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Particle size and interfacial effects on thermo-physical and heat transfer characteristics of water-based α-SiC nanofluids

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Abstract
The effect of average particle sizes on basic macroscopic properties and heat transfer performance of α-SiC/water nanofluids was investigated. The average particle sizes, calculated from the specific surface area of nanoparticles, were varied from 16 to 90 nm. Nanofluids with larger particles of the same material and volume concentration provide higher thermal conductivity and lower viscosity increases than those with smaller particles because of the smaller solid/liquid interfacial area of larger particles. It was also demonstrated that the viscosity of water-based nanofluids can be significantly decreased by pH of the suspension independently from the thermal conductivity. Heat transfer coefficients were measured and compared to the performance of base fluids as well as to nanofluids reported in the literature. Criteria for evaluation of the heat transfer performance of nanofluids are discussed and optimum directions in nanofluid development are suggested.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Nanofluids refer to suspensions of solid particles of nanometer and submicron sizes in various fluids that benefit from the high thermal conductivity of the solid phase and convective heat transfer capability typical for the liquid phase. Multiple reports on the thermal conductivity enhancement summarized in reviews [1–5] attracted significant interest in nanofluids as potential advanced heat transfer fluids. However, the addition of particles to a fluid affects not only thermal conductivity (k), but also other macroscopic properties such as viscosity (η), heat capacity (c_p) and density (ρ) that may differently affect the heat transfer characteristics of the nanofluid. Heat capacity and density depend only on the concentration of the solid phase in the fluid and are unrelated to nanoparticle morphology (size, shape etc). However both thermal conductivity and viscosity are largely determined by the shape and size of the solid particles constituting the nanofluid systems. Therefore, it is clearly important to establish the exact relationship of the size and shape of particles to macroscopic system parameters.

Particle size characterization in nanofluids by itself is a non-trivial task because of the possible size polydispersity, formation of ordered fluid layers at nanoparticle surfaces, agglomeration and clustering of individual particles through solid/solid and solid/liquid/solid interfaces. Most experimental studies use particle sizes from the manufacturer's data sheet that were determined for dry nanopowders (XRD, TEM, SEM). Characterization of particle sizes in solution with light scattering techniques estimates the average hydrodynamic diameter of species involved in Brownian motion (small angle x-ray scattering (SAXS), dynamic light scattering (DLS), laser diffraction (LD)). Therefore, if nanoparticles are agglomerated

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in suspension and/or move in clusters, sizes of individual particles and agglomerates can be confused. The variety and specifics of particle size measuring techniques used in the literature leads to the general trends that particle size effects are difficult to discern.

1.1. Particle size and thermal conductivity

Literature data on the effect of particle size on the thermal conductivity of nanofluids are limited and inconsistent. The work of Xie *et al* [6] does report data for alumina nanofluids in ethylene glycol and pump oil containing five different sizes of alumina particles (∼12–302 nm). However, they observed an increase followed by a decrease in the thermal conductivity with increasing particle size. In contrast, other studies have reported monotonic increases in the thermal conductivity with decreasing particle size, attributed to Brownian motion [7–9] or decreases due to interfacial thermal resistance [4, 10–12]. By comparing results from different research groups [6, 13–15] the review paper [4] estimated an approximate trend that indicated an increase in the thermal conductivity with increasing particle size.

The thermal conductivity enhancement in water and ethylene glycol based alumina suspensions with particle sizes from 8 to 282 nm [10] was reported to decrease as the particle size decreased below about 50 nm, which the authors attributed to phonon scattering at the solid–liquid interface. The limiting value of the enhancement for nanofluids containing large particles was described well by the volume fraction weighted geometric mean of the bulk thermal conductivities of the solid and liquid [16] or predictions of the effective medium theory [17] (EMT [18]) corrected for particle shape by Hamilton and Crosser [19] and for the interface contribution [20].

1.2. Particle size and viscosity

Regarding nanofluid viscosity, the lack of consistent data on particle size effect in the literature is even more striking [21–24]. Inconsistency of data is due to multiple factors affecting the viscosity of suspensions. The electrostatic interaction between the particles and the fluid, called the first electro-viscous effect [25, 26], results in the formation of a diffuse layer of fluid at the particle/liquid interface. This phenomenon increases the effective particle size and thus the effective volume concentration. In addition, the electrostatic interaction between the particles, called the second electro-viscous effect, is also important because it determines particle agglomeration and degrees of freedom of motion in nanofluids. As particle size decreases, the total area of the solid/liquid interface and the number of particles at the same particle volume concentration increases. Therefore both the first and second electro-viscous effects become more important for smaller particle sizes, causing the larger increase in viscosity [26]. Indeed, the larger viscosity increase from nanometer sized particles compared to micron sized particles was observed and studied by multiple groups in the 1950s and 1960s [25–29]. It was found that the Einstein [30] prediction for viscosity, derived for spherical, uncharged, hard, non-interacting particles, worked well for particle sizes between 3 and 300 μm, but the viscosity was always higher than predicted for particle sizes below 500 nm [25, 27, 28].

Since theoretical analysis [31, 32] has shown that the first electro-viscous effect alone cannot explain the experimentally observed increases in viscosity [31], the interaction of the particles (the second electro-viscous effect) must have an important role [32]. It also was shown that viscosity can be significantly modified simply by changing the pH of the suspension, which correlates to surface charges at the solid/liquid interface and interactions between particles [33]. Therefore, it is impossible to compare data and identify the true effect of the particle size on viscosity if the pH of the suspension is not controlled or if surfactants are used [14, 34–37]. In such a study [38] the authors investigated the particle size effect on the viscosity of water–Al_2O_3 (36 and 47 nm) and found that, for volume fractions >4%, viscosities for the 47 nm particle size are clearly higher than those for the 36 nm particle size, deducing that viscosity increases with increasing particle size. However no control over actual particle sizes was provided and the authors did not control the suspension pH. In addition an unknown surfactant from the manufacturer was present.

1.3. Particle size effects in heat transfer studies

Several experimental and numerical studies on convective heat transfer of nanofluids have been published [35, 39–47]. However, few of them consider particle size effects on nanofluid performance.

The evaluation of the particle size effect (45 and 150 nm alumina in water) on convective heat transfer in the developing laminar region [48] showed that both nanofluids have higher heat transfer characteristics than the base fluid and that the nanofluid with 45 nm particles showed higher heat transfer coefficients than that with 150 nm particles.

The heat transfer behavior of TiO_2/water nanofluids was studied under both laminar and turbulent flow conditions [49]. For a fixed particle concentration and Reynolds number, the small increase with increasing sizes (95, 145 and 210 nm) on the convective heat transfer coefficient was ascribed to a possible particle migration or a wide particle size distribution. However, contrary to that study [49], a study for turbulent flow [50] showed that a Al_2O_3 nanofluid with a 36 nm particle diameter, having lower viscosity, provided higher heat transfer coefficients than a similar nanofluid with a 47 nm particle diameter and higher viscosity at the same particle concentration. Such a result was explained [50] by the fact that for a given volume concentration the number of finer particles and their total contact area are higher, which could provide a more effective heat exchange between the nanoparticles and the continuous liquid phase.

Most previous studies of turbulent heat transfer in nanofluids with various particle materials and sizes (Al_2O_3 [35, 51], Cu [52], diamond [53], SiC [54], SiO_2 [55], TiO_2 [35] and ZrO_2 [51]) claim heat transfer enhancement in nanofluids when they are compared to base fluids at the same Reynolds numbers

(Re), which is a dimensionless ratio of inertial forces to viscous forces in the flow. The comparison at constant Re is not always accurate, since it may involve a faster moving nanofluid than its base fluid to compensate for the higher nanofluid viscosity. This type of comparison gives the nanofluid an unfair and unrealistic advantage. A more realistic comparison of fluid heat transfer performance is at constant pumping power, velocity, or flow rate [52, 54]. Comparing on this basis, only 3 of the 11 nanofluids previously studied had higher heat transfer coefficient than those of their base fluid [49, 52, 55]. The remaining eight nanofluids [35, 51, 53–55] had viscosity increases that canceled and sometimes overshadowed the thermal conductivity increase observed in nanofluids.

With the large discrepancy of experimental results and approaches in testing nanofluid heat transfer performance it is important to develop reliable evaluation procedures for comparing nanofluids. Two figures of merit have been developed.

Laminar flow [56]:

$$\frac{(\eta_{\text{eff}} - \eta_0)/\eta_0}{(k_{\text{eff}} - k_0)/k_0} \approx \frac{C_\eta \phi}{C_k \phi} = \frac{C_\eta}{C_k} \leqslant 4; \qquad (1)$$

Turbulent flow [57, 58]:

$$Mo = \frac{\rho^{0.8} c_p^{0.4} k^{0.6}}{\eta^{0.4}}, \qquad \frac{Mo_{\text{eff}}}{Mo_0} > 1, \qquad (2)$$

where C_η and C_k are the viscosity and thermal conductivity enhancement coefficients determined from the experimental viscosity (η_{eff}) and thermal conductivity (k_{eff}) ratios as compared to the base fluid viscosity (η_0) and thermal conductivity (k_0); ϕ represents the volume concentration of solids in the suspension. Mo_{eff} and Mo_0 stand for Mouromtseff values for the nanofluid and base fluid respectively.

In the present study experiments were performed to investigate the effects of the average particle size and the solid/liquid interface on viscosity, thermal conductivity, and ultimately heat transfer coefficient of water-based α-SiC nanofluids. Four different particle sizes (16, 29, 66 and 90 nm) were included. Prior to varying particle size, studies were conducted as to the effect of pH on viscosity and thermal conductivity for a series of nanofluids with the same concentration of 29 nm particles. The results of the present study provide careful correlation of the basic macroscopic properties involving thermal conductivity, viscosity, and heat transfer coefficient of the nanofluids with the physico-chemical parameters including particle sizes, area of solid/liquid interface, pH value and temperature of the colloidal suspensions.

2. Materials and methods

Nanofluid samples used for this study were obtained from Saint Gobain, Northboro, MA. Silicon carbide nanoparticles were prepared by milling larger pieces of SiC material. XRD analysis has confirmed that all the studied nanoparticles are α-phase SiC (hexagonal).

The volume concentration of the solid particles (ϕ) was calculated from the measured effective density (ρ_{eff}), which was determined by weighing an aliquot of suspension. From the known densities of solid (ρ_p) and liquid (ρ_0) phases, the volume concentration (ϕ) can be expressed as: $\phi = \frac{\rho_{\text{eff}} - \rho_0}{\rho_p - \rho_0}$.

The effective specific heat of the mixture $(c_p)_{\text{eff}}$ at various temperatures was calculated based on the physical principle of the mixture rule as: $(c_p)_{\text{eff}} = \frac{(1-\phi)(\rho c_p)_o + \phi(\rho c_p)_p}{(1-\phi)\rho_0 + \phi\rho_p}$.

The average particle sizes in suspension were estimated by two different techniques: the tri-laser diffraction light scattering technique using a Microtrac S3500 particle analyzer (Microtrac Inc., PA) and the dynamic light scattering technique at a 90° scattering angle using a 90Plus/BIMAS particle size analyzer (Brookhaven Instruments Corp., NY). The total surface area of dry nanopowders (after water evaporation) was measured using Brunauer–Emmett–Teller (BET) nitrogen gas adsorption on an ASAP 2010 Surface Area and Porosity System (Micromeritics Instrument Corp., GA). Dried nanoparticles were also characterized by a Model S-4700-II scanning electron microscope (SEM imaging) (Hitachi, Japan).

The pH of the nanofluids was measured with a pH meter (ExStik PH100) manufactured by ExTech Instruments. The pH of the nanofluids was adjusted with 2N solution of ammonium hydroxide (NH_4OH) or 2N solution of nitric acid (HNO_3). After pH adjustments, suspensions were sonicated (S-450, Branson Sonifier, CT) and equilibrated for ∼48 h prior to further characterization.

The relative viscosity of the nanofluids was measured using a Brookfield DV-II + rotational type viscometer with the SC4-18 spindle (Brookfield Engineering Laboratories, Inc., MA). The flow behavior of nanofluids was tested at various shear rates (39.6–264 s^{-1}) and temperatures (15–45 ± 0.2 °C). For all reported viscosity data the >10% torque criterion was satisfied.

The effective thermal conductivity of the nanofluids was measured using a thermal property analyzer (Model KD2pro, Decagon Devices, Inc.) based on the transient hot wire method.

Heat transfer measurements in the turbulent flow regime were conducted in the closed-loop heat transfer test facility that was designed and fabricated at Argonne National Laboratory [54].

3. Results and discussion

3.1. Characterization of average particle sizes in nanofluids

Because a milling approach was used for the fabrication of the SiC nanoparticles, it is expected that the nanofluids of this study have a very wide particle size distribution. Initial information on particle sizes reported by the manufacturer was ∼55, 170, 500 and 700 nm, which were measured by a particle size distribution analyzer LA-920 based on diffraction light scattering (Horiba Instruments Inc., USA). Nanofluids were characterized independently in the present study using a Microtrac S3500 diffraction light scattering setup which measured a wide particle size distribution with maxima at 120 nm, 155 nm, 288 nm and 377 nm respectively. Imaging with an SEM also showed a wide range of particle sizes with average size smaller than estimated from scattering techniques in all four nanopowders (figure 1). However, no quantitative

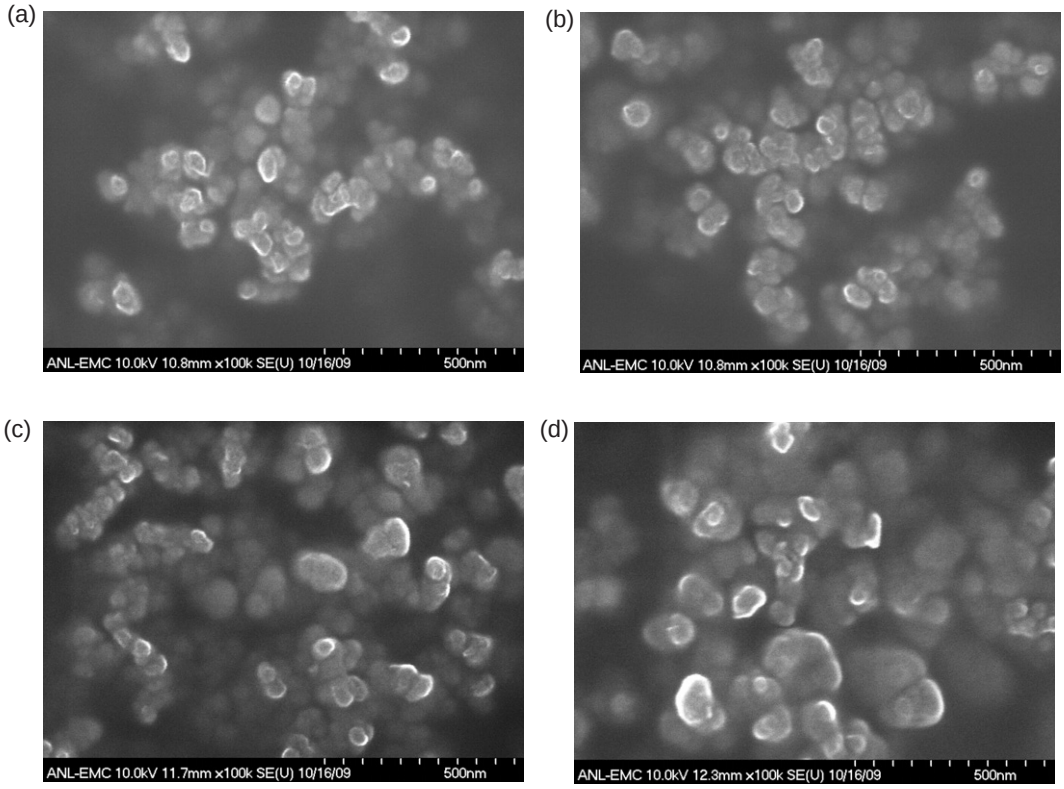

Figure 1. SEM images of SiC particles after water evaporation from diluted (∼0.001 vol%) nanofluids.

Table 1. Characterization of particle size distribution by various techniques.

Diffraction light scattering (Microtrac S3500) (nm)	Dynamic light scattering (Brookhaven 90Plus/BIMAS) (nm)	BET specific surface area, S_0, (surface area of sample/mass of sample) ($m^2\ g^{-1}$)	Average particle size from BET, d (nm)
120	44	119.3	∼16
155	149	64.8	∼29
288	215	28.6	∼66
377	250	20.8	∼90

conclusion on particle size distribution can be deduced from SEM images.

Because the size of nanoparticles defines the area of the solid/liquid interface that ultimately acts as a new phase in affecting properties of nanofluids, the specific surface area (S_0, $m^2\ g^{-1}$) was measured by BET for the nanopowders obtained after evaporating the water from the nanofluids. In this attempt to resolve the discrepancy in particle size determination, the average particle size was approximated as the diameter of spheres (d, μm) with equivalent exterior surface area: $d = \frac{6}{S_0\rho_p}$, where ρ_p is the true density of the material in g cm^{-3} (3.2 g cm^{-3} for 4H-SiC [59]).

The results show that the average particle sizes deduced from the BET total surface area are significantly smaller than those determined by the light scattering techniques (table 1). This result could be due to limitations of the light scattering techniques for characterizing polydisperse particle suspensions or simply due to the assembling of smaller nanoparticles into larger clusters moving together. Because of comparable sizes of the nitrogen molecule ($N_2 \sim 3.0$ Å) and the water molecule ($H_2O \sim 2.8$ Å), here we assume that water will penetrate wherever nitrogen does, be adsorbed on the particle surface and form solid/liquid interfaces. Therefore, in equilibrated nanofluids all surfaces measured with BET technique will acquire layers of fluid molecules that will function as an interfacial phase. Suspended nanoparticles can form larger agglomerates through the solid/liquid/solid interface; however volume and contribution of the interfacial phase will always remain proportional to the total surface area of dry powders. For that reason, in this study the average particle sizes determined from BET measurements are used for all further data analyses and correlations to the macroscopic properties of nanofluids.

3.2. Effect of pH on thermal conductivity and viscosity

The focus of the initial studies of SiC/water nanofluids was on optimization of the thermal conductivity/viscosity ratio. The effect of pH on viscosity and thermal conductivity of the suspensions was investigated for a series of nanofluids with the

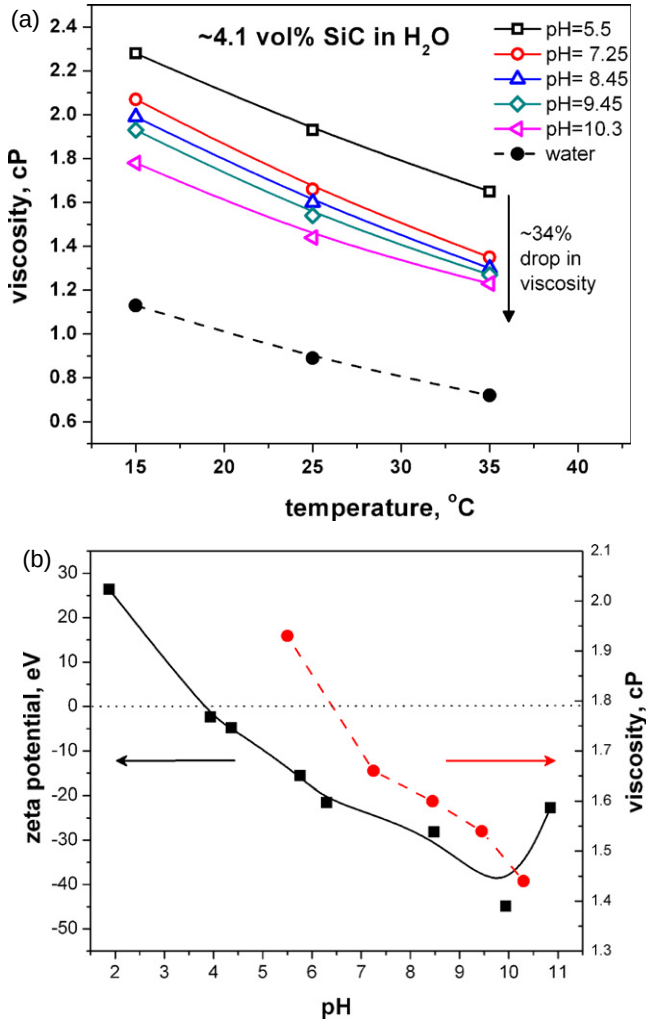

Figure 2. (a) Effect of pH on viscosity of 4.1 vol% α-SiC/H_2O nanofluids with an average particle size of 29 nm. (b) Correlation of viscosity of a nanofluid with 29 nm particles @ 25 °C with the zeta potential of the α-SiC–water interface [60].

same SiC particle size (29 nm) and volume fraction (4.1 vol%). The pH of the suspensions was adjusted and the viscosities and thermal conductivities were measured.

The thermal conductivity was not affected within the experimental uncertainty by the pH of the suspension (0.661 ± 0.005 W mK^{-1}, i.e. ~10% k increase). Conversely, variation of the pH between 5.5 and 10.3 resulted in significant (~34% versus the highest viscosity) viscosity drop (figure 2(a)). The viscosity of the SiC/water nanofluids was independent of shear rate, indicating Newtonian behavior and the absence of extended nanoparticle ensembles.

When compared to water, a SiC/water nanofluid with a pH 5.5 has an ~120% increase in viscosity, while a suspension with pH 10.3 has an only ~60% increase in viscosity. Therefore, by adjusting the pH of the suspension the viscosity increase was lower by half. This value was found to be nearly independent of temperature in the studied temperature range.

Figure 2(b) shows a relative comparison of experimental data on the viscosity and values of zeta potentials of α-SiC in distilled water at various pH values [60]. One can see that the viscosity decrease with the suspension pH is well correlated to the charge at the particle surface (zeta potential), with lower viscosity at higher surface charges (pH further from isoelectric point), indicating that the observed phenomena are most likely related to the electrostatic stabilization of the nanoparticle suspensions (second electro-viscous effect).

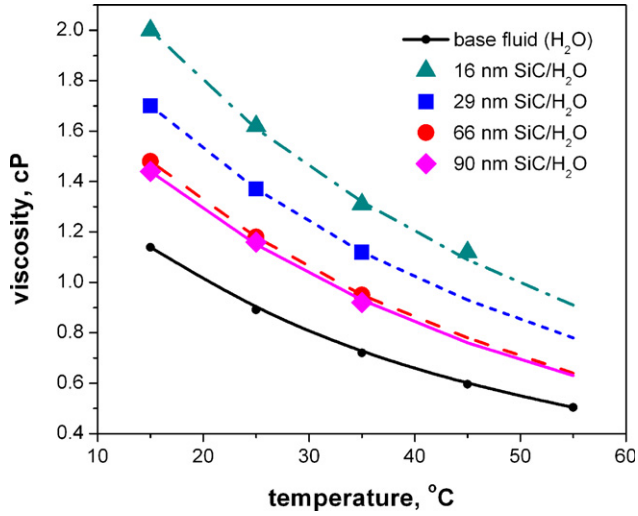

Figure 3. Viscosity of ~4.1 vol% α-SiC/H_2O nanofluids with different particle sizes at pH ~ 9.4.

Although the relationship of pH to zeta potential and viscosity is not new, and was previously described in the literature, the results of these experiments are very important for nanofluid development, since they illustrate the wide range of viscosity modifications available by simple pH adjustments with almost no effect on the nanofluid thermal conductivity. Such a variation in fluid viscosity represents a significant potential for improvement of the heat transfer performance. The results of this section were used to optimize the nanofluid pH for studying the particle size effect.

3.3. Effect of the average particle size on viscosity

In another series of experiments the average size of SiC nanoparticles was varied while the volume concentration of solids (4.1 ± 0.1 vol%) and pH (9.4 ± 0.1) were maintained constant. The viscosity of each sample, measured at various temperatures, is shown in figure 3. The experimental results clearly demonstrate that as the average particle size increases the viscosity of nanofluids with the same volume concentration of solids decreases (figure 3). Because the pH of the suspension was controlled, the observed effect can clearly be ascribed to the changing area of the solid/liquid interface and the decreasing effective volume of the nanoparticles (first electro-viscous effect). This result is in agreement with the majority of previous particle size studies on viscosity, however, the present experiments clearly show the difference in contributions and controls of the first and the second electro-viscous effects on the nanoscale. The first electro-viscous effect is clearly related to the particle size, total area of the solid/liquid interface, thickness of diffuse liquid layer around the particles, and therefore effective volume of the solid phase. The second

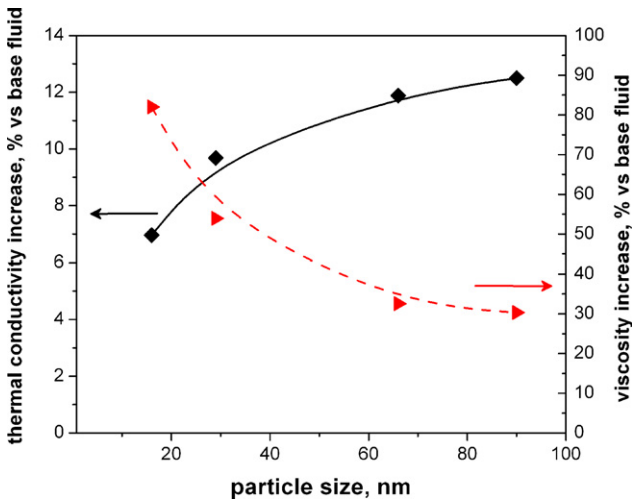

Figure 4. Dependence of thermal conductivity (at 22.5 °C) and viscosity (at 25 °C) of water-based nanofluids on the size of α-SiC for particle concentration of ∼4.1 vol% and pH ∼9.4.

electro-viscous effect is due to surface charges at the interface and particle–particle interactions controlled by the pH of the suspension.

The significance of the area of the solid/liquid interface, surface charges, and particle sizes on viscosity shown in this section indicates the path to minimizing the viscosity increase in nanoparticle suspensions: increasing particle size to minimize the area of the solid/liquid interface and optimizing the pH versus the isoelectric point to create surface charges significant for nanoparticle repulsion.

3.4. Effect of average particle size on thermal conductivity

Figure 4 represents the dependence of thermal conductivity enhancements and the viscosity decrease over the base fluid on the average particle size in 4.1 vol% SiC nanofluids with fixed pH (9.4 ± 0.1). One can see a very distinct trend in thermal conductivity enhancement: thermal conductivity increases with the increase of the average particle size. Thermal conductivity enhancements in nanofluids with larger particles are close to basic predictions of the effective medium theory and to other experimental data on SiC/H_2O (∼15% thermal conductivity enhancement at 5 vol%) [19, 61–63]. These results are in good agreement with the general trend of the particle size effect on thermal conductivity deduced from the comparison of previously published results from different groups [4, 10, 12]. The surface area of the solid/liquid interface increases geometrically as the average particle size in the nanofluids decreases. It is known [64] that interfaces act as an obstacle to heat flow, and the measure of interactions between the surface of the nanoparticles and the fluid are manifested through the interfacial thermal resistance, also known as the Kapitza resistance, $R_k = \frac{l_k}{k_0}$, where k_0 is the thermal conductivity of the matrix and l_k is the thickness of the base fluid layer excluded from thermal transport (Kapitza length), i.e. equivalent to the interface from a thermal point of view [65]. Molecular layering of the fluid at the surface of alumina [66] and silica [67] nanoparticles was demonstrated by nuclear magnetic resonance with an estimated thickness of the ordered water layer of 1.4–1.7 nm moving with the particle. The thickness and properties of this molecular layer at the interface might change with the particle type and composition of the liquid phase, but because in the present experiments only the total surface area of the nanoparticles (equivalent of average particle size) was varied (because the same milling procedures were used for all nanofluid preparation it is assumed that the average particle shape in tested suspensions is the same), the data can be used for calculating the value of Kapitza resistance for the α-SiC/H_2O interface, similar to calculations used in [33].

Assuming linearity of both EMT and interfacial contributions with the particle volume concentration, the thermal conductivity of the nanoparticle suspension can be presented as follows [33]:

$$\frac{k_{\mathrm{eff}}}{k_0} = 1 + (C_k^{\mathrm{EMT}} + C_k^{\mathrm{surface}})\phi, \qquad (3)$$

where C_k^{EMT} and $C_k^{\mathrm{surface}} = -f l_k$ are coefficients reflecting contributions to the effective thermal conductivity due to EMT (positive) and surface resistance (negative), respectively. The surface factor, f, represents the surface area of nanoparticles normalized per unit volume of nanoparticles.

Even though the shape factor of nanoparticles used for C_k^{EMT} estimation is unknown, from the experimental thermal conductivity and S_0 data of this study, one can put up a system of four equations with two unknowns (l_k and C_k^{EMT}), which results in consistent values of Kapitza length $l_k \sim 4.2 \pm 0.3$ nm and interfacial Kapitza resistance $R_k \sim (7.0 \pm 0.5) \times 10^{-9}$ m^2 K W^{-1}. These values are unique resistance characteristics of an α-SiC/H_2O interface and fall within the range of R_k reported for other water-based nanofluids [68, 69]. This also allows the estimation of $C_k^{\mathrm{EMT}} \sim 3.27$, and from the Hamilton–Crosser equation [19] it can be deduced that the average shape of SiC nanoparticles in the suspension is likely to be slightly elongated spheroids.

Based on the present experimental results, it can be concluded that the particle size is a very powerful parameter in nanofluid property management. Considering heat transfer applications, both the thermal conductivity and viscosity of nanofluids favor the use of larger particles over small ones (figure 4). However, this approach is limited by the stability of suspensions with larger particles and possible erosion damage to the equipment.

3.5. Heat transfer compared to base fluid

A series of forced convective heat transfer experiments was carried out for the 4.1 ± 0.1% SiC water nanofluids at four average particle sizes (16, 29, 66, and 90 nm) and with the pH adjusted to 9.4. The flow rates for all the studies were varied between 400 and 1400 ml min^{-1}, having Reynolds numbers based on water properties between 6000 and 20 000 with a constant inlet temperature of approximate 30 °C. The heat transfer coefficients measured at thermocouple five [54] are compared on the basis of the constant velocity for the four

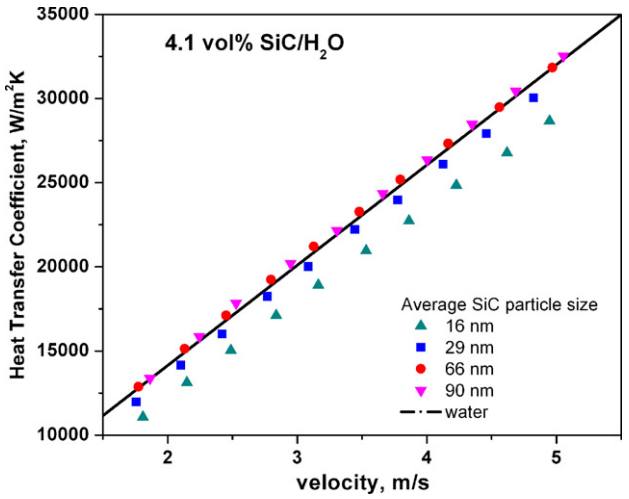

Figure 5. Heat transfer coefficient for the four different particle sized nanofluids compared to the average results obtained for water.

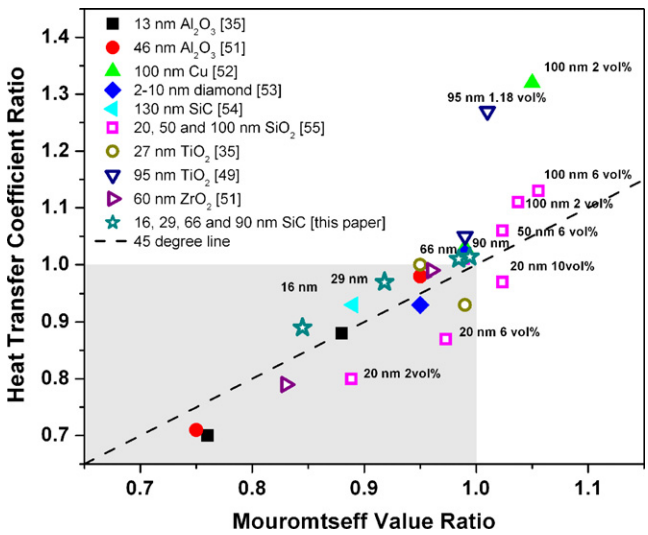

Figure 6. Experimental heat transfer coefficient ratio compared to Mouromtseff value ratio for the present particle size study and the literature data.

different particle sized nanofluids and water (figure 5). The heat transfer coefficients for nanofluids with average particle sizes of 16 and 29 nm are 10.9% and 3.3% lower than those for water, respectively, while the heat transfer coefficient for nanofluids with average particle sizes of 66 and 90 nm are 1.1% and 1.4% larger than those for water. The increased heat transfer coefficient of nanofluids with larger particles (66 and 90 nm) over the base fluid remained consistent at all inlet temperatures tested (30, 43, and 55 °C). This result suggests that heat transfer enhancement of a nanofluid is likely either independent or weakly dependent on temperature. Consistent performance at all temperatures is a valuable and important feature for heat transfer fluids in general because they typically undergo a large range of temperatures during heating and cooling cycles in real systems.

From the results of this study, it is clear that the reduction in viscosity and slight increase in thermal conductivity at larger particle sizes made some improvement to the heat transfer coefficient compared to the base fluid.

3.6. Evaluation of nanofluid heat transfer efficiencies

Buongiorno [70] theoretically assessed seven possible mechanisms, besides classical fluid dynamic equations for single phases, that could occur in nanofluids due to the addition of nanoparticles. It was concluded [70] that only thermophoresis and Brownian diffusion may be large enough to affect the heat transfer by increasing the nanoparticle contribution during heating and decreasing the nanoparticle contribution during cooling. Yu *et al* [54] tested for the effects of thermophoresis and Brownian diffusion and found no difference in heat transfer between heating and cooling, suggesting that these mechanisms are not responsible for nanofluid heat transfer enhancement beyond property effects.

The ratio of Mouromtseff values gives a comparison of nanofluid heat transfer performance to the base fluid based on fluid properties alone (equation (2)) without any additional mechanism due to the presence of nanoparticles. The heat transfer coefficients for both the base fluid and the nanofluid are evaluated from experimental data at the same velocity using the Ditter–Boelter equation [71]. Figure 6 represents a comparison of the ratios of Mouromtseff values and the experimental heat transfer coefficient ratios for the four SiC nanofluids with different average particle sizes from this study and the heat transfer studies from the literature [35, 49, 51–55]. In figure 6, heat transfer coefficient ratios above the 45° line show enhancement in nanofluid heat transfer beyond the fluid property effect, and ratios above 1.0 show enhancement over the base fluid. Interestingly, with the data available for the first time in this format, it becomes apparent that larger particles are superior to smaller particles in heat transfer for all of the studies. Studies that reported a heat transfer coefficient above the base fluid on a constant velocity basis were three of the four largest particles (100 nm Cu [52], 100 nm SiO_2 [55], and 95 nm TiO_2 [49]), and results [49, 52] showed significant enhancement. The studies that involved particles below 95 nm had heat transfer coefficients lower than the base fluid [35, 51, 53, 55].

With regard to enhancement beyond property effects, the literature data fall above and below the 45° line with the exception of the data [49, 52] which show significant enhancements. Also, the data of the present study with all of the parameters controlled show a consistent, albeit modest enhancement over property effects. The largest particles tested in the present study showed ∼1% enhancement over the base fluid. With regard to enhancement over the base fluid there are literature data in the range 0–13% enhancement, with the data of [49, 52] showing 27 and 32% enhancement respectively, which may be due to material specific or possibly electrically driven mechanisms.

The nanofluids with different SiC particle sizes were also evaluated using existing figures of merit for both laminar [56] and turbulent [57, 58] convective flow regimes (figure 7). It is clearly seen from figure 7 that larger particles are beneficial for both flow regimes. In the laminar flow regime (left Y-axis) water suspensions of SiC particles with the average

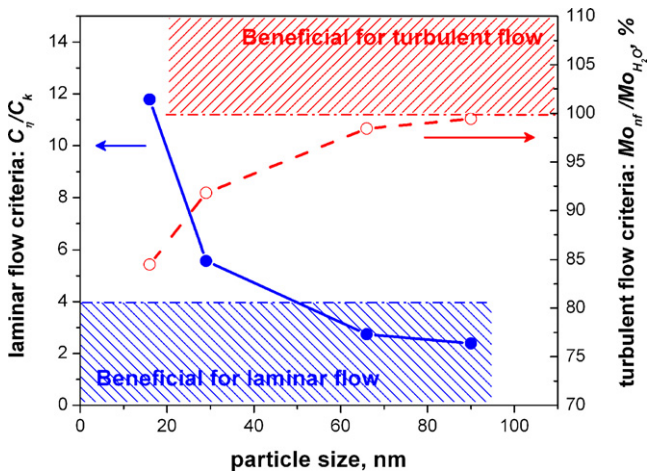

Figure 7. Property based figures of merit for laminar and turbulent flow for α-SiC/H_2O nanofluids as a function of average particle sizes.

sizes >50 nm are clearly in the benefit zone, while for the turbulent flow (right Y-axis) particles should be larger than 90 nm to show heat transfer enhancement over the base fluid performance based on fluid properties alone.

4. Conclusions

In this work the effect of particle sizes was systematically studied and related to the viscosity, thermal conductivity and heat transfer coefficient in water-based silicon carbide suspensions. It was demonstrated that in polarizable fluids a wide range of viscosity is available through the control of the surface charges on nanoparticles by concentration of specifically adsorbing ions (pH in water-based fluids) without a significant effect on thermal conductivity. With all other conditions being the same (pH and particle concentration) smaller particles result in a higher viscosity increase than larger particles due to the larger surface area of solid/liquid interfaces and increased effective volume of solids. Thermal conductivity is higher in nanofluids with larger particles, also due to effects of nanoparticle surface area; the solid/liquid interface acts as an obstacle for the heat flow with the negative contribution of the interface proportional to the total surface area of nanoparticles. Viscosity and thermal conductivity are the major parameters defining the heat transfer coefficient in SiC/water nanofluids from a properties perspective.

Experiments determining the heat transfer capabilities of $4.1 \pm 0.1\%$ SiC nanofluids with various sized nanoparticles showed that as particle size increases the heat transfer coefficient increases for the same flow velocity, as a result of lower viscosity and higher thermal conductivities. Therefore, the particle size is an important instrument for the manipulation of nanofluid properties. Limitations on using particle size for nanofluid improvements could be their stability and erosion resistance.

The heat transfer results from this study and the literature data were compared with theoretical predictions, based on fluid properties alone, using the Mouromtseff values. Much of the data were predicted by fluid properties and some reached 13% enhancement over the base fluid. The SiC data of this study showed some heat transfer enhancement above the fluid properties at larger particle sizes. These results point to the future direction of research in the field.

Analysis of experimental data based on fluid properties alone, using figures of merit, showed that the use of a SiC/water nanofluid will be beneficial in the laminar flow regime when particles are larger than ~50 nm, while for turbulent flow average particle sizes should be larger than ~90 nm.

While it is clear that the use of the larger SiC particle sizes produces a small heat transfer enhancement in the water-based nanofluid, it is expected that the enhancements in heat transfer will be larger in nanofluids with a higher base fluid viscosity.

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