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# Thermophysical profile of ethylene glycol based nanofluids containing two types of carbon black nanoparticles with different specific surface areas

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## Abstract

The paper summarized results of experimental studies on thermophysical and electrical properties of ethylene glycol based nanofluids containing carbon black nanoparticles. Two types of nanoparticles differing in size and specific surface area were used to develop nanofluids. During examination the thermal conductivity, isobaric heat capacity, mass density, nanofluid-air surface tension, dynamic viscosity, refractive index and electrical conductivity were measured in strict controlled temperature 298.15 K. Obtained results indicate that the specific surface area have great influence on these fundamental properties of nanofluids developed with carbon nanoparticles. Finally, the enhancement of electrical conductivity described in the paper is one of the highest reported for the nanofluids in available literature.

*Keywords:* nanofluid, carbon black, viscosity, thermal conductivity, heat capacity, surface tension

## Nomenclature

$A$	constant depending on the reflection plane [-]
$B$	full width at half-maximum (fwhm) [-]
$c_p$	isobaric heat capacity [ $\text{kJ kg}^{-1} \text{K}^{-1}$ ]

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$f$	frequency [Hz]
$G'$	storage modulus [Pa]
$G''$	loss modulus [Pa]
$k$	thermal conductivity [ $\text{W m}^{-1} \text{K}^{-1}$ ]
$K$	Herschel-Bulkley model consistency [ $\text{Pa s}^N$ ]
$L_a$	mean size of the "crystallinities" [nm]
$n$	refractive index [-]
$N$	Herschel-Bulkley model flow index behavior [-]
$R^2$	correlation coefficient [-]
$T$	temperature [K]
$\dot{\gamma}$	shear rate [ $\text{s}^{-1}$ ]
$\gamma$	surface tension [ $\text{mN m}^{-1}$ ]
$\eta$	dynamic viscosity [Pa s]
$\lambda$	wave length [ $\text{\AA}$ ]
$\rho$	density [ $\text{kg m}^{-3}$ ]
$\tau$	shear stress [Pa]
$\theta$	scattering angle [ $^\circ$ ]
$\tau_0$	Herschel-Bulkley model yield stress [Pa]
$\sigma$	electrical conductivity [ $\mu \text{S cm}^{-1}$ ]
$\varphi$	fraction [-]
	subscripts
bf	base fluid
m	mass
nf	nanofluid
p	particle

## 1. Introduction

One of the most important factors determining the development of society, science and technology is the efficiency of energy transfer and storage. During last decades it is observed that the energy demand has increased while the impact of the energy sources based on fossil fuels on the natural environment is adverse. Because of that, the needs to exchange "traditional" for renewable energy systems has been emphasized. Among many solutions there is solar energy and well-known collectors. However there is a problem with heat losses and poor heat transfer so this solution required improvements. At a certain point, the idea of replacing the previously used heat transfer fluids with nanofluids was born in order to increase the efficiency of these systems.

Nanofluid is a liquid-solid mixture in which there are dispersed one or various types nanometer-sized particles. One of the first experimental studies on this subject was conducted by Masuda et al. [1]. They analysed thermal properties of system containing  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$  and  $\text{TiO}_2$  particles in water. The result of their research was the improvement in thermal conductivity for  $\text{Al}_2\text{O}_3$ -water and  $\text{TiO}_2$ -water, however no increase was observed for  $\text{SiO}_2$ -water [1]. In 1995 Choi and Eastman presented results of their study where the word "nanofluid" appears for the first time [2]. They showed that thermal conductivity of fluid increases when nanoparticles are added to it and suggested many potential benefits for the scientific world in the future. Since then research work in order to find out more about the properties has risen. The current stage of knowledge on the thermo-physical properties of nanofluids can be found in recent review papers [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13]. Thermophysical properties and possible applications of nanofluids containing various types of carbon structures have been summarized in Refs. [14, 15, 16]. The review of the papers on lubrication properties of nanofluids containing carbon structures was presented in Ref. [17].

Thermal conductivity of nanofluids with different types of nanoparticles are still the subject of experimental studies. Shamaeil et al. [18] focused on the effects of temperature and volume fraction on the thermal conductivity for double-walled carbon nanotubes (DWCNTs)-ethylene glycol (EG) nanofluids. The results show that thermal conductivity ratio increases with increasing the volume fraction of particle in suspension and temperature. Analysed data also reveals that the maximum enhancement is 24.9% for 0.6% volume concentration at 325.15 K. Experimental study of thermal conductivity for nanofluid containing graphene-carbon nanotube (Gr-CNT) hybrid material in ethylene glycol was performed by Trinh et al. The sample with 0.07 vol.% Gr-CNT at 303.15 K has 18 % enhancement and in 323.15 K – 50% [19]. Harsh et

al. carried out measurements in which they studied ethylene glycol based nanofluid, with single-walled carbon nanotube (SWNT) using the transient hot wire technique. It was found out that the maximum enhancement is 14.8% for nanofluid containing 0.21% volume concentration [20]. Investigation on the thermal properties of diamond-ethylene glycol (DNP-EG) nanofluid was conducted by Yu et al. in Ref [21]. The thermal conductivity enhancement of DNP-EG nanofluids for 1.0 vol.% at 303.15 K is up to 17.23%. However, their study has been expanded to observe if thermal conductivity of nanofluids is correlative with pH values. The data shows that pH value has a direct effect on the stability of the samples - according to the measurements for DNP-EG nanofluids when pH is above value of 8.5, no sedimentation is observed within 6 months [21].

In the last years, nanofluids have achieved attention in terms of thermal conductivity studies. However properties such as viscosity of nanofluids containing carbon nanoparticles was also experimental studied. Yu et al. has conducted measurements of diamond nanoparticles in ethylene glycol at 303.15 K [21]. The data depicts that with the increase of temperature, the viscosity of nanofluids decreases rapidly. His conclusions demonstrates Newtonian behaviour due to linear relation between shear stress and shear rate.

On the other hand oscillatory measurements at 298.15 K carried out for ethylene glycol based graphite/diamonds mixture nanofluids shows that it behaves like non-Newtonian fluid [22]. Similarly the rheology results of EG-based nanofluid with single-walled carbon nanohorn presents that it has non-Newtonian shear thinning behaviour at higher nanohorn loading [23]. Recently Żyła presented results of the experimental study on the rheological behavior of ethylene glycol based nanofluids containing low concentrations (up to 1 wt.%) of carbon black nanoparticles [24]. He showed that these materials exhibit non-Newtonian viscoelastic nature.

Among all of these properties heat capacity and mass density of EG based nanofluids are also the subject of experimental studies. However most of the papers concern oxides and nitrides [25, 26, 27, 28, 29], while results on the ethylene glycol based nanofluids containing carbon nanostructures are rare. The isobaric heat capacity and mass density of EG based nanofluids containing nanodiamonds with different purities were experimentally investigated by us previously and presented elsewhere [30]. We have presented that classical models described these properties with good accuracy.

Recent results of the studies on the surface tension of EG based nanofluids starts to earn interest [31, 32, 33]. According to the best authors knowledge the surface tension of nanofluids

containing carbon nanoparticles with ethylene glycol as base fluid was not experimentally studied before.

Despite the great interest in the thermophysical properties of nanofluids containing various types of carbon based nanoparticles, there are only few papers, which reports their electrical properties. Żyła et al. [30] investigated electrical conductivity of nanodiamonds ethylene glycol nanofluids. They prepared nanofluids with two types of nanodiamond nanoparticles with different level of purity (87% – ND87, 97% – ND97). Five samples show nonlinear increase in electrical conductivity with increasing volume fraction, and the maximum value was achieved at 0.0338 volume fraction. They also noticed that there is no visible difference between nanofluids containing nanodiamonds with various purities. Different types of nanofluid based on carbon based materials and ethylene glycol were also investigated by Żyła et al. [22]. This time, they used two types of graphite and diamond mixtures, with different ash content, to prepare ethylene glycol based nanofluids. The five samples with volume fraction between 0.004–0.023 were prepared for both type of nanoparticles. Obtained results show nonlinear increase in electrical conductivity with increasing volume fraction for both types of nanofluids. They also revealed that ash content has a significant impact on the electrical conductivity of such types of nanofluids. Nanofluids prepared with nanoparticles with a higher content of ash are less electrically conductive. The maximum electrical conductivity enhancement was noted for the nanofluid with lower ash content and it was almost 200 times greater than that for pure ethylene glycol and 10 times higher than that with higher ash content at the same volume fraction.

In this paper results of the experimental studies on thermal conductivity, heat capacity, viscosity, mass density, surface tension, refractive index and electrical conductivity of nanofluids containing two different types of carbon black nanoparticles are presented.

## **2. Materials and methods**

In the following sections the description of materials and methods used in this study is presented. The information about the measuring equipment together with the uncertainty determination are also reported.

### *2.1. Nanoparticle characterization*

In this study two different types of carbon black nanoparticles were used. Both of them are

commercially available, and are labeled in this paper as CB1 (Degussa AG (Hanau-Wolfgang, Germany)), and CB2 (PlasmaChem GmbH (Berlin, Germany)). The properties of the used materials has been summarized in Table 1.

Table 1: Basic properties of the carbon black nanoparticles used in the study.

CB1	
Manufacturer	Deagussa AG
Appearance	black powder
Ash content	$\leq 2,0 \%$
CAS No.	7782-40-3
Average particle size	30 nm
BET Surface Area <sup>†</sup>	$1000 \text{ m}^2 \text{ g}^{-1}$
BET Surface Area <sup>‡</sup>	$1012 \pm 4 \text{ m}^2 \text{ g}^{-1}$
CB2	
Manufacturer	PlasmaChem
Appearance	black powder
Ash content	$< 0,02 \%$
CAS No.	7782-40-3
Average particle size	ca. 13 nm
Specific surface area <sup>†</sup>	$\text{ca. } 570 \pm 20 \text{ m}^2 \text{ g}^{-1}$
BET Surface Area <sup>‡</sup>	$450 \pm 8 \text{ m}^2 \text{ g}^{-1}$

<sup>†</sup> declared by manufacturer,

<sup>‡</sup> experimentally determined.

To evaluate the size and shape of the nanoparticles, series of scanning electron microscopy (SEM) pictures were taken with JEOL JSM-6700F field emission scanning electron microscope (JEOL, Tokyo, Japan). Figure 1 presents SEM pictures of the used carbon black nanoparticles with different magnifications. The declared size of the nanoparticles, presented in Table 1, was confirmed there.

Figure 1: Scanning electron microscope (SEM) pictures of dry nanoparticles: A) and B) present CB1 (Degussa AG (Hanau-Wolfgang, Germany)) nanoparticles with different magnification, C) and D) show CB2 (PlasmaChem GmbH (Berlin, Germany)) with different magnification.

The energy-dispersive X-ray spectroscopy microanalysis (EDS) was performed with Inca Energy 300 (Oxford Instruments, Oxford, UK) coupled to the previous described SEM. The EDS spectrum of the used nanopowders were presented in Fig. 2. Performed measurements confirm the chemical structure of carbon (C), as expected for these nanomaterials. The appearance of the silica (Si) peak can be explained by the physical support over which the analyzes were performed.

Figure 2: A) and C) the scanning electron microscope pictures of observed area of CB1 (Degussa AG (Hanau-Wolfgang, Germany)) and CB2 (PlasmaChem GmbH (Berlin, Germany)) respectively. B) and D) energy-dispersive X-ray spectroscopy microanalysis spectra of CB1 and CB2 carbon black nanopowders respectively.

Powder X-ray diffraction experiments were performed on a Bruker D8 Advance (Bruker GmbH, Karlsruhe, Germany) diffractometer working in a  $\theta-2\theta$  modified Bragg-Brentano geometry and equipped with a LynxEye fast detector. The Cu  $A\alpha 1$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ) is selected using a Ge(111) monochromator. The  $5-90^\circ$   $2\theta$  angular domain was scanned with a step size of  $0.02^\circ$  and integration time of 895 s per step. A misoriented single crystalline Si sample holder was used to avoid any background signal from it.

Both samples show similar XRD profiles which are typical of the carbon black with two main Bragg diffraction peaks for the (002) and (110) reflections at  $2\theta$  ca.  $24.5^\circ$  and  $24.6^\circ$  and ca.  $43.6^\circ$  and  $43.2^\circ$  for CB2 and CB1, respectively as presented in Fig. 3. After subtracting a baseline, the peaks were fitted with Gaussian curves and the Scherrer equation (eq. 1) was used to calculate the mean size  $L_a$  of the 'crystallites' of the two samples:

$$L_a = \frac{A\lambda}{B \cos \theta}, \quad (1)$$

where  $A$  is a constant depending on the reflection plane 0.9 and 1.84 for the 002 and 110 reflections, respectively,  $\lambda$  is the XRD wavelength,  $\theta$  is the scattering angle (in radians), and



$B$  is the full width at half-maximum (fwhm). For both samples studied,  $L_a$  is less than 1 nm in agreement with the broad XRD features shown in Fig. 3 and the disordered structure expected for carbon black particles.

Figure 3: X-Ray diffractogram for the nanopowders used in this study. Black line refers to the CB1 (Degussa AG (Hanau-Wolfgang, Germany)) nanoparticles and gray to the CB2 (PlasmaChem GmbH (Berlin, Germany)).

The Brunauer-Emmett-Teller (BET) specific surface area of both carbon black nanopowders was determined from nitrogen physisorption with a Micromeritics Gemini VII 2390t instrument using a mixture of  $N_2/He$  (30%/70%). To avoid the surface hydration, samples were previously outgassed under vacuum at 323.15 K.

The BET surface area of CB1 is experimentally evaluated to  $1012 \pm 4 \text{ m}^2 \text{ g}^{-1}$ , which is in good agreement with manufacturer data as the deviation is 1.2%. For CB2, the measured BET value is  $450 \pm 8 \text{ m}^2 \text{ g}^{-1}$  that is lower by 21% than the manufacturer data. These measurements clearly demonstrate that the carbon black nanopowders present distinct BET surface area.

## 2.2. Sample preparation

There are two methods in which a nanofluid could be prepared: (a) single step and (b) two-step preparation process. The single step method involves the direct production of nanoparticles in a base liquid and can be completed in various ways. For example by reducing the compound in the presence of another substance under microwave irradiation, a nanoparticle-source evaporation and deposition of the evaporant into a base fluid, and sonication and using the vacuum-SANSS (submerged arc nanoparticle synthesis system). Samples can also be prepared based on two-step method by dispersing the dry particles within the base fluid. Taking into account fact that preparing dry nanoparticles is often easy and cheap process, and nanoparticles are commercially available this method seems to be more attractive for potential application, which demands a large-scale production process. However two-step method has some drawback, e.g some types of nanoparticles easily aggregate and sediment because of strong van der Waals force between particles and gravity. In the present study two types of carbon black

nanoparticles were used to prepare nanofluids with two-step method. Both nanopowders (with two different specific surface area, ash content and sizes) were purchased commercially and their specification is presented in the Table 1. In the first stage of sample preparation nanoparticles were weighed on an analytical balance. After that the base fluid – ethylene glycol (EG) produced by Fisher Chemical (Loughborough, UK) with the purity over 99%, was added in order to achieve desired concentration of the particles in the suspension. For the dispersion nanoparticles, mechanical mixing and ultrasonic bath were used. Sonication is well known method of dispersing nanoparticles, as it was described in recent review paper [34]. No surfactants were used during sample preparation.

### 2.3. Stability characterization

After preparation of the samples, the stability of these type of nanofluids was evaluated. The high light absorbance of nanofluids containing carbon black nanoparticles and its high viscosity make it impossible to precise the determination of the stability of samples containing the same fraction of particles that were used in the study of thermophysical properties. The stability evaluation of these nanofluids was performed on diluted samples containing 0.02 wt.% of particles.

One of the popular methods of the stability determination of nanofluids is measuring its Zeta potential as it was described in Ref. [35]. The Zeta potential of the CB-EG nanofluids was measured with Zetasizer Nano ZS device (Malvern Instruments Ltd, Malvern, UK) at 298.15 K. The values obtained in this study were  $+57 \pm 6$  mV and  $-110 \pm 4$  mV for nanofluids containing CB1 and CB2, respectively. Nanofluids with absolute Zeta potential value higher than 60 mV are consider as very stable [35], however one must note that this method could not be fully reliable for ethylene glycol based nanofluids, therefore another type of stability examination was also performed.

The Dynamic Light Scattering (DLS) method allows the apparent size distribution of nanoparticles in suspension to be determined. This method was used with Zetasizer Nano ZS device (Malvern Instruments Ltd, Malvern, UK) for 0.02 wt.% nanofluids at 298.15 K and using a scattering angle value  $173^\circ$ . Samples were analyzed just after preparation (0 h) and 2, 6, 24 and 48 h later under static conditions. It should be noted that the size value by DLS measurements is assumed as the diameter of a perfect sphere with a translational diffusion coefficient equivalent to

that of the dispersed particle. Thus, these values are usually referred to hydrodynamic diameter or apparent size. Consequently, it should be taken into account that the stability analysis is focused on the temporal evolution of the DLS measurements rather than the values of the sizes themselves. Figure 4 presents obtained results. One could note that the apparent size of CB1 nanoparticles is two times higher than CB2 nanoparticles and it corresponds with the manufacturer declaration and SEM pictures. Nanofluids containing CB2 nanoparticles tend to agglomerate after 48 hours, however dispersion for samples with CB1 nanoparticles is stable with time during measurement. The apparent size distribution analysis presented in Fig. 4A) and Fig. 4B), show similar profiles for both samples, CB1 and CB2 nanofluids, along the observed time (2 days). Nevertheless results collected for CB1-EG sample show a low intensity peak around  $0.5 \mu\text{m}$  that appears 24 h after preparation and increases after 48 h. On the other hand, the CB2-EG nanofluid shows a similar small peak for the first time 48 h after preparation. These peaks reveal symptoms of agglomeration of nanoparticles that is occurring within the fluids, which is stronger and faster for the nanofluid containing CB1 nanoparticles.

Finally, the possible occurrence of sedimentation was not being observed until several hours after preparation of the samples that were studied. Therefore, taking into account all these factors, the examined nanofluids were thermophysically studied for the few hours after samples preparation, in conditions of stable dispersions.

Figure 4: Results of dynamic light scattering (DLS) examination of the apparent size of particles in ethylene glycol based nanofluids containing 0.02 wt.% carbon black nanoparticles. A) and B) Apparent size distribution for nanofluids containing CB1 and CB2 nanoparticles, respectively; different colors refer to time after preparation of the samples. C) Average Z-size as a function of time after preparation of the samples for the CB1-EG ( ) and CB2-EG ( ) nanofluids at 298.15 K; dotted lines indicate the value after preparation (0h).

## 2.4. Methods

Thermal conductivities of the base fluid – ethylene glycol and the elaborated nanofluids were measured at 298.15 K by means of a THW-L2 thermal conductivity device (Thermtest Inc., Canada) based on the transient short hot wire (THW) technique, that was already used in examination of nanofluids [36, 37, 38]. The THW-L2 apparatus allows obtaining direct thermal

conductivity data of liquids in agreement with ASTM D7896-14 standard [39]. This device is coupled with an EchoTerm Dry Bath which maintains the sample at the test temperature. The wire welded in a sensor has a length of 60 mm with a diameter of 0.1 mm. It is vertically inserted in the sample, and the whole set is introduced in the dry bath. Before starting the measurements, the used software performs a power test for each family of fluids under study and from this test, a heating power and a short power input time are selected avoiding natural convection phenomena in the measurements. Five measurements were carried out to ensure the repeatability of the experimental values, the duration of each measurement being 10 seconds. The declared accuracy in thermal conductivity values by the manufacturer is 5%.

Experimental isobaric heat capacities for CB-EG nanofluids were determined by a quasi-isothermal temperature modulated differential scanning calorimetry technique. For this purpose, a differential scanning calorimeter, Q2000 (TA Instruments, New Castle, USA) was used at 298.15 K as measurement temperature. Samples are housed into  $T_{zero}$  aluminum pans and weighted before and after performing the tests to validity the measurement. Sinusoidal temperature modulations around the studied temperatures were selected to obtain the isobaric heat capacities. More details about the  $c_p$  determination by using this device were previously reported [28, 40], the total relative uncertainty being lower than 3%.

The mass density of the investigated nanofluid was determined using an automatic oscillating U-tube densitometer DMA 4100M (Anton Paar, Graz, Austria). The sample was inserted into a U-shaped borosilicate glass tube and constant temperature 298.15 K was maintained. Three series of measurements were carried out, ten measurements in each series. The uncertainty of the values obtained with this equipment was determined on the base of (a) the deviation between the experimental data and the corresponding value in the literature and (b) standard deviation of one hundred measuring points distilled water at 298.15 K. For these one hundred measurements, the obtained mass density of water was  $0.9969 \text{ g cm}^{-3}$  with standard deviation  $0.0002 \text{ g cm}^{-3}$ . According to the literature [41], the water density is  $0.997066 \pm 0.000001 \text{ g cm}^{-3}$ . Considering the literature values and the obtained experimental results the relative uncertainty of mass density measurements could be declared as 0.1%.

Surface tension between air and nanofluid measurements were performed with Drop Shape Analyzer DSA25 (KRÜSS GmbH, Hamburg, Germany). This equipment uses the analysis of the

shape of the drop and based on the Young–Laplace equation allows to determine the surface tension of the fluid. In order to measure surface tensions of nanofluids a drop "hang" for 10 minutes to equalize nanofluids temperature with ambient air temperature. After such stabilization the surface tension of a droplet was measured. To obtain reliable results after each measurement a drop was dropped and another one was produced with the waiting time for temperature stabilization. The experimental study included three series of measurements that followed each other. Environmental chamber (KRÜSS GmbH, Hamburg, Germany) allows to perform measurements in strict controlled temperature, 298.15 K. Relative uncertainty of obtained surface tension values was estimated as 1% as presented elsewhere [31, 33].

The equipment used to perform the rheological properties examination was a rheometer HAAKE MARS 2 (Thermo Electron Corporation, Karlsruhe, Germany). Two types of measurements were performed: (a) the rotational tests in control rate mode, and (b) the oscillatory tests at constant oscillation frequency 1.0 Hz. Taking into account fact that CB-EG nanofluids exhibit a non-Newtonian character, as presented in previous study [24], the cone-plate measuring geometry was used; the diameter of the rotor is 60 mm, and cone angle is  $2^\circ$ . Constant temperature of the sample, 298.15 K, was maintained by the Peltier system coupled with a Phoenix 2 thermostat (Thermo Electron Corporation, Karlsruhe, Germany); this system allows to keep the temperature of the sample with the accuracy of 0.1 K. Rotational measurements were performed at shear rate range from  $0.1 \text{ s}^{-1}$  to  $1000.0 \text{ s}^{-1}$  and oscillatory tests in deformation range from 0.1% (0.001) to 1000.0% (10.000). The step of shear rate and deformation increases was planned on the logarithmic scale. This measuring system was previously used, therefore more detailed description of the uncertainty discussion could be find elsewhere [30, 42].

The refractive index was measured with the Refractometer Abbemat 550 (Anton Paar OptoTec GmbH, Seelze-Letter, Germany). A  $500 \mu\text{L}$  sample was placed in the refractometer using the HTL Discovery Comfort DV1000 single channel pipette (HTL Lab Solutions, Warsaw, Poland) with an accuracy of volume of 0.5%. The sample temperature was stabilized at 298.15 K with an accuracy of 0.03 K for 5 minutes. The refractive index value was determined as the average of a series of three measurements performed for various samples. The relative uncertainty of measurement of refractive index performed at this station was determined at less than 1% of the measured value, as shown in Ref. [43].

The electrical conductivity of carbon black ethylene glycol was measured with MultiLine

3410 conductivity meter (WTW GmbH, Weilheim, Germany) working with conductivity probe TetraCone 925 (WTW GmbH, Weilheim, Germany). Measurement was performed for 10 minutes, at constant temperature 298.15 K with 0.2 K accuracy. The value of electrical conductivity was obtained as the average value. The temperature was stabilized at least 10 minutes before measurement. Relative uncertainty of this measuring setup was evaluated to 1% of the measured value as presented in Ref. [42].

### 3. Results and discussion

In the following subsections the obtained results have been presented along with the discussion and graphic representation.

#### 3.1. Thermal conductivity

The experimental value of atmospheric pressure thermal conductivity of ethylene glycol at 298.15 K, Table 2, agrees well with those of Lide [44], with a deviation of  $-2.2\%$ , Akilu et al. [45], with a deviation of  $-1\%$ , and Żyła et al. [30], with an agreement of  $+1.8\%$ . As it can be observed, these deviations are lower than uncertainty in thermal conductivity measurements.

Results of experimental study on the thermal conductivity,  $k$ , of CB1-EG and CB2-EG nanofluids were summarized in Table 2 and presented in Fig. 5, where it is shown how does the value of thermal conductivity change with increasing mass fraction of nanoparticles. The picture depicts that the highest enhancement of thermal conductivity ( $4.8\%$ ) was observed for nanofluids containing CB1 nanoparticles with the highest examined mass concentration 2 wt.%. Additionally it was presented that the dependence of the thermal conductivity on the mass fraction of these materials could be modeled with linear function:

$$\frac{k_{nf}}{k_{bf}} = 1 + 2.42\varphi_m, \quad (2)$$

where  $k$  is thermal conductivity,  $\varphi$  is fraction, and subscripts  $nf$ ,  $bf$ ,  $m$  stands for nanofluid, base fluid and mass respectively. In the case of the fitting the linear function (2) to the experimental data the coefficient of determination  $R^2 = 1$ .

Thermal conductivity for the second type of investigated nanofluids is definitely lower than in case of CB1-EG nanofluids. The enhancement comparing with the pure ethylene glycol is

approximately 1 %. Results of the experimental study of thermal conductivity of CB2-EG nanofluids could be also modeled with linear function in form of:

$$\frac{k_{nf}}{k_{bf}} = 1 + 0.69\varphi_m. \quad (3)$$

Table 2: Experimental values of the thermal conductivity,  $k_{nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$k_{nf}$	$k_{nf} / k_{bf}$	$k_{nf}$	$k_{nf} / k_{bf}$
-	W m <sup>-1</sup> K <sup>-1</sup>	-	W m <sup>-1</sup> K <sup>-1</sup>	-
0.000	0.248	1.0000	0.248	1.0000
0.005	0.251	1.0121	0.250	1.0081
0.010	0.254	1.0242	0.250	1.0081
0.020	0.260	1.0484	0.251	1.0121

Figure 5: Dependence of thermal conductivity,  $k$ , and thermal conductivity ratio,  $k_{nf} / k_{bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG (○) and CB2-EG (●) nanofluids at 298.15 K. Points represent experimental data, dotted and solid line – linear function (2) and (3) respectively.

The first type of carbon black nanopowder has approximately two times greater value, it is  $1012 \pm 4 \text{ m}^2 \text{ g}^{-1}$  and  $450 \pm 8 \text{ m}^2 \text{ g}^{-1}$  for CB1 and CB2 respectively. Furthermore the average particle size which is greater for the first type can contribute to better thermal conductivity for this nanofluid. The effect of particle size on the thermal conductivity was analyzed for alumina nanofluids where diameter in the range from 8 to 282 nm was taken [46]. Beck et al. came to the conclusion that the thermal conductivity decreases as the particle size is below value of 50 nm. The improvement in the thermal conductivity due to the increasing specific surface area (SSA) is well recognized in the literature. From the results by studying carbon nanofluids [47, 48, 49, 50, 51], it was found as higher thermal conductivity belongs to carbon-based nanoadditives with higher specific surface area. Nowadays, it is extensively recognized that the presence of a nanolayer at the solid–liquid interface and nanoparticle aggregation entails the main way for thermal conductivity enhancement in nanofluids. The liquid molecules close to particle surfaces allow the formation of layered structures and act much like a solid [47]. Therefore, the dimension (thickness) of the

interfacial layer has a noticeable effect on the enhanced thermal conductivity of the carbon-based nanofluids [48]. Thus, nanoadditives with higher specific surface area lead to decreases on the contact resistance, and consequently superior thermal conductivity enhancements [47, 49]. In addition, it should be noted that all surfaces measured with BET measurement in equilibrated suspensions will acquire layers of fluid molecules that will behave as an interfacial phase [50]. Suspended nanoparticles can form larger agglomerates through the solid/liquid/solid interface, but volume and contribution of the interfacial phase will always remain proportional to the total surface area of dry powders. Thus, the microscopic properties corresponding to the SSA values for the CB1 and CB2 nanopowders correlate to the macroscopic effect in the thermal conductivity enhancements of the nanofluids. Thermal conductivity is superior in nanofluids with larger nanoparticle surface area, CB1-EG.

### 3.2. Isobaric heat capacity

The experimental isobaric heat capacity value for EG at 298.15 K, presented in Table 3, is in good agreement with literature data [52, 53, 54, 55, 29]. Deviations of +0.1%, +0.6%, +0.8%, +0.4% and -1.4% were obtained with respect to the values from Nikolaev and Rabinovich [52], Murthy and Subrahmanya [53], Stephens and Tamplin [54], Zaripov [55], and Żyła et al. [29], respectively.

Figure 6: Dependence of heat capacity,  $c_p$ , and heat capacity ratio,  $c_{p,nf}/c_{p,bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG (●) and CB2-EG (○) nanofluids at 298.15 K. Points represent experimental data, solid lines – equation (4).

Table 3: Experimental values of the isobaric heat capacity,  $c_{p,nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$c_{p,nf}$	$c_{p,nf}/c_{p,bf}$	$c_{p,nf}$	$c_{p,nf}/c_{p,bf}$
-	$\text{kJ kg}^{-1} \text{ K}^{-1}$	-	$\text{kJ kg}^{-1} \text{ K}^{-1}$	-
0.000	2.424	1.0000	2.424	1.0000



0.005	2.383	0.9831	2.399	0.9893
0.010	2.357	0.9724	2.371	0.9777
0.020	2.302	0.9497	2.311	0.9530

The experimental results for the different-loaded CB-EG nanofluids are presented in Table 3. It can be observed that the isobaric heat capacity decreases slightly with the increasing fraction of both CB nanoparticles. These decreases reach 5.0% and 4.6% for the nanofluids with the highest examined CB1 and CB2 concentration, respectively. The literature about nanofluids evidenced different effects in the isobaric heat capacity of the base fluid by the dispersion of nanoadditives. According to Riazi et al. [56], when the base fluid is water or a glycol, it tends to decrease with the increasing nanoadditive loading. Nevertheless, it is usual to observe increases in the isobaric heat capacity of nanofluids based on molten salts or ionic liquids caused by the nanoparticle addition. Therefore, our reported experimental behaviour in Fig. 6 is in accordance with the described behaviours for glycol-based nanofluids.

The decreases caused by the addition of CB1 and CB2 are similar among them, 0.7% being the maximum deviation. Likewise, systematic lower values appear by the dispersion of CB1, the nanoadditive with higher specific surface area. The correlation from Xuan and Roetzel [57, 30] describes the isobaric heat capacity of nanofluids on the basis of the mass fraction of nanoparticles in the dispersion:

$$c_{p,nf} = \varphi_m c_{p,p} + (1 - \varphi_m) c_{p,bf}, \quad (4)$$

where  $c_p$  is the specific heat capacity,  $\varphi_m$  is the nanoadditive mass fraction and subscripts  $nf$ ,  $p$  and  $bf$  stand for nanofluid, nanoparticles and base fluid, respectively.

Experimental isobaric specific heat capacities of  $1.017 \text{ kJ kg}^{-1} \text{ K}^{-1}$  and  $1.148 \text{ kJ kg}^{-1} \text{ K}^{-1}$  were obtained for the CB1 and CB2 raw nanopowders at 298.15 K. The heat capacity of CB1 is slightly lower than that of CB2, which contributes to further reducing the heat capacity of the base fluid in which it is dispersed, as indicated above. Deviations between experimental and predicted values by using these experimental values here reported as  $c_{p,p}$  are lower than 4%. Likewise, the  $c_{p,p}$  is usually taken in the literature as the value of the heat capacity for the bulk material [29].

Taking into account the reported  $c_p$  value of  $0.716 \text{ kJ kg}^{-1} \text{ K}^{-1}$  for amorphous carbon by

Takahashi and Westrum [58] or  $0.770 \text{ kJ kg}^{-1} \text{ K}^{-1}$  for dense graphite by Lutcov et al. [59], deviations between experimental and predicted values for the 0.005 and 0.010 nanoadditive mass fractions lower than 2.2% are obtained. For the highest nanoadditive mass concentration the deviation increases up to 3.7%.

### 3.3. Mass density

Table 4 shows the obtained experimental results of mass density measurements for two different nanofluids at 298.15 K. The density measurement of the base fluid is highly consistent with our previous work [30] with a deviation less than 0.02%. In Fig. 7, a clear increase in the density of the nanosuspension can be observed as the mass fraction of the nanoparticles increases. No significant differences between mass density values of CB1-EG and CB2-EG were noted.

Table 4: Experimental values of the mass density,  $\rho_{nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$\rho_{nf}$	$\rho_{nf} / \rho_{bf}$	$\rho_{nf}$	$\rho_{nf} / \rho_{bf}$
-	$\text{g cm}^{-3}$	-	$\text{g cm}^{-3}$	-
0.000	1.1095	1.0000	1.1095	1.0000
0.005	1.1121	1.0023	1.1099	1.0004
0.010	1.1137	1.0038	1.1134	1.0035
0.020	1.1156	1.0055	1.1189	1.0085

Figure 7: Dependence of mass density,  $\rho$ , and mass density ratio,  $\rho_{nf} / \rho_{bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG ( $\circ$ ) and CB2-EG ( $\bullet$ ) nanofluids at 298.15 K.

### 3.4. Surface tension

Results of experimental study on the surface tension,  $\gamma$ , of CB1-EG and CB2-EG based nanofluids were summarized in Table 5. The value of surface tension decreases for both nanofluids with increasing mass fraction of nanoparticles as it is presented on graph in Fig. 8.

Table 5: Experimental values of the surface tension,  $\gamma_{nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$\gamma_{nf}$	$\gamma_{nf} / \gamma_{bf}$	$\gamma_{nf}$	$\gamma_{nf} / \gamma_{bf}$
-	mN m <sup>-1</sup>	-	mN m <sup>-1</sup>	-
0.000	47.53	1.0000	47.53	1.0000
0.005	46.92	0.9872	47.83	1.0063
0.010	45.57	0.9588	47.12	0.9914
0.020	39.76	0.8365	46.25	0.9731

Figure 8: Dependence of surface tension,  $\gamma$ , and surface tension ratio,  $\gamma_{nf} / \gamma_{bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG (○) and CB2-EG (●) nanofluids at 298.15 K. Points represent experimental data, dotted and solid line – linear function (5) and (6) respectively.

Based on these results it can be concluded, that the surface tension of the CB1-EG is decreasing more significantly that in case of CB2-EG. On the base of the results presented in Fig. 8 one might note that the surface tension of nanofluids with mass concentration 2 wt.% is 39.76 mN m<sup>-1</sup> and 46.25 mN m<sup>-1</sup> for CB1 and CB2 respectively. Experimental data could be modeled with linear functions, as follow:

$$\frac{\gamma_{nf}}{\gamma_{bf}} = 1 - 7.14\varphi_m, \quad (5)$$

$$\frac{\gamma_{nf}}{\gamma_{bf}} = 1 - 1.13\varphi_m, \quad (6)$$

for CB1-EG and CB2-EG respectively. The coefficient of determination  $R^2 = 1$  for both fitted curves.

The difference in surface tension could be related to average particle size which value for CB1 is almost twice higher than for CB2. Similar conclusions were obtained in [60], where Chinnam et al. were analyzing the surface tension of four types of nanofluids in base fluid of 60% propylene glycol and 40% water by mass. On the other hand it should be noted that the specific surface area of CB1 is also two times higher that in case of CB2, which could affect the surface tension.

### 3.5. Dynamic viscosity

Dependence of the shear stress and dynamic viscosity on the shear rate of both examined nanofluids has been presented in Fig. 9. Both, CB1-EG and CB2-EG nanofluids exhibit a non-Newtonian shear thinning behavior. Similar nature of ethylene glycol based nanofluids containing carbon structures (nanohorns, graphite/diamond mixture, nanodiamonds) has been already presented in literature [23, 22, 30]. One might note that this kind of behavior could be modeled with Herschel-Bulkley (H-B) model as follow:

$$\tau = \tau_0 + K \dot{\gamma}^N, \quad (7)$$

where:  $\tau_0$  is yield stress,  $K$  – consistency,  $N$  – dimensionless flow index behavior. Figure 9 presents both the experimental results and model (7) fitting. Herschel-Bulkley rheological parameters together with correlation coefficient,  $R^2$ , were summarized in Table 6.

Figure 9: Dependence of shear stress ( $\tau$ ), and dynamic viscosity ( $\eta$ ) on shear rate (flow and viscosity curves respectively) for ethylene glycol based nanofluids containing two types of carbon black nanoparticles at 298.15 K. Symbols represent measuring points collected with different concentration of nanoparticles, as follow: triangle ( $\triangle, \blacktriangle$ ) – 0.50 wt.%, diamond ( $\diamond, \blacklozenge$ ) – 1.00 wt.%, squares ( $\square, \blacksquare$ ) – 2.00 wt.%; solid (red) lines – results of the (7) model, dotted lines - pure ethylene glycol according to Ref. [61]. Empty and solid symbols refers to CB1-EG and CB2-EG nanofluids respectively.

Table 6: Herschel-Bulkley rheological parameters,  $\tau_0$ ,  $K$ ,  $N$ , determined on the base of flow curves presented in Fig. 9, of nanofluids containing two types of carbon black nanoparticles at 298.15 K and correlation coefficient,  $R^2$ , of fitting.

$\varphi_m$	CB1-EG				CB2-EG			
	$\tau_0$	$K$	$N$	$R^2$	$\tau_0$	$K$	$N$	$R^2$
–	Pa	Pa s <sup>N</sup>	–	–	Pa	Pa s <sup>N</sup>	–	–
0.005	0.12	0.047	0.90	0.99994	0.08	0.034	0.93	0.99995
0.010	0.75	0.046	0.94	0.99905	0.31	0.045	0.90	0.99992

0.020	3.70	0.153	0.91	0.97881	1.21	0.049	0.93	0.99969
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### 3.6. Viscoelastic structure

Non-Newtonian shear thinning behavior could indicate that these nanofluids has viscoelastic structure. In the series of oscillatory experiments at constant frequency 1 Hz both the storage ( $G'$ ) and the loss ( $G''$ ) moduli were determined in function of deformation. Results of this study have been presented in Fig. 10. It could be noted that the viscoelastic structure is fragile and  $G'$  starts decreasing at low deformation value (0.01). However a short linear viscoelastic region could be observed for the lowest examined deformations (up to 0.01). In this region both  $G'$  and  $G''$  are constant and could be calculated as an average in this deformation range. Results of such evaluation has been presented in Table 7 and shown as solid lines in Fig. 10.

Figure 10: Storage ( $G'$ ) and loss ( $G''$ ) moduli as a function of deformation ( $\gamma$ ) for various mass fractions of nanoparticles in ethylene glycol based nanofluids containing two types of carbon black nanoparticles at temperature 298.15 K and oscillation frequency of 1.0 Hz. Symbols represent measuring points collected with different concentration of nanoparticles, as follow: triangle ( $\triangle, \blacktriangle$ ) – 0.50 wt.%, diamond ( $\diamond, \blacklozenge$ ) – 1.00 wt.%, squares ( $\square, \blacksquare$ ) – 2.00 wt.%; solid lines – average  $G'$  and  $G''$  in the range of deformations from 0.001 to 0.01. Empty and solid symbols refers to CB1-EG and CB2-EG nanofluids respectively.

Table 7: Values of storage ( $G'$ ), loss ( $G''$ ) modulus of CB1-EG and CB2-EG nanofluids at 298.15 K. Values determined on the base of experimental results presented in Fig. 10 in region of deformation (lower than 0.010) where both  $G'$  and  $G''$  were constant.

$\varphi_m$	CB1-EG		CB2-EG	
	$G'$	$G''$	$G'$	$G''$
–	Pa	Pa	Pa	Pa
0.005	0.397	0.360	0.601	0.458
0.010	23.004	2.585	5.717	1.449
0.020	93.647	8.671	55.726	6.579

Existence of the maximum in loss moduli observed for both examined nanofluids could be

explained by the "overshoot phenomenon" as it was described in details by Pastoriza-Gallego et al. [62] and Hermida-Merino et al. [63].

### 3.7. Refractive index

Summary results of refractive index,  $n$ , measurements for examined ethylene glycol based carbon black nanofluids with different mass fractions of nanoparticles are shown in Fig. 11 and Table. 8. The refractive index of the studied nanofluids increases linearly with the mass fraction of the nanoparticles. The mathematical analysis carried out using the least squares fitting showed a correlation coefficient,  $R^2$ , equal to 1 for both linear functions fitted experimental data. The linear dependence describing increase of the refractive index value for both examined types of nanofluids could be presented as:

$$\frac{n_{nf}}{n_{bf}} = 1 + 0.0914\varphi_m, \quad (8)$$

$$\frac{n_{nf}}{n_{bf}} = 1 + 0.1410\varphi_m, \quad (9)$$

where equation (8) refers to the CB1-EG and eq. (9) to CB2-EG nanofluids respectively.

Table 8: Experimental values of refractive index,  $n_{nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$n_{nf}$	$n_{nf} / n_{bf}$	$n_{nf}$	$n_{nf} / n_{bf}$
-	-	-	-	-
0.000	1.43028	1.0000	1.43028	1.0000
0.005	1.43092	1.0004	1.43167	1.0010
0.010	1.43138	1.0008	1.43238	1.0015
0.020	1.43301	1.0019	1.43419	1.0027

Figure 11: Dependence of refractive index,  $n$ , and refractive index ratio,  $n_{nf} / n_{bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG ( $\circ$ ) and CB2-EG ( $\bullet$ ) nanofluids at 298.15 K. Points represent experimental data, dotted and solid line – linear function (8) and (9) respectively.

### 3.8. Electrical conductivity

The electrical conductivity of two types of carbon black ethylene glycol based nanofluids was measured at 298.15 K and its results were presented in Fig. 12 and Table 9. The obtained results clearly indicate that both types of carbon black have impact on electrical properties of ethylene glycol. Increase in mass fraction (in whole examined range) of nanoparticles in EG causes enhancement of electrical conductivity for CB1-EG nanofluid as well as CB2-EG nanofluid. Observed changes in values of electrical conductivity for CB2-EG nanofluids are similar to those presented for ethylene glycol based nanofluids containing another carbon nanostructures (graphite/diamond with low ash content) presented elsewhere [22]. On the other hand electrical conductivity enhancement for CB1-EG nanofluids is high and according to the best authors knowledge it is the highest increase in electrical conductivity reported for nanofluids based on ethylene glycol for such low load of nanoparticles. The similar results in electrical conductivity enhancement of nanofluids was achieved for ethylene glycol with 7.5 wt.% silicon nitride nanoparticles ( $\text{Si}_3\text{N}_4$ ) at 298.15 K, which is almost four times higher mass fraction than used in this study. Such huge increase in electrical conductivity of CB1-EG nanofluids can be caused by extremely high specific surface area of used carbon black nanoparticles (Table 1) and creation of conduction path for fluids with higher content of CB1 as schematically shown in Fig. 13. For low mass fraction of nanoparticles in ethylene glycol, electrical conduction can increase by formation of electrical double layer (EDL) around nanoparticles [64]; for higher content of CB1, the conduction path can start to create.

Electrical conductivity enhancement of both type of CB-EG nanofluids can be described with good accuracy with the power function. It was found that results for CB1-EG nanofluids can be fitted by following equation (with  $R^2 = 0.97247$ ):

$$\frac{\sigma_{nf}}{\sigma_{bf}} = 1 + 1.62 \cdot 10^7 \varphi_m^{1.73}, \quad (10)$$

and for CB2-EG nanofluids (with  $R^2 = 0.99999$ ):

$$\frac{\sigma_{nf}}{\sigma_{bf}} = 1 + 2.59 \cdot 10^4 \varphi_m^{1.22}. \quad (11)$$

Table 9: Experimental values of electrical conductivity,  $\sigma_{nf}$ , of CB-EG nanofluids at 298.15 K.

$\varphi_m$	CB1-EG		CB2-EG	
	$\sigma_{nf}$	$\sigma_{p,nf} / \sigma_{p,bf}$	$\sigma_{p,nf}$	$\sigma_{p,nf} / \sigma_{p,bf}$
-	$\mu S cm^{-1}$	-	$\mu S cm^{-1}$	-
0.000	0.21	1.00	0.21	1.00
0.005	83.13	395.86	8.52	40.57
0.010	1365.25	6501.19	19.73	93.95
0.020	3941.79	18770.43	45.69	217.57

Figure 12: Dependence of electrical conductivity,  $\sigma$ , and electrical conductivity ratio,  $\sigma_{nf} / \sigma_{bf}$ , on the mass fraction,  $\varphi_m$ , of the particles for the CB1-EG (○) and CB2-EG (●) nanofluids at 298.15 K. Points represent experimental data, dotted and solid line – functions (10) and (11) respectively.

Figure 13: Presentation of idea of creating conduction path in CB1-EG nanofluids. A) pure base fluid, B) nanofluid with low content of CB1, C) nanofluid with middle content of CB1, D) nanofluid with high content of CB1. Black dots present dispersed CB1 nanoparticles, red dots present creation of potential conduction path through dispersed CB1 nanoparticles.

## 4. Conclusions

The paper presents a comprehensive experimental characterization about thermal conductivity, isobaric heat capacity, mass density, nanofluid-air surface tension, dynamic viscosity and viscoelastic structure of ethylene glycol based nanofluids containing two types of carbon black nanoparticles. The electric properties like refractive index and electrical conductivity were also evaluated. This experimental study was performed at well controlled temperature 298.15 K and varying the mass content in CB nanoparticles up to 2 wt.%.

It was presented that the specific surface area of particles in nanofluids has great influence on the fundamental physical properties. From the application point of view, the nanofluids containing nanoparticles with higher specific surface area seems to be more interesting according to their higher thermal conductivity and lower surface tension and isobaric heat capacity. On the other hand one must note that the dynamic viscosity increases significantly for these materials,



which make it harder to apply in real-life heat exchanging systems.

The remarkable electrical conductivity enhancement reported in this paper (according to the best authors knowledge one of the highest reported in literature) in CB1-EG nanofluids could be interesting from the viewpoint of potential application of these materials in ink jet printed electronics, where currently expensive silver nanoparticles are used. More potential applications of the nanofluids with enhanced electrical conductivity could be found in recent review paper [8].

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## **Competing interests**

The authors declare that they have no competing interests.

## **Declaration of interests**

**The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.**

**The authors declare the following financial interests/personal relationships which may be considered as potential competing**

interests:

### **Credit Author Statement**

- **Jolanta Sobczak: Investigation, Formal analysis, Writing - Original Draft,**
- **Javier P. Vallejo: Investigation, Formal analysis, Writing - Original Draft,**
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- **Gawel 'Zyl a: Conceptualization, Methodology, Investigation, Formal analysis, Validation, Resources, Writing - Original Draft, Writing - Review & Editing, Visualization, Supervision.**

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#### Highlights

- • Two types of carbon black nanoparticles were used in EG based nanofluids.
- • Thermal conductivity, heat capacity, mass density, surface tension were examined.
- • Electrical conductivity and refractive index were investigated as well.
- • Flow and viscosity curves were presented with Herschel-Bulkley model fitting.
- • Specific surface area of particles influence the physical properties of nanofluids