

# INFLUENCE OF PARTICLE PROPERTIES ON CONVECTIVE HEAT TRANSFER OF NANOFLUIDS

Mikkola V.<sup>1</sup>, Puupponen S.<sup>1</sup>, Granbohm H.<sup>2</sup>, Saari K.<sup>1</sup>, Ala-Nissila T.<sup>3,4</sup> and Seppälä A.<sup>1\*</sup>

\*Author for correspondence

<sup>1</sup>Aalto University School of Engineering, Department of Mechanical Engineering, Thermodynamics and Combustion Technology, P.O. Box 14400, FI-00076 Aalto, Finland

<sup>2</sup>Aalto University School of Chemical Technology, Department of Chemistry and Materials Science, Advanced and Functional Materials, P.O. Box 16200, FI-00076 Aalto, Finland

<sup>3</sup>Aalto University School of Science, Department of Applied Physics and COMP CoE, P.O. Box 11000, FIN-00076 Aalto, Finland

<sup>4</sup>Loughborough University, Departments of Mathematical Sciences and Physics, Loughborough, Leicestershire LE 11 3TU, United Kingdom.

E-mail: [ari.seppala@aalto.fi](mailto:ari.seppala@aalto.fi)

## ABSTRACT

An experimental study is performed in order to examine how particle properties such as size and thermal conductivity affect the convection heat transfer of nanofluids. For this purpose, we prepare and study self-synthesized water-based nanofluids with different kinds of particles: polystyrene, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and micelles. Concentrations of the nanofluids vary in the range of 0.1–1.8 vol-% and particle sizes between 8–58 nm. Full-scale convective heat transfer experiments are carried out using an annular tube heat exchanger with the Reynolds numbers varying in the range of 1000–11000. The pressure losses are also taken into account in the analysis in order to assess the feasibility of the nanofluids for practical forced convection heat transfer applications. The fluids are thoroughly characterized: viscosities, thermal conductivities, densities, particle size distributions, shapes and zeta potentials are all determined experimentally. In many previous studies, anomalous enhancement in convective heat transfer is observed based on comparison of the Nusselt numbers with equal Reynolds numbers. Also in this work, the nanofluids exhibit Nusselt numbers higher than water when compared on this basis. However, this comparison neglects the impact of differences in the Prandtl numbers, and therefore the altered thermal properties of nanofluids are not properly taken into account. In this study, no difference in Nusselt numbers is observed when the Prandtl number is properly considered in the analysis. All nanofluids performed as the Gnielinski correlation predicts, and the widely reported anomalous convective heat transfer enhancement was not observed with any nanoparticle types. Instead, we show that the convection heat transfer behavior of nanofluids can be explained through the altered thermal properties alone. However, addition of any type of nanoparticles was observed to change the fluid properties in an unfavorable manner: the viscosity increases significantly, while only moderate enhancement in the thermal conductivity is obtained. The more viscous nanofluids reach lower Reynolds numbers than water with equal pumping powers resulting in lower heat transfer coefficients. However, the increase in viscosity, and therefore also the deterioration of the convective heat transfer, is less pronounced for the nanofluids with smaller particle size indicating that small particle size is preferable for convective heat transfer applications.

## INTRODUCTION

Nanofluids are a modern class of heat transfer fluids, in which typically solid particles with diameters of 1–100 nm are suspended in a liquid medium. The concept of nanofluids was first proposed by Choi *et al.* in 1995 [1] and since then nanofluid research has been thriving. According to the literature, addition of nano-sized particles has been claimed to cause anomalous enhancement in thermal conductivity and convective heat transfer performance of the base fluid. Several experiments suggest that the increment of thermal conductivity is significantly larger than the predicted enhancement according to the well-known Maxwell equation for thermal conductivity of heterogeneous solutions [2–5]. In addition, the convective heat transfer performance of nanofluids has been stated to increase even beyond the effect of the enhanced thermal conductivity [6–11].

The thermal properties of nanofluids are very different from those of conventional heat transfer fluids even with relatively low particle concentrations of only a few vol-%. Typically, the addition of nanoparticles has been observed to increase the following three properties by tens of percents: thermal conductivity, convective heat transfer and viscosity. However, an ongoing debate about the magnitudes of these changes exists, since the results of different groups are often contradictory. In some publications, an anomalous behavior in convective heat transfer has not been observed at all [12–17]. In spite of the large body of research, no theory has been able to provide a solid and well-established explanation for the physical basis of the possible anomalous heat transfer enhancement of nanofluids.

The aim of this study is to experimentally scrutinize the influence of particle properties such as size and thermal conductivity on convective heat transfer of nanofluids. Nine water-based nanofluid samples are prepared, characterized, measured and analyzed. The convective heat transfer is studied with an annular tube heat exchanger with Reynolds numbers varying in the range of 1000–11000. In addition to the convective heat transfer, the analysis includes the change in the required pumping power due to increased viscosity and friction factor caused by the nanoparticles. The nanofluids are also thoroughly characterized; particle sizes, shapes, fluid stabilities, viscosities,

densities and thermal conductivities are all determined experimentally.

## MATERIALS AND METHODS

Several different types of water-based nanofluids were investigated in the present study. The thermal properties of the particle materials studied varied in a wide range (Table 1). For example, the thermal conductivities varied in the range of 0.16 – 36 W/mK and the specific heats in the range of 745 – 2090 J/kgK. The influence of thermal properties of the particle material on the convective heat transfer behavior of nanofluids was evaluated by measuring and comparing two equal concentrations (0.5 vol-% and 1.0 vol-%) of Al<sub>2</sub>O<sub>3</sub> and polystyrene nanofluids (PS2) with similar particle size distributions (~10 nm). Thus, the influence of concentration and particle size was attempted to be kept similar in order to obtain a fair comparison between the two types of nanofluids with different thermal properties of particle materials. The polystyrene nanofluids were self-prepared using a method adopted from Kaiyi and Zhaoqun [18], and a commercial dispersion of Al<sub>2</sub>O<sub>3</sub>(aq) (Nanostructured & Amorphous Materials Inc.) was used for the Al<sub>2</sub>O<sub>3</sub> nanofluid preparation. In addition, a polystyrene nanofluid sample with slightly larger particle size of ~17 nm was prepared and measured (PS1). The influence of concentration on convective heat transfer of nanofluids was evaluated by preparing and measuring three different concentrations of SiO<sub>2</sub> nanofluids. The particle size of the SiO<sub>2</sub> nanofluids (~50 nm) was also significantly larger than those of the other nanofluids (~10 nm) thus providing means to evaluate the influence of the particle size on convective heat transfer. The SiO<sub>2</sub> nanoparticles were self-synthesized using the Stöber method [19]. In addition to these solid-particle nanofluids, a micelle-in-water fluid (~10 nm) was prepared in order to evaluate the influence of the differing particle structure. The micelles were formed using polysorbate20 (Tween20, 81.9 w-%) and sorbitan trioleate (Span85, 18.9 w-%) surfactants.

Particle size distributions were determined with Dynamic Light Scattering (DLS) method using the Malvern Zetasizer Nano ZS apparatus. The results were also verified with the Tecnai F-20 G2 200 kV FEG transmission electron microscope (TEM). The DLS measurements were conducted at temperatures of 20°C and 60°C in order to study the stability of the fluids in the temperature range used in the convective heat transfer measurements. The size distribution of each sample was also verified with DLS after the convective heat transfer measurements. In addition to the particle size distributions, DLS was used to determine zeta potentials of the nanofluids. The zeta potentials were also measured at 20°C and 60°C.

The viscosities were measured with two different types of viscometers in order to ensure the measurement reliability and to compare the functionality of the different measurement methods. The two measurement devices were a Haake falling ball type C viscometer and a Brookfield DV3TLVCJ0 cone/plate rheometer. Based on measurement repetition, the maximum errors for these two measurement methods were estimated to be 0.5% and 1.5%, respectively. The temperature range in both viscosity measurements was 20°C-60°C, which was roughly equal to the temperature range of the convective heat transfer measurements.

The thermal conductivities were determined with the C-therm TCi-3-A thermal conductivity analyzer, based on modified transient source plane technique. According to the manufacturer, the uncertainty of the device was 3%. The thermal conductivities were measured at room temperature.

The specific heats of the nanofluids  $c_{p,nf}$  were obtained according to Eq. (1) as mass-weighted averages of specific heats of the nanoparticles  $c_{p,s}$  and the base fluid (water)  $c_{p,bf}$ .

$$c_{p,nf} = \frac{1}{\rho_{nf}} [\rho_s \phi c_{p,s} + \rho_{bf} (1 - \phi) c_{p,bf}], \quad (1)$$

where  $\phi$  is the volume fraction of the nanoparticles and  $\rho_s$  and  $\rho_{bf}$  are the densities of the particles and the base fluid, respectively. The densities of the nanofluids were determined using VWR Hydrometers.

**Table 1.** The particle materials and their thermal properties

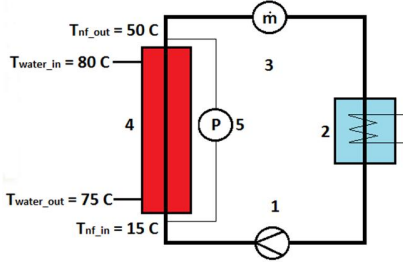
Material	Thermal conductivity (W/mK)	Density (kg/m <sup>3</sup> )	Specific heat (J/kgK)
Polystyrene	0.16[20]	1053[21]	1210[22]
SiO <sub>2</sub>	1.38[22]	2220[22]	745[22]
Al <sub>2</sub> O <sub>3</sub>	36.0[22]	3970[22]	765[22]
Tween20	0.20	1100	2010
Span85	0.17	1000	2090[23]

## CONVECTIVE HEAT TRANSFER MEASUREMENTS

The convective heat transfer experiments were conducted using an annular type heat exchanger, in which the nanofluid samples flowed in the inner tube and hot water flowed in the outer section (Fig. 1). The inner and outer tubes of the heat exchanger were 1.47 m long acid-resistant steel pipes with inner diameters of  $d_i = 6$  mm and  $d_o = 13$  mm, respectively. The thickness of the inner pipe, which corresponds to the wall separating the two fluids, was 1 mm. Thus, the outer diameter of the inner tube was  $d_{io} = 8$  mm. The temperature of the incoming nanofluid was set to 15-20°C. The cooling was arranged using a heat exchanger with cold water flowing in the external side. The outlet temperature of the heated sample varied between 45°C and 78°C, depending on the flow rate. The volumetric flow rate of the nanofluids was varied in the range of 0.13-2.17 l/min. The flow rates were controlled with pump frequency controllers. The hot water in the outer section entered into the heat exchanger at the temperature of 80°C and cooled to 75-80°C, depending on the flow rate of the nanofluid. The flow rate of hot water was kept constant at ~8 l/min in all the measurements. In order to prevent natural convection, the warming nanofluid was arranged to flow upwards in the vertically positioned heat exchanger. Consistently, the water flow on the external side was set to flow downwards.

The temperatures of the nanofluids were measured with two K-type thermocouples (accuracy  $\pm 0.05$  K) at the inlet point and another two at the outlet. Before reaching the outlet thermocouples, the fluids were strangled in a narrow gap of 1 mm in diameter in order to ensure complete mixing of the fluid. With such an arrangement, cross-sectional temperature gradients were minimized thus improving the quality of outlet temperature measurement. The temperature of the hot water was measured

with one thermometer on each side of the tube. The apparatus for measuring pressure losses (Yokogawa DP Harp pressure transmitter, uncertainty 0.04 %) was connected to each side of the inner tube of the heat exchanger, with a distance of 1.68m. The velocities of the nanofluid and water flows were measured with an Optiflux 4000 electromagnetic flow sensor (uncertainty 0.20%). Based on measurement repetition, the maximum experimental errors were estimated to be 1% for both heat transfer coefficients and pumping powers. However, the accuracies of the velocity and pressure loss measurements are limited with very low flow rates (laminar regime) causing the uncertainty for pumping powers to approach 5%.



**Fig 1.** A schematic of the convection heat transfer measurement apparatus: pump (1), cooler (2), flow meter (3), tube-in-tube type heat exchanger (4) and pressure meter (5)

### Calculation of heat transfer coefficient

Heat transfer coefficients were determined based on the measured inlet and outlet temperatures, mass flows and fluid properties. First, a logarithmic temperature difference is calculated using the definition.

$$\theta_{ln} = \frac{(T_{water,in} - T_{nf,out}) - (T_{water,out} - T_{nf,in})}{\ln \frac{T_{water,in} - T_{nf,out}}{T_{water,out} - T_{nf,in}}}, \quad (2)$$

where  $T$  (K) are inlet and outlet temperatures of fluids. The subscript  $nf$  refers to the nanofluid. The conductance  $G$  of the heat exchanger is defined as the ratio of the heat transfer power  $\phi$  and the logarithmic temperature difference  $\theta_{ln}$  as

$$G = \frac{\phi}{\theta_{ln}} = \frac{\dot{m}c_p\Delta T}{\theta_{ln}}, \quad (3)$$

where  $\dot{m}$  is the mass flow,  $c_p$  is the specific heat and  $\Delta T$  the temperature change of the fluid. Conductance per length can be also expressed as

$$\frac{1}{G/L} = \frac{1}{\pi d_i h_i} + \frac{\ln(\frac{d_o}{d_i})}{2\pi \lambda_{tube}} + \frac{1}{\pi d_{io} h_{io}}, \quad (4)$$

where  $d_i$  and  $d_{io}$  are the inner and outer diameters of the inner tube, respectively,  $h_i$  and  $h_{io}$  are the inner and outer heat transfer coefficients, respectively, and  $\lambda$  is the thermal conductivity of the tube material (15 W/mK). The heat transfer coefficient of nanofluid  $h_i$  can be calculated after  $h_{io}$  is obtained using well-known correlations for the Nusselt number of turbulent flow. In this work, the Dittus-Boelter correlation for cooling fluids [6] was used to determine the Nusselt number of the external water side

$$Nu_{DB} = 0.023Re^{0.8}Pr^{0.3}, \quad (5)$$

where  $Re$  is the Reynolds number and  $Pr$  is the Prandtl number of the hot water flow. A hydraulic diameter  $d_h = d_o - d_i$  is applied to the Reynolds number. The Nusselt number was further

corrected to correspond to the geometry of the duct between the annular tubes using a method suggested by Petukhov and Roizen [24].

$$Nu_{ann} = \frac{h_{io}d_h}{\lambda_{water}} = 0.86Nu_{DB} \frac{d_o^{0.16}}{d_{io}} \quad (6)$$

## RESULTS AND DISCUSSION

Nine different nanofluids were characterized and measured to study the effect of particle properties on convective heat transfer performance of the nanofluids. The main properties of the nanofluids are summarized in Table 2.

**Table 2.** The concentration ( $C$ ) and the main material properties of the nanofluids. The particle size was measured with DLS and reported as the peak value of the number distribution. The viscosity ( $\eta$ ), thermal conductivity ( $\lambda$ ), density ( $\rho$ ) and zeta potential ( $\zeta$ ) values were measured at 25 °C. The specific heats ( $c_p$ ) were determined using Eq. (1).

Particle material	$C$ (vol-%)	Particle size (nm)	PdI	$\zeta$ (mV)	$\frac{\lambda_{nf}}{\lambda_w}$	$\frac{\eta_{nf}}{\eta_w}$	$\frac{\rho_{nf}}{\rho_w}$	$\frac{c_{p,nf}}{c_{p,w}}$
SiO <sub>2</sub>	0.09	52	0.04	-50.2	1.00	1.04	1.00	1.00
SiO <sub>2</sub>	0.45	58	0.06	-43.9	0.99	1.08	1.01	0.99
SiO <sub>2</sub>	1.81	47	0.08	-32.3	0.99	1.22	1.02	0.97
Micelle	0.5	8	0.40	-10.9	1.00	1.01	1.00	1.00
Polystyrene	1.0	17	0.10	-53.9	1.00	1.03	1.00	0.99
Al <sub>2</sub> O <sub>3</sub>	0.5	10	0.23	52.3	1.01	1.09	1.01	0.99
Al <sub>2</sub> O <sub>3</sub>	1.0	10	0.26	50.6	1.02	1.21	1.02	0.97
Polystyrene	0.5	12	0.09	-57.8	0.99	1.04	1.00	1.00
Polystyrene	1.0	12	0.12	-40.9	0.98	1.09	1.00	0.99

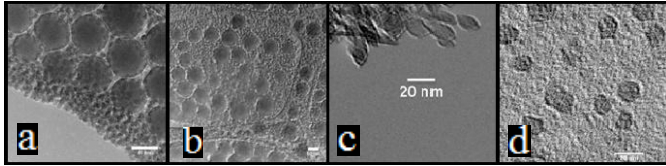
### Structure

The zeta potentials are presented in Table 2. The zeta potentials of SiO<sub>2</sub>, polystyrene and Al<sub>2</sub>O<sub>3</sub> samples were considered to be sufficient for stability since the absolute values exceeded the stability limit of 30 mV. However, for the micelle nanofluid a relatively low zeta potential of -10.9 mV was obtained and thus its stability was considered to be uncertain.

The particle sizes and polydispersity indices (PdI) measured with DLS are presented in Table 2. No significant differences were observed in particle size distributions measured at the two temperatures: 25 °C and 60 °C. The size distributions of the fluids were also measured after the convective heat transfer measurements in order to ensure the dispersion stability during the experiments. All nanofluids except for the micelle fluid remained unchanged during the heat transfer measurements. The size distribution of the micelle sample could not be verified after the heat transfer experiments, since parallel measurements yielded inconsistent results. This indicated that the fluid composition had slightly altered during the heat transfer measurements. Micelles are not strongly bound particles but rather loose assemblies of amphiphilic molecules, and therefore their size and shape may change in flowing systems.

Since DLS assumes the particles to be spherical in shape, the size distributions and shapes for the solid-particle nanofluids were also verified with TEM. The TEM images are presented in Fig 2. The SiO<sub>2</sub> and polystyrene particles were observed to be approximately spherical (Figs. 2a-b and d) and alumina particles were somewhat oval-shaped (Fig. 2c). The TEM images of

polystyrene and  $\text{Al}_2\text{O}_3$  particles were in good agreement with the DLS data, thus confirming the particle size distributions measured. However, the  $\text{SiO}_2$  particles (Figs. 2a and b) can be divided into two groups in terms of size: small particles with an average diameter of  $\sim 10$  nm and large particles with an average diameter of  $\sim 90$  nm. These results differ significantly from the DLS measurements that indicated a distribution with only one peak at a diameter of  $\sim 50$  nm. However, DLS measures the suspension whereas dry samples are imaged in TEM. Therefore, the differences may have been caused by the drying and storage of the TEM samples.



**Fig 2.** TEM-images of  $\text{SiO}_2$  (a,b),  $\text{Al}_2\text{O}_3$  (c) and polystyrene particles (d). The scale bars are 50 nm (a,b) and 20 nm (c,d).

### Viscosity

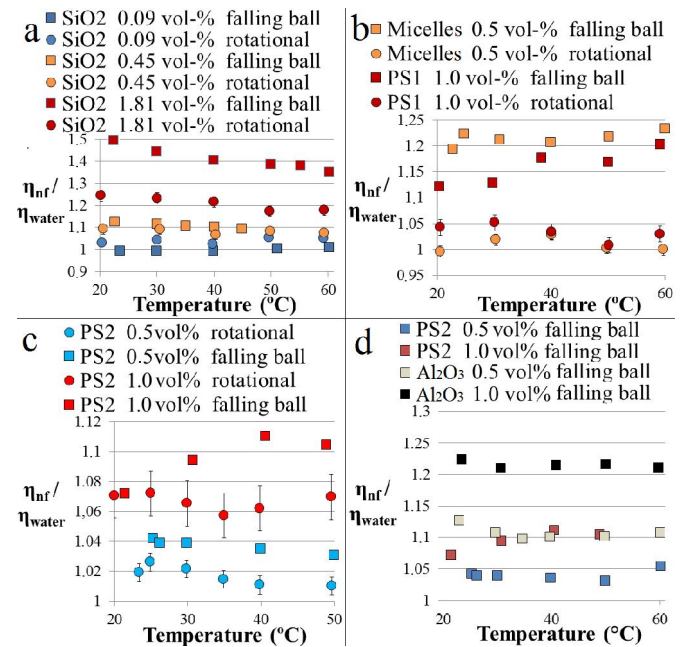
The relative viscosities measured at 25 °C are presented in Table 2. In addition, the relative viscosities are plotted as a function of temperature in Fig 3. The viscosities were measured with two measurement devices: a Haake Type C falling ball viscometer and the Brookfield DV3TLVCJ0 cone/plate rotational rheometer. The results differed significantly depending on which measurement technique was used, as can be seen in Fig. 3. In all the measurements except for one (0.09 vol-%  $\text{SiO}_2$ ), the relative viscosities were lower when measured with the rotational rheometer. The relative magnitude of the difference seems to increase with increasing particle concentration. The largest difference of 20.5% was observed for the 1.81 vol-%  $\text{SiO}_2$  nanofluid. A definite reason for these differences could not be concluded. Nevertheless, the sensitivity of the falling ball viscometer to any larger particles or agglomerates may result in excessively high values. In addition, micelles may break in the rheometer due to vigorous stirring resulting in anomalously low values. Similar uncertainties in viscosity measurements could be one of the reasons for inconsistent results in the literature [25-49]. Furthermore, errors in viscosity measurements could also distort the analysis of convective heat transfer experiments, since the Reynolds number is heavily dependent on the viscosity. In this work, the rotational viscometer was considered to be a more reliable measurement device and thus these results were used in the data analysis of the convective heat transfer experiments. However, the viscosities of  $\text{Al}_2\text{O}_3$  nanofluids could not be measured with the rotational viscometer, since the  $\text{Al}_2\text{O}_3$  particles agglomerated rapidly due to the presence of the chamber material (Aluminium). Therefore, the falling ball viscometer values were used in the analysis of  $\text{Al}_2\text{O}_3$  and PS2 nanofluids in order to obtain a fair comparison.

The addition of nanoparticles caused the fluid viscosities to increase considerably, as expected. A particle fraction of only 0,5 vol-% resulted in viscosity increase of up to 10 % (rotational viscometer). However, the viscosity increase cannot be explained with the concentration alone, but also the particle properties have an influence. The highest increases in viscosities

were measured for the  $\text{SiO}_2$  nanofluids with the largest particle sizes of  $\sim 50$  nm, whereas equal concentrations of very small  $\sim 10$  nm particles ( $\text{Al}_2\text{O}_3$ , polystyrene, micelle) increased the viscosity significantly less. This indicates that small particle size is preferable in terms of viscosity. In addition, the viscosities of  $\text{Al}_2\text{O}_3$  nanofluids were higher than those of the polystyrene nanofluids although both the concentrations and particle size distributions were similar. Therefore, the viscosity of nanofluids seems to depend on other parameters as well.

Typically, the relative viscosity of nanofluids was independent on the temperature, as proposed in several publications [25,26,50]. However, the relative viscosity of the 1.81 vol-%  $\text{SiO}_2$  nanofluid was observed to decrease with increasing temperature (Fig. 3a). This phenomenon was observed with both measurement devices. Similar behavior was also observed by Sundar *et al.* [27].

The Newtonian behavior of the samples was verified with the rotational viscometer. However, the range of shear rates studied (1125-1875 1/s) was relatively high and narrow, since sufficiently high shear rates are required for low viscosity fluids, such as the water-based fluids studied herein. On the contrary, in the falling ball viscometer the fluid is stationary, and the ball descends slowly through the fluid. Therefore, non-Newtonian behavior with very low shear rates cannot be ruled out as a reason for the differences between the results obtained with the two devices, since the measurement conditions of the devices differ from each other in terms of shear rates. In fact, some previous articles have proposed that nanofluids exhibit shear thinning behavior particularly in the regime of very low shear rates [29,52].



**Fig 3.** The relative viscosities of the nanofluids. The error bars are estimated based on the differences between parallel measurements. In case of the falling ball viscometer, the differences were insignificant.

### Thermal conductivity

The relative thermal conductivities of the samples are presented in Table 2. The addition of the nanoparticles caused the thermal conductivities of the fluids to change in an expected manner. The particle material with the highest thermal conductivity ( $\text{Al}_2\text{O}_3$ ) caused the highest enhancement (2%) and the particle material with the lowest thermal conductivity (polystyrene) caused the largest decrease (2%). The change in the thermal conductivity was observed to increase with the particle concentration. However, the differences observed in the thermal conductivities were rather small and, in fact, within the uncertainty of the measurement device (3%). The thermal conductivity enhancements reported in literature for 0.5 – 1 vol-%  $\text{Al}_2\text{O}_3$  nanofluids are typically higher than those measured herein, but also similar values have been reported [4]. Most of the measured conductivities give slightly lower values than the well-known Maxwell equation for dispersed spheres in a base fluid (Table 3). However, the difference between the measurements and the model is minute.

**Table 3.** Difference between measured thermal conductivities of solid particle nanofluids (values presented in Table 2) and those from the Maxwell equation.

Particle material	C (vol-%)	Particle size (nm)	Difference %
$\text{SiO}_2$	0.09	52	-0.1%
$\text{SiO}_2$	0.45	58	-1.4%
$\text{SiO}_2$	1.81	47	-2.6%
Polystyrene	1.0	17	+1.0%
$\text{Al}_2\text{O}_3$	0.5	10	-0.4%
$\text{Al}_2\text{O}_3$	1.0	10	-0.9%
Polystyrene	0.5	12	-0.5%
Polystyrene	1.0	12	-1.0%

### Convective heat transfer

The Nusselt numbers are plotted as a function of the Reynolds numbers (averaged along the tube length) in Fig. 4. Typically, the nanofluids reached higher Nusselt numbers than water with an equal Reynolds number and the difference increased with increasing concentration. The largest difference of 12.5-20.5% was obtained for the sample with the highest particle fraction: 1.81 vol-%  $\text{SiO}_2$  nanofluid. Similar behavior has been reported widely in the literature [6-8,16,52-55]. However, this presentation method has been criticized in several recent publications [14-17,52,56-58]. The standard method does not take the pumping power into account and therefore the suitability of the fluids for practical forced convection applications cannot be properly assessed. In addition, the method disregards the effect of the Prandtl number leaving the altered thermal properties of nanofluids incompletely accounted for. Therefore, the method is also unable to state whether the measured convective heat transfer is truly unexpected, or simply a result of the altered fluid properties. To account for this, a direct comparison between the experimental results and the Gnielinski correlation [7] is presented. The correlation is valid in both the

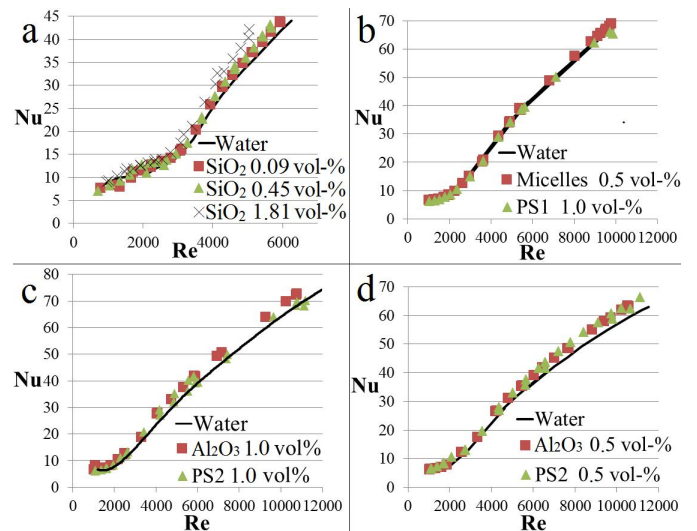
transition and turbulent regimes for  $2300 < Re < 5 \times 10^6$  thus covering the majority of the experimental results [7].

$$Nu_{correlation} = \frac{\left(\frac{f}{2}\right)(Re-1000)Pr}{1+12.7\left(\frac{f}{2}\right)^{1/2}(Pr^{2/3}-1)} \quad (7)$$

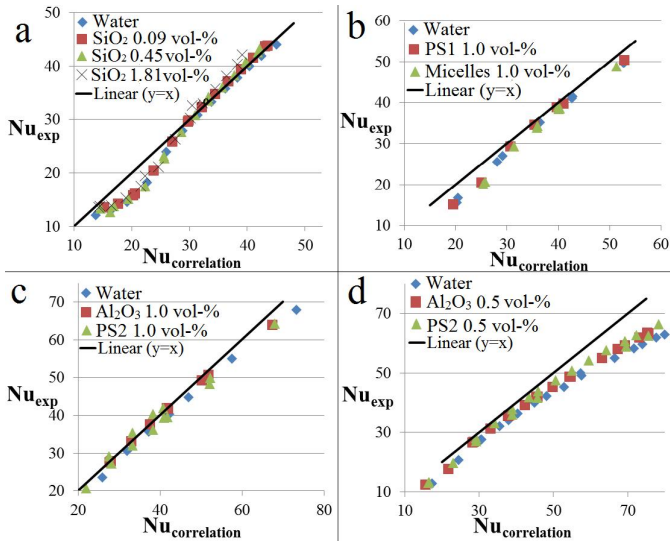
Experimental friction factors were not used in Eq. (7), since purely correlation-based reference values were desirable. Instead, the friction factors for this purpose were determined based on the Blasius Law for turbulent flow in a pipe [59].

$$f = 0.316Re^{-0.25} \quad (8)$$

In Fig. 5, the measured  $Nu$  ( $Nu_{exp}$ ) are presented as a function of  $Nu$  calculated based on the Gnielinski correlation ( $Nu_{correlation}$ ). The correlation seems to be able to explain the heat transfer behavior of all the nanofluids regardless of the different particle properties. In particular with higher Nusselt numbers, where the flow is approaching a fully turbulent regime, the experimental values agree with the predicted values accurately. In addition, the nanofluids and water behave almost similarly when presented with this method that takes both  $Re$  and  $Pr$  into account. The Nusselt numbers of all nanofluids were within 5% from that of water, and no anomalous heat transfer behavior was observed. In Fig. 5d, the experimental results do not follow the correlation very well, but the nanofluids show slightly deteriorated heat transfer performance instead. However, the same deterioration of the heat transfer performance can be observed for the reference water sample as well. The difference between the predicted and measured values in Fig. 5d can be attributed to a thin thermal resistance layer on the surface of the measurement tube caused by an earlier measured unstable polystyrene sample. However, regardless of the resistance layer, the measurements are comparable within the same measurement set. Due to the similar results between the nanofluids and the water reference, no anomalous heat transfer behavior was observed for these fluids either.



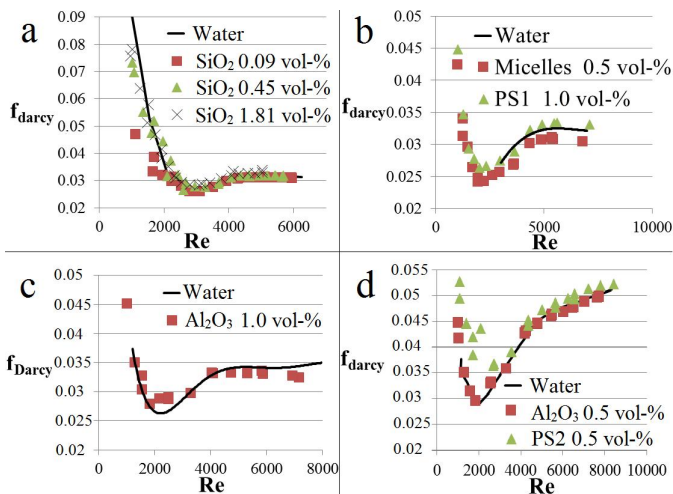
**Fig 4.** The Nusselt numbers as a function of the Reynolds numbers.



**Fig 5.** The experimental results compared to the Gnielinski correlation.

### Friction factors

The Darcy friction factors are presented in Fig. 6. In all cases, the friction factors of the nanofluids were approximately equal to those of water with equal  $Re$ . In the turbulent flow regime, all the data are in agreement. Likewise, the behavior in the laminar and transition flow regimes is similar for all fluids measured. The pressure loss measurements of 1 vol-% polystyrene nanofluid (PS2) failed due to the aforementioned fluid instability and these results are not presented here. Also the subsequently measured 0.5 vol-%  $Al_2O_3$  and polystyrene fluids exhibited increased pressure losses (Fig. 6d). Therefore, the friction factors of these fluids can be compared only with each other and with the corresponding water reference. Overall, the differences in the friction factors of the nanofluids and water were rather small and thus, all differences were interpreted to be within the measurement uncertainty. Therefore, the nanofluids were observed to behave as conventional fluids in terms of the friction factors.



**Fig 6.** The friction factors of the nanofluids.

### Convective heat transfer efficiency

Figure 7 describes the effective heat transfer performance of the fluids: the heat transfer coefficients (HTC) are presented as a function of the pumping power. On this basis, the addition of nanoparticles seems to deteriorate the heat transfer performance. In all cases, the nanofluids showed lower or similar performance to that of water. The differences in convective heat transfer efficiencies arise from the differences in fluid properties, since all the nanofluids were observed to perform as conventional fluids in terms of both the convective heat transfer and the friction factors. The addition of nanoparticles was observed to change the fluid properties in an unfavorable manner regardless of the particle type: the viscosities increased significantly in all cases, while only moderate enhancements in the thermal conductivities were observed. With equal pumping powers, the more viscous nanofluids reach lower Reynolds numbers than water resulting in lower heat transfer coefficients (Fig. 7). The viscosity increases with concentration, which causes the heat transfer efficiency to deteriorate further. Similar decrease of heat transfer efficiency with increasing concentration has been also reported in some earlier publications [50,60,61]. The  $SiO_2$  nanofluids with the highest viscosities (and the largest particle sizes) show the most notable deterioration in the heat transfer performance, whereas the other fluids with slightly lower viscosities perform almost equally with water. In some previous publications, small particle size has been reported to be preferable for the convection heat transfer of nanofluids, and these results show partial support on the claim [28,29,53,62]. However, the  $SiO_2$  nanofluids differ from the other samples in terms of the particle material as well, and thus no firm conclusions of the particle size effect can be drawn. The alumina and polystyrene nanofluids performed roughly equally and therefore, the thermal conductivity of the particle material does not seem to have a notable impact on the convection heat transfer efficiency with the rather small concentrations studied herein ( $\leq 1$  vol-%).

Although anomalous enhancement in forced convection heat transfer of nanofluids was not observed, enhancement in thermal conductivity caused by the nanoparticles could still be harnessed to improve the heat transfer fluids. Significant increment in thermal conductivity might be obtained for instance by using metallic nanoparticles [62]. However, the addition of the nanoparticles would result in practical enhancement only if the negative effects caused by increasing viscosity and decreasing specific heat could be retained low.

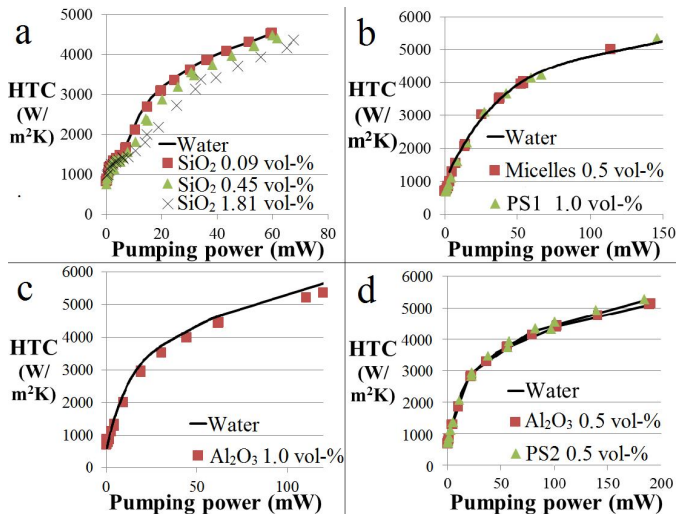


Fig 7. Effective heat transfer performance of the nanofluids.

## CONCLUSIONS

Influence of particle properties on convective heat transfer of nanofluids was experimentally studied. Nine different nanofluids were prepared and measured with an annular tube heat exchanger. The nanofluids were also thoroughly characterized: particle size distributions, shapes, fluid stabilities, viscosities, densities and thermal conductivities were all determined experimentally.

The standard analysis method of presenting the Nusselt numbers solely as a function of the Reynolds numbers indicates enhancement in the convection heat transfer. However, since the Nusselt number is dependent on both the Reynolds and Prandtl numbers, the standard method is found to be questionable and to favor more viscous nanofluids excessively. Indeed, the convection heat transfer enhancement that the standard method suggests can be explained by considering also the Prandtl numbers in the analysis. The convection heat transfer of all the nanofluids followed the well-known Gnielinski correlation for turbulent flow, and no anomalous heat transfer enhancement was observed for any nanofluids regardless of the particle type. In addition, the nanofluids were observed to behave similarly to conventional fluids in terms of friction factors. Therefore, both the convection heat transfer and the friction behavior of nanofluids can be explained through the altered thermal properties.

The addition of any type of nanoparticles was observed to change the fluid properties in an unfavorable manner in terms of forced convection heat transfer applications: the viscosity increases significantly due to the nanoparticles. The viscosity of nanofluids was observed to depend mainly on the concentration and the particle size, but also other parameters seem to have an influence. In addition, the two devices used in the viscosity measurements were observed to yield contradictory results. The values obtained with the falling ball viscometer were typically higher than those obtained with the rotational viscometer. The relative magnitude of the difference increased with increasing particle concentration, and the largest difference observed was 20.5%. Only moderate enhancements in the thermal conductivities were obtained (up to 2%), depending on the

particle type. The influence of particle material on the thermal conductivity of nanofluids was observed to be as expected: the highest conductive particle materials cause the highest enhancement, and vice versa.

The nanofluids exhibited heat transfer efficiency similar or lower than water, when compared with equal pumping powers. The differences in the heat transfer efficiencies were analyzed to be caused mainly by the differences in the viscosities. Increasing the nanoparticle concentration decreased the convective heat transfer efficiency in all cases. However, the decrease in the heat transfer efficiency was less pronounced for nanofluids with smaller particle size indicating that a very small particle size would be preferable for forced convective heat transfer applications. The thermal conductivity of particle material did not have a notable impact on the convection heat transfer efficiency with the relatively low concentrations studied herein ( $\leq 1$  vol-%).

## ACKNOWLEDGEMENTS

The research was funded by the Aalto Energy Efficiency Research Programme (EXPECTS-project) and the Academy of Finland through its COMP CoE grants 251748 and 284621 (T. A.-N.).

## REFERENCES

- [1]Choi, S. U. S. and Eastman, J. A., Enhancing thermal conductivity of fluids with nanoparticles, *ASME International mechanical engineering congress & exposition*, 1995
- [2]Sundar, L. S., Singh, M.K., Sousa, A.C.M., Investigation of thermal conductivity and viscosity of  $Fe_3O_4$  nanofluid for heat transfer applications, *International communications in heat and mass transfer*, vol. 44, pp. 7-14, 2013
- [3]Yu, W., Xie, H., Chen, L., Li, Y., Investigation of thermal conductivity and viscosity of ethylene glycol based ZnO nanofluid, *Thermochimica Acta*, Vol. 491, pp. 92-96, 2009
- [4]Yu, W., France, D.M., Routbort, J.L. and Choi, S.U.S., Review and comparison of nanofluid thermal conductivity and heat transfer enhancements, *Heat transfer engineering*, Vol. 29, 2008
- [5]Esfé, M., Wongwises, S., Naderi, A., Asadi, A., Safaei, M.R., Rostamian, H., Dahari, M. and Karimpour, A., Thermal conductivity of Cu/TiO<sub>2</sub>-water/EG hybrid nanofluid: Experimental data and modeling using artificial neural network and correlation, *International communications in heat and mass transfer*, Vol. 66, pp. 100-104, 2015
- [6]Michaelides, E. E., *Nanofluidics: Thermodynamic and transport properties*, Springer, 2014. ISBN: 978-3-319-05620-3
- [7]Kakac, S., Yener, Y. and Pramuanjaroenkij, A., *Convective heat transfer* (Third edition), CRC Press, Taylor&Francis Group, 2014. ISBN: 978-1-4665-8344-3
- [8]Sarkar, J., A critical review on convective heat transfer correlations of nanofluids, *Renewable and sustainable energy reviews*, Vol. 15, pp.3271-3277, 2011
- [9]Anoop, K. B., Sundarajan, T. and Das, S.K., Effect of particle size on the convective heat transfer in nanofluid in the developing region, *International journal of heat and mass transfer*, Vol. 52, pp. 2189-2195, 2009
- [10]Chen, H., Yang, W., He, Y., Ding, Y., Zhang, L., Tan, C., Lapkin, A.A. and Bavykin, D.V., Heat transfer and flow behaviour of aqueous suspensions of titanate nanotubes (nanofluids), *Powder Technology*, Vol. 183, pp. 63-72, 2008
- [11]Gupta, S., Arora, N., Kumar, R., Kumar, S. and Dilbaghi, N., A comprehensive review of experimental investigations of forced convective heat transfer characteristics for various nanofluids, *International journal of mechanical and materials engineering*, 2014
- [12]Akhavan-Zanjani, H., Saffar-Avval, F., Mansourkiaei, M., Ahadi, M. and Sharif, F., Turbulent convective heat transfer and pressure drop of graphene-water nanofluid flowing inside a horizontal circular tube, *Journal of dispersion science and technology*, Vol. 35, pp. 1230-1240, 2014
- [13]Williams, W., Buongiorno, J. and Hu, L., Experimental investigation of turbulent convective heat transfer and pressure loss of alumina/water and

zirconia/water nanoparticle colloids (nanofluids) on horizontal tubes, *Journal of heat transfer*, Vol. 130, pp. 2008

[14]Haghighi, E. B., Single phase convective heat transfer with nanofluids: An experimental approach (Doctoral thesis), *KTH industrial engineering and management, Department of energy technology, Division of applied thermodynamics and refrigeration*, 2015. ISBN: 978-91-7595-414-1

[15]Mikkola, V., Impact of concentration, particle size and thermal conductivity on effective convective heat transfer performance of nanofluids (Master's thesis), *Aalto University School of Engineering, Department of Energy Technology*, 2015

[16]Mikkola, V., Puupponen, S., Saari, K., Ala-Nissila, T. and Seppälä, A., Thermal properties and convective heat transfer of phase changing paraffin nanofluids, *International Journal of Thermal Sciences*, Vol. 117, pp. 163-17, 2017.

[17]Mikkola, V., Puupponen, S., Granbohm, H., Saari, K., Ala-Nissila, T. and Seppälä, A., Convective Heat Transfer Performance of Polystyrene, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Micelle Nanofluids, 12th International Conference on Heat Transfer, Fluid Mechanics and Thermodynamics (HEFAT12) Proceedings, 2016

[18]Kaiyi, L., Zhaoqun, W., A novel method for preparing monodispersed polystyrene nanoparticles, *Front. Chem. China* 2007 2(1) 17-20.

[19]Stöber, W., Fink, A., Bohn, E., Controlled growth of monodisperse silica spheres in the micron range, *Journal of colloid and interface science*, 26, 62 (1968), DOI:10.1016/0021-9797(68)90272-5

[20]Carwile LCK, Hoge HJ. Thermal conductivity of polystyrene: Selected values, Technical Report 62-27-PR, 1966.

[21]Fox, T. G. and Flory, P.J., Second-order transition temperatures and related properties of polystyrene. I. Influence of molecular weight, *Journal of Applied Physics*, Vol. 21, pp. 581-591, 1950

[22]Incropera, F. P. and De Witt, D. P., Fundamentals of Heat and Mass Transfer (third edition), *John Wiley & Sons, Inc.*, 1990, ISBN: 0-471-51729-1

[23]Puupponen, S., Seppälä, A., Vartia, O., Saari, K. and Ala-Nissila, T., Preparation of Paraffin and Fatty Acid Phase Changing Nanoemulsions for Heat Transfer, *ThermoChimica Acta*, Vol. 601, pp. 33-38, 2015

[24]Cengel, Y. A., Heat transfer - A practical approach, 2. Edition, *McGraw-Hill*, pp. 444, 2003 ISBN: 0-07-115150-8

[25]Esfé, M. and Saedodin, S., An experimental investigation and new correlation of viscosity of ZnO-EG nanofluid at various temperatures and different solid volume fractions, *Experimental thermal and fluid science*, Vol. 55, pp. 1-5, 2014

[26]Chen, H., Ding, Y., He, Y. and Tan, C., Rheological behavior of ethylene glycol based titania nanofluids, *Chemical physics letters*, Vol. 444, pp. 333-337, 2007

[27]Syam Sundar, L., Venkata Ramana, E., Singh, M.K. and De Sousa, A.C.M, Viscosity of low volume concentrations of magnetic Fe<sub>3</sub>O<sub>4</sub> nanoparticles dispersed in ethylene glycol and water mixture, *Chemical physics letters*, Vol. 554, pp. 236-242, 2012

[28]Esfé, M. H. and Saedodin, S., Turbulent convection heat transfer and thermophysical properties of MgO-water nanofluid with concentration of different nanoparticles diameter, an empirical study, *Journal of thermal analysis and calorimetry*, Vol. 119, pp. 1205-1213, 2015

[29]He, Y., Jin, Y., Chen, H., Ding, Y., Cang, D. and Lu, H., Heat transfer and flow behaviour of aqueous suspensions of TiO<sub>2</sub> nanoparticles (nanofluids) flowing upward through a vertical pipe, *International journal of heat and mass transfer*, Vol. 50, pp. 2272-2281, 2007

[30]Mishra, P.C., Mukherjee, S., Nayak, S.K., Panda, A., A brief review on viscosity of nanofluids, *International nano letters*, Vol. 4, pp. 109-120, 2014

[31]Jarahmejad, M., Hanghigi, E.B., Saleemi, M., Nikkam, N., Khodabandeh, R., Palm, B., Toprak, M.S. and Muhammed, M., Experimental investigation on viscosity of water-based Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> nanofluids, *Rheologica acta*, Vol. 54, pp. 411-422, 2015

[32]Corcione, M., Empirical correlating equations for predicting the effective thermal conductivity and dynamic viscosity on nanofluids, *Energy conversion and management*, Vol. 52, pp. 789-793, 2011

[33]Said, Z., Sajid, M.H., Alim, M.A., Saidur, R. and Rahim, N.A., Experimental investigation of the thermophysical properties of Al<sub>2</sub>O<sub>3</sub>-nanofluid and its effect on a flat plate solar collector, *International communications in heat and mass transfer*, Vol. 48, pp. 99-107, 2013

[34]Fedele, L., Colla, L. and Bobbo, S., Viscosity and thermal conductivity measurements of water-based nanofluids containing titanium oxide nanoparticles, *International journal of refrigeration*, Vol. 35, pp. 1359-1366, 2012

[35]Suganthi, K. S., Leela Vinodhan, V. and Rajan, K.S., Heat transfer performance and transport properties of ZnO - ethylene glycol and ZnO -ethylene glycol - water nanofluid coolants, *Applied energy*, Vol. 135, pp. 548-559, 2014

[36]Pastoriza-Gallego, M. J., Casanova, C., Legido, J.L. and Pineiro, M.M. CuO in water nanofluid: Influence of particle size and polydispersity on volumetric behavior and viscosity, *Fluid phase equilibria*, Vol. 300, pp. 188-196, 2011

[37]Namburu, P. K., Kulkarni, D.P., Dandekar, A. and Das, D.K., Experimental investigation of viscosity and specific heat of silicon dioxide nanofluids, *IET micro & nano letters*, Vol. 2, s. 67, 2007]

[38]Yu, L. Liu, D. and Botz, F., Laminar convective heat transfer of alumina-polyalphaolefin nanofluids containing spherical and non-spherical nanoparticles, *Experimental thermal and fluid science*, Vol. 37, pp. 72-83, 2012

[39]Venerus, D.C. et al., Viscosity measurements on colloidal dispersions (nanofluids) for heat transfer applications, *Applied rheology*, Vol. 20, 2010

[40]Jeong, J., Li, C., Kwon, Y., Lee, J., Kim, S.H. and Yun, R., Particle shape effect on the viscosity and thermal conductivity of ZnO nanofluids, *International journal of refrigeration*, Vol. 36, pp. 2233-2241, 2013

[41]Moosavi, M., Goharshadi, E.K. and Youssefi, A., Fabrication, characterization, and measurement of some physicochemical properties of ZnO nanofluids, *International journal of heat and fluid flow*, Vol. 31, pp. 599-605, 2010

[42]Nguyen, C.T., Desgranges, F., Galanis, N., Roy, G., Mare, T., Boucher, S., Angue Mintsa, H., Viscosity data for Al<sub>2</sub>O<sub>3</sub>-water nanofluid - hysteresis: is heat transfer enhancement using nanofluids reliable?, *International journal of thermal sciences*, Vol. 47, pp. 103-111

[43]Said, Z., Saidur, R., Hepbasli, A. and Rahim, N.A., New thermophysical properties of water based TiO<sub>2</sub> nanofluid - The hysteresis phenomenon revisited, *International communications in heat and mass transfer*, Vol. 58, pp. 85-95, 2014

[44]Li, X., Zou, C., Wang, T., Lei, X., Rheological behavior of ethylene glycol-based SiC nanofluids, *International journal of heat and mass transfer*, Vol. 84, pp. 925-930, 2015

[45]Pastoriza-Gallego, M.J., Lugo, L., Legido, J.L. and Pineiro, M. M., Rheological non-Newtonian behaviour of ethylene glycol-based Fe<sub>2</sub>O<sub>3</sub> nanofluids, *Nanoscale research letters*, Vol. 6, pp. 1-7, 2011

[46]Mariano, A., Pastoriza-Gallego, M.J., Lugo, L., Camacho, A., Canzonieri, S. and Pineiro, M.M., Thermal conductivity, rheological behaviour and density of non-Newtonian ethylene glycol-based SnO<sub>2</sub> nanofluids, *Fluid phase equilibria*, Vol. 337, pp. 119-124, 2013

[47]Sadri, R., Ahmadi, G., Togun, H., Dahari, M., Kazi, S.N., Sadeghinezhad, E. and Zubir, N., An experimental study on thermal conductivity and viscosity of nanofluids containing carbon nanotubes, *Nanoscale research letters*, Vol. 9, 2014

[48]Sundar, L.S., Sharma, K.V., Naik, M.T. and Singh, M.K., Empirical and theoretical correlations on viscosity of nanofluids: A review, *Renewable and sustainable energy reviews*, Vol. 25, pp. 670-686, 2013

[49]Yiamsawas, T., Dalkilic, A.S., Mahian, O., Wongwises, S., Measurement and correlation of the viscosity of water-based Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> nanofluids in high temperatures and comparisons with literature reports, *Journal of dispersion science and technology*, Vol. 34, pp. 1697-1703, 2013

[50]Saarinen, S., Puupponen, S., Meriläinen, A., Joneidi, A., Seppälä, A., Saari, K. and Ala-Nissila, T., Turbulent heat transfer characteristics in a circular tube and thermal properties of n-decane-in-water nanoemulsion fluids and micelles-in-water fluids, *International journal of heat and mass transfer*, Vol. 81, pp. 246-251, 2015

[51]Chen W., Yang, W., He, Y., Ding, Y., Zhang, L., Tan, C., Lapkin, A.A. and Bavykin, D.V., Heat Transfer and Flow Behaviour of Aqueous Suspensions of Titanate Nanotubes (Nanofluids), *Powder Technology*, Vol. 183, pp. 63-72, 2007

[52]Wu, Z., Wang, L. and Sunden, B., Pressure drop and convective heat transfer of water and nanofluids in a double-pipe helical heat exchanger, *Applied thermal engineering*, Vol. 60, pp. 266-274, 2013

[53]Meriläinen, A., Seppälä, A., Saari, K., Seitsonen, J., Ruokolainen, J., Puisto, S., Rostedt, N. and Ala-Nissila, T., Influence of particle size and shape on turbulent heat transfer characteristics and pressure losses in water-based nanofluids, *International journal of heat and mass transfer*, Vol. 61, pp. 439-448, 2013

[54]Fotukian, S.M. and Esfahany, M. N., Experimental study of turbulent convective heat transfer and pressure drop of dilute CuO/water nanofluid inside a circular tube, *International communications in heat and mass transfer*, Vol. 37, pp. 214-219, 2010

[55]Dalkilic, A.S., Kayaci, N., Celen, A., Tabatabaei, M., Yildiz, O., Daungthongsuk, W., Wongwises, S., Forced convective heat transfer of



nanofluids – A review of the recent literature, *Current nanoscience*, Vol. 8, pp. 949-969, 2012

[56] Yu, W., France, D.M., Timofeeva, E.V., Singh, D. and Routbort, J.L., Thermophysical property-related comparison criteria for nanofluid heat transfer enhancement in turbulent flow, *Applied physics letters*, Vol. 96, 2010

[57] Yu, W., Comparative review of turbulent heat transfer of nanofluids, *International journal of heat and mass transfer*, Vol. 55 pp. 5380-5396, 2012

[58] Buschmann, M. H., Nanofluid Heat Transfer In Laminar Pipe Flow with Twisted Tape, *Heat Transfer Engineering*, DOI: 10.1080/01457632.2016.1177381, pp. 162-176, 2017

[59] White, F. M., Viscous fluid flow (third edition), *McGraw-Hill*, 2006. ISBN: 007-124493-X

[60] Ferrouillat, S., Bontemps, A., Poncelet, O., Soriano, O. and Gruss, J., Influence of nanoparticle shape factor on convective heat transfer and energetic performance of water-based SiO<sub>2</sub> and ZnO nanofluids, *Applied thermal engineering*, Vol. 51, pp. 839-851, 2013

[61] Bayat, J. and Nikseresht, A. H., Thermal performance and pressure drop analysis of nanofluids in turbulent forced convective flows, *International journal of thermal sciences*, Vol. 60, pp. 236-243, 2012

[62] Mehrali, M., Sadeghinezhad, E., Rosen, M.A., Latibari, S.T., Mehrali, M., Metselaar, H.S.C., Kazi, S.N., Effect of specific surface area on convective heat transfer of graphene nanoplatelet aqueous nanofluids, *Experimental thermal and fluid science*, Vol. 68, pp. 100-108, 2015

[63] Eastman, J. A., Choi, S. U. S., Li, W. Yu., and Thompson, L. J., Anomalous increased effective thermal conductivities of ethylene glycol-based nanofluids containing copper nanoparticles, *Applied Physics Letters*, Vol. 78, 2001, pp. 718-720