



Investigation on stability and viscosity of SiO₂–CH₃OH (methanol) nanofluids[☆]



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ABSTRACT

This work studied the stability and viscosity of methanol based nanofluids with SiO₂ nanoparticles at various volume concentrations and temperatures in order to investigate the enhancement in heat transfer. SiO₂ nanoparticles of 5–15 nm sizes were suspended into methanol base fluid at five different concentrations which were 0.005, 0.01, 0.05, 0.10 and 0.15 vol%. The nanofluid preparation was completed through sonication using an ultrasonic homogenizer. The viscosity of the prepared nanofluids was measured by LVDV III ultra-programmable viscometer at five different temperatures (5, 10, 15, 20 and 25 °C). The measurement deviations were about 3.96% and 2.4% for pure methanol and water, respectively. The results of zeta potential and UV-Vis spectrometer showed that SiO₂–methanol nanofluids were stable. It was found that shear stress and viscosity increased with volume concentration and shear rate and decreased in terms of temperature. These results showed that SiO₂–methanol nanofluids appeared as a non-Newtonian fluid. The maximum viscosity increment was 13.5% at 0.15 vol% base fluid and temperature of 25 °C. Therefore, this study revealed that viscosity enhancement depends on both volume concentration and temperature.

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1. Introduction

Nanofluid is an advanced fluid that contains a suspension of metallic or non-metallic nanoparticles with a typical size of 1–100 nm in a base fluid. The concept of nanofluids was introduced by Choi in 1995 [1]. It is a new category of heat transfer fluids due to its significant enhancements on thermophysical properties and heat transfer. It is necessary to identify the thermophysical properties such as thermal conductivity, viscosity, density, and specific heat of nanofluids for heat transfer applications. The thermophysical and transport properties of nanofluids are influenced by several factors, which affect the heat transfer coefficient and thermal performance of the system. Therefore, researchers are trying to improve the performance of nanofluids in many industrial applications such as power generation, heating or cooling processes, chemical processes and microelectronic manufacturing [2–5].

Viscosity of nanofluids is one of the important parameters in all thermal applications involving fluid flow. Besides, it is important to calculate the pressure drop, accurate pumping power, Prandtl and Reynolds numbers and convective heat-transfer coefficient in forced convection. Viscosity measures interface friction, which is the internal resistance

of fluid layers. According to the literature, nanofluid viscosity depends on many parameters such as particle volume concentration, particle size, temperature, extent of clustering, surfactants and properties of base fluids [6,7]. Several researchers have studied viscosity for different nanoparticles suspended in different nanofluids (i.e. water, engine oil, ethylene glycol, ionic liquid, etc.) and found nanofluid viscosity as a function of nanoparticle volume concentrations [8,9]. They also reported that heat transfer might increase while pressure dropped and volume concentration increased.

Tavman et al. [10] measured the effective viscosity of SiO₂–water using Vibro-Viscometer at different particle concentrations and temperatures. They found that effective viscosity of nanofluids increased with increasing particle concentration and decreased with increase in temperature but their results could not be predicted by Einstein's model [11]. Similar result has also been observed by other researchers [12,13]. Jin et al. [14] measured the viscosity of mineral oil–SiO₂ nanofluids at different temperatures (10–80 °C). They found that the viscosity of nanofluids increased by 21% and 38% in the presence of 0.01% and 0.02% SiO₂ nanoparticles, respectively.

Namburu et al. [15] experimentally investigated the viscosity of SiO₂ nanoparticles of different sizes with 60:40 of ethylene glycol to water within the particle volume concentration range of 0% to 10% and temperature range of –35 to 50 °C. According to their results, nanofluids showed a non-Newtonian behavior at lower temperatures and Newtonian behavior at higher temperatures. The results also showed that viscosity increased with volume concentrations and it was 1.8 times higher than the base fluids at 10 vol%. Anoop et al. [16] investigated the rheological behavior of mineral oil–SiO₂ nanofluids at a shear rate

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Nomenclature

| | |
|-----|--------------------------------------|
| SEM | Scanning electron microscope |
| EDS | Energy-dispersive X-ray spectroscopy |
| TEM | Transmission electron microscope |

Greek symbols

| | |
|-----------|-------------------------------|
| μ | Viscosity (mPa.s) |
| φ | Volume concentration (vol%) |
| ρ | Density (kgm^{-3}) |

Subscripts

| | |
|------|---------------|
| Bf | Base fluid |
| Nf | Nanofluids |
| np | Nanoparticles |

of 100 kPa and a pressure of 42 MPa within a temperature range of 25 °C to 140 °C. Their results showed the non-Newtonian behavior of the nanofluid at elevated temperatures and pressures. Contradictory results were found for SiO₂–alcohol and the results showed that nanofluids displayed a Newtonian behavior at a high shear rate of $2 \times 10^5 \text{ s}^{-1}$ [17].

Water is commonly used as a heat transfer fluid in different heat exchangers in industries, automobiles, solar collectors, heat pipes and HVAC (heating, ventilation and air conditioning) systems. Methanol is useful in gravity aided and pool boiling application at low temperatures [18]. The main objective of this study is to experimentally investigate the stability and viscosity of SiO₂–methanol nanofluids. This experimental study was conducted at five volume concentrations (0.005, 0.01, 0.05, 0.10 and 0.15 vol%) and temperatures (5, 10, 15, 20 and 25 °C). This study promoted the use of methanol based nanofluids in different types of heat pipes such as flat plate, conventional, oscillating, sorption, micro/minature, vapor-dynamic thermosyphon heat pipes and HVAC (heating, ventilation and air conditioning) systems [2,19]. Previous studies [20,21] have only discussed the thermal conductivity of SiO₂–methanol nanofluids. Therefore, to the best of the authors' knowledge, there have not been any studies on the viscosity of SiO₂–methanol nanofluids. Therefore, the present study helped identify the effects of nanoparticle concentration, temperature on viscosity, rheological characteristic and stability of SiO₂–methanol nanofluids.

2. Materials and methods

2.1. Materials and preparation of nanofluids

In this study, SiO₂ nanoparticles were selected due to their chemical stability, availability and large-scale production in the industry. The SiO₂ nanoparticles (purity 99.5%) were procured from Sigma Aldrich, USA. Methanol (CH₃OH) with a purity of 99.8% was procured from R&M Chemical and used as the base fluid due to its low melting point (−97.6 °C) and boiling point (64.7 °C). SiO₂–methanol nanofluids at different volume concentrations were prepared by using a two-step method. The nanoparticles were suspended into the base fluid (methanol) followed by shaking in an incubator. Then, the suspension was mixed using an ultra-sonication homogenizer in order to overcome the strong cohesion force between the particles and encourage even distribution of particles. The sonication process was maintained at constant temperature of 20 °C with the help of refrigerated thermal bath. The experimental condition of this investigation is presented in Table 1.

2.2. Characterization

It is important to understand the behavior of nanoparticles and nanofluids prepared in order to apply nanofluids in practical situations.

Table 1
Experimental conditions.

| Base fluid | Methanol |
|-----------------------------|----------------------------------|
| Nanoparticle | SiO ₂ |
| Nanoparticle type | Spherical |
| Nanoparticle size (nm) | 5–15 |
| Shaking | 15 min at 150 rpm |
| Volume concentration (vol%) | 0.005, 0.01, 0.05, 0.10 and 0.15 |
| Ultra-sonicator | Time [min] 120 |
| | Power [W] 500 |
| | Frequency [kHz] 20 |
| | Pulse [s] 2 |
| | Term [s] 2 |

The shapes of nanoparticle were identified with a scanning electron microscope (SEM) with Phenom ProX. The SEM and energy-dispersive X-ray spectroscopy (EDS) were taken at the acceleration voltage of 15 KV which are shown in Fig. 1 (a) and (b). In the SEM image, dry particles are displayed in micrographs. Highly agglomerated particles in the size range of micrometer were observed under atmospheric condition and most of the nanoparticles were spherical. Smaller particles were found to have larger surface-to-volume ratio, which showed higher agglomeration than that of large particles. Fig. 1 (b) shows the percentage of chemical compounds in SiO₂ nanoparticles with impurities. The weight percentage of silicon (Si) and oxygen (O) was 33.8% and 64.8% while the atomic percentage was 20.1% and 78.2%, respectively with carbon (C).

A LIBRA 120, transmission electron microscope (TEM) manufactured by Zeiss, Germany was used to analyze particle dispersion. TEM was used to check the particle size, shape, and distribution of 0.05 vol% of SiO₂–methanol nanofluids. The TEM image presented that the nanoparticles agglomerated into small aggregates and some particle clusters were formed, as shown in Fig. 2. The nanoparticles might have agglomerated during dry sample preparation, which affected the results of TEM image eventually.

The nanoparticle distribution in suspension was analyzed by Zetasizer ZS from Malvern. The volume distribution of nanoparticles within the 0.05 vol% of SiO₂–methanol nanofluids is presented in Fig. 3. The average SiO₂ nanoparticle size in the solution was around 190 nm which was larger than the original size of 5–15 nm. It might be attributed to very high surface energy of the nanoparticles which leads to agglomeration of the particles. Such agglomeration can be difficult to reverse by ultra-sonication.

2.3. Stability of nanofluids

In this investigation, a UV-Vis spectrophotometer (Lambda 35 model, PerkinElmer) with an absorption range of 190 nm to 1100 nm was used to study the stability of the nanofluid. The inspection range was from 200 nm to 800 nm. The UV-Vis spectrometer works according to the principles of Beer–Lamberts law. The Beer–Lamberts law relates absorption of light to amount of materials in a medium. The lesser the suspended particles in the solution, the lesser the light absorption.

Zetasizer ZS from Malvern was used to measure the size distribution of nanoparticles in nanofluids. This instrument is capable of measuring sizes from nanometer to several microns using dynamic light scattering. The same Zetasizer was also used to measure the zeta potential of the nanofluid at different concentrations of methanol nanofluids using electrophoretic light scattering. Stokes law suggests an equation for calculation of sedimentation velocity of small spherical particles to evaluate the stability of nanoparticles inside a base fluid, as below.

$$v = \frac{r^2}{9\mu} (\rho_{np} - \rho_{bf}) \quad (1)$$

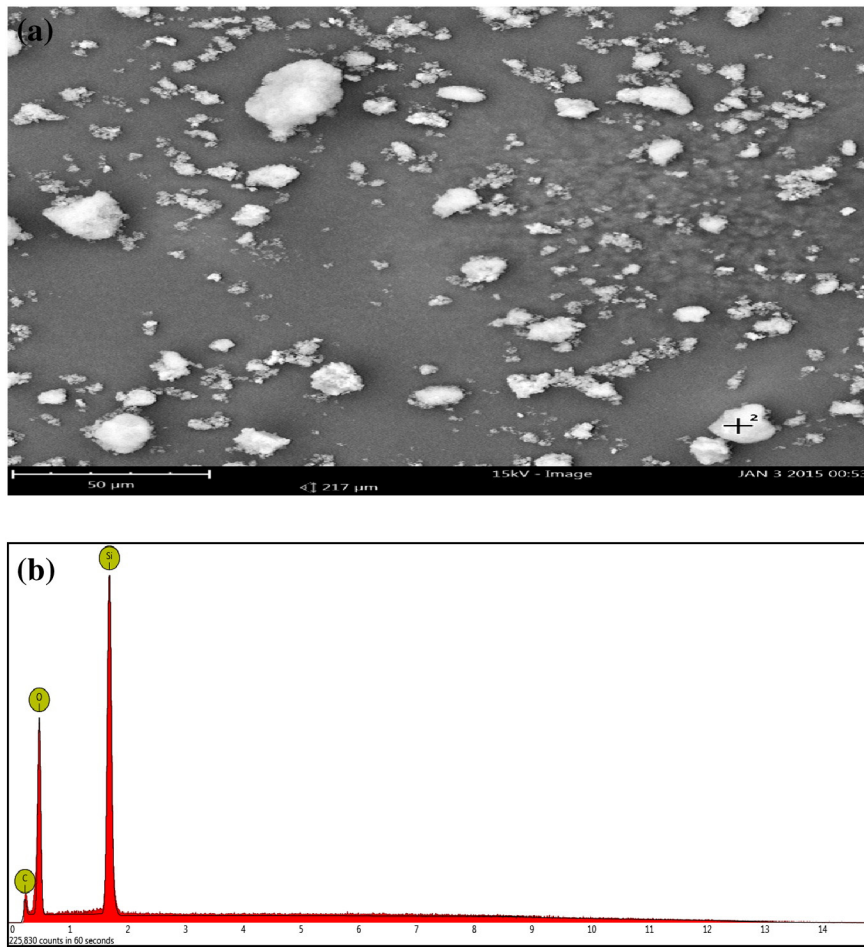


Fig. 1. (a) SEM and (b) EDS images of SiO₂ nanoparticles.

2.4. Measurement of effective viscosity

The most commonly used devices to measure viscosity are the LVDV series viscometers. These devices are suitable to measure low viscosity. A viscometer named LVDV III ultra-programmable was used to measure the viscosity of methanol based nanofluids. A rotating spindle was submerged into the nanofluids and the viscous effect was developed against the spindle due to deflection of a calibrated spring with the help of a low adapter. The device was connected to a computer to store and collect data with the help of Rheocalc 32 software. In this experiment, the

viscosity of 0.005, 0.01, 0.05, 0.1 and 0.15 vol% of SiO₂–methanol was measured at temperatures between 5 and 25 °C by increasing the temperature by 5 °C every time. A refrigerated circulating bath was used to control the system temperature. The measured data were recorded five times and the corresponding averages were plotted. Then, the measured viscosities for 0.0005–0.15 vol% of SiO₂–methanol based nanofluids at 25 °C were compared with the existing values. These models used are as follows:

In 1906, Einstein [11] suggested an equation which infinitely dilutes suspension for spherical particle and low volume concentration as follows:

$$\mu_{nf} = (1 + 2.5\phi)\mu_{bf} \quad (2)$$

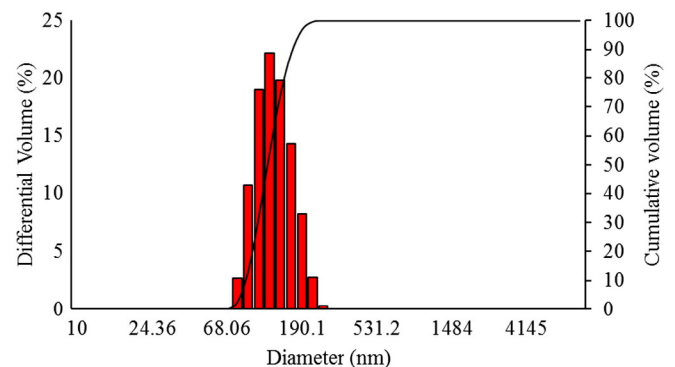


Fig. 3. Nanoparticle size distribution within the 0.05 vol% of SiO₂–methanol nanofluids.

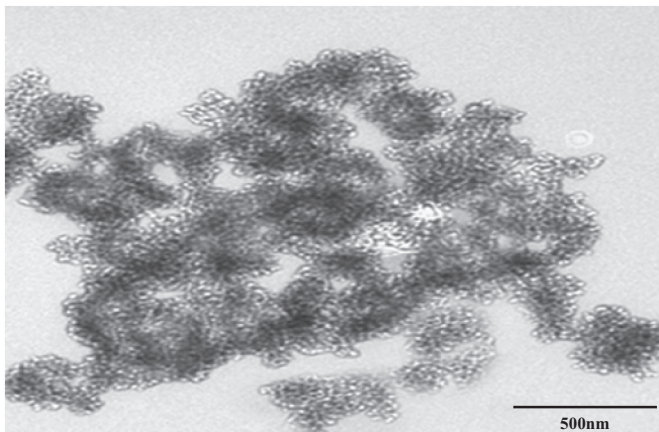


Fig. 2. TEM image of SiO₂–methanol nanofluids.

In 1952, Brinkman [22] modified Einstein's formula to a more generalized form considering Brownian motion, very small particles and spherical nanoparticles, as follows:

$$\mu_{nf} = \frac{1}{(1-\phi)^{2.5}} \mu_{bf} \quad (3)$$

In 1977, Batchelor [23] proposed a formula for viscosity of nanofluids with rigid and spherically shaped particles as follows:

$$\mu_{nf} = (1 + 2.5\phi + 6.2\phi^2) \mu_{bf} \quad (4)$$

Furthermore, Song et al. [24] developed a model with their experimental results for the measurement of SiO₂–water nanofluids which is presented as follows:

$$\mu_{nf} = (1 + 56.5\phi) \mu_{bf} \quad (5)$$

3. Results and discussions

3.1. Stability analysis

The photographs of SiO₂–methanol nanofluids of different volume concentrations left to right (0.005, 0.01, 0.05, 0.10 and 0.15 vol%) were captured right after preparation and 24 h, as shown in Fig. 4. Generally, sedimentation was observed at the bottom of the sample bottles. The photos were inspected by naked eyes and there appeared to be no sedimentation.

The SiO₂–methanol nanofluid's stability was analyzed with zeta potential and the results are shown in Fig. 5. The zeta potential values for 0.005–0.15 vol% SiO₂–methanol nanofluids varied in the range of –30 mV to –40 mV in both samples (right after preparation and after 24 h). The error bar presents the minimum and maximum values of the results. The agglomeration cluster size increased when zeta potential became closer to zero. Generally, a nanofluid is considered stable when its absolute value of zeta potential is above ± 30 mV [25]. The zeta potential results showed that the SiO₂–methanol nanofluids were considered stable.

Fig. 6 shows the spectrum image of SiO₂–methanol at different volume concentrations right after preparation by using ultrasonic agitation. It also showed that the peak absorbance of SiO₂ nanoparticle suspension on methanol appeared in the range of 215 nm to 222 nm. The absorption range of the nanofluid was 0.39 to 1.19. The absorption peak of all volume concentrations varied within a small range and thus the nanofluids were considered stable. A similar absorption peak was observed in one of the previous studies on Al₂O₃–methanol nanofluids

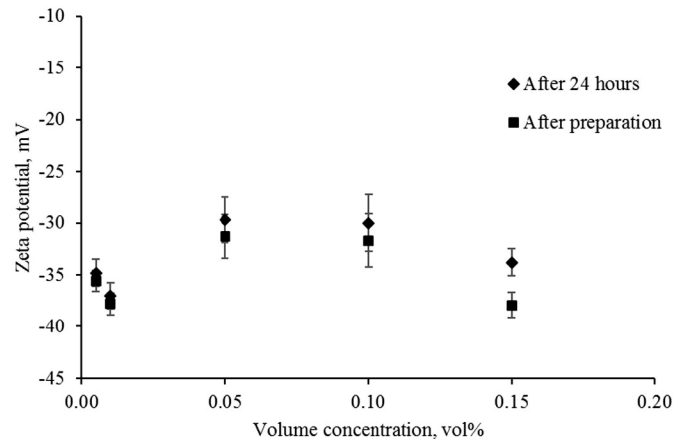


Fig. 5. Zeta potential of SiO₂–methanol nanofluids.

[26]. Furthermore, the absorption strength of 0.005 vol% nanofluid was lower. This was because the 0.005% nanofluid created more 'particle free region' in the base fluid. The absorption strength of other volume concentrations was relatively higher than that of 0.005% nanofluid.

3.2. Viscosity of nanofluids

3.2.1. Validation of instruments

The viscosity of water and pure methanol was measured with a viscometer in the temperature range of 5–25 °C. Each measurement was repeated five times and averaged in order to obtain reliable results. The standard deviation was $\pm 1\%$ for LVDV III ultra-programmable viscometer. The experimental data were then compared with the reference data for water [27] and methanol [28]. Fig. 7 presents the results for three sets of measurements for both water and methanol. The difference between the experimental values and the reference line was 3.96% for methanol and 2.4% for water.

3.2.2. The effect of temperature and volume concentration on viscosity

The viscosity of SiO₂–methanol nanofluids as a function of temperature and volume concentration is illustrated in Fig. 8. The results indicated that the viscosity of SiO₂–methanol nanofluids in every volume concentration was higher than that of pure methanol. As expected, adding SiO₂ nanoparticles and increasing the volume concentrations increased the viscosity of methanol considerably. For example, the viscosity was 0.78 mPa·s and 0.82 mPa·s for 0.005 vol% and 0.15 vol% respectively at 5 °C. Similar results were found at all volume concentrations. It was observed that the viscosity of nanofluids with higher SiO₂

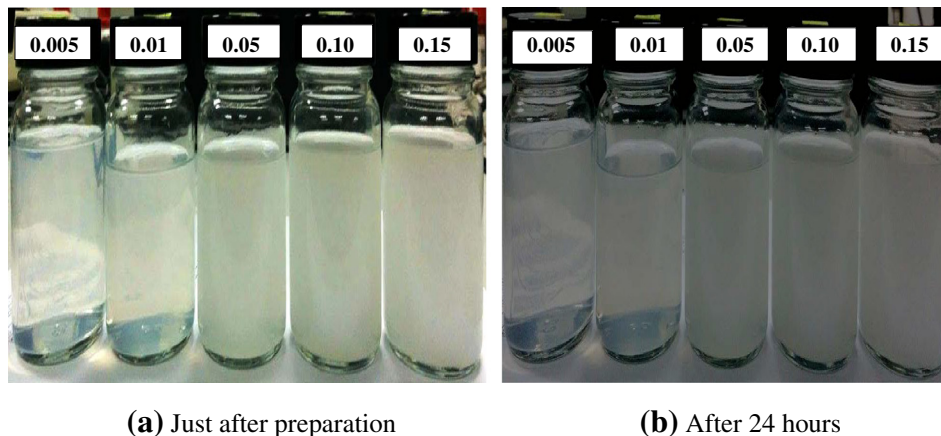


Fig. 4. Photograph of methanol based nanofluids just after preparation and after 24 h preparation.

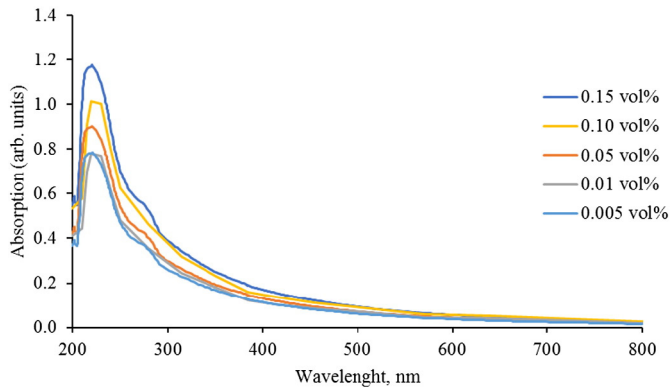


Fig. 6. UV-Vis spectrum of SiO₂-methanol nanofluids.

loading was greater than those with lower solid particle concentrations. This revealed that particle concentration was an influential parameter on the viscosity of SiO₂-methanol nanofluids. This occurred due to stronger internal shear force among nanoparticles. This increased the number of nanoparticles, formed bigger agglomerates and a larger force was required to disperse them [29]. Fig. 8 also shows that viscosity decreased with increased temperature. The 0.005 vol% nanofluids at 25 °C are 20.5% lower than those at 5 °C. Viscosity values of 0.62 mPa.s and 0.78 mPa.s are at 25 °C and 5 °C respectively at 0.005 vol%. Similar results were found for 0.01, 0.05, 0.10 and 0.15 vol% nanofluids respectively. Viscosity increased with concentration while decreased with temperature nonlinearly. Masuda et al. [30] who investigated the concentration and temperature dependence of viscosity of aqueous nanofluids containing SiO₂ also found the same trend. Viscosity was found to decrease with temperature as high temperature reduced the intermolecular and inter-particle adhesion forces of fluids [31].

The relative viscosity of SiO₂-methanol nanofluids in the temperature range of 5–25 °C and volume concentration of 0.005–0.15 vol% is shown in Fig. 9. The average increase for viscosity of SiO₂-methanol nanofluid were 5.1, 6.7, 8.5, 10.2, and 13.5% for particle concentrations of 0.005, 0.01, 0.05, 0.10 and 0.15 vol%, respectively at 25 °C. The relative viscosity of SiO₂-methanol nanofluids was temperature independent within the particle concentration range of 0.005 to 0.15 vol% and temperature range of 5–25 °C. However, the relative viscosity with 0.15 vol% solid particle slightly reduced by 7% in the temperature range of 5–25 °C. The results are in agreement with the result by Niu et al. [32]. They found an increase of around 1.08 times in viscosity for SiO₂-water nanofluids at 0.10 vol% and 25 °C.

3.2.3. Rheological behavior of nanofluids

The shear stress versus the shear rate of SiO₂-methanol nanofluids at different temperatures in 0.05 vol% are shown in Fig. 10. The flow

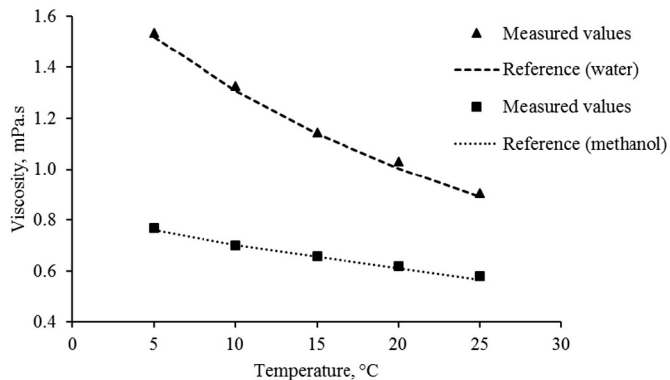


Fig. 7. Viscosity of water and methanol measured by viscometer compared with reference data.

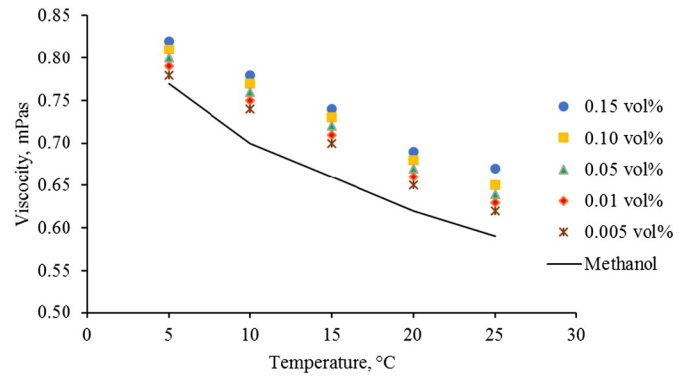


Fig. 8. Viscosity of SiO₂-methanol nanofluids at different temperatures and particle concentrations.

curves in Fig. 10 show that all processing temperatures did not show any hysteresis by increasing and then decreasing the shear rate. This showed that reversible shear rate-shear stress paths overlapped, which is in agreement with the results reported by Alphonse et al. [33]. The same results were also observed for 0.005, 0.01, 0.10 and 0.15 vol%. The nonlinear increment trend of shear stress vs. shear rate exhibited the characteristic of a non-Newtonian fluid. Mostafizur et al. [34] reported a typical non-Newtonian (shear thickening) behavior for Al₂O₃-methanol and TiO₂-methanol nanofluids. Meanwhile, this study showed that shear stress decreased by increasing temperature. This is in agreement with the results of the previous investigations [34].

The viscosity versus shear rate of SiO₂-methanol nanofluids for 0.05 vol% as a function of temperature is illustrated in Fig. 11. The results showed that viscosity increased with shear rate and decreased with temperature. As shown in Fig. 11, a shear thickening or dilatant behavior was observed. The shear thickening behavior could be due to agglomeration of nanoparticles or realignment in the direction of the shearing force, which led to less viscous drag. Previous studies of Al₂O₃-methanol and TiO₂-methanol nanofluids found the similar trend that viscosity increased with shear rate [34].

3.2.4. Comparison of experimental values and theoretical correlations

Fig. 12 shows the comparison between these results and correlations obtained from this study with the results from the studies on SiO₂ nanofluids at 25 °C. The results showed that the experimental value was higher than that of the existing correlations. The experimental values were 13% higher than Einstein's [11] equation and 5% than Song et al.'s [24] equation. These equations were in generalized model and developed for base fluid water. Mahbubul et al. [35] reported that these equations were not appropriate to evaluate the viscosity of all base fluids. Higher viscosity may occur due to agglomeration of nanoparticles. Fig. 12 illustrates that increment of viscosity was lower than

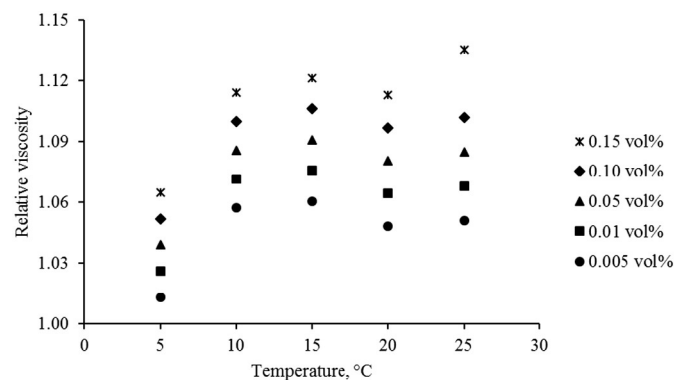


Fig. 9. Relative viscosity of SiO₂-methanol nanofluids at different temperatures and particle concentrations.

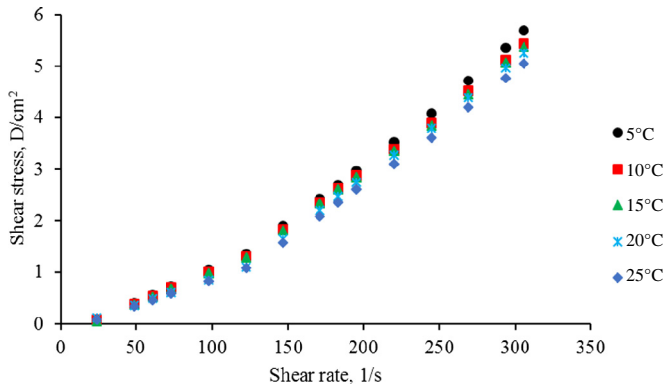


Fig. 10. Shear stress vs shear rate of SiO₂-methanol nanofluids for 0.05 vol% as a function of temperature.

that obtained by Jamshidi et al. [12]. They studied water, ethylene glycol and transformer oil with SiO₂ nanoparticles and their results were 21%, 7% and 2% higher than these experimental results. Fig. 12 also shows that increased viscosity was lower than the previous experiments [34]. Therefore, the viscosity of nanofluids depends on types of nanoparticle, particle size and viscosity of base fluids [36,37].

Therefore, a new correlation was proposed hereby based on the experimental results for measuring the viscosity of methanol based nanofluids:

$$y = 0.2861x^2 + 0.4752x + 1.0562, R^2 = 0.9648$$

The correlation was valid for a particle volume concentration of 0.005–0.15 vol% and temperature of 25 °C. The dotted black line in Fig. 12 shows the comparison between the measured value and value obtained from the proposed correlation. The results showed that the correlation coefficient (R^2) value of the present correlation was about 0.9648 which is close to 1.

4. Conclusions

In this study, the viscosity of SiO₂-methanol nanofluids was investigated at different volume concentrations and temperatures. The experimental results were then compared with the existing correlations. The findings of this study are summarized as follows:

- (a) The results of zeta potential and UV-Vis spectrometer showed that SiO₂-methanol nanofluids were considered stable right after preparation and after 24 h.

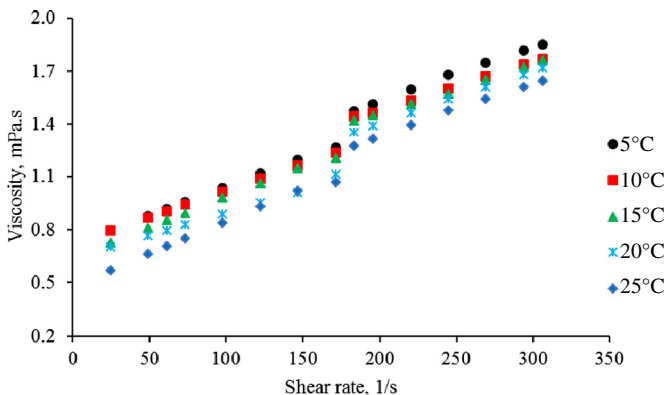


Fig. 11. Viscosity vs shear rate of SiO₂-methanol nanofluids for 0.05 vol% as a function of temperature.

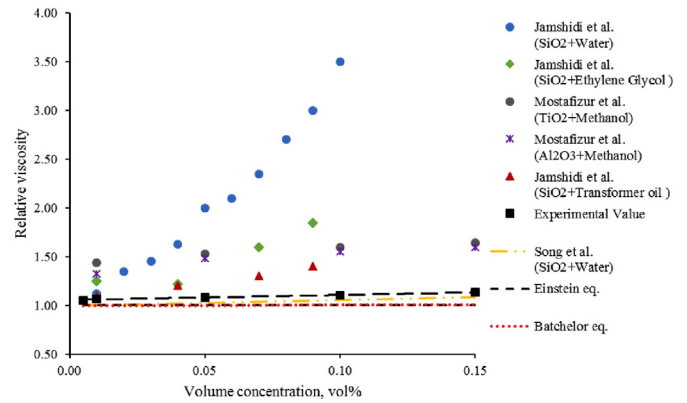


Fig. 12. Comparison between experiment, correlations and results obtained from other researchers of SiO₂ nanofluids.

- (b) Viscosity increased with volume concentrations. However, it decreased with temperatures.
- (c) Rheological behavior of SiO₂-methanol nanofluids was found to be non-Newtonian with shear thickening or dilatant.
- (d) Viscosity increased by about 1.13 times with respect to base fluids at a nanoparticle volume concentration of 0.15 vol% and at temperature 25 °C.
- (e) The existing correlations for predicting the viscosity of SiO₂-methanol nanofluids yielded lower viscosity values than the experimental values but 5% and 13% higher than Song et al.'s [24] and Einstein's equation [11], respectively.

Acknowledgments

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