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Viscosity and thermal conductivity of MgO–EG nanofluids

Experimental results and theoretical models predictions

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Abstract Nanofluids are a group of novel engineering materials that are increasingly being used, particularly in the processes of heat exchange. One of the most promising materials in this group is magnesium oxide–ethylene glycol (MgO–EG) nanofluid. The literature informs that this material is characterized by an significant increase in thermal conductivity with low dynamic viscosity increase. The aim of this paper is to provide experimental data on the dynamic viscosity and thermal conductivity of nanofluids containing MgO nanoparticles with 20 nm average size and ethylene glycol as base fluid. To determine dynamic viscosity and thermal conductivity of samples, a HAAKE MARS 2 rheometer (Thermo Electron Corporation, Karlsruhe, Germany) and KD2 Pro Thermal Properties Analyzer (Decagon Devices Inc., Pullman, Washington, USA) were used. Additionally, a comparison of the experimental results and the predictions of theoretical models was presented. It was presented that the vast majority of theoretical models does not describe in a correct way both viscosity and thermal conductivity. It was also shown that the enhancement of this basic physical properties might be described with good result with second degree polynomials. Finally, evaluation of the heat transfer performance was presented.

Keywords Nanofluid · Thermal conductivity · Viscosity · MgO · Ethylene glycol

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Introduction

Nanofluids, suspensions of nanoparticles in a liquid base, are an interesting group of materials which, due to the increased thermal conductivity, are used in various industries [1–3], especially in energy sector [4–6] and the automotive industry [7, 8].

The increase in thermal conductivity of nanofluids containing various types of nanoparticles was intensively studied [9–25]. However, when planning the practical use of nanofluids, it should be taken into account that with increasing concentration of the nanoparticles in a liquid its viscosity increases [19, 20, 26]. In some cases, an increase in viscosity may also be associated with a change of liquid from Newtonian to non-Newtonian [27–33]. It was also demonstrated that the addition of nanoparticles changes the electrical properties of ethylene glycol [34–37].

Xie et al. [38] pointed out that MgO nanofluids present higher thermal conductivity among ethylene glycol-based nanofluids containing oxide nanoparticles and low viscosity. These properties make MgO–EG nanofluids particularly interesting from the point of view of future applications. Adio et al. [39] present results of their work on experimental investigation and model development for effective viscosity of magnesium oxide–ethylene glycol nanofluids. They used dimensional analysis, FCM-ANFIS and GA-PNN techniques to prepare model of viscosity depend on temperature and volume fraction of nanoparticles. Other properties of this material, which are extensively studied, are the pH and electrical conductivity. Adio et al. [40] present results of experimental investigation into the pH and electrical conductivity of MgO–ethylene glycol nanofluids containing nanoparticles with various diameters, and in other paper, they presented factors affecting this properties [41]. Thermal transport properties of MgO–EG nanofluids was the object of study to Yu et al. [42]. Attempt to modeling, the thermal conductivity of MgO–EG nanofluids was conducted by Esfe et al. [43]. They used artificial neural network and presented new model of thermal conductivity enhancement depending on temperature, volume fraction, and particle diameter.

Not only ethylene glycol seems to be an interesting base fluid for MgO nanoparticles. Shoghl et al. [44] present results of experimental studies on electrical conductivity, viscosity, and density of MgO–water nanofluids. Menlik et al. [45] described the possibility of use MgO–water nanofluids in heat pipe. Esfe et al. [46], and Davarnejad and Jamshidzadeh [47] present results of experimental studies and CFD modeling of heat transfer in MgO–water nanofluids under turbulent flow.

MgO nanoparticles can also be used in the advanced heat transfer systems. For example, Manikandan and Rajan [48] describe the opportunities and benefits from use the MgO nanoparticles suspended in Therminol 55. The development of increasingly simple and effective methods of obtaining MgO nanoparticles [49] in various media gives hope for wider use of these materials in industrial processes.

This paper presents the results of experimental research on viscosity and thermal conductivity of MgO–EG nanofluids and also compares the experimental data with the theoretical models used to describe this basic thermophysical properties of nanofluids.

Materials and methods

MgO nanoparticles

The nanoparticles used in the study are a commercially available MgO nanopowder produced by PlasmaChem GmbH (Berlin, Germany) with >99% purity. The particle average size declared by the manufacturer is 20 nm, and specific surface ca. 50 m^2 g^{-1}. The thermal conductivity of MgO was determined by Hofmeister [50], and it is 50.1 W m^{-1} K^{-1} at temperature 298 K. Scanning electron microscope (SEM) picture of dry MgO nanoparticles was taken using a VEGA3 microscope (TESCAN Brno, s.r.o., Brno, Czech Republic). Figure 1 presents SEM image of nanoparticles used to prepare nanofluids. SEM piture presents that particle size corresponding with supplier information.

Sample preparation

Nanofluids were prepared using two-step method through the dispersion of nanoparticles in a base fluid—ethylene glycol (POCH, Avantor Performance Materials Poland, Gliwice, Poland). Samples were prepared in different mass concentration from 0 to 20% with 5% step. Then, after taking into account the density of nanoparticles (3.58 g cm^{-3} [51]) and base fluid, ethylene glycol (1.114 g cm^{-3} [52]) mass concentrations were recalculated to volumetric fractions.

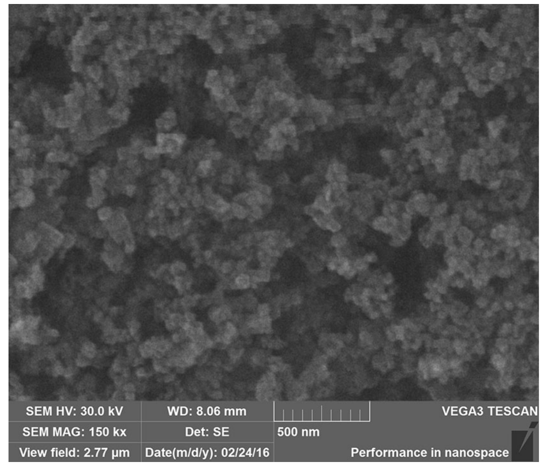


Fig. 1 SEM image of dry MgO nanoparticles

Nanoparticle dispersion process was assisted by mechanical stirring for 30 min in Genius 3 Vortex (IKA, Staufen, Germany), and the sonication for 200 min in ultrasound wave bath Emmi 60 HC (EMAG, Moerfelden-Walldorf, Germany). All samples were prepared at room temperature not exceeding 298.15 K, and due to the possibility of agglomeration and sedimentation of nanoparticles in suspension, all measurements were performed immediately after sonication. Provenance and purities of the used materials are listed in Table 1.

Dynamic viscosity measuring system

HAAKE MARS 2 rheometer (Thermo Electron Corporation, Karlsruhe, Germany), with the minimum measurable torque of 0.5 μNm, was used to determine dynamic viscosity of nanofluids. Temperature was stabilized with use of a Peltier element coupled with a Phoenix 2 thermostat (Thermo Electron Corporation, Karlsruhe, Germany). Double cone measurement geometry with 60 mm diameter and cone angle 1° was used. Dynamic viscosity of samples was measured in the range of shear rates from 100 to 1000 s^{-1} at a constant temperature of 298.15 K. In addition, to ensure the stability of the measurement conditions, measuring geometry was isolated from the environment by glass rings. All rheological measurements were performed immediately after preparation of samples.

Viscosity of pure ethylene glycol determined in this system was 16.9 mPa s with 5% relative standard uncertainty as presented in Ref. [26]. This result correspond with

Table 1 Provenance and purity of the used materials

Product	Provenance	Mass fraction purity
Ethylene glycol	Avantor performance materials Poland	>0.99
MgO nanopowder	PlasmaChem GmbH	>0.99

results presented by Bohne et al. [53], which reported that viscosity of ethylene glycol at 298.15 K is 16.63 mPa s with 5% uncertainty.

Thermal conductivity measuring system

KD2 Pro thermal properties analyzer (Decagon Devices Inc., Pullman, Washington, USA) was used to determine thermal conductivity of nanofluids. Detailed description of the calibration process of this measuring system was presented in Ref. [26]. This device uses the transient line heat source method to measure thermal conductivity, resistivity, diffusivity, and specific heat of liquid samples. The dependence of thermal conductivity of MgO-EG nanofluids on the volume fraction of nanoparticles was measured at constant temperature of 298.15 K, stabilized in a water bath MLL 547 (AJL Electronic, Cracow, Poland). After preparation of the samples, the probe was putted inside, and material was thermostated to a temperature of 298.15 K. After 15 min and reaching a predetermined temperature of the sample material was hold another 15 min with probe inside at constant temperature in order to avoid temperature gradients within the sample. The measurement started exactly 30 min after preparation of the sample. The thermal conductivity values presented in this paper were determined as the average of ten measurements, and the time between successive measurements was 15 min, which corresponds to the recommendations of the manufacturer of equipment. All measurements of thermal conductivity were performed immediately after preparation of samples.

Results and discussion

Viscosity

Figure 2 presents dependence of viscosity on share rate for MgO–EG nanofluids with various volume fractions of nanoparticles. It might be noticed that the dynamic viscosity is considered constant in the examined range of shear rates. Therefore, it can qualify this nanofluids as Newtonian materials. The increase in the volume fraction of nanoparticles increases the viscosity of the suspension, but does not change its nature. It can therefore be assumed that the viscosity of nanofluids is constant and designate it as the average of these measurements. The viscosity and the viscosity enhancement of nanofluids relative to the base fluid are listed in Table 2 and presented in Fig. 3.

On the beginning of XX century, Einstein [54] presented first theoretical prediction of viscosity of suspensions. This model was introduced for spherical particles in low volume fractions and has form of:

$$\frac{\eta_{\mathrm{nf}}}{\eta_{\mathrm{bf}}} = 1 + 2.5\varphi_{\mathrm{v}}, \qquad (1)$$

where η_{nf} and η_{bf} are dynamic viscosities of nanofluid and base fluids, respectively, and φ_{v} is volume fraction.

Einstein's model was the starting point to create other models. Among the most widely used is model presented in 1952. Brinkman presented an expression for the viscosity of suspensions of finite fraction by considering the effect of the addition of one solute-molecule to an existing solution [55]:

$$\frac{\eta_{\mathrm{nf}}}{\eta_{\mathrm{bf}}} = \frac{1}{(1-\varphi_{\mathrm{v}})^{2.5}}. \qquad (2)$$

Batchelor [56] took under consideration the hydrodynamic interaction between particles in a statistically homogeneous suspensions and proposed equation:

$$\frac{\eta_{\mathrm{nf}}}{\eta_{\mathrm{bf}}} = 1 + 2.5\varphi_{\mathrm{v}} + 6.2\varphi_{\mathrm{v}}^2. \qquad (3)$$

Coricione [57] presented empirical correlating equation for predicting dynamic viscosity of nanofluids in form:

$$\frac{\eta_{\mathrm{nf}}}{\eta_{\mathrm{bf}}} = \frac{1}{1 - 34.87\left(d_{\mathrm{p}}/d_{\mathrm{f}}\right)^{-0.3}\varphi_{\mathrm{v}}^{1.03}}, \qquad (4)$$

where d_{bf} is the equivalent diameter of a base fluid molecule:

$$d_{\mathrm{bf}} = 0.1\left(\frac{6M}{N\pi\rho_{\mathrm{bf}}}\right)^{\frac{1}{3}}, \qquad (5)$$

where M is molecular weight of base fluid and N is Avogadro number.

On the other hand, it was already presented that nanoparticles in nanofluids form aggregates. When assume that hydrodynamic forces are too weak to break aggregates and aggregates form spherical flow units, the viscosity ratio

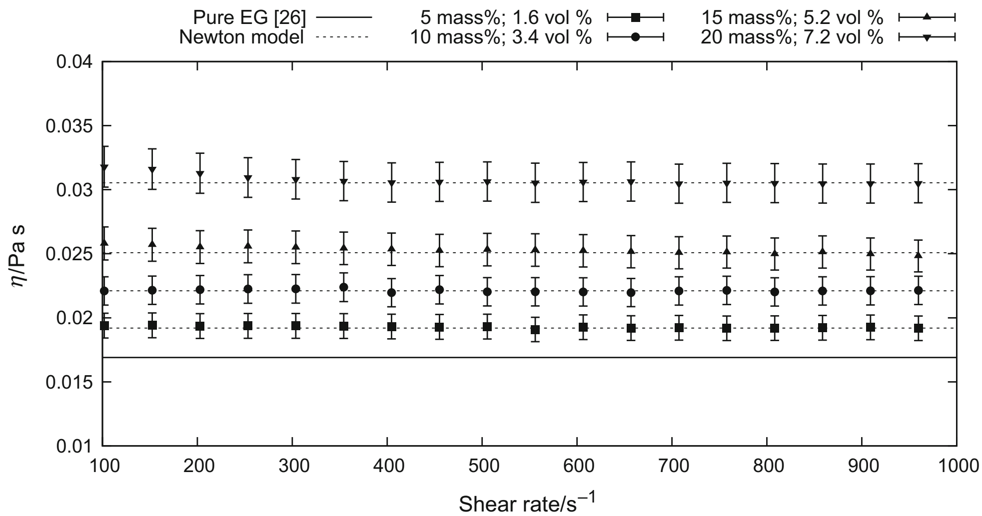


Fig. 2 Dynamic viscosity curves of MgO–EG nanofluids at 298.15 [K]. *Symbols* represent measuring points, lines – the theoretical model fits

Table 2 Experimental values of the viscosity, η_{nf}, of MgO–EG nanofluids at temperature $T = 298.15$ K for various mass fractions φ_m, and volume fractions φ_v

φ_m –	φ_v –	η_{nf}/Pa s	η_{nf}/η_{bf} –
0.00	0.000	0.01690	1.0000
0.05	0.016	0.01920	1.1361
0.10	0.034	0.02211	1.3083
0.15	0.052	0.02508	1.4840
0.20	0.072	0.03054	1.8071

The estimated standard uncertainty $u_r(\eta)$ =5% and $u(T) = 0.10$K

might be modeled with modified Krieger–Dougherty equation:

$$\frac{\eta_{nf}}{\eta_{bf}} = \left(1 - \frac{\varphi_v}{\varphi_c}\left(\frac{d_a}{d_p}\right)^{3-D}\right)^{-[\eta]\varphi_c}, \quad (6)$$

where φ_c is critical particle packing fraction with the value approximately 0.605, D is so called fractal index with the value in the range from 1.8 to 2.5 [58], $[\eta]$ is the intrinsic viscosity with the value of 2.5 for spherical particles, d_a is effective radius of aggregates, and d_p is radius of particle. Equation (6) for nanofluids reduces to:

$$\frac{\eta_{nf}}{\eta_{bf}} = \left(1 - \frac{\varphi_v}{0.605}\left(\frac{d_a}{d_p}\right)^{1.2}\right)^{-1.5125}, \quad (7)$$

as presented in details in Refs. [58–60]. Fitting Eq. (7) to experimental data presented in Table 2 shows that $d_a/d_p = 2.3$.

It might be noticed that for volume fraction lower than 0.04 Batchelor model (3) might be modified with effective volume fraction:

$$\varphi_a = \varphi_v\left(\frac{d_a}{d_p}\right)^{3-D}, \quad (8)$$

to the form:

$$\frac{\eta_{nf}}{\eta_{bf}} = 1 + 2.5\varphi_v\left(\frac{d_a}{d_p}\right)^{3-D} + 6.2\left(\varphi_v\left(\frac{d_a}{d_p}\right)^{3-D}\right)^2. \quad (9)$$

After substituting all values of parameters equation (9) reduces to:

$$\frac{\eta_{nf}}{\eta_{bf}} = 1 + 6.79\varphi_v + 45.76\varphi_v^2. \quad (10)$$

As shown in Fig. 3, this fitting works good in the volume fraction of particles less than 0.04.

Chow [61] proposed variable degree volume fraction polynomial to model the η_{nf}/η_{bf} ratio for suspensions of equal size spherical particles in form of:

$$\frac{\eta_{nf}}{\eta_{bf}} = 1 + \sum_{i=1}^{N} C_i\varphi_v^i, \quad (11)$$

where N is polynomial degree, and C_i are the correlation coefficients. To properly model the experimental data, a second degree polynomial was employed:

$$\frac{\eta_{nf}}{\eta_{bf}} = 1 + 6.62\varphi_v + 61.60\varphi_v^2. \quad (12)$$

Fitting parameters were calculated using OriginPro 9.1 (OriginLab Corporation, Northampton, USA) with coefficient of determination $R^2 = 0.9998$.

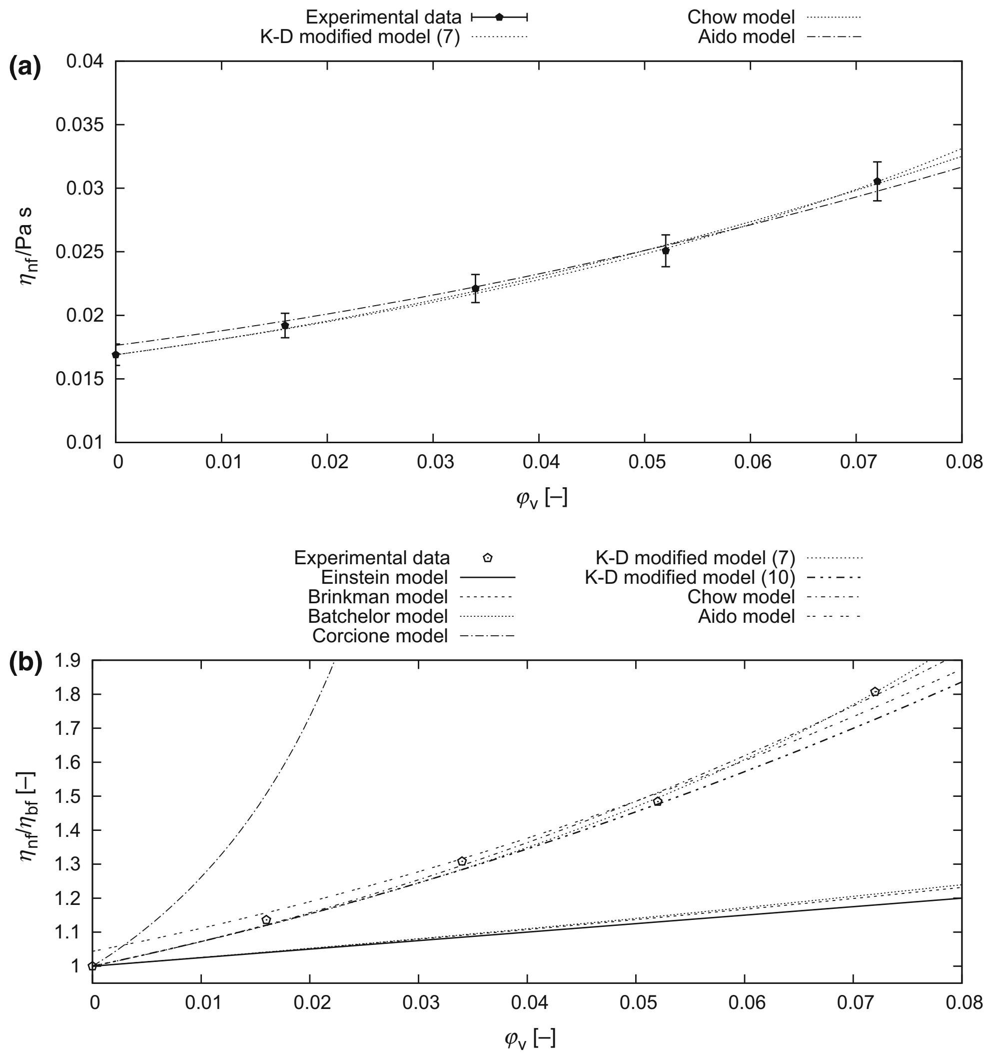


Fig. 3 **a** Dependence of viscosity of MgO–EG nanofluids on volume fraction of particles, and **b** viscosity enhancement of MgO–EG nanofluid at 298.15 K. *Symbols* represent measuring points, *lines*—theoretical models

Adio et al. [39] proposed another model introduced specially for MgO–EG nanofluids on the basis of nonlinear regression modeling:

$$\frac{\eta_{\mathrm{nf}}}{\eta_{\mathrm{bf}}} = 1 + a_0\varphi_{\mathrm{v}} + a_1\left(\frac{T'}{T'_0}\right)\varphi_{\mathrm{v}} + a_2\left(\frac{d_{\mathrm{p}}}{h}\right)\varphi_{\mathrm{v}} + a_3\left(\left(\frac{d_{\mathrm{p}}}{h}\right)\varphi_{\mathrm{v}}\right)^2 + a_4\left(\left(\frac{T'}{T'_0}\right)\varphi_{\mathrm{v}}\right)^2 + a_5\varphi_{\mathrm{v}}^2 + a_6\left(\frac{T'}{T'_0}\right)^2\varphi_{\mathrm{v}}^{1/3}, \quad (13)$$

where a_0–a_6 are empirical constants given as $a_0 = 7.0764, a_1 = -0.1246, a_2 = -0.0346, a_3 = -0.0024, a_4 = -1.2357, a_5 = 53.6946$ and $a_6 = 0.0436, T'$ is working temperature in Celsius degrees, T'_0 is reference temperature in Celsius degrees taken as 20 °C, h is thickness of the capping layer (nanolayer) taken as 1 nm. They assumed that this correlation is valid for volume fraction of MgO nanoparticles ≤5%, temperature between 20–70 °C and particle size between 21 and 125 nm.

Figure 3 presented both measuring points, and theoretical models fits. It may be noticed that only the modified K-D (7), Chow (12) and Adio (13) models correctly describes MgO–EG nanofluid in examined volume fraction range.

Thermal conductivity

Figure 4 presents results of ten subsequent measurements of thermal conductivity, and the average calculated based on them. These results show that MgO–EG nanofluids were stable several hours after preparation.

The results of experimental studies are summarized in Table 3. Additionally, there is a column that presents thermal conductivity enhancement in this material. The obtained results show that the thermal conductivity increases with increasing fraction of nanoparticles in suspensions.

Figure 5 present dependence of thermal conductivity on volume fraction of particles of MgO–EG nanofluids at constant temperature 298.15 K.

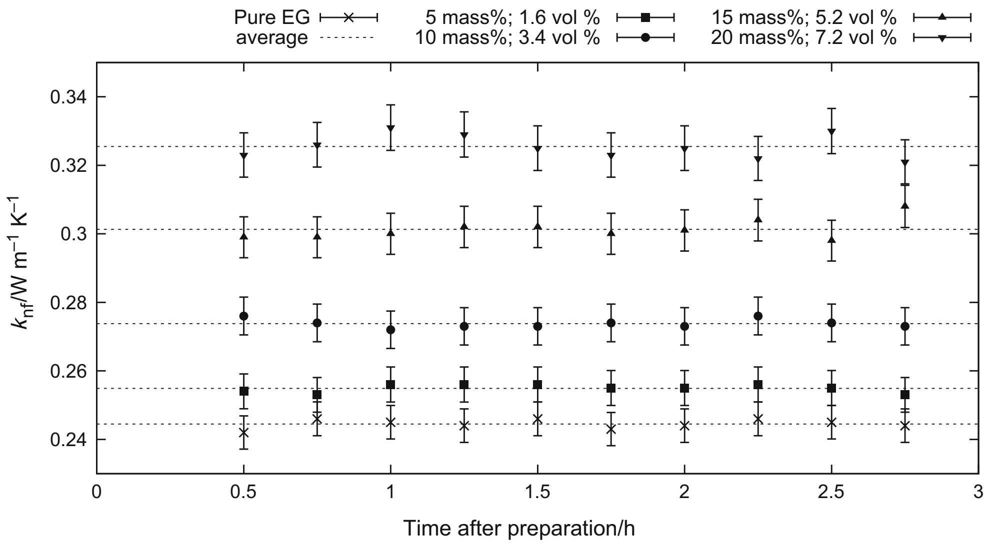


Fig. 4 Time dependence of thermal conductivity for various volume fractions of nanoparticles

Table 3 Experimental values of the thermal conductivity, k_{nf}, of MgO–EG nanofluids at temperature $T = 298.15K$ for various mass fractions φ_m, and volume fractions φ_v

φ_m –	φ_v –	k_{nf} W m^{-1} K^{-1}	k_{nf}/k_0 –
0.00	0.000	0.2445	1.0000
0.05	0.016	0.2549	1.0425
0.10	0.034	0.2738	1.1198
0.15	0.052	0.3013	1.2323
0.20	0.072	0.3255	1.3313

The estimated standard uncertainty $u_r(k_{nf}) = 2\%$ and $u(T) = 0.10K$

Historically the first theoretical model describing thermal conductivity of suspension of large spherical particles was introduced by Maxwell [62]:

$$\frac{k_{nf}}{k_{bf}} = \frac{k_p + 2k_{bf} + 2(k_p - k_{bf})\varphi_v}{k_p + 2k_{bf} - (k_p - k_{bf})\varphi_v}, \quad (14)$$

where k_{nf}, k_{bf}, and k_p are thermal conductivities of nanofluid, base fluid, and particles, respectively.

Another commonly used model was introduced by Jeffrey [63]. This model was also introduced for suspensions of spherical particles in low fractions, and it assumes that the increase in thermal conductivity is accurate to φ_v^2:

$$\frac{k_{nf}}{k_{bf}} = 1 + 3\beta\varphi_v + \left(3\beta^2 + \frac{3\beta^3}{4} + \frac{9\beta^3}{16}\frac{\gamma+2}{2\gamma+3} + \frac{3\beta^4}{64} + \ldots\right)\varphi_v^2, \quad (15)$$

where $\beta = \frac{\gamma-1}{\gamma+2}$, and $\gamma = \frac{k_p}{k_{bf}}$.

Yamada and Ota [64] introduced a similar model in form of:

$$\frac{k_{nf}}{k_{bf}} = \frac{1 + K\alpha + K(1-\alpha)\varphi_v}{1 + K\alpha - (1-\alpha)\varphi_v}, \quad (16)$$

where $\alpha = \frac{k_{bf}}{k_p}$ and K factor depends on shape of particles. For spherical particles $K = 2\varphi_v^{-0.2}$, and for cylindrical particles $K = 2\varphi_v^{0.2}(a/b)$, and $2a$ and b are the length and radius of cylindrical particle, respectively.

Another modification of Maxwell model dedicated for carbon nanotubes based composites was presented by Xue [65]

$$\frac{k_{nf}}{k_{bf}} = \frac{1 - \varphi_v + 2\varphi_v \frac{k_p}{k_p - k_{bf}} \ln\frac{k_p + k_{bf}}{2k_{bf}}}{1 - \varphi_v + 2\varphi_v \frac{k_{bf}}{k_p - k_{bf}} \ln\frac{k_p + k_{bf}}{2k_{bf}}}. \quad (17)$$

This model relates the thermal conductivity enhancement only with volume fraction, and other factors like geometry of particles were neglected.

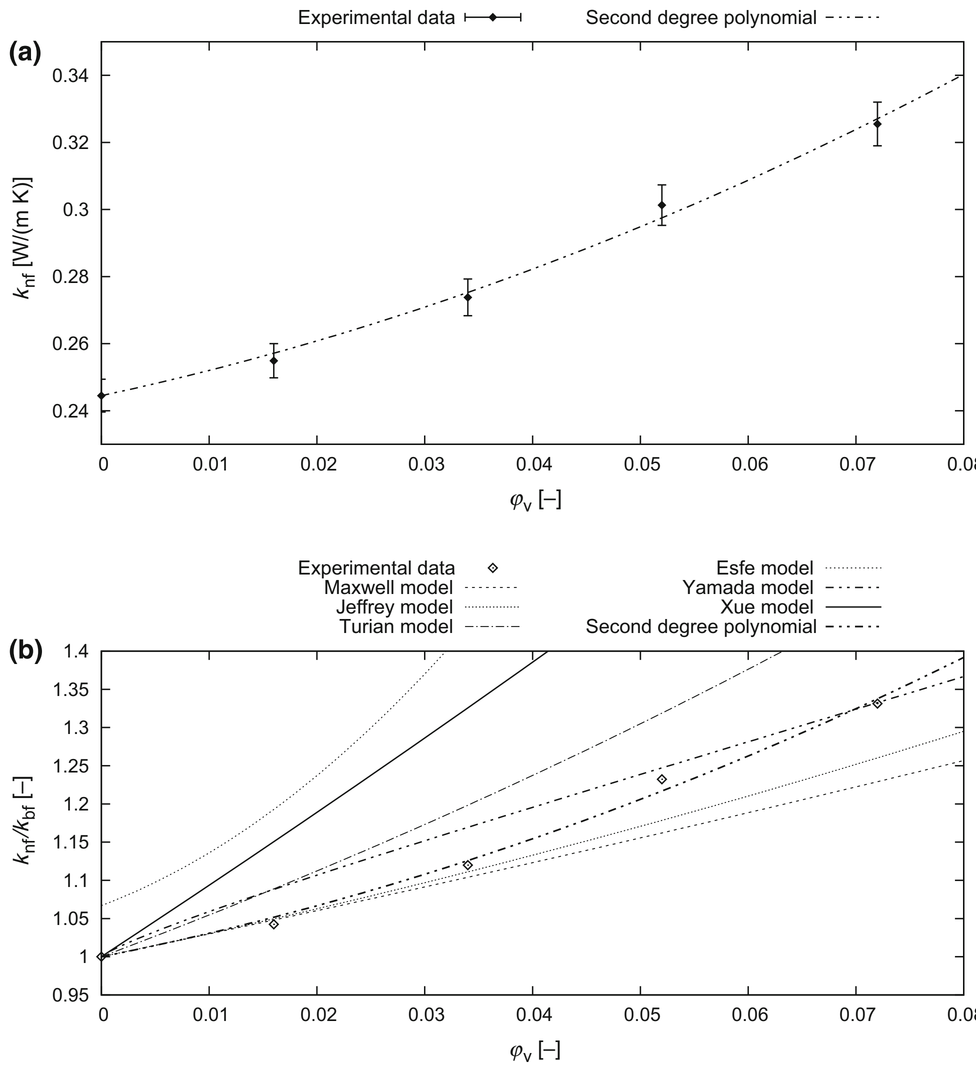
Turian et al. [66] presented model based on experimental results for thermal conductivities of suspensions of particulate coals in water, in fuel oil and in other liquids:

$$\frac{k_{nf}}{k_{bf}} = \frac{k_p^{\varphi_v} k_{bf}^{1-\varphi_v}}{k_{bf}}. \quad (18)$$

Esfe et al. [43] used artificial neural network to predict thermal conductivity enhancement in MgO–EG nanofluids. They introduced empirical correlation to calculate the thermal conductivity ratio for MgO–EG nanofluids. The correlation as a function of volume fraction, temperature, and particle diameter can be written as follows:

$$\frac{k_{nf}}{k_{bf}} = A + (B \cdot T') + (C \cdot \varphi_v) + (D \cdot d_p) + (E \cdot (T'^2)) + (F \cdot (\varphi_v^2)) + \left(G \cdot d_p^2\right). \quad (19)$$

Fig. 5 **a** Dependence of thermal conductivity of MgO–EG nanofluids on volume fraction of particles, and **b** Thermal conductivity enhancement of MgO–EG nanofluid at 298.15 K. *Symbols* represent measuring points, *lines*—theoretical models



This model was proposed for volume fractions less than 5%, the temperature range between 25 and 55 °C, and particle size between 20 and 60 nm. Constants of proposed correlation were calculated by them as: $A = 1.1461, B = 0.0052, C = 5.3056, D = -0.0159, E = -7.09 \cdot 10^{-5}, F = 160, G = 3.83 \cdot 10^{-4}$.

Also, as in the case of an increase in viscosity, thermal conductivity enhancement can be described using a second degree polynomial:

$$\frac{k_{nf}}{k_{bf}} = 1 + 2.82\varphi_v + 25.97\varphi_v^2. \tag{20}$$

Fitting parameters were calculated using OriginPro 9.1, $R^2 = 0.9999$.

Figure 5b presents experimental results and theoretical models for thermal conductivity enhancement depending on volume fraction. As presented on Fig. 5a, only Eq. (20) models thermal conductivity of MgO–EG nanofluids correctly.

Evaluation of heat transfer performance: an engineering approach

Based on viscosity and thermal conductivity enhancement, it is possible to perform evaluation of heat transfer performance. When consider using nanofluids in laminar flow, it might be compared on the base of ratio of enhancement in viscosity and thermal conductivity as presented by Prasher et al. [67]:

$$\frac{C_\eta}{C_k} = \frac{(\eta_{nf} - \eta_{bf})/\eta_{bf}}{(k_{nf} - k_{bf})/k_{bf}}. \tag{21}$$

When this ratio is less than 4, nanofluid might be consider as beneficial for use in energy transport systems.

Benefits from use particular nanofluid for heat transfer performance in turbulent flow might be evaluated on the base of the Mouromtseff number [68]:

$$Mo = \frac{\rho^{0.8} k^{0.67} c_p^{0.33}}{\eta^{0.47}}, \tag{22}$$

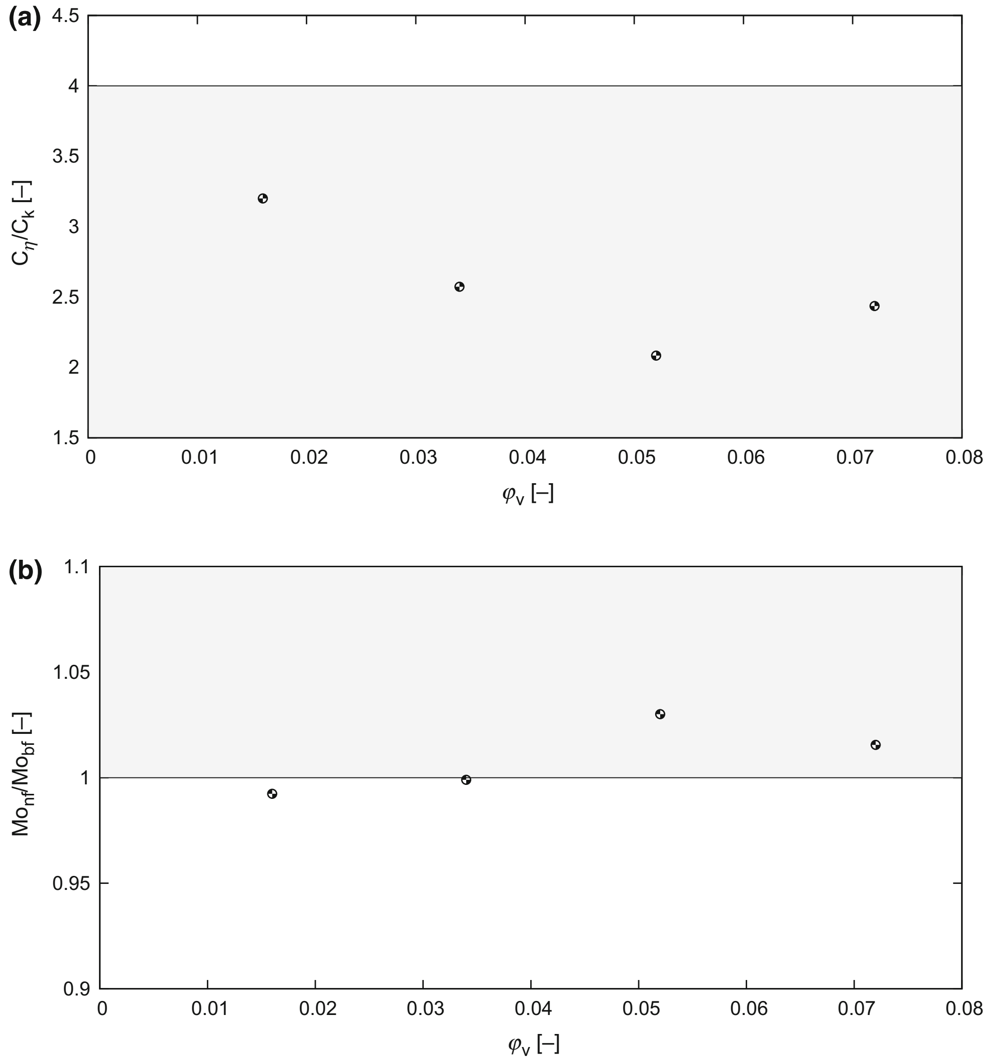


Fig. 6 Dependence of **a** C_η/C_k, and **b** Mo_{nf}/Mo_{bf} ratios on volume fraction of particles in MgO–EG nanofluids at constant temperature 298.15 K. *Grey areas* are beneficial for **a** laminar, and **b** turbulent flows

where ρ is density and c_p is specific heat (at constant pressure). Nanofluids with higher Mo number present better heat transport capabilities. When the ratio of Mo number for nanofluid is higher than base liquid, then material might evaluate as beneficial for turbulent flow applications:

$$\frac{Mo_{nf}}{Mo_{bf}} > 1. \tag{23}$$

Viscosity and thermal conductivity necessary to calculate the Mo number was presented in this paper, and density and specific heat of nanofluids might be calculated as presented by Pak and Cho [69]:

$$\rho_{nf} = (1 - \varphi_v)\rho_{bf} + \varphi_v\rho_p, \tag{24}$$

$$c_{p,nf} = (1 - \varphi_v)c_{p,bf} + \varphi_v c_{p,p}. \tag{25}$$

The values of specific heat of MgO and ethylene glycol are well known and reported in the literature [51, 70], so it is possible to compare nanofluids with pure ethylene glycol based on their thermal properties. Figure 6 presents results of this evaluation. It is clearly visible that the greatest benefits for both applications for laminar and turbulent flows exhibit nanofluid with 0.052 volume fraction of particles.

Conclusions

The paper presents results of research on basic thermo-physical properties of MgO–EG nanofluids. It was presented that with increasing volume fraction of nanoparticles in suspension viscosity of the material increase. At the same time, it was confirmed that the material exhibit Newtonian nature. It was also confirmed that the empirical model proposed by Adio et al. (13) describes the enhancement of the viscosity in MgO–EG nanofluids correctly. It was presented that it is possible to model an increase of the viscosity by using modified K-D model (7) a second degree polynomial (12). It has been shown that the enhancement of thermal conductivity can be modeled also by using a second degree polynomial (20), and the classical models of thermal conductivity do not describe the MgO–EG nanofluids correctly. Comparison of MgO–EG nanofluids with pure ethylene glycol from the point of view of the benefits for applications in laminar and turbulent flows exhibits that nanofluid with 0.052 volume fraction of particles presents the best heat transfer capabilities.

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