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Effect of Environmental Factors on Dynamic Viscosity of Zirconia and Silica Nanofluids: Experimental Insights and Theoretical Predictions

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Abstract

Zirconia (ZrO_2) and silica (SiO_2) nanoparticles suspended in water are the focus of this investigation on the influence of environmental variables on the dynamic viscosity of these nanofluids. Two different viscometers (a falling ball and a capillary) were used to measure the range of temperatures from 30 to 60 °C and the percentage of particles from 4 to 15.4%. The results demonstrate that, similar to their base fluids, nanofluids' viscosity reduces as temperature rises. Surfactants are added to nanofluids to improve their stability at room temperature; however, this is likely at the expense of an increase in viscosity. However, the modified Krieger-Dougherty relation provides reasonably accurate estimation of nanofluid viscosity within a narrow limit of solid size of particle to cumulative size, while relations attained from the lenient liquid concept, like Einstein's and Bachelor's, fail to predict nanofluid viscosity for solid concentrations above 1.5 wt.%.

Keywords

Viscosity, Krieger-Dougherty, Nanofluid, Zirconia, Silica, Surfactant

Introduction

The dispersion of nanoparticles in a colloidal solution defines a nanofluid. The thermophysical parameters of a fluid are often modified by the existence of nanoparticles [1]. Over the past decade, scientists have worked hard to better understand the nanofluids because of the numerous mysteries surrounding their purportedly special properties and promising uses. When multi-walled carbon nanotubes were dispersed into oil at a concentration of 1% by volume, the authors found that the oil's thermal conductivity improved by a factor of 2.5 [2]. Nanoparticle characteristics such nanoscale size and high specific surface area explains the unusual behavior of nanofluids documented in the scientific literature. High heat conductivity, homogeneity, long-term stability, and resistance to channel clogging are only some of these features [3-5].

Due to the increasing complexity of thermal management responsibilities brought on by technological progress, a high-performing coolant is essential. High temperatures degrade the durability of modern electronic systems, therefore limiting their operation temperature is a common way to restrict device performance [6]. While increasing the device's size can boost its heat transfer surface and facilitate greater heat diffusion, this is rarely a cost or resource-effective strategy. This highlights the need for research into innovative heat transfer fluids that exhibit good rheological behavior in addition to improved thermal properties [7]. Heat transfer, power generation, microelectronics, and heating, ventilation, and air conditioning could all benefit from coolants with enhanced thermal properties [8].

Dispersing nanoparticles in a heat carrier fluid may produce a heat exchange fluid [9]. The thermal conductivity of nanoparticles, including metals, metal oxides, and carbides, is dramatically greater than that of conventional base fluids. It's hardly surprising that the nanofluid has better heat conductivity than the original fluid [10, 11]. When nanoparticles are added to these simple fluids, they change consistency, becoming thicker and denser. Viscosity is crucial to cooling systems because it determines pumping energy and pressure drop [12]. While there is a wealth of information on nanofluids' thermal conductivity, few research have addressed their viscosity [13].

At temperatures between from 20 to 70 °C, the viscosity of water mixed with 10 vol.% γ -Al_2O_3 and SiO_2 nanofluids was measured. Based on the results, nanofluids composed of γ-ZrO_2 and SiO_2 dissolved in water had a viscosity roughly 4 times more than that of base fluid [14, 15]. The concentrations of TiO_2 nanoparticles in a water-based nanofluid were measured at 0.25, 0.60, and 1.18 vol.% (95, 145, and 210 nm). It was discovered that the higher viscosity of the nanofluids was caused by a combination of factors, including particle concentration and particle size [16]. The 95 nm particle size nanofluid's viscosity was enhanced by 4, 5.5, and 11% points at 25 °C when the particle concentration was 0.25, 0.60, and 1.18 vol.%, respectively [17]. The nanofluid's viscosity also increased by 5.3% and 7.2% for 95 nm and 210 nm particle sizes, respectively, when 6 volumes were added.

Researchers examined the consistency of a TiO_2 nanofluid in water at various temperatures (15 - 35 °C) and particle volume fractions (0.2 - 2%) [18, 19]. The viscosity of a TiO_2 water nanofluid was found to be 4 - 15% points greater than that of base fluid at particles size of 1.5 vol.% and temperatures between 20 and 40 °C. Increasing the particle load from 0.2 to 2% vol. resulted in a 4 - 12% increase in the viscosity at 25 °C. Particle size distribution of sub-micron SiO_2 between 27 and 55 °C and 0.27 and 1.39 vol.% [20]. Increasing the SiO_2-water particle concentration from 0.27 to 1.39 vol.% resulted in a rise in relative viscosity from 1.03 to 1.14.

Over a range of 10 – 70 °C and four different nanoparticle concentrations, the temperature dependence of the viscosity of a SiO_2-water nanofluid was studied (1, 10, 20, and 35 wt.%) [21]. The results demonstrated that nanofluids exhibited temperature-independent relative viscosity. Between 10 and 60 °C, the nanofluid exhibited behavior not dissimilar to that of the basic fluid at 1 wt.%, with only a slight increase in viscosity (up to 1 %) [22]. However, the increase in viscosity was significantly larger at maximum concentrations and temperatures. The viscosity of SiO_2-water based nanofluids was observed to rise with temperature and particle concentration [23]. Nanofluids made from ZrO_{2-}water have their characteristics analyzed. For concentrations of volume larger than 4%, the 47-nm samples' viscosities were obviously greater than those of the 36-nm samples. It has been found that the viscosities of both particle sizes are roughly the same for particle loadings lower than 4% [24].

ZrO_2 nanoparticles were suspended in an ethylene glycol and water solution, and their rheological and heat transport properties were studied (45 and 55 vol.%) [25]. Viscosity was discovered to be significantly affected by particle loading (1 - 2 vol.%), as well as temperature (10 - 60 °C) [26]. They demonstrated that increased viscosity was associated with increased particle concentration in nanofluids. For instance, at T = 10 °C, the nanofluid containing 2 vol.% ZrO_2 had a viscosity that was almost 46% higher than that of the nanofluid containing 1 vol.% [27, 28].

This study employed a Höppler (falling ball) and a capillary viscometer to precisely measure the viscosity of ZrO_2 and SiO_2 nanofluids in water at temperatures between 20 and 60 °C. We also looked into how changing the particle concentration, temperature, or surfactant affected the nanofluid's viscosity. Finally, the results of the studies were interpreted using published correlations.

Experimentation

Each nanofluid system is described in detail and its constituent materials are characterized in table 1. All of the nano-

Table 1: Properties of materials of estimated specimens.

Material concentration (wt.%)		Surfactant	pH	Particle size by SEM (nm)	Particle distribution/ most common particle size by DLS (nm)	Morphology
ZrO2 (I)	8 - 16	Octylsilane	9.1	180 ± 20	120 - 20/230	Spherical
ZrO2 (II)	4 - 12	Octylsilane	9.4	210 ± 30	160 - 720/320	Spherical + elongated
ZrO2 (III)	Dispersia,12 wt.% (230, 260, and 290 nm)	Unknown	6.2	220 ± 30	80 - 450/210	Inhomogeneous
				290 ± 25	100 - 700/350	Inhomogeneous
				330 ± 35	100 - 900/380	Inhomogeneous
SiO2 (I)	4 - 12	Polycarboxylate	7.2	40 ± 10	70 - 250/140	Spherical
SiO2 (II)	12	Trioxadecaneacid	8	40 ± 10	50 - 650/200	Spherical
SiO2 (III)	12	No surfactant	-	40 ± 10	60 - 300/90	Spherical

fluids were aqueous solutions that included stabilizers like surfactant and pH buffers alongside the nanoparticles. The pH of the ZrO_2 nanofluids was about 9, and the surfactant of choice was octylsilane. However, polycarboxylate and trioxadecane acid were used to stabilize SiO_2 nanoparticles. These surface modifiers had not had their concentrations determined. The first stage in making any of the nanofluids was using high-energy tip sonication to disperse nanoparticles throughout a base fluid. Particles with varying surface chemistry in the zeta potential stable region were tested at a range of pH levels.

Characterization

Dynamic light scattering (DLS) have been used to examine ZrO_2 and SiO_2 nanoparticles and assess their size and shape. Each nanofluid was drop-cast into its respective sample container, and after 6 h of drying in a vacuum oven, the solvent had been removed. For the DLS analysis, all materials were diluted to 1 wt.% with base fluid (water) [28]. The DLS results for all of the ZrO_2 and SiO_2 nanofluids employed in this work are shown below. About 290 nanoparticles were counted from micrographs to estimate the nanoparticles' dry size. ZrO_2 (I) has a dry primary particle size of around 180 nm. The particle size of ZrO_2 (II) was somewhat greater than that of ZrO_2 (I). Figure 1 displays DLS results showing that the hydrodynamic particle size distribution spans a range of 120 to 520 nm, with a median DLS size of 180 nm. Particle size was found to be greater in ZrO_2 (II) than in ZrO_2 (I). Micrographs of dry ZrO_2 (III) nanoparticles. ZrO_2 (III) nanoparticles of three sizes were synthesized to investigate the impact of particle size. The flake-like appearance of the particles is supported by the fact that the greatest dimension can be used to determine their dry size. These nanoparticles were analyzed, and their diameters were determined. Cluster formations caused by adsorbed molecular species give these nanoparticles slightly greater average hydrodynamic diameters compared to dry particles. SiO_2 (I), SiO_2 (II), and SiO_2 (III) nanofluids with a dry particle size of 30 nm. SiO_2 (I), SiO_2 (II), and SiO_2 (III) nanofluids have particle sizes that are four to eight times bigger than the dry size of particle, as shown in table 1. This proves that nanoparticles have accumulated in the nanofluid, maybe during the nanoparticle's formation and manufacturing process.

Viscosity measurements

Both Höppler (falling ball) and capillary viscometers were used to measure viscosities between 30 and 60 ˚C. These viscometers had standard operating procedure (SOP)-reported uncertainties of 0.5 - 1% (from now on referred to as "Capillary") and 0.5 - 2%. Connecting the viscometers to a thermostatic bath ensured that the samples being tested were always at the same temperature. The water's viscosity was measured before and after each series of tests to make sure the results were reliable. Atmospheric pressure was used for all readings. For 15 min before testing, every nanofluids was immersed in an ultrasonication immersion to achieve accurate viscosity values.

Results and Discussion

Instrumental validation

Both viscometers were used to test the viscosity of distilled water across a temperature spectrum from 10 to 80 ˚C. To ensure accuracy, each measurement was taken at ten times and took the mean. For Höppler, it was 2%, but for capillary it was only 1%. The data from three different measurements with both viscometers are shown in figure 2. Both viscometers show discrepancies between experiment and standard of less than 2%. There is also less than a 1% difference between the results of these two viscometers.

Measurements for nanofluids

Water-based ZrO_2 and SiO_2 nanofluids had their impacts on temperature, nanoparticle size, nanoparticle concentration, and surfactants defined by their absolute and relative viscosity.

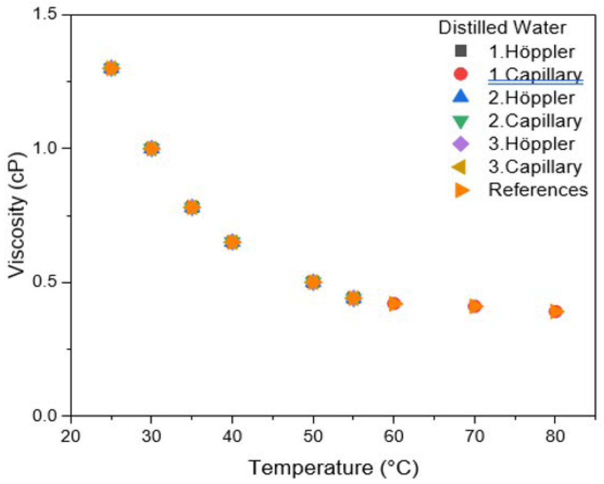


Figure 2: Distilled water viscosity as measured using Höppler and Capillary viscometers and compared to standard values.

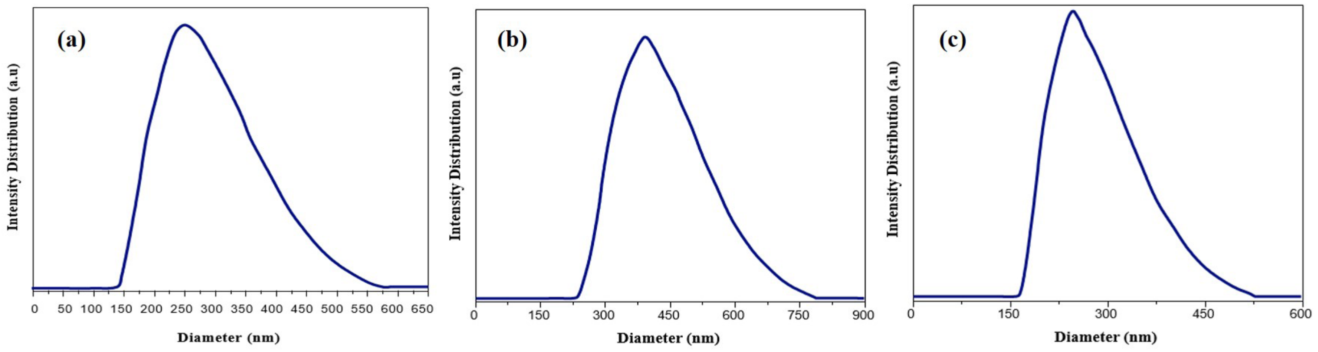


Figure 1: Analyzing nanofluids for particle size dispersal with DLS: **(a)** 230 nm, **(b)** 260 nm, and **(c)** 290 nm.

Before starting any nanofluid experiments, we measured the viscosity of distilled water at 30 ˚C to ensure the accuracy of our instruments. Each viscometer measured the sample three times (within a 2% standard deviation) and the average was used to get the final value. Since the discrepancies between the capillary and Höppler viscometer readings were so small (less than 1.1%), we simply averaged the two and used those numbers to characterize nanofluids.

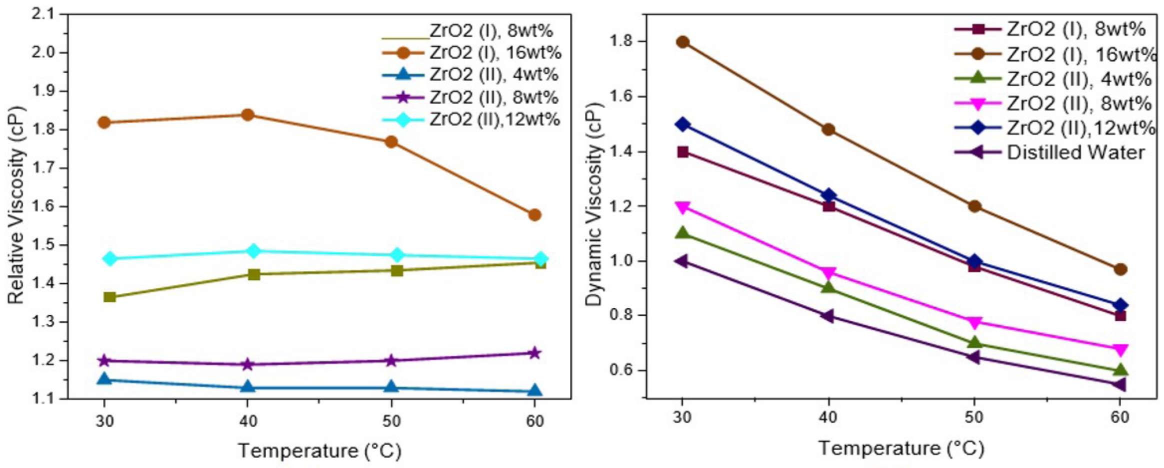
The effect of particle concentration and temperature on viscosity

At varying temperatures and solid particle densities, the viscosity of nanofluids composed of ZrO_2 (I, II, and III) and SiO_2 (I, II, and III) was determined. Figure 3 and figure 4 show the relative and absolute viscosities of ZrO_2 and SiO_2 nanofluids at varying temperatures. Viscosity was determined between 20 and 60 ˚C for ZrO_2 (I) at 8 and 16 wt.% and ZrO_2 (II) at 4, 8, and 12 wt.%. Water's viscosity was significantly raised after being exposed to ZrO_2 nanoparticles, as predicted. All ZrO_2-water nanofluids were discovered to have a viscosity that decreased with raising temperature, just like the base fluid.

Compared to nanofluids with lower concentrations of solid particles, those with larger ZrO_2 loadings exhibited higher viscosity (Figure 3). It's proof that the number of particles in a nanofluid has a key influence on its thickness. The relative and dynamic viscosity of ZrO_2 nanofluids between 30 and 60 ˚C is shown in figure 3a and figure 3b. The average ZrO_2-nanofluid viscosity increased by 8, 18, 37, 47, and 80% at 30 ˚C for particle concentrations of 4, 8, 12, 8, and 16 wt.%. Nanofluids made of ZrO_2 and water have a relative viscosity that is independent of both temperature (30 - 60 ˚C) and particle concentration (4% - 12%). ZrO_2 (I), which has a solid particle content of 16 wt.%, exhibits a moderate 15% decrease in relative viscosity between 30 and 60 ˚C.

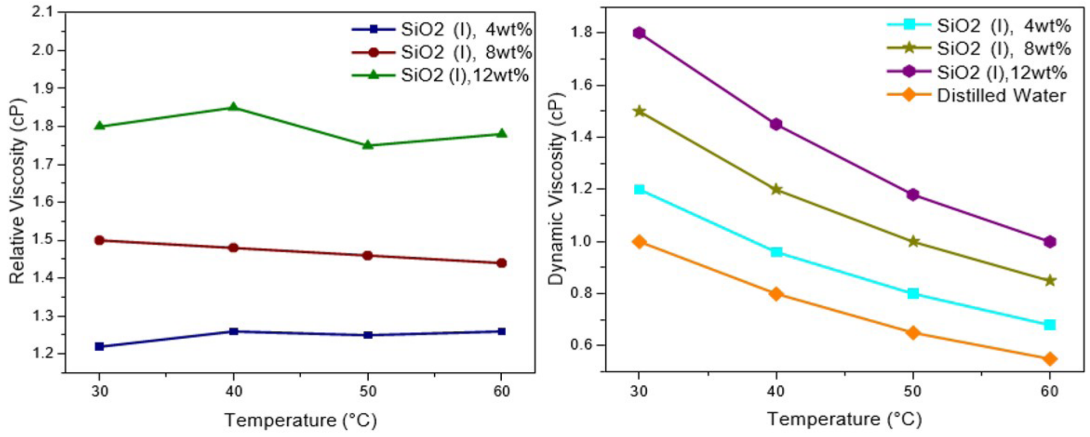
Comparable studies were conducted on SiO_2 nanofluids with solid contents of 4, 8, and 12 wt.% between 30 and 60 ˚C. Both ZrO_2 nanofluids and SiO_2 nanofluids exhibited a rise in viscosity with increasing particle loading and decreasing temperature. For example, the maximum viscosity was observed at 30 ˚C in the nanofluid containing 12 wt.% SiO_2 nanoparticles.

Highly concentrated nanofluids have a higher viscosity



(a) (b)

Figure 3: (a) Relative viscosity and **(b)** Dynamic viscosity for ZrO_2 nanofluids in water, considering varying particle concentrations and temperatures.



(a) (b)

Figure 4: (a) Relative viscosity and **(b)** Dynamic viscosity for nanofluids based on SiO_2 and water at various particle concentrations and temperatures.

because of the greater internal shear force among the nanoparticles. Authors claim that more nanoparticles mean bigger agglomerates and more energy is needed to disperse them. As the temperature of a nanofluid increases, the intermolecular and inter-particle adhesion forces weaken, resulting in a decrease in viscosity similar to that of a normal liquid.

SiO_2 nanofluids exhibited temperature-insensitive relative viscosities, similar to those of ZrO_2 nanofluids. Both SiO_2-water and ZiO_2-propylene glycol nanofluids have a consistent relative viscosity over a temperature range of 30 - 60 °C and 30 - 60 °C, respectively, as demonstrated by the authors' experiments. SiC-water and ZrO_2-water nanofluids were both found to have a relative viscosity that rise from 28 to 72 and 20 to 70 °C, respectively, as the temperature was raised. ZrO_2 (I) 16 wt.% and the results of this study were indistinguishable in any other way. ZnO-water nanofluids were shown to have a steady relative viscosity between 10 and 35 °C, but a significant decrease between 35 and 60 °C. This was attributed, at least in part, by the researchers to the lubricating properties of the nanoparticles. Nanoparticles in a nanofluid can adsorb to a solid surface if the fluid is compressed between two solids. The adsorption process lowers friction. Because of this, the fluidity of the nanofluid is altered, and its viscosity is decreased. When the temperatures of both the nanofluid and the base fluid are increased, the viscosity difference between them decreases even further. The nanofluid's low viscosity between 35 and 50 °C was the result.

SiO_2-nanofluid with 4, 8, and 12 wt.% increased its viscosity relative to water by 18, 52, and 81%, on average, at T = 30 °C. ZrO_2 (II) had much smaller increases of 8, 18, and 47% at the same temperature and particle concentrations. Consistent with expectations, SiO_2 nanofluids had greater viscosities than ZrO_2 nanofluids. However, different findings have been reported in several studies for similar particle concentrations of ZrO_2 and SiO_2.

The impact of particle size on viscosities

The impact of particle size on the viscosity of ZrO_2 (III) water-based nanofluids was investigated by analyzing samples with a 12-wt.% particle concentration and three variant initial size of particle. As can be shown in figure 5, the 230 nm ZrO_2 nanofluid had a lower viscosity than the 260 nm and 290 nm variants. However, it is not possible to extrapolate a broad trend from these data. The nanofluids with a 260 nm size of particle had a viscosity that was between 0.4 and 7% greater than the nanofluid with a 290 nm particle size between 30 and 60 °C. It's important to keep in mind that the inhomogeneous shape of these nanofluids may have influenced the research and helped to explain this seemingly conflicting conclusion.

The literature revealed the exact opposite relationship between nanofluid viscosity and particle size. Some research has found a modest connection between particle size and viscosity in SiO_2-water nanofluids, despite claims to the contrary. They discovered that when the particle size of ZrO_2-water nanofluids (1 - 6 wt.%) increased, the fluid's viscosity decreased. SiO_2 nanofluids in ethylene glycol-water (60:40 by weight) were tested, and their viscosity was measured as a function of particle size. Their research also demonstrated that larger particles resulted in lower viscosity.

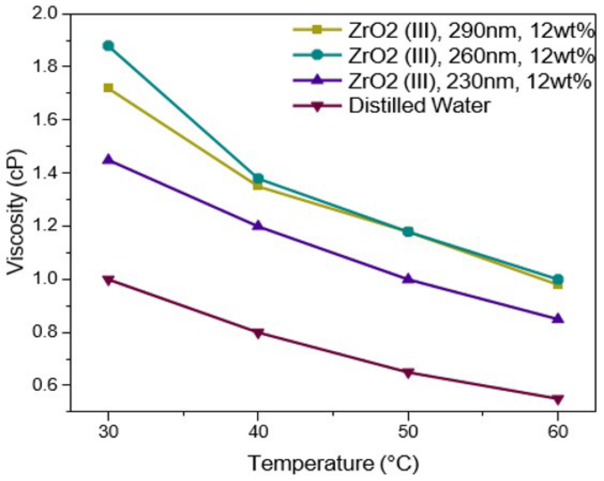


Figure 5: The relationship between temperature and the dynamic viscosities of ZrO_2-water containing 12 wt.% solid size of particles.

The viscosity of a nanofluid may appear to be affected by particle size, but it is important to also consider the impact of the other factors. Because nanoparticles tend to agglomerate and form clusters, studying the solitary impact of particles on the viscosity of nanofluids is difficult. In other words, nanoparticle clustering and agglomeration would increase the hydrodynamic particle size of nanoparticles, leading to a higher viscosity. To prevent accumulation and clumping, the well-dispersion method might be used. Furthermore, the particle size distribution is modified and clusters are dispersed into finer particles due to ultrasonic processing.

The effect of surfactant on viscosity

Pipeline pressure drops more when nanoparticles are introduced to a base fluid because of the increased viscosity of the resulting nanofluids. The flow properties may also be affected by the increasing viscosity. When added to an ethylene glycol-water base fluid, the concentration of ZrO_2 increases the pressure drop by a factor of 4.7. Since surfactants are able to maintain their drag-reducing action under strong shear forces, they are widely used as a useful strategy to minimize drag in the nanofluid. Incorporating a cationic surfactant into water-coated carbon nanotubes has been shown to lower the pressure drop by as much as 30%.

Suspension stability is crucial in any real-world application. For the aim of stability, surfactants are frequently added to nanofluids. Surfactants are essential for enhancing the stability and dispersibility of nanofluids, but they may also affect the viscosity in unexpected ways.

The purpose of this analysis was to examine the impact of various surfactants on the viscosity of SiO_2 water nanofluids. The impacts of various surfactants on SiO_2 water nanofluids at 30 - 60 °C are displayed in figure 6.

The highest viscosities were observed in SiO_2 (II) nanofluid (using trioxadecane acid Surfactant) and the lowest were found in SiO_2 (III) nanofluid (without surfactant) (Figure 6). When compared to SiO_2 (I) and SiO_2 (III) nanofluids, the

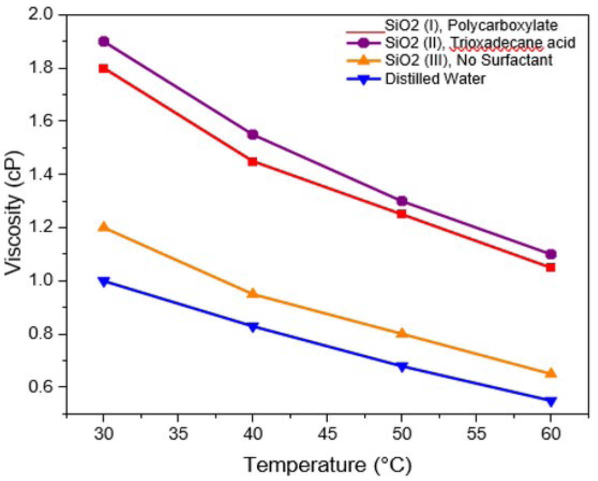


Figure 6: Changing viscosity of SiO_2-water nanofluids at various temperatures and surfactant concentrations.

viscosity of SiO_2 (II) nanofluids was ranging from 5.9% to 62.0% higher. Intriguingly, the SiO_2 (III) nanofluid, when utilized without a surfactant, exhibited just a marginal increase in viscosity compared to water. This suggests that the surfactant, rather than particle size or shape, may play a more significant role in determining the final viscosity. The stability and viscosity of a silica nanofluid containing SiO_2 nanoparticles (25 nm diameter) were studied, and the impact of anionic surfactant and ultrasonic processing was examined. They demonstrated that adding a surfactant to the nanofluid raised its viscosity, but that nanofluids without surfactants sedimented at a faster rate. They found that the best strategy to boost nanofluid stability was to combine the use of a surfactant with an ultrasonic bath.

Comparisons between experimental results and values calculated using the correlations are used to probe the validity of the correlations. Figure 7a and figure 7b demonstrates comparable findings, which agree with the SiO_2-water nanofluids (Particle loads between 4% - 12% or 0.7 - 2.3 vol.%). At 30 °C, figure 8a and figure 8b displays the outcomes for ZrO_2-water nanofluids with particle loadings ranging from 3% to 14% (0.8 - 4% vol.). For these nanofluids, the viscosity predicted by the Batchelor, Brinkman, Einstein, and Krieger-Dougherty correlations (shown in both pictures) are grossly inadequate. For most ZrO_2 nanofluids, the experimentally determined values are significantly lower than those predicted by the Nguyen (using a diameter of 36 nm for ZrO_2) and Williams (using a diameter of 47 nm for ZrO_2) correlations, respectively. The correlation suggests that ZrO_2 nanofluids with concentrations below 6 wt.% will have a high viscosity. The majority of these equations neglect everything saves the volume fraction's effect. Most ZrO_2 and SiO_2 nanofluids' viscosity may be estimated using correlations shown in figure 7 that consider the effect of solid particle and aggregate sizes and concentration. The viscosity of a nanofluid based on ZrO_2 and water, for instance, can be predicted with an error of 6% using the Rudyak correlation. The modified Krieger-Dougherty correlation is the most accurate method for predicting the viscosity of SiO_2-water nanofluids. The aggregation factor (ξ), compare the aggregated nanoparticle size to the basic nanoparticle size., plays a crucial role in the predictions made with the modified Krieger-Dougherty correlation. Its study, however, employed DLS analysis to evaluate this worth (Table 1). As can be observed in figure 8, the optimal operating range for modified Krieger-Dougherty for ZrO_2 and SiO_2 is between 4 and 5. Despite ZrO_2 (I) and (II) having values outside the aforementioned range (1.8 and 1.96, respectively), the viscosity of these nanofluid samples is still overestimated by 23 - 33% using modified Krieger-Dougherty. The experimental parameter Df (fractional index) was considered to be 1.9 for ZrO_2 nanofluids and 17 for SiO_2 nanofluids. However, the viscosity of SiO_2 nanofluids with a value of close to 4.79 can be predicted using the modified Krieger-Dougherty correlation.

Conclusion

Two separate viscometers (Höppler and capillary) were used to measure the viscosity of ZrO_2 (8 - 16 wt.%) and SiO_2 (4 - 12 wt.%) nanofluids in water over the temperature between of 30 to 60 °C. Once solid particles were introduced and the temperature was dropped, both nanofluids became more viscous. The relative viscosity of these nanofluids hardly

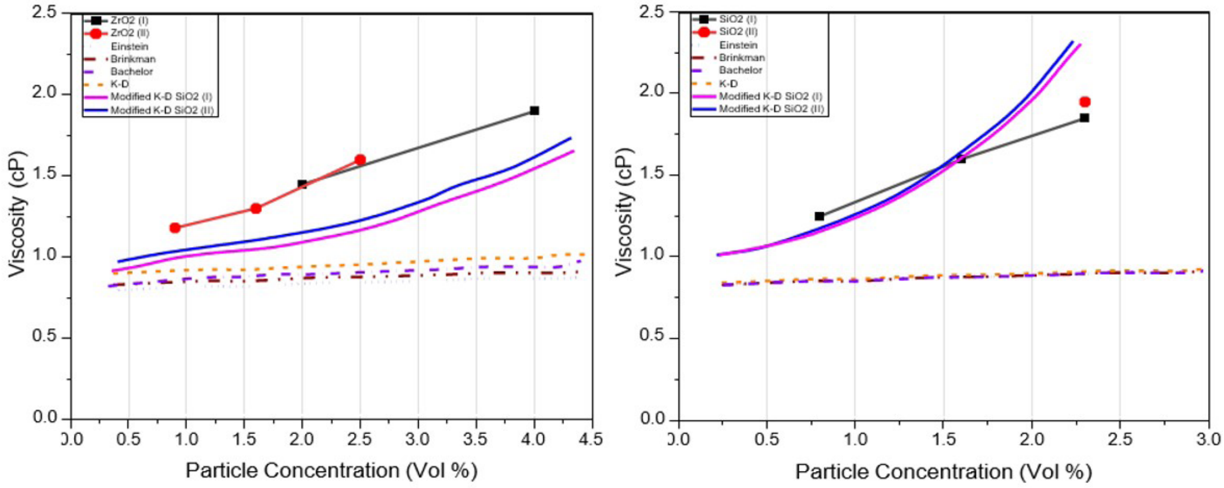


Figure 7: Experiments and correlations comparing ZrO_2-water and SiO_2-water nanofluids at T = 30 °C.

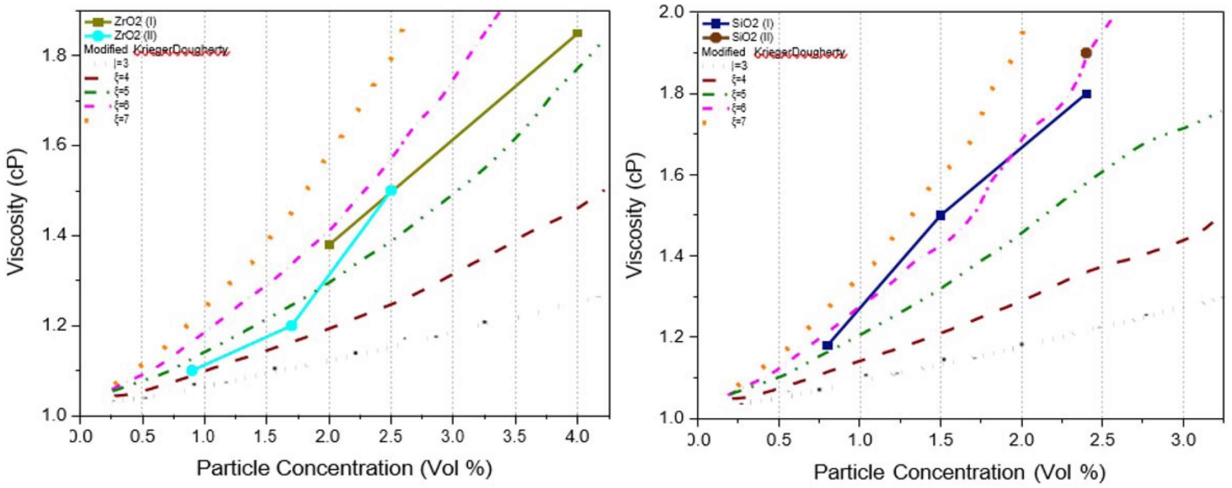


Figure 8: Comparison of the viscosity of ZrO_2-water and SiO_2-water nanofluids using a modified Krieger-Dougherty correlation at T = 30 °C and varying ratios of aggregate to primary nanoparticle size.

changes between 30 and 60 °C. When comparing nanofluids with equal loadings of SiO_2 and ZrO_2 nanoparticles, the SiO_2 nanoparticle-containing fluids exhibited approximately greater viscosity. Adding surfactants to a base fluid to make nanofluids more stable at room temperature will likely make them more viscous. More viscosity was seen in polycarboxylate surfactant-treated SiO_2 nanofluids compared to those treated with trioxadecane acid, demonstrating that the type of surfactant used may affect the degree to which the viscosity increases. Predicting the nanofluid's viscosity is challenging, however certain connections have been presented. Although certain correlations do exist that take into consideration the effect of nanoparticle aggregate size in nanofluid, most of these models tend to either underestimate or exaggerate the experimented results of the current analysis. Experimental measurements of the fraction index and the proportion of the accumulated to initial nanoparticle size allow for an accurate prediction of the nanofluid's viscosity using a modified Krieger-Dougherty correlation, with an error of no more than 6%. The modified Krieger-Dougherty correlation is useful for uniform nanofluids, but it is ineffective for nanofluids with non-uniform shape and varying aggregated-to-primary nanoparticle-size ratios.

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None.

Conflict of Interest

None.

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