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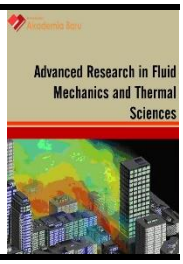

Empirical Viscosity Modeling for SiO_2 and Al_2O_3 Nanofluids using the Response Surface Method

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Faiza Mohamed Nasir[1,2,*], Mohd. Zulkifly Abdullah[2], Mior Firdaus Mior Abdul Majid[1]

[1] Mechanical Section, Malaysian Spanish Institute, Universiti Kuala Lumpur, Kulim Hi-Tech Park, 09000 Kedah, Malaysia
[2] School of Mechanical Engineering, Kampus Kejuruteraan, Universiti Sains Malaysia, Nibong Tebal, 14300 Pulau Pinang, Malaysia

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ABSTRACT

The ability of nanofluids, an engineered fluid, to effectively remove heat has been proven to exceed that of a conventional fluid. However, dynamic viscosity may put a limitation on this ability. This paper presents the results of the experimental measurement of the dynamic viscosity for water-based nanofluids and the development of empirical viscosity models using the response surface method (RSM). The nanofluids that are being considered in this work are silicon dioxide (SiO_2)-water and aluminum oxide (Al_2O_3)-water at a concentration of 0.01, 0.055 and 0.1 vol.%. Experiments were designed and analyzed according to the face-centered central composite design (CCD) in the RSM. ANOVA was used to evaluate the significance of the independent factors, which are the nanoparticle concentration and temperature. Empirical models to predict the dynamic viscosity of both nanofluids at a specific temperature and volume concentration were developed and validated. Excellent fits of the models were demonstrated by their high coefficient of determination, R^2. Results indicate that dynamic viscosity increases with nanoparticle concentration and decreases with temperature. It is also observed that the addition of less or equal than 0.1 vol.% of SiO_2 in water would not significantly change the viscosity.

1. Introduction

Nanofluids are defined as engineered materials that comprise of nanoparticles, base fluid(s), and optional stabilizers (surfactants/dispersants). Many studies have proved the capability of these innovative materials in enhancing heat transfer performance. Their superb heat dissipation ability is said to be attributed to their improved thermal conductivity. Nanofluids have higher thermal conductivities than that of their base fluids as demonstrated by these investigations [1–5]. In a study by Aberoumand and Jafarimoghaddam [5], it was found that by adding 1% weight concentration of CuO nanoparticles to engine oil, thermal conductivity can be enhanced by 49%. Dispersing 5%

* *Corresponding author.*
E-mail address: faiza@unikl.edu.my

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volumetric fraction of TiO_2 nanoparticles in deionized water increased the thermal conductivity in the range of 29.70% - 32.80% depending on the size of the nanoparticle [6].

Besides enhancing the thermal conductivity, nanofluids have been shown to have a superior convective heat transfer coefficient. Published experimental works reported enhancement of convective heat transfer coefficient and Nusselt number when nanofluids were used as the working fluid [7-9,10-12]. Viscosity plays an essential role in convective heat transfer performance as they affect the rheological behavior and the fluid dynamic of the nanofluids.

Experimental studies showed that the viscosity of a base fluid increases when nanoparticles are dispersed in the fluid. In a study of Esfe [13], it has been shown that by adding 1.5 vol.% CuO nanoparticle concentration in ethylene-glycol would increase its viscosity by 82.46% at a fluid temperature of 50°C. Nguyen *et al.,* [14] found that Al_2O_3-water nanofluid has dynamic viscosity that is 5.3 higher than that for water at a volumetric concentration of 12%. It is theorized that the enhancement in viscosity is due to the increase in overall drag effect of the medium, dominance of forces (such as electroviscous, solvation, hydrophilic, hydrophobic) caused by the reduction in interparticle distance and particle agglomeration [15].

There are many experimental studies dedicated to investigating the dynamic viscosity of nanofluids. These studies reported findings for nanofluids containing nanoparticles such as Al_2O_3, TiO_2, CuO and carbon nanotubes (CNT), that are dispersed in water, ethylene glycol (EG) or a mixture of water and EG at specified ratio [13,16-23]. The consensus of the researchers who investigated the rheological behavior of the nanofluids is that nanofluid viscosity increases when the concentration of nanoparticle increases and they are decreasing as the temperature increases.

Several of the commonly used classical models for the viscosity of nanofluids are presented in Table 1. The majority of these models relate the viscosity of the nanofluid only to the volume concentration of the nanoparticles, and they have been proven to be inadequate in describing the nanofluid viscosity with the changes in nanoparticle concentrations and temperature [24,25]. Efforts were made by researchers to develop standard models that can accurately predict nanofluids' viscosities by fitting the models to either their experimental data or other published data. However, this poses a challenge as there are inconsistencies in literature results [26].

Hence it is the objective of this study to develop empirical models to estimate nanofluids' viscosity that are dependent on the temperature as well as the concentration of the nanoparticle. It is also the aim of this work to develop models that are simple to use, without the need to acquire the base fluid's properties.

Table 1
Commonly used classical models to determine the dynamic viscosity of nanofluids

Model's name	Equation
Einstein model	$\mu_{nf} = \mu_{bf}(1 + 2.5\varphi)$
Brinkman model	$\mu_{nf} = \mu_{bf}(1 - \varphi)^{2.5}$
Batchelor model	$\mu_{nf} = \mu_{bf}(1 + 2.5\varphi + 6.5\varphi^2)$
Wang *et al.,* [31] model	$\mu_{nf} = \mu_{bf}(1 + 7.3\varphi + 123\varphi^2)$
Pak and Cho [32] model	$\mu_{nf} = \mu_{bf}(1 + 39.11\varphi + 533.9\varphi^2)$

2. Experimental Setup

2.1 Nanofluid Preparation

The nanofluids were prepared by diluting commercial water-based dispersion purchased from US Research Nanomaterials, Inc. The purchased nanofluids dispersions are SiO_2-water and Al_2O_3-water

with nanoparticle mass concentration of 25% and 20%, respectively. According to the manufacturer's specifications, the sizes of the nanoparticles are 5-35 nm for SiO_2 and 10 nm for Al_2O_3. The mass concentration was converted to volume concentration φ using the following equation.

$$\varphi = \frac{\omega \rho_{bf}}{\left(1-\frac{\omega}{100}\right)\rho_{np}+\frac{\omega}{100}\rho_{bf}} \quad (1)$$

where ω is the weight concentration of the original dispersion, ρ_{bf} and ρ_{np} is the density for the base fluid and the nanoparticle, respectively. Dilution of the original nanoparticle dispersion to the desired volumetric concentration was conducted by adding a certain amount of distilled water, ΔV, that is calculated from Eq. (2).

$$\Delta V = V_2 - V_1 = V_1\left(\frac{\varphi_1}{\phi_2} - 1\right) \quad (2)$$

where V is the fluid volume and φ is the volume concentration of the nanoparticle (subscript 1 is for original dispersion, and subscript 2 is for the required dispersion).

In this study, three different volumetric concentrations of the nanofluids were prepared; 0.01 vol.%, 0.055 vol.%, and 0.1 vol.%. There is no stabilizing agent added to the SiO_2 nanofluid. However, difficulty in maintaining stability in Al_2O_3 nanofluid prompted the addition of 3 vol.% of polyvinylpyrrolidone (PVP) to the solution. The diluted dispersions were then undergoing homogenization using probe sonicator for two hours with an ultrasonic pulse at 60 kHz to break down agglomeration of the nanoparticles.

2.2 Viscosity Measurement

The viscosity of the nanofluids was measured using a ball-bearing rotational rheometer (RheolabQC) manufactured by Anton Paar. The rheometer has a viscosity measuring range of 1 – 10^9 mPa.s, a temperature range of minus 20 to 180°C, shear stress range of 0.5 – 30,000 Pa, and shear rate range of 10^{-2} to 4000 1/s. The viscosity is measured at different temperatures as dictated by the design of the experiments (30°C, 40°C, and 50°C). The rheometer is fitted with a Peltier temperature system that ensures a quick heating rate of the sample. The system employs air cooling of the sample at a rate of 4 K/min.

2.3 Design of Experiments

In order to develop the empirical models to predict the nanofluids' viscosities, a face-centered central composite design (CCD) of the RSM was used. The design was used to plan the experiments with the nanoparticle volume concentration, and operating temperature is selected as the two independent variables or factors, which resulted in eight numbers of experimental runs to be conducted. The factors and their levels are outlined in Table 2.

The experimental results were then analyzed using the Design-Expert software. In order to evaluate the significance of the developed models, analysis of variance (ANOVA) were conducted for both nanofluids. The models' fit qualities and statistical significance were assessed by the coefficient of determination R^2 and the F-test, respectively.

Table 2
Surface design factor levels

Factor	Name	Low	High
A	Temperature (°C)	30	50
B	Volume concentration (%)	0.01	0.10

3. Results and Discussion

The results of the experimental measurement of the viscosities for SiO_2 and Al_2O_3 nanofluids are illustrated in Figure 1 to Figure 3. It is observed from these figures that adding a small amount of SiO_2 nanoparticles in water would slightly decrease the viscosity. At 30°C, the viscosity decreases by 3% as the nanoparticle concentration increases from 0.01% to 0.055 and 0.1%. At a higher fluid temperature of 50°C, decrement in viscosity in the range of 5-6% was observed.

Published works established that the addition of nanoparticles into a conventional fluid would increase the dynamic viscosity of the fluid. However, in this study, contradicting behaviour is noted, albeit at a much smaller extent. This behaviour may be due to the different methods in preparing the nanofluids and their homogenization procedures. At a glance, it can be seen that the viscosity of SiO_2-water nanofluid is more or less similar to that for water. Hence it can be deduced that adding less or equal than 0.1 vol.% SiO_2 nanoparticles to water would insignificantly alter its viscosity.

Significant enhancement in viscosity is observed when Al_2O_3 was added to water. For example, the addition of 0.1 vol.% Al_2O_3 nanoparticles to the water would increase the viscosity by 51.5% at a fluid temperature of 30°C. Similar findings were observed at an operating temperature of 50°C. In a study by Pastoriza-Gallego *et al.,* [27], they observed that by adding 0.029 vol.% of Al_2O_3 nanoparticle into the water, viscosity increases by almost 100%. Lee *et al.,* [28] reported 74% enhancement in the viscosity of the nanofluids when 0.1 vol.% Al_2O_3 nanoparticles were dispersed and homogenized in water.

When a certain number of nanoparticles is added to the base fluid, the distance between the particles in the fluid reduces, hence increasing the overall drag effect of the particles. The reduction of this distance would also cause the inter-particle forces to be very significant. Consequently, these effects make it difficult for the fluid to move.

The relationship of nanofluids' viscosity with temperature is shown in Figure 3, where an inverse relation is observed. As the temperature increases, the dynamic viscosity of both SiO_2 and Al_2O_3 nanofluids decreases. Similar findings were also observed by [28–30]. As the temperature increases from 30°C to 50°C, the viscosity of SiO_2-water and Al_2O_3-water nanofluids decrease by at least 33.73% and 32.3%, respectively. Higher temperatures would cause an intense movement of particles, which then weakened the molecular forces. Therefore, the fluid will be easier to flow, and the viscosity will be reduced.

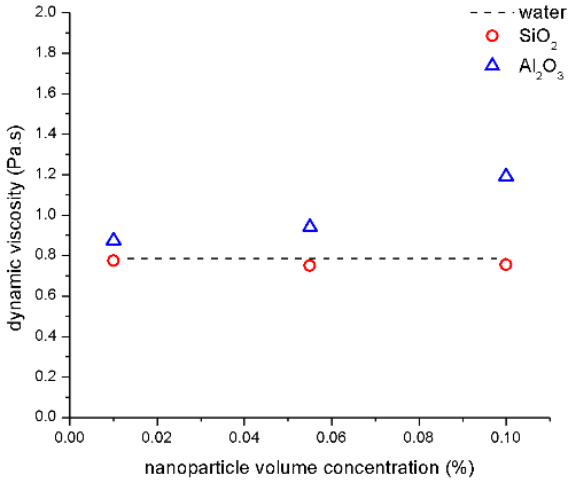

Fig. 1. Dynamic viscosity of SiO_2 and Al_2O_3 at a temperature of 30°C

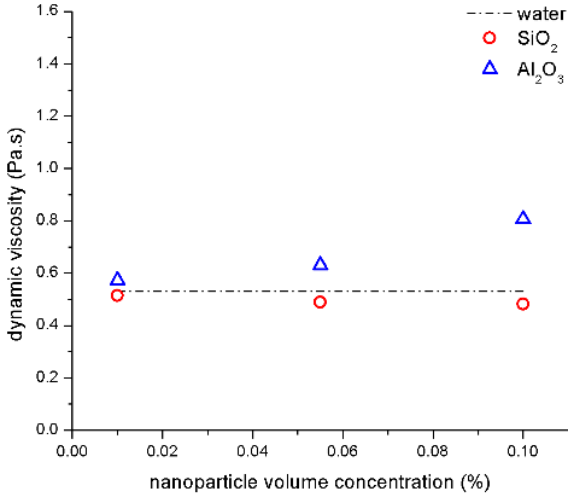

Fig. 2. Dynamic viscosity of SiO_2 and Al_2O_3 at a temperature of 50°C

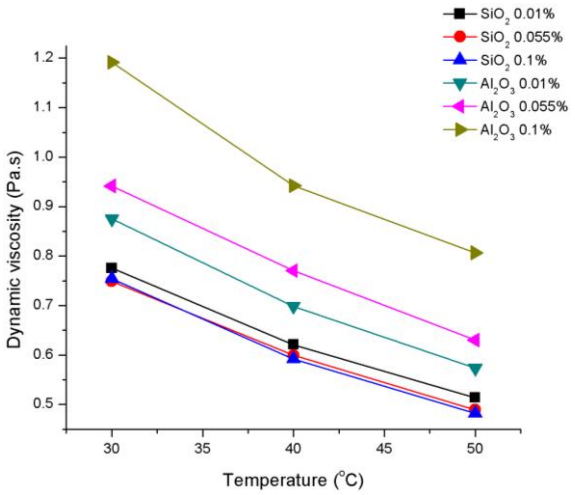

Fig. 3. Dynamic viscosity of SiO_2 and Al_2O_3 nanofluids at different fluid temperatures

3.1 Analysis of Variance (ANOVA)

3.1.1 Viscosity of SiO_2

The result indicates that viscosity of SiO_2 nanofluid can be adequately described by a quadratic relationship, as shown by the resulted empirical model that is

$$\mu_{nf} = 0.001544 - 0.000033 \cdot T - 0.000736 \cdot \phi - 5.94 \times 10^{-6} \cdot T \cdot \phi + 2.49 \times 10^{-7} \cdot T^2 + 0.006085 \cdot \phi^2 \quad (3)$$

where T is the temperature (°C) and ϕ is the nanoparticle volume concentration (%). The ANOVA analysis of the model is presented in Table 3. The model is considered to be significant in predicting the viscosity of the SiO_2 nanofluid as it has high F-value and p-value that is less than 0.05.

From the table, it was found that temperature and the nanoparticle volume concentration affect the viscosity of SiO_2 significantly. However, there is no significant effect of their interaction with the viscosity. The fit quality of the model is excellent as it has a high value of adjusted R^2 of 0.9994. The small difference between the adjusted and the predicted R^2 proved that the model is capable of predicting SiO_2 viscosity accurately without overfitting. Adequate precision of 109.8457, which is higher than 4, ensures that the model can be used to navigate the design space.

Table 3
ANOVA output for the quadratic model of SiO_2 viscosity

Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	1.070×10^{-7}	5	2.139×10^{-8}	2271.84	0.0004
A-temperature	1.050×10^{-7}	1	1.050×10^{-7}	11146.31	< 0.0001
B-volume concentration	1.126×10^{-9}	1	1.126×10^{-9}	119.53	0.0083
A x B	2.857×10^{-11}	1	2.857×10^{-11}	3.03	0.2237
A^2	8.265×10^{-10}	1	8.265×10^{-10}	87.77	0.0112
B^2	2.025×10^{-10}	1	2.025×10^{-10}	21.50	0.0435
Residual	1.883×10^{-11}	2	9.417×10^{-12}		
Cor Total	1.070×10^{-7}	7			
Adjusted R^2	0.9994				
Predicted R^2	0.9968				
Adeq. Precision	109.8457				

3.1.2 Viscosity of Al_2O_3

The RSM analysis showed that the relationship of the viscosity of Al_2O_3 nanofluid with temperature and volume concentration of the nanoparticle could be described by a linear model. Eq. (4) is the resulted empirical model to predict the viscosity of Al_2O_3-water nanofluids.

$$\mu_{nf} = 0.001336 - 0.000017 \cdot T + 0.002940 \cdot \phi \quad (4)$$

It is demonstrated in Table 4 that this model is significant in predicting the viscosity for Al_2O_3 nanofluid, as implied by its high F-value and small p-value of less than 0.05. The importance of the effects of the temperature and the nanofluid's concentration on the viscosity is highlighted by the small numbers of p-values. It is also observed that there is no effect of the interaction of these factors on the viscosity. The model has a high value of adjusted R^2, and its predicted coefficient of determination varies by less than 0.2 from the adjusted value. These indicate that the linear model is capable of predicting the viscosity of Al_2O_3 nanofluid accurately.

Table 4
ANOVA output for the linear model of Al_2O_3 viscosity

Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	2.709×10^{-7}	2	1.354×10^{-7}	65.30	0.0003
A-temperature	1.659×10^{-7}	1	1.659×10^{-7}	79.97	0.0003
B-volume concentration	1.050×10^{-7}	1	1.050×10^{-7}	50.64	0.0008
Residual	1.037×10^{-8}	5	2.074×10^{-7}		
Cor Total	2.812×10^{-7}	7			
Adjusted R^2	0.9484				
redicted R^2	0.8996				
Adeq. Precision	21.4110				

3.2 Response Surface

Figure 4 and Figure 5 show the contour plots of the viscosity for SiO_2 and Al_2O_3 nanofluid, respectively. The plots illustrate the dependence of the dynamic viscosity on the operating temperature and nanoparticle volume concentration. The dynamic viscosity of the SiO_2 nanofluids will be the smallest when the operating temperature and the nanoparticle concentration is at its highest.

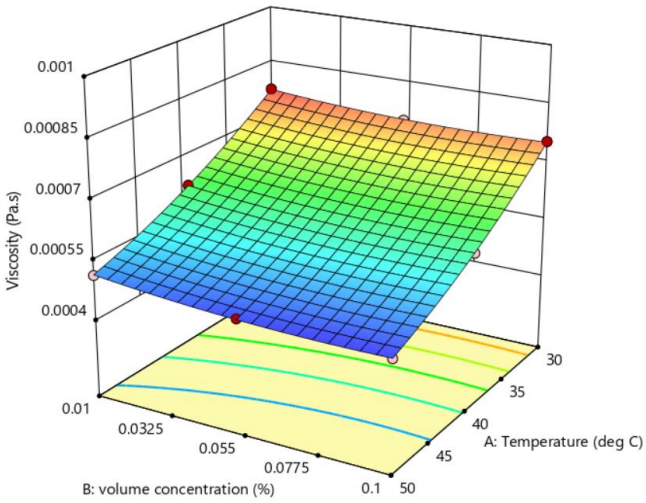

Fig. 4. Contour plot of SiO_2 viscosity

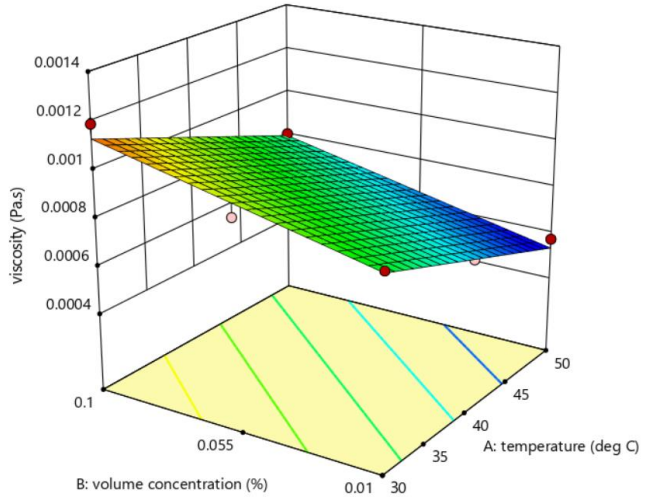

Fig. 5. Contour plot of Al_2O_3 viscosity

For Al_2O_3 nanofluids, the dynamic viscosity of the nanofluids is minimized when the operating temperature is at its highest, and the nanoparticle concentration is at its lowest.

3.3 Confirmation of the Models

Further validation and confirmation of the accuracy of the developed empirical models were performed by conducting additional experimental measurements. The viscosities of the nanofluids were measured at additional two different temperatures, 35°C and 45°C. The results are shown in Table 5.

Table 5
Actual and predicted values for SiO_2 and Al_2O_3 nanofluids' viscosities

Temp (°C)	Vol. concentration (%)	SiO_2-water viscosity (mPa·s)			Al_2O_3-water viscosity (mPa·s)		
		Actual	Predicted	Error	Actual	Predicted	Error
35	0.1	0.661	0.660	0.033%	1.056	1.035	1.970%
45	0.1	0.527	0.524	0.680%	0.874	0.865	1.067%

The difference between the actual (measured) viscosities and the predicted viscosities by the empirical models is less than 2%, thus confirming the reliability and accuracy of the developed models.

4. Conclusions

This paper reports the experimental measurement of dynamic viscosity of two water-based nanofluids; SiO_2 and Al_2O_3. The experiments were designed using a face-centered central composite design. Results indicate that the viscosity decreases when the fluid temperature increases. It is also observed that SiO_2 nanoparticle concentration did not significantly change the viscosity. However, an increase in nanoparticle concentration caused a pronounced enhancement in the viscosity of Al_2O_3. From the ANOVA results, it is noted that the interaction of the concentration and the temperature plays an essential role in the viscosity of SiO_2-water nanofluids, but not for the Al_2O_3 nanofluids. The developed empirical models have been validated and are confirmed to be able to predict the nanofluids' viscosity with less than 2% error. The models are suitable to be used for SiO_2 and Al_2O_3water-based nanofluids, at a concentration of less than 0.1 vol.% and temperature of less or equal to 50°C. The models are simple to be used as it does not require the knowledge of the base fluid properties.

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