. Figure 1 shows the scanning electron microscope (SEM) picture of used nanoparticles.

In the experiments ethylene glycol (EG), with purity over 99% produced by Fisher Chemical (Loughborough, UK), was employed as a base fluid. All samples were prepared in 50 mL vials using two-step method. Firstly, the specific amount, which was weighted on an analytical balance Pioneer Semi-Micro PX225DM (Ohaus, Poland) with an accuracy of 0.01 mg, of nanopowder was putted into the vial. The second step of the preparation consisted adding the correct amount of EG in order to obtain the right concentration. After that, sample was mechanical mixed on IKA Vortex 3 shaker (IKA, Staufen, Germany) for 30 minutes. In order to ensure well dispersion of nanoparticles and avoid agglomerates for a period of 200 min samples were kept in Emmi 60HC ultrasonic water bath (EMAG, Moerfelden-Walldorf, Germany). The final step included immersion each sample in a high-energy ultrasonic probe (Sonics, Vibracell VCX 130, Sonics & Materials Inc., Newtown, USA) for 5 minutes. The samples were prepared in various mass concentrations of the particles from 1 to 5 mass% with 1 mass% step. During preparation process, no surfactants were used and the
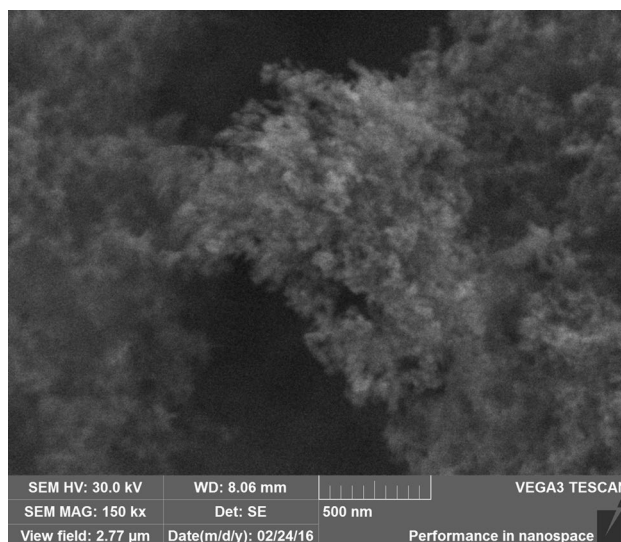

SEM HV: 30.0 kV	WD: 8.06 mm		VEGA3 TESCAN
SEM MAG: 150 kx	Det: SE	500 nm	
View field: 2.77 µm	Date(m/d/y): 02/24/16		Performance in nanospace

Fig. 1 Scanning electron microscope (SEM) picture of dry SiO_2 nanoparticles. Reproduced with permission from Ref. [19], license number 5080150950041

measurements were conducted immediately after sample preparation.

Fourier Transform Infrared (FTIR) measurements

Samples were analyzed by using a Thermo Scientific Nicolet iS5 with ATR iD5, with resolution of 4 cm^{-1}, in a range of wave number from 4000 to 400 cm^{-1} and with a scan number of 64. The attenuated total reflection (ATR) is a technique of infrared spectroscopy. The ATR analysis is based on internal reflection at a diamond with high refractive index placed in direct contact with the sample. The radiation passes through the diamond and is refracted. Then, it strikes the sample several times and penetrates for about 2 µm and is reflected, recrossing the crystal and reach the detector DLaTGS.

Mass density measurements

Density measurements of the studied nanofluids were performed using an automatic oscillating U-tube density meter DMA 4100 M (Anton Paar, Graz, Austria). The nanofluid samples were measured over a temperature range of 285.15–320.15K with a step of 5K. Three measurement series were conducted for each temperature with ten measurements in each one. The uncertainty of the measured values derived from this instrument was determined by the deviation between the experimental data and the corresponding value in the literature and the standard deviation of one hundred measurement points of distilled water at 298.15 K. For these one hundred measurements, the water mass density obtained was 0.9969 g cm^{-3} with a standard deviation of 0.0002 g cm^{-3}. The literature [33] states that the density of water is 0.997066±0.000001 g cm^{-3}. Taking into account the literature values and the achieved experimental results, the relative uncertainty of the density measurements can be determined as 0.1%.

Surface tension measurements

Surface tension measurements were performed on a PI-MT1A tensiometer (Polon-Izot, Warsaw, Poland). To measure surface tension, this device uses the Du Noüy ring method. The surface tension of the studied nanofluids was conducted in the temperature range 285.15–320.15K with a step of 5K. Ten surface tension measurements were taken for each temperature and the given experimental values were calculated as the average. The uncertainty of this device was determined as 0.03 mNm^{-1} [27]. Taking into account all these factors, the relative uncertainty was estimated as 1%. Additionally the results obtained with the commercial device at the room temperature were compared with home-made

tensiometer construction of which is presented elsewhere [34].

Refractive index measurements

The refractive index was determined using Refractometer Abbemat 550 (Anton Paar OptoTec GmbH, Seelze-Letter, Germany). After setting the air reference in measuring chamber a 500 µL of a sample was placed with HTL Discovery Comfort DV1000 single channel pipette (HTL Lab Solution, Warsaw, Poland)—an accuracy of volume is 0.5%. The value of the refractive index was calculated based on the average of three series of measurements. The relative uncertainty of used device is less than 1% as it was presented elsewhere [28].

UV–Vis measurements

Light extinction of silicon dioxide–ethylene glycol nanofluids was measured with a UV–Vis spectrometer Evolution 220 (ThermoFisher SCIENTIFIC, Waltham, MA, USA) with an absorbance range of 190 nm to 1100 nm. The research was carried out with inspection range from 190 to 800 nm with a 1 nm gap and an integration time of 0.30 s. The scanning speed was set to 200.00 nm min^{-1}. The tested samples were contained in 3 mL quartz cuvettes.

Results

In this section, the results of measurements of fundamental physical properties of SiO$_2$-EG nanofluids at the temperature of 298.15K were presented and discussed. The complete results of measurements in the temperature range 283.15–308.15K are included Appendix 1.

FTIR

Figure 2 shows the FTIR spectra of SiO$_2$-EG and pure ethylene glycol as a reference sample. All samples show the identical characteristic spectra for ethylene glycol, therefore analyzed spectra show vibrations: 3299 cm^{-1} (OH group), 2937 cm^{-1} (CH$_2$/CH$_3$ group), 1410 cm^{-1} (CH$_3$ group), 1366 cm^{-1} (CH$_3$/CH$_2$ group), 1083 cm^{-1} (CH$_2$/CH group). This proves that the samples are stable and no new chemical bonds are formed.

Mass density

The Table 1 shows the obtained mass density values of nanofluids with different concentrations at 298.15K. The base fluid density measurements stand in high agreement
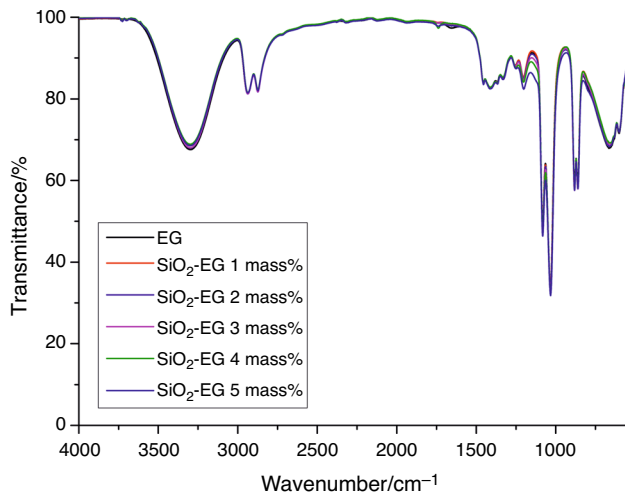
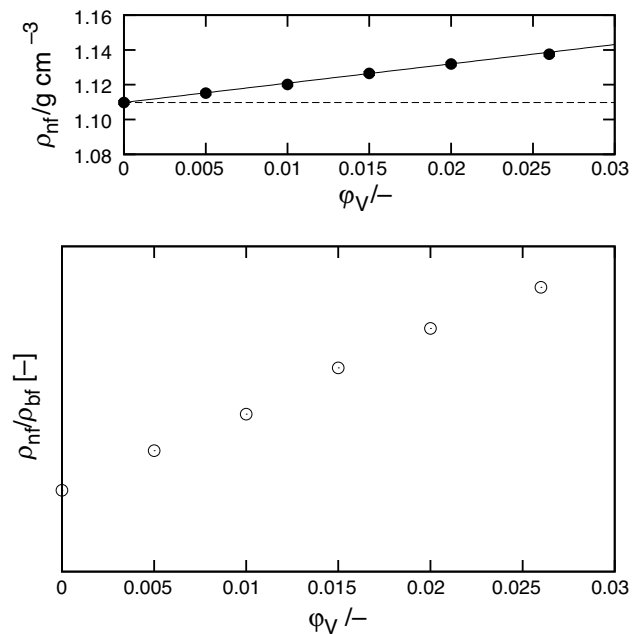Fig. 2 Fourier transform infrared (FTIR) spectra of SiO$_2$-EG nanofluids and pure ethylene glycol

Fig. 3 Dependence of density, ρ, and density ratio, ρ_{nf}/ρ_{bf}, on the volume fraction, φ_V, of the particles for the SiO$_2$-EG nanofluids at 298.15 K. Dotted line presents the value of pure ethylene glycol, solid line shows function (1)
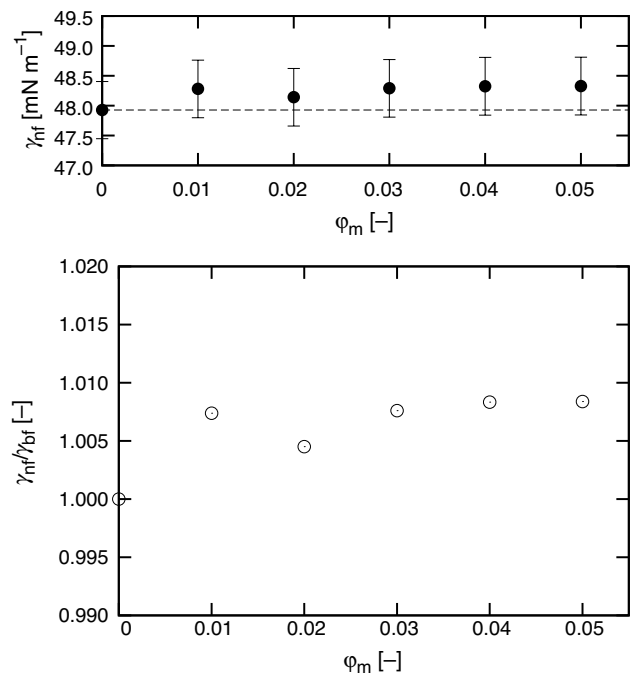
with our previous works [32, 35]. In Fig. 3, a linear increase in the density of the nanosuspension can be observed with increasing mass fraction of nanoparticles.

More than 20 years ago, Pak and Cho [36] proposed the following equation to describe this dependence:

$$\rho_{nf} = (1 - \varphi_V)\rho_{bf} + \varphi_V\rho_p, \qquad (1)$$

where ρ_{nf} is density of nanofluid, ρ_{bf} is density of the base fluid and ρ_p is density of particles, φ_V is the volume fraction corresponding to the mass fraction φ_m by:

$$\varphi_V = \frac{\varphi_m}{\rho_p\left(\frac{\varphi_m}{\rho_p} + \frac{1-\varphi_m}{\rho_{bf}}\right)}. \qquad (2)$$

This correlation was based on ideal gas mixing theory. According to the authors, this equation allows to model the

Fig. 4 Dependence of surface tension, γ, and surface tension ratio, γ_{nf}/γ_{bf}, on the mass fraction, φ_m, of the particles for the SiO$_2$-EG nanofluids at 298.15 K. Dotted line presents the value of surface tension of pure ethylene glycol

Table 1 Experimental values of the density, ρ, surface tension, γ, and refractive index, n, of SiO$_2$-EG nanofluids at pressure $p=0.10$ MPa and temperature $T=298.15$ K for various mass fractions φ_m, and volume fractions φ_V of nanoparticles

φ_m / −	φ_V / −	ρ_{nf} / g cm^{-3}	γ_{nf} / mN m^{-1}	n_{nf} / −
0.00	0.000	1.1098	47.926	1.43025
0.01	0.005	1.1152	48.280	1.43047
0.02	0.010	1.1202	48.142	1.43061
0.03	0.015	1.1265	48.290	1.43077
0.04	0.020	1.1319	48.325	1.43092
0.05	0.026	1.1375	48.328	1.43106

The estimated standard relative uncertainty $u_r(\varphi_m) = 0.01$, $u_r(\varphi_V) = 0.001$, $u_r(\rho) = 0.001$, $u_r(\gamma) = 0.01$, $u_r(n) = 0.01$, $u(p) = 0.01$ MPa and $u(T) = 0.10$ K

density of nanofluids within a few percent accuracy. The experimental results obtained for SiO_2-EG are consistent with this model as can be seen in Fig. 3.

Surface tension

The surface tension values of SiO_2-EG nanofluids at 298.15K are presented in Table 1. The addition of nanoparticles to the base fluid slightly affects the surface tension, which can be observed in Fig. 4. However, the data obtained are within the measurement uncertainty for the literature value of ethylene glycol surface tension 48.02 mNm^{-1} [37]. Despite an increase in the mass concentration of the nanoparticles, the surface tension emerges around 48.3 mNm^{-1}. Based on these data, it can be concluded that the addition of SiO_2 nanoparticles at the concentrations tested to ethylene glycol does not affect the surface tension of the base fluid significantly.

Additionally, the surface tension of the samples was examined with a tensiometer built using arduino and 3D printing technology. The construction and its operation is presented in the recent paper [34]. Surface tension measurements were performed at 298.15 ± 2.00 K, and the obtained results of the measurements performed with this device are shown in Fig. 5. Taking into account, the
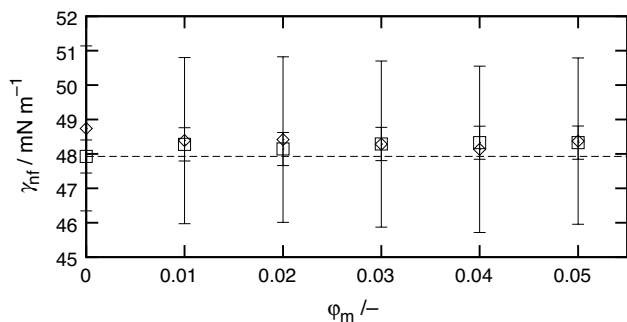
measurement uncertainties, one could note that the results of measurements of the surface tension observed with this tensiometer stands in high agreement with measurements made on a commercial device.

Absence or slight increase in surface tension is also observed for other nanoparticles in ethylene glycol based nanofluids such as nitrides [27, 38] or oxides [25, 26].

Refractive index

Despite the great interest in the thermophysical properties of nanofluids containing SiO_2, there is lack of information about their optical properties designated in the experimental way. However, in some publications, this topic is presented from the theoretical perspective [39, 40]. The obtained results of refractive index, n, analyzed for SiO_2-EG nanofluids with different concentrations of nanoparticles are summarized in Table 2. The measured value of refractive index increases with the higher mass fraction of
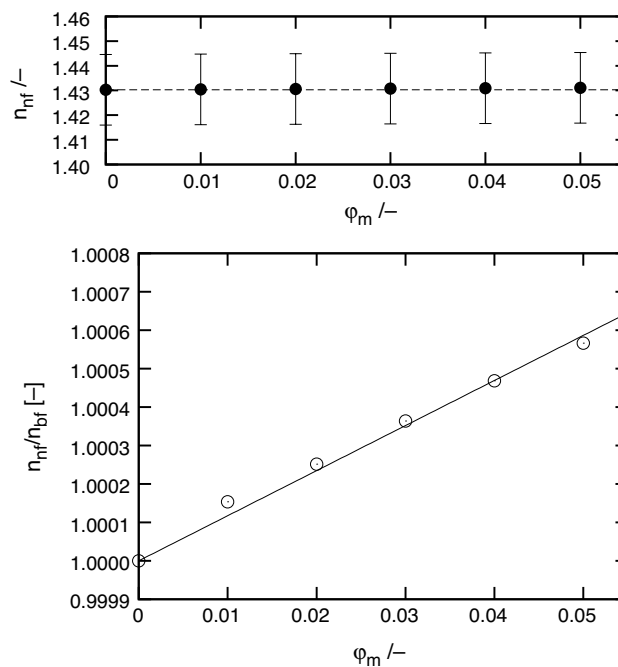

Fig. 5 Dependence of surface tension, γ, on the mass fraction, φ_m, of the particles for the SiO_2-EG nanofluids at 298.15 K. Square points present results obtained with commercial equipment, diamonds—results obtained with self-made station. Dotted line presents the value of surface tension of pure ethylene glycol measured with commercial equipment

Fig. 6 Dependence of refractive index, n, and refractive index ratio, n_{nf}/n_{bf}, on the mass fraction, φ_m, of the particles for the SiO_2-EG nanofluids at 298.15 K. Dotted line presents the value of refractive index of pure ethylene glycol, solid line shows Eq. (3)

SiO$_2$ nanoparticles in the sample. Using the least squares fitting method, in which correlation coefficient, R^2, is 1, a linear function was fitted to the data according to the equation:

$$\frac{n_{\text{nf}}}{n_{\text{bf}}} = 1 + 0.01173\varphi_{\text{m}}. \tag{3}$$

Furthermore, the graph in Fig. 6 depicts fitted linear function to the experimentally determined values of *n*.

Extinction

Figure 7 shows the extinction spectra obtained for silicon dioxide-ethylene glycol nanofluids with various mass concentration of nanoparticles. A maximum extinction peak was observed at a wavelength of about 200 nm. As it could be noted, with an increasing amount of SiO$_2$ nanoparticles, the light extinction of SiO$_2$-EG nanofluid increased until it reached a maximum value for concentration 5 mass%. The maximum extinction for an increasing mass concentration of the nanofluid increase from 0.79 to 1.70, respectively. The fewer particles dispersed in the solution, the lower the light extinction of the nanofluids. The presence of nanoparticles increases the scattering and therefore the extinction. This proves that even a greater amount of SiO$_2$ does not cause the particles to easily aggregate in suspension, reducing the extinction of particle light during UV–Vis measurements. Additionally, the stability of these nanofluids is confirmed by the fact that the extinction peak
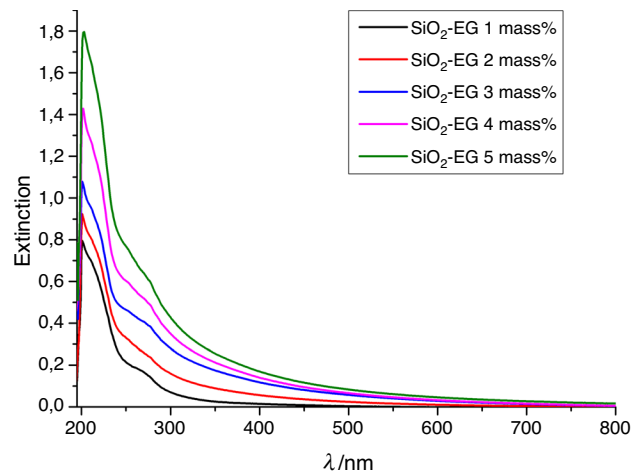

Fig. 7 Extinction spectra for SiO$_2$-EG nanofluids with various mass concentration of nanoparticles

varies to a small extent in the entire range of the tested concentrations. In colloidal solutions, not the absorbance value is measured, but the absorption and dispersion due to the presence of nanoparticles. Therefore, in this case, the concept of extinction is more appropriate. The presence of nanoparticles increases the scattering and therefore the extinction.

See Appendix Fig. 8 and Table 2

Fig. 8 Dependence of **a** density, **b** surface tension, **c** refractive index on the mass fraction, φ_m, of the particles for the SiO_2-EG nanofluids at temperature range of 283.15–318.15 K
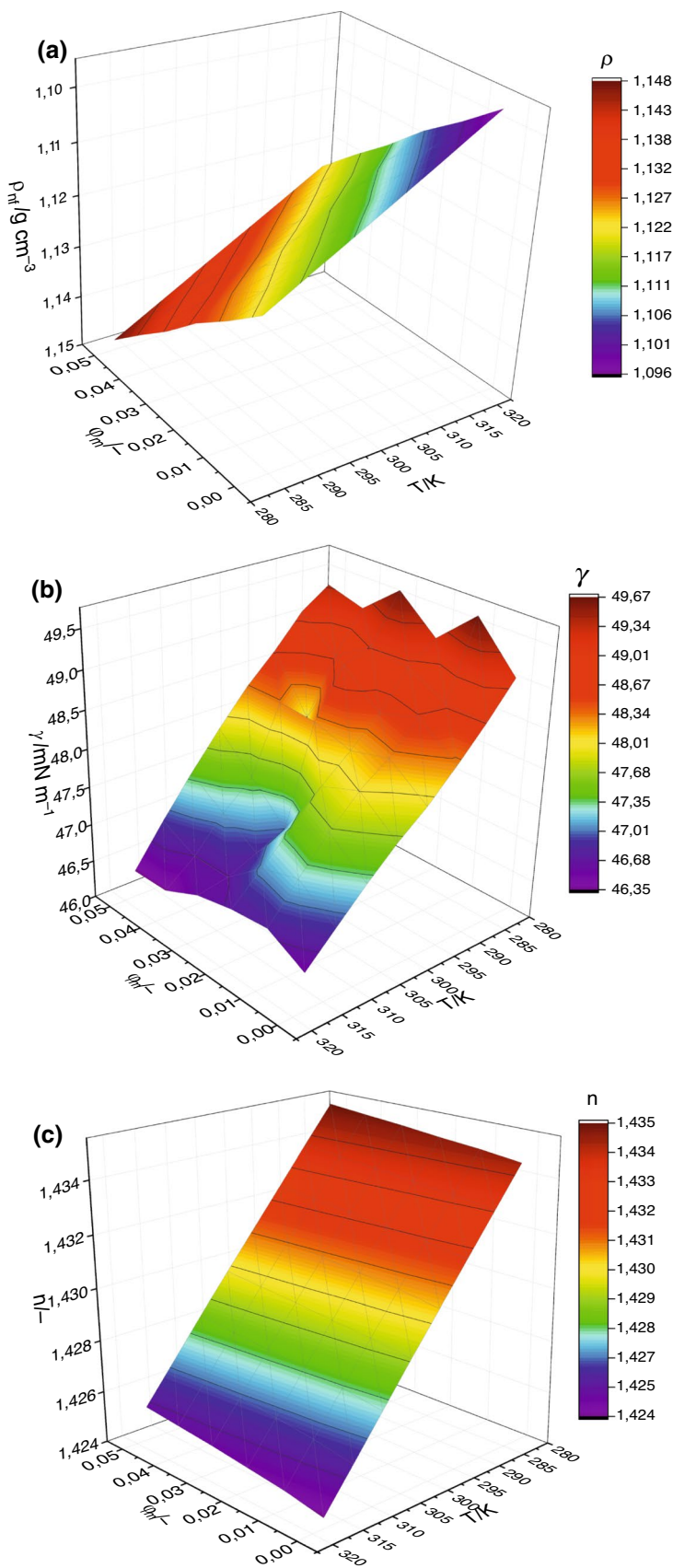
Table 2 Experimental values of the density, ρ, surface tension, γ, and refractive index, n, of SiO$_2$-EG nanofluids at pressure p=0.10 MPa and various temperatures from T=283.15 K to T=318.15 K and different fractions of nanoparticles: mass, φ_m, and volume, φ_V

T / K →		283.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15
φ_m / −	φ_V / −	ρ / g cm^{-3}							
0.00	0.000	1.1202	1.1167	1.1133	1.1098	1.1063	1.1028	1.0992	1.0957
0.01	0.005	1.1257	1.1222	1.1187	1.1152	1.1118	1.1082	1.1047	1.1012
0.02	0.010	1.1307	1.1272	1.1237	1.1202	1.1167	1.1132	1.1097	1.1062
0.03	0.015	1.1369	1.1334	1.1300	1.1265	1.1229	1.1194	1.1159	1.1123
0.04	0.020	1.1423	1.1388	1.1354	1.1319	1.1284	1.1249	1.1213	1.1177
0.05	0.026	1.1480	1.1445	1.1410	1.1375	1.1340	1.1305	1.1270	1.1235
φ_m / −	φ_V / −	γ / mN m^{-1}							
0.00	0.000	49.109	48.678	48.286	47.926	47.624	47.273	46.890	46.504
0.01	0.005	49.670	48.707	48.657	48.280	48.014	47.646	47.218	46.758
0.02	0.010	49.210	48.946	48.490	48.142	47.823	47.442	47.069	46.704
0.03	0.015	49.632	49.024	48.895	48.290	47.015	46.837	46.669	46.595
0.04	0.020	49.229	48.704	48.003	48.325	47.459	47.116	46.679	46.370
0.05	0.026	49.365	49.109	48.694	48.328	47.883	47.406	46.901	46.357
φ_m / −	φ_V / −	n / −							
0.00	0.000	1.43455	1.43312	1.43169	1.43025	1.42882	1.42734	1.42586	1.42440
0.01	0.005	1.43472	1.07498	1.43189	1.43047	1.42904	1.42759	1.42613	1.42468
0.02	0.010	1.43484	1.43343	1.43202	1.43061	1.42919	1.42775	1.42630	1.42486
0.03	0.015	1.43499	1.43358	1.43218	1.43077	1.42936	1.42792	1.42648	1.42504
0.04	0.020	1.43511	1.43372	1.43232	1.43092	1.42950	1.42806	1.42662	1.42518
0.05	0.026	1.43524	1.43386	1.43246	1.43106	1.42965	1.42823	1.42677	1.42532

The estimated standard relative uncertainty $u_r(\varphi_m) = 0.01$, $u_r(\varphi_V) = 0.001$, $u_r(\rho) = 0.001$, $u_r(\gamma) = 0.01$, $u_r(n) = 0.01$, $u(p) = 0.01$ MPa and $u(T) = 0.10$ K

Conclusions

The paper summarizes results of the experimental study on the surface and optical properties of SiO$_2$-EG nanofluids. The mass density, surface tension, refractive index and extinction of the nanofluids with various mass fractions have been examined. It was presented that mass density could be modeled with well-known Pak and Cho correlation. In case of surface tension, it was presented that fraction of the nanoparticles have marginal influence on this property; the surface tension of nanofluids increases after adding the nanoparticles, but taking into account the uncertainty of measurements this increase is insignificant. The refractive index increase with the nanoparticle fraction and could be described with linear function. It was also presented that in these nanofluids absorbance of light is the highest for wavelength approximately 200 nm, and its value depends on the mass fraction of the particles. This paper brings a contribution to the knowledge of the physical properties of the SiO$_2$-EG; however, one could note that there are still some gaps in this area. Future research could be focused on the heat capacity, contact angle and heat transfer of this type of nanofluids.

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